

Assessment of NORM in fruits and vegetables from local markets of Hartbeespoort, Mahikeng and Pretoria

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DECLARATION

I, Veronica Kgabisang Gouws, hereby declare that this dissertation is a presentation of my original research work for the degree of MSc in Applied Radiation Science and Technology and has not been submitted to this institution or any other South African institution of higher learning. Every contribution of others involved in this dissertation has been acknowledged. All sources used have been referenced. The work was done under the supervision of Prof. M. Mathuthu of the Centre for Applied Radiation Science and Technology at the North-West University, South Africa, Dr R.D Mavunda of South African Nuclear Energy Corporation SOC LTD, South Africa, and Dr. M. Madhuku of iThemba Laboratory for Accelerator-based Sciences (iThemba LABS), South Africa.

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ABSTRACT

This study's aim was to determine the gamma activity concentration of NORM of (^{238}U , ^{234}U , ^{235}U and ^{232}Th), the gross alpha and gross beta activity concentration, estimation of absorbed dose rate, annual effective dose equivalent, internal (H_{int}) and external hazard indexes (H_{ext}), annual ingestion dose and the excess lifetime cancer risk (ELCR) in fruits and vegetables collected from Hartbeespoort, Mahikeng, and Pretoria markets. The measured results will help the regulators to formulate guidelines and regulations to ensure that fruits and vegetables were safe for human consumption. For this purpose, the agricultural sample types: 4 fruits and 7 vegetables were collected. The samples were prepared and analysed at the South African Nuclear Energy Corporation (Necsa) RadioAnalysis Laboratory using different techniques. Gross alpha and gross beta activity concentration measurements were performed using the Oxford series 5 proportional gas-flow counter in fruit and vegetables. The neutron activation analysis (NNA) was used to identify naturally occurring radioactive materials (NORM) in fruits and vegetables using a combination of high-resolution gamma-ray spectrometry and a high purity germanium (HPGe) detector.

The gross alpha analysed in fruits from Hartbeespoort, Mahikeng and Pretoria showed a significant difference ($p < 0.05$). The gross beta analysis in fruit with showed there is no significant difference ($p > 0.05$). The gross alpha and beta in vegetables showed no significant difference from Mahikeng, Pretoria and Hartbeespoort ($p > 0.05$). The activity concentrations of ^{238}U , ^{234}U and ^{232}Th in fruits and vegetables were lower than the world values of 35 Bq.kg^{-1} and 30 Bq.kg^{-1} (UNSCEAR, 2000), respectively. The activity concentration of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria showed there is significant difference ($p < 0.05$). The current study showed lower annual ingestion doses of ^{238}U and ^{232}Th which are lower than the world annual ingestion dose of $290 \mu\text{Sv.y}^{-1}$ (UNSCEAR, 2000). The annual ingestion dose of ^{238}U and ^{232}Th showed no significant difference ($p > 0.05$) in fruits. The annual ingestion dose of ^{238}U showed no significant difference ($p > 0.05$) in vegetables and ^{232}Th showed a significant difference in vegetables ($p < 0.05$).

The AEDE results from the current study for both fruits and vegetables were lower than the world safety limit of 0.48 mSv.y^{-1} (UNSCEAR, 2000). The absorbed dose rates were lower than the world safety limit of 59 nGy.h^{-1} (ICRP, 1993). The excess lifetime cancer risk in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria were lower than the world

safety limit of $2.9 \times 10^{-4} \text{ mSv.y}^{-1}$ (UNSCEAR, 2000) and showed no significant difference ($p > 0.05$) in fruits and a significant difference ($p < 0.05$) in vegetables excess lifetime cancer risk from the results obtained in Hartbeespoort, Mahikeng and Pretoria. Therefore, it can be concluded that consumption of fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria markets may not pose health threats to human beings.

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LIST OF ABBREVIATIONS

HPGe	Hyper-Pure Germanium Detector
NORM	Naturally Occurring Radionuclide Materials
LC	Critical Level
IAEA	International Atomic Energy Agency
UNSCEAR	The United Nations Scientific Committee on the Effects of Atomic Radiation
NAA	Neutron Activation Analysis
PGNAA	Prompt Gamma Neutron Activation Analysis
DGNAA	Delayed Gamma Neutron Activation Analysis
INAA	Instrumental Neutron Activation Analysis
RNAA	Radiochemical Neutron Activation Analysis
ICRP	International Commission for Radiation Protection
NECSA	South African Nuclear Energy Corporation
MDA	Minimum Detectable Activity
RINGA	Routine Instrumental Neutron Gamma Analysis
NCRP	National Council on Radiation Protection
SANAS	South African National Accreditation Standards
ELCR	Excess Lifetime Cancer Risk
WHO	World Health Organization

CHAPTER ONE: INTRODUCTION AND PROBLEM STATEMENT

1.1. Natural background radiation

Humans are continuously exposed to Naturally Occurring Radioactive Materials (NORM) through the air, food, water, soil and our bodies (Alao, 2011). Most of NORM form natural background radiation in the environment. NORM are natural source of radioactivity and can be terrestrial or cosmic origin (Canbazoglu and Dogru, 2013). Since the formation of the earth, primordial radionuclides were formed and cosmogenic radionuclides were formed by cosmic rays interacting with elements in the atmosphere. The radioactive nuclei interactions of cosmic ray particles in the atmosphere are ^3H , ^7Be , ^{14}C and ^{22}Na . The health hazards of primordial radionuclides together with some of their radioactive decay products are of great concern in radiation protection (Papadopoulos et al., 2013).

Radiation emitted from naturally occurring radionuclide materials is called terrestrial background radiation. The level of concentrations of naturally occurring radionuclides and their associated external exposure in the environment are widely different because of the different background levels, and climate change (Keser et al., 2011). Materials in which NORM can be found at different concentrations are rock, soil, underground water, and air in certain locations of the earth. The naturally occurring radionuclides present in the environment come from the decay of thorium ^{232}Th and uranium ^{238}U , with daughter and granddaughter decays together with primordial potassium ^{40}K which are present in the environment (Bajoga et al., 2015). Some studies have described various aspects related to NORM, such as measuring techniques, exposure characteristics and regulation policies (Alharbi, 2016).

Primordial radionuclides are also called terrestrial radionuclides which are found in nature and radionuclide isotopes have long half-lives primarily (Bayrak et al., 2018). Many countries such as USA, Canada, Australia, Switzerland, Slovakia, Slovenia, Czech Republic, UK have monitored the natural distribution of radioactivity (Xhixha, 2012). The age of primordial radionuclides (^{40}K , ^{238}U , ^{235}U , ^{232}Th) is comparable to the earth age and the products of their radioactive decay are the reason for all studies in the context of radiation protection as a result of their long half-lives and health hazards (Alharbi, 2016).

1.2. Radionuclides of Anthropogenic Sources

TENORM (Technologically enhanced NORM) are found as by-products of industrial activities, mining and milling of metalliferous and non-metalliferous ores, extraction of radioisotopes and treatment of water (IAEA, 2003). In the last decades, this industrial activity increased the production of NORM and long-term radiological hazards. As a result of nuclear weapon tests, effluents discharge from nuclear facilities, and reactor accidents, anthropogenic radionuclides may be released into the environment (Bajoga et al., 2015). These man-made materials produced from the discharged sources can be retained in environmental materials as well as on the ground soil.

The production of waste from the results of these activities may alter the level of radioactivity of the soil that is used for agricultural purposes, the levels of radioactivity may increase in food grown on such soils that are contaminated by tailings from industrial mining and milling operations (Aswood et al., 2017).

1.3. Pathway exposure to NORM and other elements

Heavy metals have always been present in the environment since the formation of the earth (Asaduzzaman et al., 2015). Heavy metals are also contaminants of the food supply due to long biological half-lives and contribute to environmental pollution (Görür et al., 2012). They can affect living organisms (plants and humans) only to a small degree (Lubis et al., 2019). The environmental heavy metals found in water systems can be due to agricultural activities which is attributed to man-made sources (Amin, 2017).

The terrestrial radionuclides total exposure from ingestion has been found to be 0.29 mSv according to (UNSCEAR, 2000) report. The exposure of 0.17 mSv was found from radionuclide ^{40}K and that from the series of thorium and uranium was found to be 0.12 mSv of which 0.01 mSv came from inhalation of terrestrial radionuclides (Görür et al., 2012). The food safety increasing demand has increased research regarding the risk associated with consumption of foodstuffs contaminated by pesticides, radionuclides, heavy metals and toxins (Lubis et al., 2019). The annual average exposure in the world to natural background radiation is 2.4 mSv (UNSCEAR, 2008). Indoor rates are larger than outdoor rates (UNSCEAR, 2000).

1.4. Source of exposure of humans and the environment to natural and artificial radionuclides

Radioactive contamination is any increase in the background natural radiation from human activities by using natural occurring or artificially produced radioactive substances. It is as a result of the loss of control during the production of radioisotopes which could be occasional or accidental (Ogundare and Adekoya, 2015). Human activities in mining and milling of mineral ores, processing and enrichment, nuclear fuel fabrications, and handling of the fuel cycle tail end products (Nieder et al., 2018), release natural radionuclides into the environment. The radioactive materials are released into the environment as a result of these mining activities which enhance effective dose to the population through exposure to internal or external gamma radiation through inhalation and ingestion of radionuclides (Banzi et al., 2017). Anthropogenic radionuclides such as ^{137}Cs found in the atmosphere due to nuclear weapon tests and radioactive accidents has a half-life of 30.17 years (Kam et al., 2016).

During the period of plant growth, fertilizers are added to increase the quality of food crops depending on the soil type, the plant type, the parts of plants used and the certain growth periods of the crop (Van et al., 2018). There are two types of fertilizers which include manure fertilizers which are from plants and animals products, and chemical fertilizers which are from the industrial products (Liu et al., 2017). Radionuclides which exists in fertilizers are from series of uranium (^{235}U and ^{238}U), series of thorium (^{232}Th), and potassium (^{40}K) (Hegedűs et al., 2017).

Fertilizers can be broken into N (nitrogen), P (phosphor in P_2O_5), K (potassium in K_2O), and NPK (the compound of N, P, and K) (Liu et al., 2017). As a result of production technology, many elements have been found in chemical fertilizers. Supplementary materials added during process of production contains calcium (Ca), chlorine (Cl), boron (B), zinc (Zn) and heavy metals which are nutrient elements, therefore the activity concentrations of radionuclides depend on these three parts, the fertilizers source (phosphate mine) and production technology (supplementary materials) (Van et al., 2018). Phosphate rock is a raw material for the production of all phosphate products (Aswood et al., 2013) and it contains some quantity of natural radionuclides (El-Bahi et al., 2017).

By adding fertilizers in the soil, the activity concentration of radionuclides increases during cultivation, leading to radiological hazards due to intake of foodstuffs (Alharbi and El-Taher, 2013). As the water is added during cultivation, fertilizers leach to the soil and there is absorption of large amount of fertilizers through the soil-plants root transfer system in which the other amount of fertilizers remains in the soil. Radionuclides in fertilizers may results to radiological hazards due to external exposure by handling, packing and transportation of fertilizers (Van et al., 2018).

NORM can pose a significant radiological hazard to humans and the environment. The dose from external exposure is mainly derived from gamma radiation due to its long distance travelling and high penetrating power (Sánchez et al., 2012). External exposure to gamma radiation comes from primordial radionuclides and it exists in all soils, building materials, in indoor environment and is responsible for external exposure to humans (IAEA, 2015). External exposure is exposure resulting from a source that comes from outside the body (Alharbi, 2016) and it contributes minimal exposure to the general populace due to low average intensity and limited penetration in the body (Banzi et al., 2017).

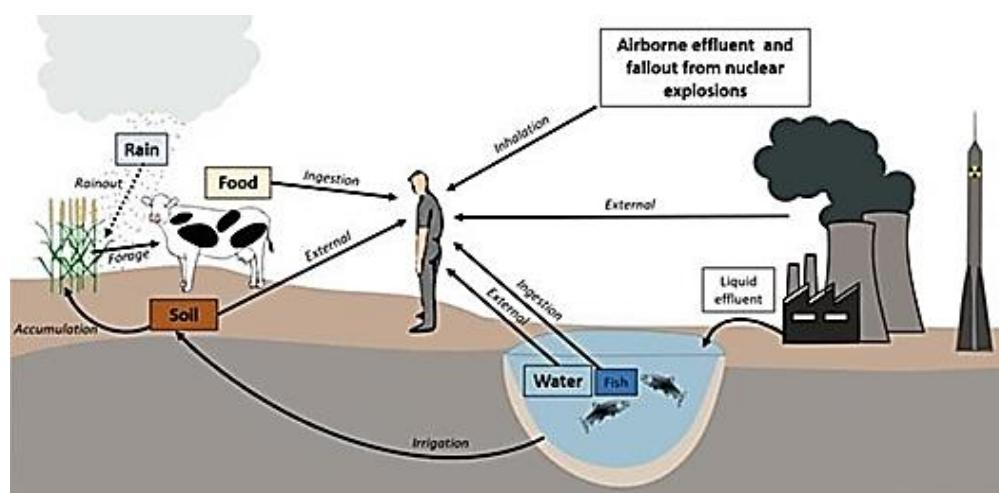


Figure 1.1: Major exposure pathways of radionuclides (Nieder et al., 2018).

The source can be removed by controlling the exposure, i.e., to reduce the exposure duration, source distance and shielding. The occupancy time for external exposure is lower than the occupancy time for internal exposure inside the human body (Banzi et al., 2017). Alpha or beta particles emanated from radionuclides may accumulate inside the body through absorption or inhalation and gamma rays which produces the essential source of external exposure to humans from some of the nuclear species. (Al-Hamzawi, 2017).

NORM are released to the environment in different ways and give rise to human radiation doses. Natural processes such as weathering deposition and wind erosion release NORM which cause human exposure to radiation posing radiological hazards and potential health risk (Shanthi et al., 2010). Through ingestion and inhalation, internal exposure occurs when a radionuclide member of NORM enters the human body (Awudu et al., 2012). The internal exposure from food is more important because it is related to the concentration of radionuclides in food from the soil through roots uptake (Banzi et al., 2017). Inhalation exposure dose comes from dust particles that exist in the air including radionuclides from ^{238}U and ^{232}Th decay series (Canbazoglu and Dogru, 2013).

The main reservoir for radioactivity contamination is the soil (Asaduzzaman et al., 2015). There have been studies which concerns NORM in soil media which gives information about the nature and background radiation levels in a certain area (Santawamaitre et al., 2010). The natural radionuclides in the soil such as ^{40}K and ^{226}Ra are metabolically transferred into soil and plants and find their way into water (Harb, 2015). Most soils contain ^{40}K and nuclides of the uranium and thorium series in which their concentrations vary as shown by many studies. The concentration in plants is proportional to the concentration of radionuclides in the soil as it is postulated under equilibrium conditions (Chakraborty et al., 2013).

The concentration and the distribution of radionuclides is important because it give information about the monitoring of environmental contamination by natural radioactivity through agricultural land producing foodstuff (Asaduzzaman et al., 2015). The status of the soil on which food crops are grown will determine, to a significant extent, the quality of foodstuffs produced (Görür et al., 2012). The concentration variation of radionuclides on the surface of the soil depends on the mineralogical composition the soil, chemical and physical properties, conditions in meteorological, and the transfer of material to deeper layers of the soil (Asaduzzaman et al., 2014). It has been shown in previous studies that surface soil is the largest radioactive source providing radionuclides to plants (Le et al., 2017).

Water is also considered as a nutritional requirement used for irrigation in fruits and vegetables and contributes to exposure to naturally occurring radionuclides (Machraoui et al., 2019). The effective dose received from drinking water is about 0.1 mSv, which is less than 5% of the total effective dose received from all-natural sources. Aquatic environment provide a large fraction of diet through aquatic foods of some individual and fish contamination constitute a significant pathway of radiocaesium transfer to human being (Alao, 2011).

Air, water, and soil transfer radioactivity to plants. The radionuclides present in the environment can be transferred into plants by indirect uptake from soil through roots and direct methods by absorption through aerial (stem, leaves) parts of the plants (Abojassim et al., 2016). Natural radionuclides from the soil find their way to the plants through roots, stem and end up in the leaves during absorption of nutrients dissolved in water (Sowole and Olaniyi, 2018). Then, the radioactivity is transferred to the human diet (Governorte of the Hong Kong, 2013).

Food may be contaminated from fallout of short-lived and long-lived radionuclides (Zehring, 2016). It has been estimated that during intake of food, at least one-eighth of the mean annual effective dose is as a result of natural sources (Giri et al., 2013). Natural radioactivity measurements have been carried out in different countries to determine in the base line data from the natural radiation levels (Ahmad et al., 2015). Several studies have been performed in different countries to determine the radionuclide concentration in different food samples and dose assessment from consumption of that foodstuff by the population (Awudu et al., 2012).

Concentrations of natural radioactivity in food are commonly in the range of 40–600 Bq.kg⁻¹ (Shahbazi-Gahrouei et al., 2013). Food, water, and air also contain trace amounts of alpha emitters from the uranium, thorium, and actinium series. Some of the radon (²²²Rn, and to a lesser extent ²²⁰Rn and ²¹⁹Rn) gas diffuses into the food supply (Awudu et al., 2012). The radioactivity of these radionuclides in food are related to the safety of population (Salih, 2018). The base line data can be used to assess any changes in radioactivity background levels occurring as a result of radioactive materials fallouts in future measurements (Al-Hamzawi, 2017).

1.5. Problem Statement

Nuclear technologies, industrial products and by-products may contribute to an increase in terrestrial radiation. Human interference may lead to an increase in levels of NORM through dumping, extracting, or concentrating radioactive materials from uranium mines (Taqi et al., 2016). Radioactivity from these mining activities and mineral resources can contaminate food (fruits and vegetables) after it has been discharged into the environment. The contamination of fruits and vegetables may also be direct and indirect (Al-Absi et al., 2015) from the uranium series radionuclides through the use of fertilizers and may lead to elevation of these nuclides in fruits and vegetables because fertilizer contains many toxicity elements and phosphate fertilizers contains significant amount of natural radionuclide because of phosphate rock (Van et al., 2018).

Fruits and vegetables are important in our bodies and the presence of these radionuclides ^{238}U , ^{232}Th and ^{40}K have radiological implications on people consuming these food sources. The fruits and vegetables may become contaminated with these natural radionuclides through uptake from the shoots of the plants from the soil together with the nutrients throughout mineral uptake. Thus, consuming the contaminated fruits and vegetables may increase the amount of radioactivity inside a person and causing the accumulation of radionuclides in human organs, and hence increasing the exposure to radiation, subsequently increasing the health risks associated with them. The vegetables from local markets of Hartbeespoort, Mahikeng and Pretoria in South Africa may contain ^{238}U , ^{232}Th , and ^{40}K radionuclides. Determination of activity concentrations of these radionuclides in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria provides the radiological baseline information that can be used to estimate exposure of human beings to NORM in the future. Elongated exposure to NORM may lead to a life-time cancer risk.

1.6. Aims and objectives

The main aim of this study was to assess the levels of NORM in locally produced fruits and vegetables from the local markets in Hartbeespoort, Mahikeng and Pretoria in South Africa.

The specific objectives of the study:

- Determine radioactivity levels of ^{238}U , ^{235}U , ^{234}U and ^{232}Th in fruits and vegetables from Hartbeespoort; Mahikeng and Pretoria local markets, using gamma spectrometry;
- Estimate the exposure dose of ^{238}U , ^{235}U , ^{234}U and ^{232}Th radionuclides concentrations in fruits and vegetables;
- Evaluate the exposure risk to members of the public due to consumption of NORM in fruits and vegetables; and
- Evaluate the Excess Lifetime Cancer Risk to the members of the public.

CHAPTER TWO: LITERATURE REVIEW

2.1. Radioactivity

Radioactivity is the emission of alpha, beta and gamma radiation from unstable isotopes such as a long-live uranium (^{238}U), thorium (^{232}Th) and (^{40}K) radioactive elements (Gaafar et al., 2016). The materials such as rocks, soils, plants, sand, water, air and building materials present natural radioactivity geological formations and in the earth's environment (Bayrak et al., 2018). The study of the radioactivity concentration in plants is of interest because it provides more information in monitoring environmental radioactivity and to measure the level of radiation doses to which humans are exposed (Asaduzzaman et al., 2015).

2.2. Exponential law of radioactive decay

Radioactive decay is a process where an unstable nucleus loses energy as radiation is emitted. The parent nuclide transforms to a daughter nuclide during the loss of energy (Cappellaro, 2012). According to the radioactive decay law, an unstable nucleus transforms into another nucleus which is a random process (Sagiron, 2012). In a radioactive material, the radioactive decay per unit time is directly proportional to the total number of radioactive compounds in the sample (Kónya and Nagy, 2018). The change in N is given by:

$$dN = -\lambda dt; N(0) = N_0 \quad (2.1)$$

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

$$d[\log N] = -\lambda dt \quad (2.3)$$

$$\log N = \log N_0 - \lambda t \quad (2.4)$$

$$N = N_0 e^{-\lambda t} \quad (2.5)$$

where N is the number of atoms present at time t , N_0 is the number of atoms initially present, λ is decay constant (s^{-1}) and t is time in seconds.

Radiation is measured using the quantity called activity (Magagula, 2015). The activity (A) is defined as the rate of decay of a sample measured by the number of disintegrations per second. The unit of activity, Ci (curie) is equivalent to 3.7×10^{10} disintegrations per second and the SI unit of radioactivity is the Becquerel (Bq). The activity, the number of atoms, and decay constant relationship is shown in the following Equation (2.6).

$$A = -\frac{dN}{dt} = \lambda N \quad (2.6)$$

where A is the activity of the nuclide (disintegrations/second), and λ and N are defined in equation (2.1) and (2.5) above. A is equal to the number N of radioactive nuclei disintegrating in a given time. From equation (2.6), A and N are always proportional (Santawamaitre et al., 2010).

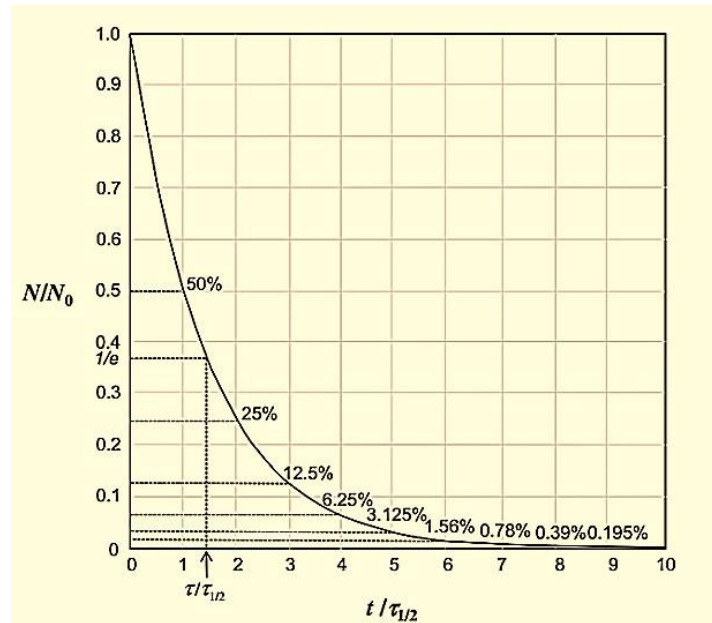


Figure 2.1: The radioactive decay curve shows the radionuclides numbers of fractions surviving until the time $\frac{t}{\tau_{1/2}}$, throughout ten half-lives. The number of radionuclides is decreased to less than 0.8 % of the initial value after seven half- lives and after ten half-lives, less than 0.1 % can be deduced (De Sanctis et al., 2016).

2.3. Half-life

The half-life of a radionuclide is the rate at which the nuclide undergoes radioactive transition (Van Rooyen, 2012). The half-life and decay constant relationship can be obtained from Equation (2.7). To calculate the half-life, Equation (2.7) must be solved

for time t , when the activity A , is equal to one-half its initial activity A_0

$$A = A_0 e^{-\lambda t} \quad (2.7)$$

$$\frac{A}{A_0} = e^{-\lambda t} \quad (2.8)$$

$$\ln\left(\frac{A}{A_0}\right) = -\lambda \quad (2.9)$$

$$t = \frac{-\ln \frac{A}{A_0}}{\lambda} \quad (2.10)$$

$$t_{1/2} = \frac{-\ln \frac{1}{2}}{\lambda} \quad (2.11)$$

$$t_{1/2} = \frac{\ln 2}{\lambda} = \frac{0.693}{\lambda} \quad (2.12)$$

2.4. Decay chain

Radioactive decay series is defined as groups of isotopes which represent several stages of radioactive decay by which the group of heavier members are transformed into lighter ones, in which the lightest are stable (Radioactive series, 2019). There is emission of various radiation types during the decay. From less than a second to billions of years, a half-life varies in different types of the atoms, for example, it will take 4.5 billion of years for the ^{238}U atom to spontaneously disintegrate, and it will take the ^{239}Pu atom up to 24,000 years for half the mass of it to spontaneously disintegrate (Sagirov, 2012). The radionuclide species making up the primordial radioactive decay chains headed by ^{232}Th and ^{238}U , together with their associated daughter and granddaughter decay members constitute a significant fraction of the naturally occurring radioactive materials (NORM) which are present in the wider environment (Shams et al., 2017).

2.5. The decay series of NORM

There are three natural decay series that include the heavy elements, from thallium to uranium; their initial nuclides are ^{238}U , ^{235}U , and ^{232}Th isotopes, and via alpha and beta decays, they end up as lead isotopes (^{206}Pb , ^{207}Pb , and ^{208}Pb , respectively). The Uranium-238 series is found naturally with a half-life 4.5 billion years. The decay series has a total of 14 steps, emitting 8 alpha particles and 6 beta particles accompanied by gamma radiation, until stable ^{206}Pb is formed (Hussain and Hussain, 2011). The Actinium series starts with ^{235}U and ends with stable ^{207}Pb which has a half-life of 700 million years. Only ^{235}U can be measured within this series, although ^{227}Th , ^{223}Ra , and ^{219}Rn can be measured with more difficulty. The uncertainty may be high, the measurement of daughter nuclides may give useful information by confirming the direct measurements of ^{235}U (Kónya and Nagy, 2018).

There are thorium isotopes with shorter half-lives in three natural decay chains, ^{234}Th (21.4 d) and ^{230}Th (7.54×10^4 y) in the ^{238}U , ^{228}Th (1.9 y) in the ^{232}Th , ^{231}Th (1.06 d) in the ^{235}U (Xhixha, 2012). The isotopes of thorium in natural decay series are all present. The series has ten decay stages with six alpha particles and four beta particles (Kónya and Nagy, 2018). Natural potassium with a natural abundance of 0.0118% has three modes of decay which are positron emission, electron capture and beta emission. ^{40}K radionuclides, by the emission of beta particle, disintegrate into ^{40}Ca (the ground state). The following are the decays series of natural radionuclides ^{238}U , ^{235}U , ^{232}Th and ^{40}K (Alharbi, 2016).

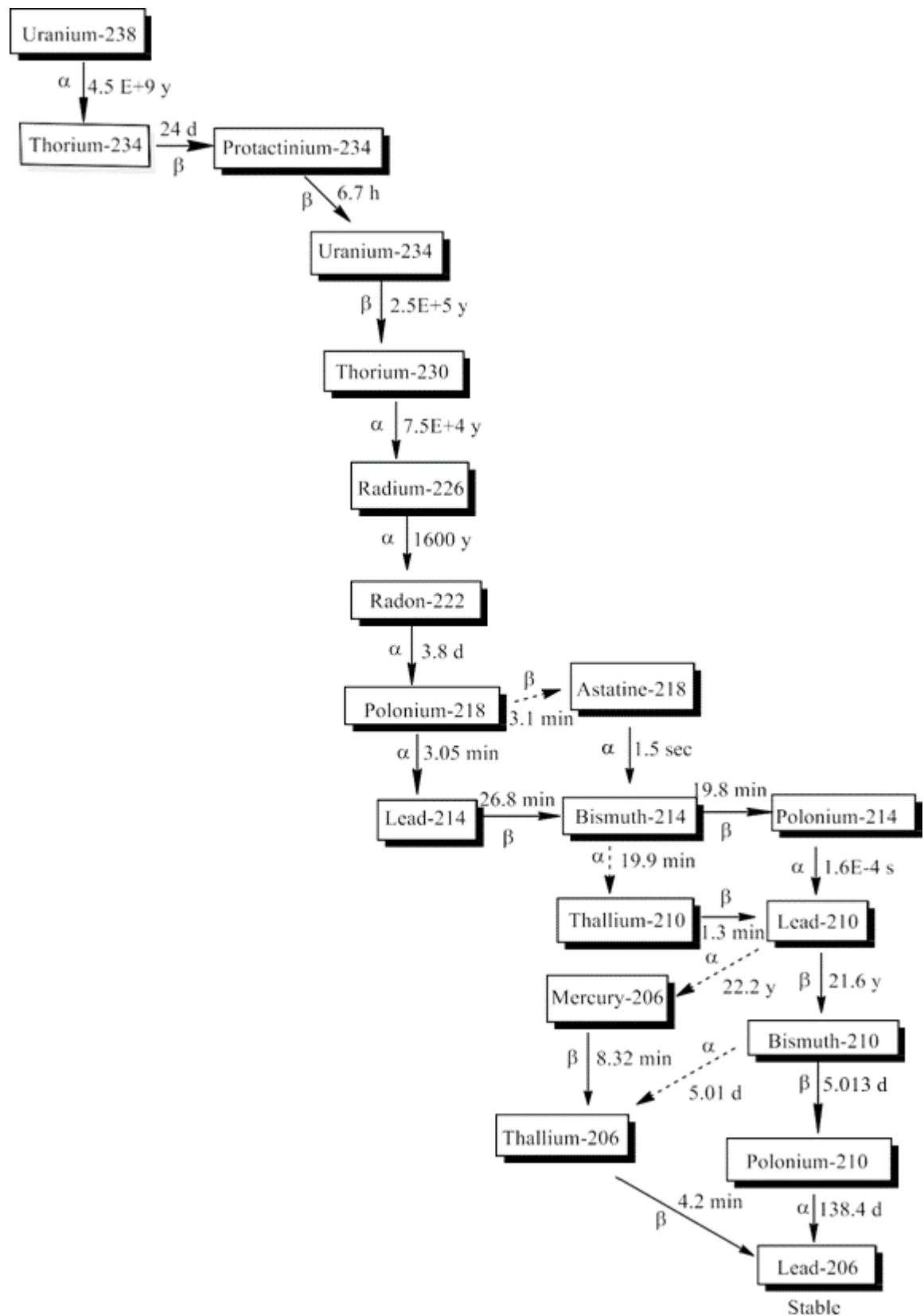


Figure 2.2: ^{238}U decay series (Pallavicini, 2011).

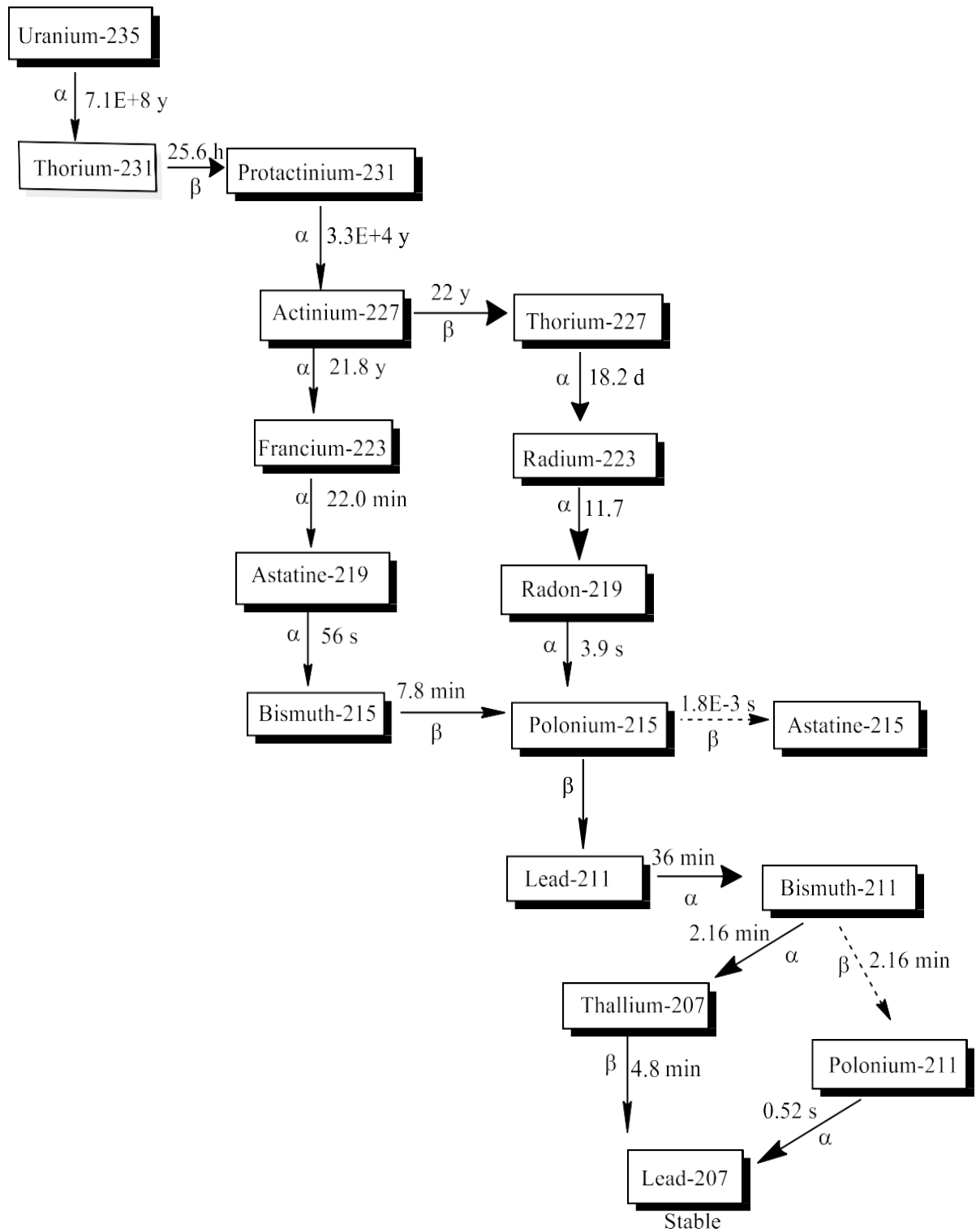


Figure 2.3: ^{235}U decay series (Pallavicini, 2011).

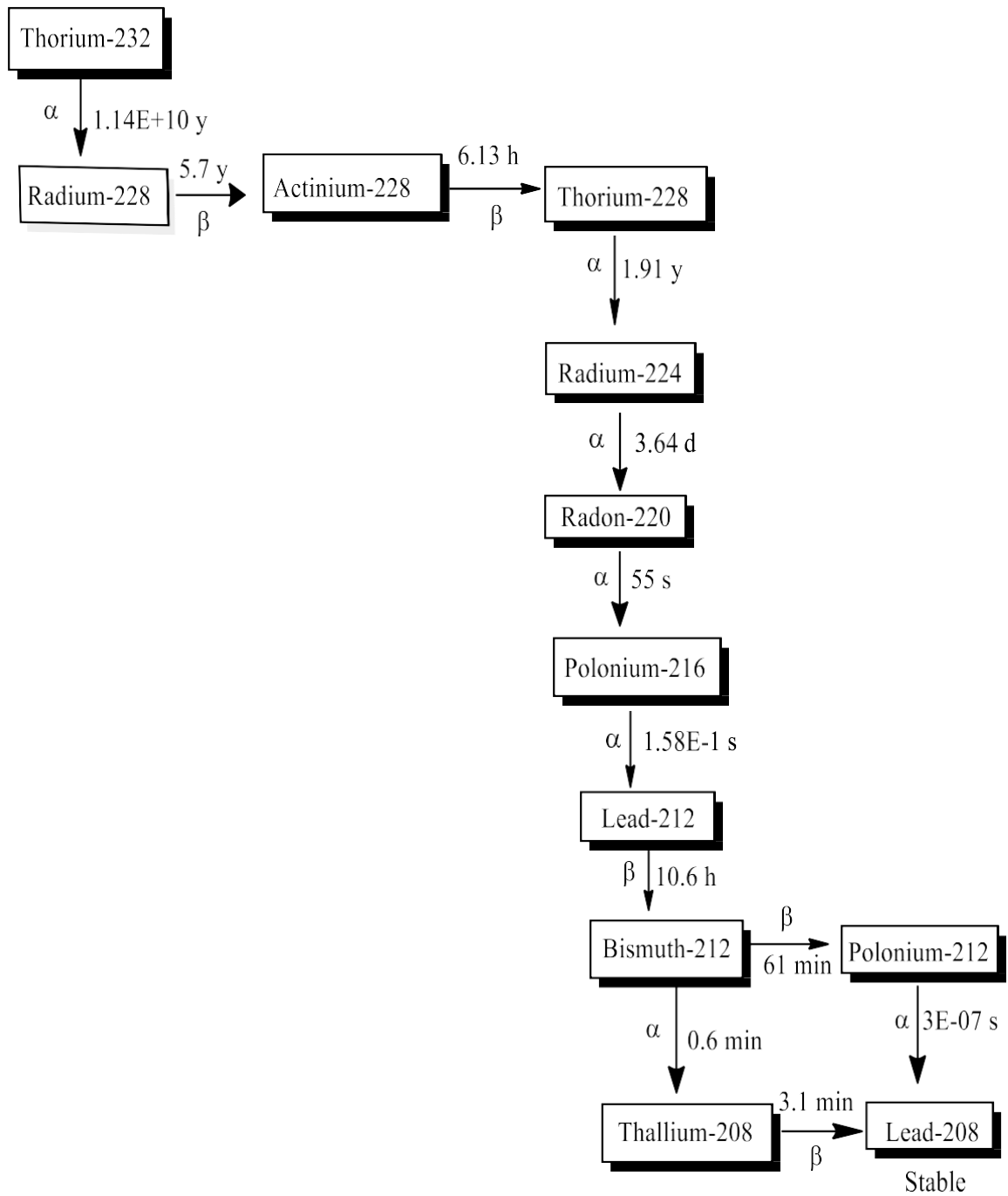


Figure 2.4: ^{232}Th decay series (Pallavicini, 2011).

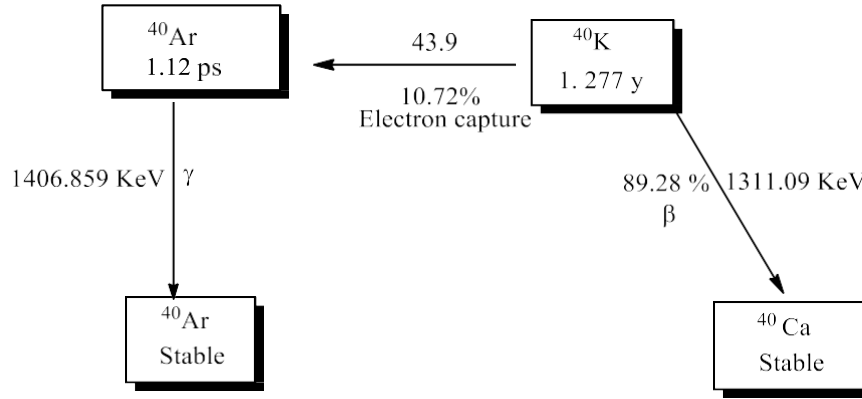


Figure 2.5: ^{40}K decay mode (Xhixha, 2012).

2.6. Nuclear decay types

Radioactive atoms are said to be unstable and they emit or give off excess energy or mass to reach stability. The three main types of ionizing radiation are called alpha (α), beta (β) and gamma (γ) radiation (Nieder et al., 2018).

2.6.1. Alpha decay

An Alpha particle is a helium particle which consists of two protons and two neutrons (Cherry et al., 2012). The particle moves slowly at about $20,000 \text{ km.s}^{-1}$ and can penetrate a paper sheet and thus they have very low penetration power compared to beta particles (Salih, 2018). Naturally occurring radionuclides with atomic number greater than 82 and artificially produced transuranic elements with atomic number greater than 92 undergo alpha decay and emit alpha particle. Alpha particle emission leads to the reduction of the final nuclear mass and charge from the initial nucleus (Pallavicini, 2011). The daughter nuclide may reach the ground state by energy release through electromagnetic gamma radiation emission (Santawamaitre et al., 2010). The following nuclear equation shows the alpha decay:

$${}^A_Z M \rightarrow {}^{A-4}_{Z-2} M + \alpha \quad (2.13)$$

The following equation is an example of alpha decay of ^{235}U :

$${}^{235}_{92}\text{U} \rightarrow {}^{231}_{90}\text{Th} + {}^4_2\alpha \quad (2.14)$$

The energy range of alpha particles is from 4 to 8 MeV with a high atomic mass of 4 in

which they travel in straight lines through matter and deposit their energy over short distances (Royal Society of Chemistry, 2019). The following figure (2.6) shows alpha decay of ^{235}U :

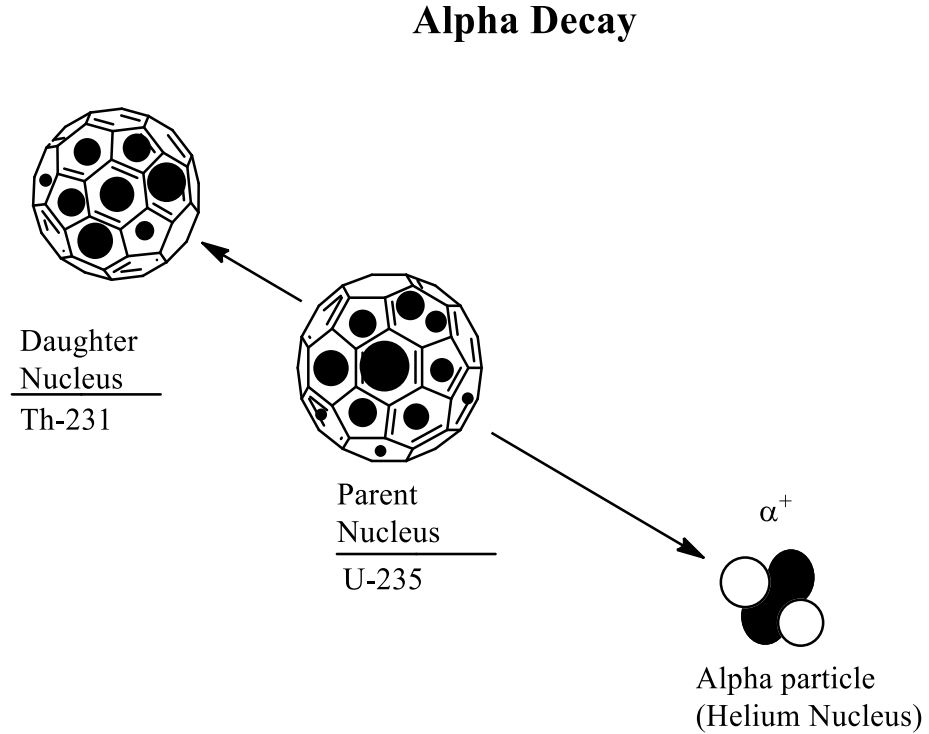


Figure 2.6: Alpha decay (ISU, 2011).

2.6.2. Beta decay

Beta particles are charged particles which are characterized by β^- , β^+ and electron capture emission. The beta negative decay (β^-) is when the electron is emitted from the nucleus of the radioactive nuclide with an excess of neutrons. Positive beta particles or positrons (β^+) can occur when an unstable nucleus emits an electron and an antineutrino, and a neutron in the nucleus becomes a proton remaining in the product nucleus (Lilley, 2013).

The process of emission of beta decay can be described as follows:

$$\beta^- \text{ decay: } n \rightarrow p^+ + \beta^- + \bar{\nu} \quad (2.15)$$

$$\beta^+ \text{ decay: } p^+ \rightarrow n + \beta^+ + \nu \quad (2.16)$$

$$EC: p^+ + e^- \rightarrow n + \nu \quad (2.17)$$

where $\bar{\nu}$ is the electron antineutrino, and ν the electron neutrino. A positron is an antiparticle having the same rest mass but opposite electron charges. Neutral particles with a zero or a very small mass are neutrinos and antineutrinos.

The β^- is the decay of nucleons in the nucleus, that is, of neutrons into protons:

$${}_0^1n \rightarrow {}_1^1p + {}_{-1}^0e + \bar{\nu} \quad (2.18)$$

Or

$${}_1^1p \rightarrow {}_0^1n + {}_{+1}^0e + \nu \quad (2.19)$$

The decay equation of beta negative decay can be written as follows:

$${}_{19}^{40}K \rightarrow {}_{20}^{40}Ca + {}_{-1}^0\beta^- + {}_0^0\nu \quad (2.20)$$

The β^- particle moves very fast at 280,000 km/s and its a high-energy particle. Beta particle can be stopped by a few millimeters of air or a sheet of aluminium. Figure (2.7) illustrates the decay of negative Beta:

Beta Decay

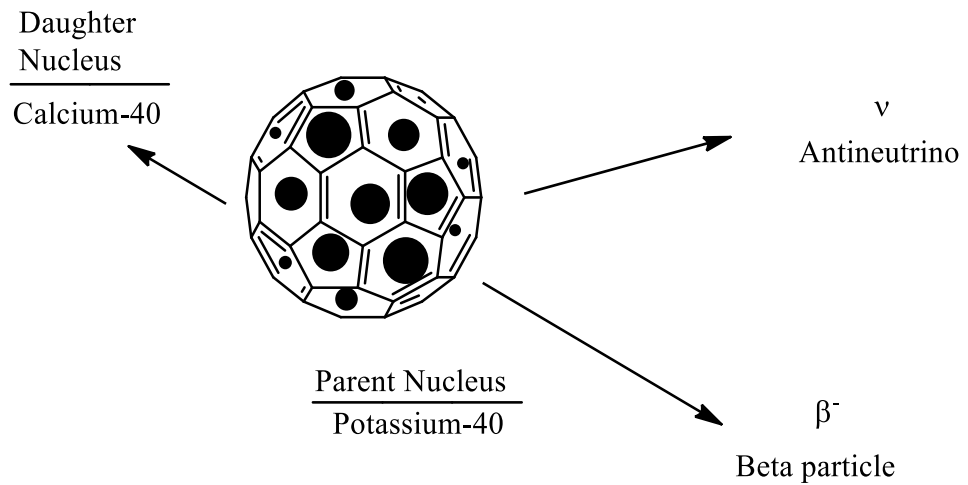


Figure 2.7: Beta decay (ISU, 2011).

2.6.3. Gamma decay

Gamma radiation (γ) is the de-excitation of the nucleus by the emission of a γ photon. It originate from the excited nuclei after alpha and beta decay which release energy via gamma ray emission from higher to lower energy state (L'Annunziata and Kessler, 2012). During gamma emission, the number of protons and neutrons do not change. A γ photon has a high electromagnetic radiation energy with a high frequency (wavelength of 10^{-10} m) and it can be attenuated by lead or concrete. The ranges of energy are between 0.1 to 10 MeV (Royal Society of Chemistry, 2019). If the initial energy of the nucleus is E_i at rest, then it decays to E_f , the conservation of the total mass-energy emitted by gamma rays is given in the following equation:

$$E_i = E_f + E_\gamma + E_r, \quad (2.21)$$

where E_γ is the gamma ray energy and E_r is the recoil energy of the nucleus. The difference between the initial and final state of gamma-ray energy is:

$$\Delta E = E_i - E_f = E_\gamma \quad (2.22)$$

Gamma Decay

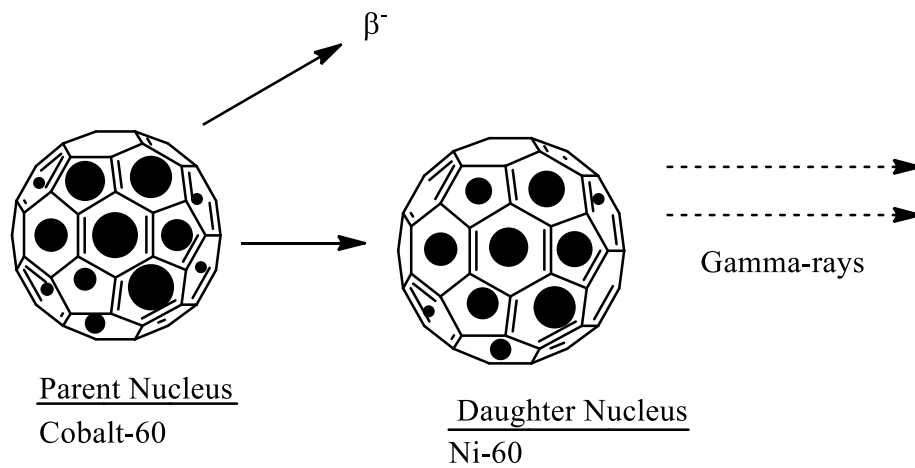


Figure 2.8: Gamm Decay (ISU, 2011)

2.7. Electron capture

Electron capture (EC) is similar to the decay of a positron which can take place when energy transformation is insufficient to create an electron pair (Santawamaitre et al., 2010). Electron capture is a decay process in which an electron is absorbed by the nucleus and combines with a proton to form a neutron and a neutrino (Chan, 2012). From the atomic nucleus, the neutrino is ejected:



During the process of electron capture, the total number of nucleons (protons + neutrons) will remain the same (De Sanctis et al., 2016). The following is an example of electron capture in which beryllium atom (with 4 protons and 3 neutrons) becomes lithium atom (with 3 protons and 4 neutrons):



2.8. Internal conversion

Internal conversion is a process of radioactive decay by which a gamma photon from the nucleus interacts with and transfers all of its energy to a tightly bounded (K-shell) orbital electron. The electron is ejected from the atom with considerable energy, equal to that of the initial gamma photon minus the binding energy. The electron is emitted from an atom with high amount of energy and the resulting high-speed electrons from internal conversion are not beta particles (De Sanctis et al., 2016). The difference in energy between the excited state and the energy state which is lower is transferred directly to one of the orbital electrons of the atom, which can be in the electron shell of a K- or L in internal conversion (Santawamaitre et al., 2010). The atomic number does not change during the process of internal conversion and no transmutation of one element to another. The electron can be ejected from the atom if this energy is greater than the binding energy for this particular electron by the following equation (Saha, 2012):

$$E_{e^{-}} = \Delta E - E_b \quad (2.25)$$

where ΔE is the transition energy and E_b is the binding energy of the electron in the atom.

2.9. Interaction of gamma rays with matter

The activity concentration measurements of radionuclides are based on gamma-ray interaction with matter. There are three mechanism by which gamma-rays interact with matter. These mechanisms are photoelectric effect, Compton scattering and pair production (Tsoulfanidis, 2010).

2.9.1. Photoelectric effect

Photoelectric effect is a phenomenon where a photon undergoes an interaction with an electron which is bound in an atom and this occur at low energies. The incident photon disappears, and a photoelectron is ejected by the atom from one of its bound shells (Kónya and Nagy, 2018). The resultant vacancy will be filled by a free electron from the other shell of the atom, which then emits characteristics X-rays. The kinetic energy of the ejected photoelectron is equal to the incident photon energy minus the binding energy of the photoelectron in its original shell. Photoelectrons are emitted by the photoelectric effect if the photon energy exceeds a threshold energy, called the binding energy of the electron or the work function (L'Annunziata and Kessler, 2012). The photoelectron appears with an energy given by:

$$E_e = h\nu - E_b \quad (2.26)$$

where $h\nu$ and E_b represent the energy of the photon and the binding energy.

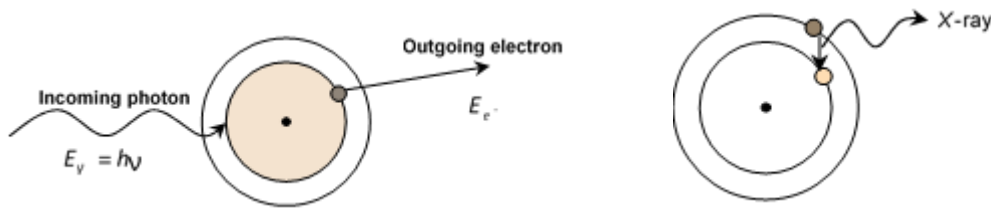


Figure 2.9: Schematically diagram of the photoelectric effect process (Knoll, 2010).

2.9.2. Compton scattering

Compton scattering is the inelastic scattering of a photon by a charged particle, usually an electron. In Compton scattering, the incident gamma ray photon is deflected through an angle θ with respect to its original direction. This deflection results in a decrease in energy of the photon and is called the Compton effect. The photon of frequency f collides with an electron at rest in Compton scattering. Upon collision, the photon bounces out of the electron, giving away its initial energy (given by Planck's formula ($E = hf$)), while

the electron gains momentum, the photon cannot lower its velocity (Kónya and Nagy, 2018). As a result of momentum conservation law, the photon must lower its momentum given by:

$$P_{\text{photon}} = \frac{E}{c} = \frac{hf}{c} = \frac{h}{\lambda} \quad (2.27)$$

The decrease in photon's momentum is converted into its decreasing frequency (increasing in wavelength ($\Delta\lambda = \lambda' - \lambda$)). The wavelength shift increased with scattering angle according to the Compton formula:

$$\Delta\lambda = \lambda' - \lambda = \frac{h}{m_e c} (1 - \cos(\theta)), \quad (2.28)$$

where λ' is the initial wavelength of photon, λ is the wavelength after scattering, h is the Planck constant $-6.626 \times 10^{-34} \text{ J}$, m_e is the electron rest mass (0.511 MeV) and c is the speed of light, θ is the scattering angle.

The incident gamma-ray photon is deflected through an angle θ with respect to its original direction. This deflection results in a decrease in energy of the photon. The minimum change in wavelength ($\lambda' - \lambda$) for the photon takes place when $\theta = 0$, ($\cos(180) = 1$). The maximum change in wavelength ($\lambda' - \lambda$) for the photon takes place when the angle $\theta = 180^\circ$ ($\cos(180) = -1$), the photon transfers to the electron as much momentum as possible (Lindsay, 2011). The maximum change in wavelength can be derived from Compton formula:

$$\Delta\lambda = \frac{2h}{m_e c} = \frac{2 \times (6.626 \times 10^{-34} [\text{J.s}])}{9.1 \times 10^{-31} [\text{kg}] \times 3 \times 10^8 \left[\frac{\text{m}}{\text{s}} \right]} = 4.86 \times 10^{-12} \text{ m} \quad (2.29)$$

Compton scattering contributes to the full energy peak in a spectrum by multiple of Compton scattering under the condition of that the last interaction is a photonelectric effect and all the proceeding of Compton interactions takes place in the Germanium crystal (Khandaker, 2011).

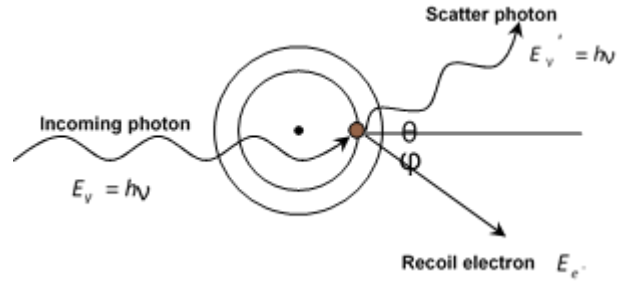


Figure 2.10: The process of Compton scattering schematic diagram (Knoll, 2010).

2.9.3. Pair production

Pair production is a phenomenon where energy is directly converted to matter. There are two ways in which pair production can be viewed, such as a particle and antiparticle (Khandaker, 2011). The probability mechanisms of pair production depends on the gamma-ray energy above threshold energy. The threshold energy of these reaction is 1.022 MeV which occurs when the incident photon interacts in the electric field surrounding the nucleus. The threshold energy for electron-positron pair production is double because 0.511 MeV energy is equivalent to the single electron rest mass. After the creation of the electron and positron pair, their kinetic energy will be lost by collisions through ionization with electrons in the surrounding material (Santawamaitre et al., 2010).

Pair production involves the following equation of the creation of mass from energy:

$$E_{pair} = m_e^- c^2 + m_e^+ c^2 = 2mc^2 = 1.022 \text{ MeV} \quad (2.30)$$

The photon may interact with the nucleus if it enters matter with 1.022 MeV excess energy. From the law of total mass-energy conservation, the following equation describe the energies that are carried away:

$$h\nu = 2M_e C^2 + K.E_{-e} + K.E_{+e} \quad (2.31)$$

The original gamma energy ($h\nu$) should be equal to the total rest-mass energy of both the electron and positron ($2M_e C^2 = 1022 \text{ keV}$). The kinetic energy $K.E_{-e}$ and $K.E_{+e}$ occurs when the energy is greater than 1022 keV. The $K.E_{+e}$ positron annihilate produce two

photons of energy 511 keV with atomic electrons in the detector (L'Annunziata and Kessler, 2012). The pair-production process provides the γ -ray energy total absorption. The initial gamma must have the energy of 1.022 MeV to create both negative and positive electrons because it takes much more energy for the event to happen using the law conservation of mass (Khandaker, 2011).

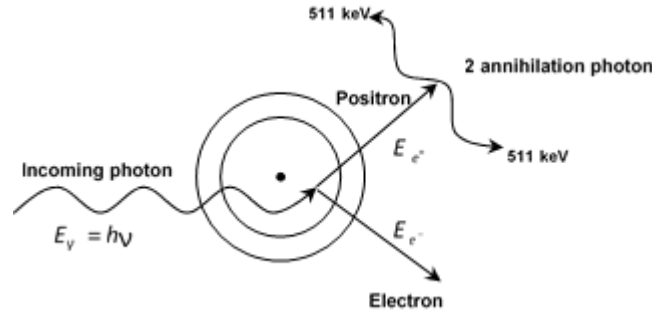


Figure 2.11: Pair production process schematic diagram (Knoll, 2010).

Figure 2.12 below illustrates how the probability of the three-principle gamma interacting with matter depend on the incident photon and the atomic number of the absorber. The left line represents the energy at which photoelectric absorption and Compton scattering are equal and the right line represents Compton scattering and pair production at which their energy are equal (Knoll, 2010).

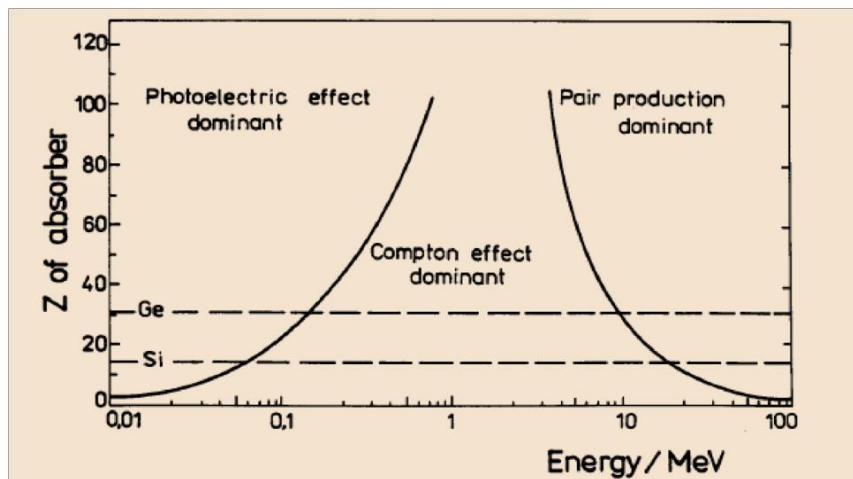


Figure 2.12: Three important gamma-ray interaction processes (Knoll, 2010).

2.10. Radioactive equilibrium

The parent radionuclide activity concentration in natural decay series is estimated from its daughters (Siegel, 2013).

$$\lambda_A N_A = \lambda_B N_B \quad (2.32)$$

Each nuclide activity is given by:

$$A_A = \lambda_A N_A \text{ and } A_B = \lambda_B N_B \quad (2.33)$$

Equilibrium conditions are used to determine the detection method or when there are difficulties in the measurements (Siegel, 2013). For example, in gamma spectrometry, the ^{226}Ra activity concentration is measured using the decay products of ^{214}Pb and ^{214}Bi after secular equilibrium. This is inaccurate when measuring ^{226}Ra from its gamma peak 186.1 keV that interferes with ^{235}U peak 185 keV (Alharbi, 2016).

2.10.1. Secular equilibrium

Secular equilibrium is a condition in which the activity between a long-lived parent radionuclide and short-lived daughter radionuclide are equal. The secular equilibrium criteria depend on the parent nuclide must have half-life longer than the daughter nuclide (L'Annunziata and Kessler, 2012). The activities of the product and the parent nuclide will be the same if product nuclide activity grows in to reach the parent radionuclide (Van Rooyen, 2012). Examples are the relations between the long-lived isotopes of uranium and thorium, ^{238}U , ^{235}U and ^{232}Th , and their decay products:

$$\lambda_p \ll \lambda_D \quad (2.34)$$

where λ_p and λ_D are the decay constant of the parent and daughter nuclide. More radiation is produced by the daughter nuclide. The activities of the parent and daughter become equal after about 7 half-lives of the daughter (D) (Lilley, 2013). When secular equilibrium is reached, the parent and daughter activity become equal, the following equation (2.35) can be generalized as follows:

$$\lambda_p N_p = \lambda_D N_D \dots \lambda_n N_n \quad (2.35)$$

2.11. Analytical techniques applied in NORM measurements

Alpha spectrometry, beta spectrometry and gamma spectrometry are the detection techniques used to measure the radioactivity of radionuclides within the soil, sediment, rocks, products, by-products, waste, and residues during the decay of radioactive radionuclides. Mass spectrometry is also used to measure the mass concentration and then converted to activity concentration of long-lived radioisotopes. The above-mentioned measuring methods involve the process of chemical separation except for gamma spectrometry. Gamma-ray spectrometry can be used for measuring the activity concentration of a large number of natural radionuclides (Alharbi, 2016). The calibration of the detector is performed by counting standard source. The measured energy of a gamma-ray corresponds to the type of element and its isotope, while the number of counts correspond to the abundance of the radioactive source present in the measured sample (Joel et al., 2017).

Gross alpha and beta analysis are another technique used to measure the overall activity of alpha and beta emitters. Gross alpha and beta measurements do not give information about the isotopic composition of the sample and cannot be used to estimate radiation exposure (Hoang, 2016). The gross alpha and beta methods of analysis used are the alpha spectroscopy, liquid scintillation spectroscopy or proportional counter (L'Annunziata and Kessler, 2012). The gross alpha radioactivity values depend on geological area characteristic, the mineral component content and the activities types on an area (Ogundare and Adekoya, 2015).

2.12. Principle of Gamma-ray spectrometry

There are different types of detector used either for ionization or excitation. The common materials used for semiconductors are germanium (Ge) and Silicon (Si). Semiconductor detectors are used for gamma spectrometry because of certain advantages such as high energy resolution, good stability, excellent timing characteristics and simplicity of operation. Germanium (Ge) detectors have a large energy resolution which detect activity concentration measurements of different radioisotopes of different gamma emission. The High Purity Germanium (HPGe) detector has been used in many environmental studies while the Coaxial detector has been used to quantify the ^{238}U in the soil on 63 keV gamma peak of its daughter radionuclides ^{234}Th (Canberra, 2011). The detector works based on gamma ray interacting with matter to produce signals from

particles entering the system. The energy of the gamma-ray is transferred to electrons creating electron pairs during the interaction (Tsoulfanidis, 2010). The following schematic diagram displays the electronic system of the HPGe detector semi-conductor detector.

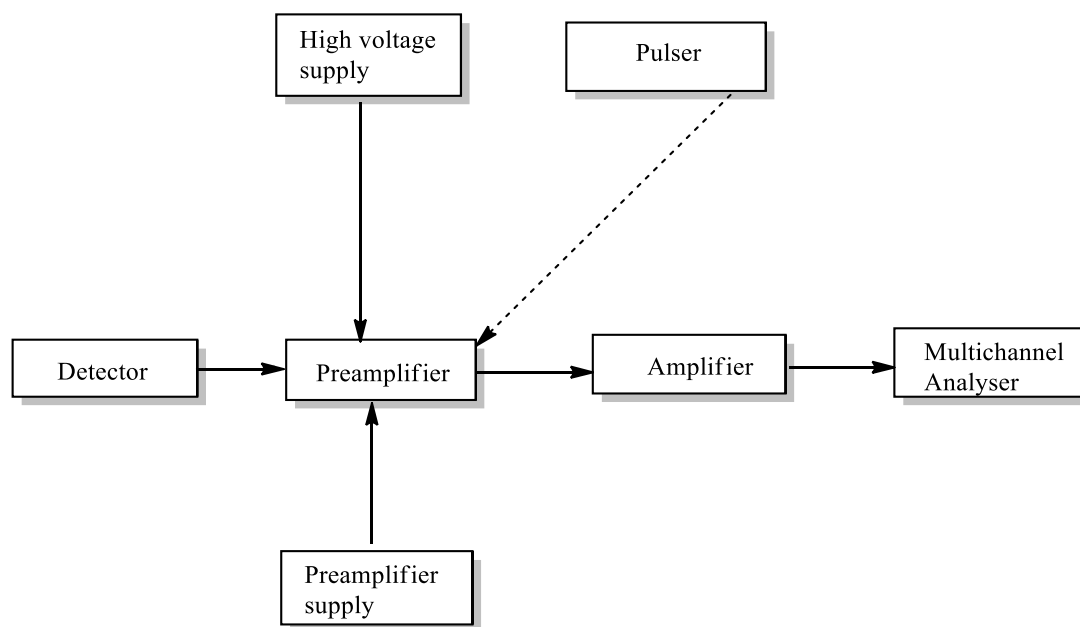


Figure 2.13: An electronic system schematic for gamma-ray spectrometry

(Tsoulfanidis, 2010).

There is intrinsic uncertainty in radioactivity analysis (Alharbi, 2016). Uncertainty can be due to many components such as matrix, weighing, the content of water, radioactivity and homogeneity, instrumentations which include the special acquisition, background and efficiency calibration, and properties of nuclear which include radioactive decay, emission probability and the half-life (Xhixha et al., 2013). Long time of counting, large sample size and sample-detector distances that are small are recommended for better statistics when using the detector in gamma spectrometry. However, a sample with a large volume results in self-absorption (Huy et al., 2013) and large counting time causing short-lived radionuclides to decay during the measurements (Abo-Elmagd et al., 2010).

2.13. Neutron activation analysis (NAA)

Neutron activation analysis was first introduced by George Von Hevesy and Hilde Levi in 1936. It is a technique to measure amounts of chemical elements in a sample, based on the conversion of stable nuclei to other, mostly radioactive nuclei, by neutron irradiation of the material (Heimann and Barron, 2014). The technique is based on the

nuclear reaction between neutrons and target nuclei (Hamidatou et al., 2013). Neutron activation analysis (NAA) is also a technique used for qualitative and quantitative elemental analysis of multiple major, minor, and trace elements in samples and is the useful method for the determination of elements from various sources in the ppb-ppm range without or with chemical separation (University of Missouri-Columbia, 2017).

Samples are bombarded with neutrons in order to transform stable isotopes of most element constituting samples into radioactive isotopes by neutron capture. The target is bombarded by neutrons such that neutron capture occurs, and atom will either remain stable or become unstable. If the nuclei is in an excited state and undergo β -decay emitting γ -rays, this can be detected using an HPGe detector (Chan, 2012). The activated nuclides decay according to a characteristic half-life emitting beta particles only, but other nuclides emit gamma-photon with specific energies.

2.13.1. Types of Neutron Activation Analysis

Instrumental neutron activation analysis (INAA) is based on short-lived radionuclide production by nuclear reactions, such as reactor neutrons (i.e., thermal neutrons). INAA has advantages such as better sensitivity, low detection limit, simple and no reagent blank correction (Sathyapriya et al., 2017). The method of INAA has been used to determine ^{232}Th by targetting ^{233}Pa produced in the (n, γ) reaction (Heimann and Barron, 2014). The decay of the short-lived of ^{233}Th with half-life 22.3 minutes produces ^{233}Pa with half-life of 27.0 days. In INAA, the sample does not undergo chemical separation and can be used when other radioactive isotopes do not interfere with measurement of the element of interest.

Radiochemistry neutron activation analysis (RNAA) involves sample decomposition and separation of elements. Through separation method, the interfering elements are separated from elements of interest (Hamidatou et al., 2013). INAA can be classified into two: prompt gamma ray neutron activation analysis (PGNAA) where measurements are performed immediately after irradiation and delayed gamma ray neutron activation analysis (DGNAA) where measurements follow after radioactive decay.

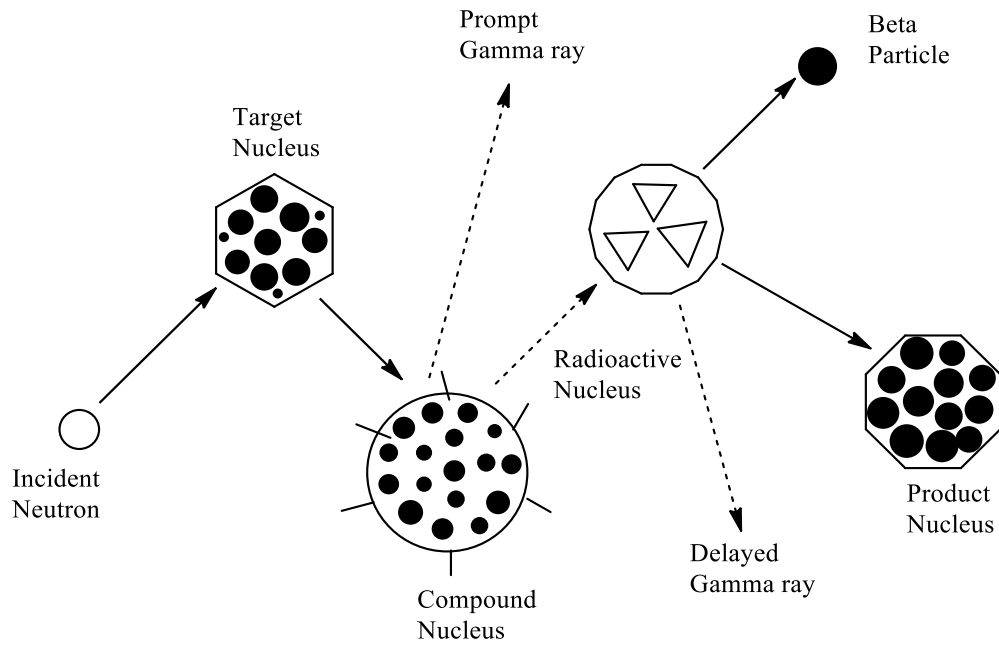
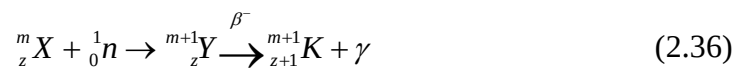


Figure 2.14: Method of NAA (University of Missouri-Columbia, 2017).

2.13.2. Neutron sources

The probability of a neutron interacting with a nucleus for a particular reaction is dependent on the kind of nucleus involved and the energy of the neutron. Isotropic neutron sources are most frequently used as neutron sources. They consist of an alpha emitting radioactive nucleus mixed with beryllium. The neutron sources are provided by neutron chain reactors utilising the fission reaction and neutron irradiation is commonly used as an activation technique in a nuclear reactor. These neutron sources are applied in accelerators where a convenient target material is bombarded by accelerated charged particles and are produced in a nuclear reaction (Chan, 2012). The nuclear activation process can be expressed using the following equation:



where;

X - radioisotope with atomic number z and mass number m of stable isotope.

Y - radioisotope which emit beta particle accompanied by gamma-rays.

n - neutrons.

γ - gamma-ray released.

The number of atoms at time t after irradiation can be expressed as:

$$N_y(t) = N_y(t_0)e^{-\lambda_y(t-t_0)} \quad (2.37)$$

For a nuclear reaction of the type (n, γ) , the induced activity (A_t) in the disintegration per second at the end of irradiation of time (t) is given by:

$$A_t = N\sigma\phi(1 - e^{-\lambda t}) = N\sigma\phi \left(1 - e^{-\frac{0.693}{t_{1/2}} t} \right) \quad (2.38)$$

where σ is thermal neutron capture cross section in cm^2 . The unit of cross-section is the barn ($1 \text{ barn} = 10^{-24} \text{ cm}^2$), ϕ is thermal neutron flux in neutrons $\text{cm}^{-2} \text{ sec}^{-1}$, λ is decay constant of the product nuclide ($\lambda = \frac{0.693}{t_{1/2}}$), $t_{1/2}$ is half-life of the product nuclide, N is number of atoms in the target.

2.14. Biological effects of radiation

Radiation is harmful to the life of all living organisms, depending on the dose, it can cause cancer, genetic and organs damages, cell killing and death (Nieder et al., 2018). The human body has many organs of different sizes, functions, and sensitivities (Hussain and Hussain, 2011).

The effects of radiation are classified based on their nature and timing after early or delayed exposure, e.g., somatic or hereditary, stochastic, or deterministic. Stochastic effects refer to random and unpredictable effects usually following chronic exposure to low dose radiation. Hereditary effects and carcinogenesis following diagnostic imaging is of a stochastic nature, deterministic (non-stochastic) effects are non-random and have a highly predictable response to radiation. Cancer may result when humans are exposed to certain levels of ^{238}U , ^{232}Th , and ^{40}K for a long period of time (Elgazzar and Kazem, 2015).

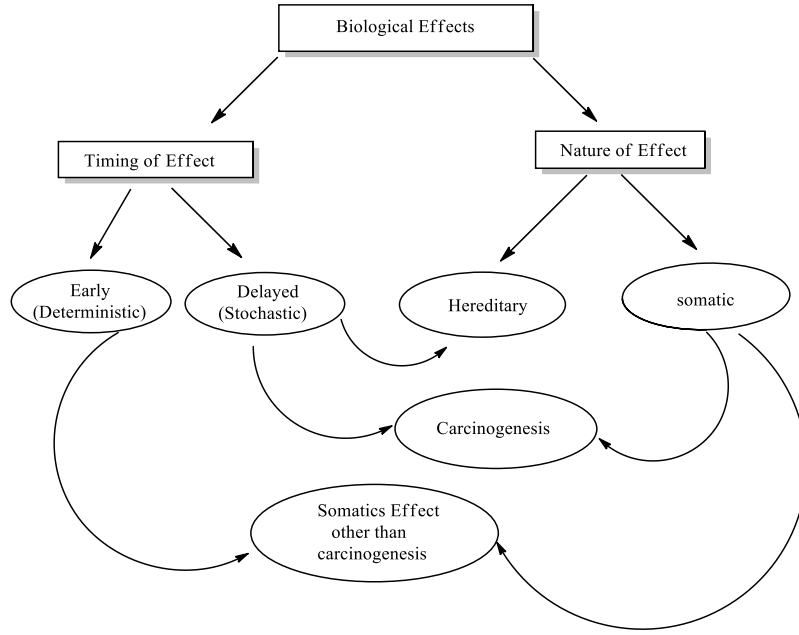


Figure 2.15: The various biological effects of ionizing radiation (Elgazzar and Kazem, 2015).

2.14.1. Absorbed dose and equivalent dose

Absorbed dose is a one of the quantities that is considered on biological effects of radiation. It measures the energy that is deposited into a medium by any type of radition. Absobed dose is the measure of the energy absorbed per unit mass from ionizing radiation into the material which is measured using the quantity gray Gy . $1 Gy = 1 J$ of absorbed energy per unit mass (kg) (Andiscoa et al., 2014). The ICRP introduced another unit which is equivalent dose in order to account for relative biological effectiveness (RBE). The equivalent dose is measured in Sievert (Sv). The equivalent dose used for biological damage to the tissues, is obtained when the absorbed dose is multiplied by a dimensionless, weighting factor (w_R) for the particular radiation (RSNA, 2018).

At any point in tissue, the dose equivaleng is given as follow:

$$H_T = \sum_R w_R D_{T,R} , \quad (2.39)$$

where;

H_T -is equivalent dose.

w_R - is the radiation weighting factor.

$D_{T,R}$ - is the absorbed dose averaged over the tissue or organ.

Different types of radiation are provided with their weighting factors used to correlate to the damage they cause from the energy they deposit on biological tissue (De Sanctis et al., 2016). The type of biological effects of ionising radiation vary with their energy. High linear energy transfer (LET) in a certain type of radiation will cause more damage than low LET of radiation type. The biological effects of radiation are influenced by other factors such as the rate of energy transfer (Balarane and Oladele, 2012).

Tables 2.1: Radiation weighting factors w_R of various radiation types (Balarane and Oladele, 2012).

Radiation weighting factors	
Type and energy range	w_R
Photons, all energies	1
Electrons and muons, all energies	1
Neutrons, energy <10 keV	5
10 keV-100 keV	10
>100 keV-2 MeV	20
>2 MeV-20 MeV	10
>20 MeV	5
Protons, other than recoil, energy > 2 MeV	5
Alpha particles, fission fragments, heavy nuclei	20

2.14.2. Effective dose

Effective dose is used to check for damage of radiation on different sensitivities to different tissues. Effective dose is obtained by multiplying the equivalent dose for a by weighting factor w_T of the relevant tissue (De Sanctis et al., 2016).

$$H_E = \sum_T w_T H_T, \quad (2.40)$$

where H_E effective dose, H_T is the equivalent dose in specific tissue or organ, and w_T is the tissue weighting factor. The SI unit of effective dose is Sievert (Sv).

The weighting factors w_T of certain tissues and organs are shown in Table 2.2 The equivalent dose older unit and effective dose is the rem (Roentgen Equivalent in Man), with the symbol of rem. One rem equals to one hundredth of a Sievert (1 rem = 0.01 Sv, = 10 mSv, or equivalently, 1 Sv = 100 rem).

Table 2.2: Weighting factors (w_T) of some of the tissues and organs (De Sanctis et al., 2016).

Tissues/organ	w_T
Gonads	0.20
Red bone marrow, colon, stomach, lung	0.12
Bladder, breast, liver, oesophagus, thyroid	0.05
Skin, bone surface	0.01
Other	0.05

2.15. Radiological Dose Assessment

2.15.1. Internal (H_{int}) and External Hazard Index (H_{ext})

The radiological hazards were presented in terms of single quantities called hazard index and the measured specific activity concentrations were used to assess the radiological hazards in terms of external hazards index (H_{ext}) and Internal hazards index (H_{int}), which were obtained using equations 2.41 and 2.42 respectively (Abdel-Rahman and El-Mongy, 2017).

$$H_{ext} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (2.41)$$

$$H_{int} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (2.42)$$

where A_U , A_{Th} , and A_K are the activity concentration in $Bq.kg^{-1}$ of ^{238}U , ^{232}Th and ^{40}K respectively. The radiation exposure due to radioactive substance from any material must be limited $1.5 mGy.y^{-1}$, the external hazard index, H_{ext} should obey the following

relation, $H_{ext} \leq 1$ (UNSCEAR, 2000).

2.15.2. Annual ingestion dose

Ingestion dose depends on the rate of consumption of the foodstuff and the concentration of the radionuclide involved (Canbazoglu and Dogru, 2013). The ingested radiation doses were obtained from the determined activity concentration of the radionuclides in the foodstuff ($Bq.kg^{-1}$) and multiplying by the annual consumption rate of food ($kg.y^{-1}$). The average consumption rate of fruits is $20 kg.y^{-1}$ while green vegetables are $15 (kg.y^{-1})$ for adults as reported by (UNSCEAR, 2000). The following equation was used to calculate the effective ingestion dose:

$$D_{IND} = A \times C \times FDC, \quad (2.43)$$

where D_{IND} is the annual ingestion dose ($\mu Sv.y^{-1}$), A is the activity concentration ($Bq.kg^{-1}$) of the radionuclide in fruits and vegetables, C is the consumption rate and FDC is the ingestion dose coefficient which equals to $0.28 \mu Sv.Bq^{-1}$ for ^{238}U , $0.23 \mu Sv.Bq^{-1}$ for ^{232}Th and $0.0062 \mu Sv.Bq^{-1}$ for ^{40}K (Aswood et al., 2017).

2.15.3. Estimation of absorbed dose rate

The absorbed dose rate is the measure of the number of radionuclides absorbed by the body for a given period (UNSCEAR, 2000). The activity concentration of ^{238}U , ^{232}Th and ^{40}K determined were converted into doses rate ($nGy.h^{-1}$) applying conversion factors of 0.462, 0.604 and 0.0417 for uranium, thorium and potassium respectively (UNSCEAR, 2000). These factors were calculated applying the following relation:

$$D_{rate} (nGy.h^{-1}) = (0.462A_U + 0.604A_{Th} + 0.0417A_K), \quad (2.44)$$

where, A_U , A_{Th} and A_K are the activity concentrations ($Bq.kg^{-1}$) of uranium, thorium, and potassium in the samples, respectively.

2.15.4. Annual effective dose equivalent (AEDE)

To estimate annual effective doses for indoor occupancy, the absorbed dose was

multiplied by the radiation weighting factor and the tissue weighting factors (KANT et al., 2015). The average of the annual effective dose equivalent received by public members is evaluated by using a value of 0.7 Sv.Gy^{-1} used for the conversion coefficient from the absorbed dose rate to an annual effective dose received by adults and 20 % for the outdoor occupancy (UNSCEAR, 2000). The annual effective doses were determined by the following equations:

$$Indoor(mSv) = (Absorbed \text{ Dose})nGh^{-1} \times 8760h \times 0.8 \times 0.7 \times 10^{-6} \text{ Sv.Gy}^{-1} \quad (2.45)$$

$$Outdoor(mSv) = (Absorbed \text{ Dose})nGh^{-1} \times 8760h \times 0.2 \times 0.7 \times 10^{-6} \text{ Sv.Gy}^{-1} \quad (2.46)$$

2.15.5. Excess Lifetime Cancer Risk (ELCR)

The excess lifetime cancer risk deals with the probability of developing cancer over a lifetime at a given exposure level (Adewumi, 2016). For low doses, ICRP fatal cancer risk factor is 0.05 Sv (ICRP-60, 2005).. The risk factor states that the probability of a person dying of cancer increases by 5% for a total dose of 1 Sv received during his lifetime. Therefore, the probability of death from cancer due to natural incidence increases from about 25% to 30% following a total lifetime exposure of 1 Sievert. The following equation was used to calculate the lifetime cancer risk (UNSCEAR, 2000):

$$ELCR(mSv.y^{-1}) = AEDE \times DL \times RF \quad (2.47)$$

where *AEDE* is the annual effective dose equivalent, *DL* is the average duration of life (estimated to be 70 years) *RF* is risk factor (Sv), fatal cancer risk per Sievert, for stochastic effects, the ICRP 106 uses *RF* as 0.05 Sv^{-1} for the public (UNSCEAR, 2000).

CHAPTER THREE: METHODOLOGY

3.1. Geography of the study areas

3.1.1. Hartbeespoort

Hartbeespoort is situated in the province of North West in South Africa. It is situated at the Magaliesberg mountain slope and the banks of the Hartbeespoort Dam. It falls under Madibeng Local Municipality which is located in the Bojanala Platinum District Municipality of the North West province between the majestic ancient Magaliesberg and the Witwatersrand mountain range. The Municipality includes the urban areas (Brits, Hartbeespoort and Skeerpoort). It has 43 villages and 9 000 farm areas. Madibeng is centrally situated approximately 50 km from Pretoria, 55 km from Johannesburg and 60 km from Rustenburg.

3.1.2. Mahikeng

Mahikeng is also situated in the North West province of South Africa, and it lies on the border of Botswana and South Africa, about 260 km west of Johannesburg. Mahikeng Local Municipality is 25 kilometres south of the Botswana Border. Mahikeng used to be known as the City Council of Mafikeng. The total area of the Mahikeng Local Municipality is approximately 3703 km² (Balarane and Oladele, 2012). It is divided into 28 wards consisting of 102 Villages and Suburbs. The population of the Municipality is estimated at 271 501 people. Approximately 75% of the area is rural. The average temperature in Mahikeng is 32⁰C in summer with less rainfall, and cold winter.

3.1.3. Pretoria

Pretoria is the Capital city of the Republic of South Africa and in the Northern part of Gauteng province with a population of about 750,000. The city is rich in the production of mineral resources and mining activities. The average temperature in summer is 29° C with much more rainfall compared to winter. It has a high agricultural land used for crop farming.

3.2. Materials and method

Fruits and Vegetables were purchased from Hartbeespoort, Mahikeng and Pretoria local markets. The samples were purchased between April and May 2019. The fruit and vegetable samples included potatoes, cucumbers, tomatoes, cabbage, spinach, carrots, pumpkin, oranges, apples, bananas, and pineapples as shown in the tables below.

Table 3.1: Food categories of fruits and vegetables.

Food Category	No of samples	Sample Name	Scientific Name
Fruits	1	Oranges	Citrus sinensis
	2	Apples	Malus domestica
	3	Bananas	Musa acuminata
	4	Pineapples	Ananas comosus
Vegetables	5	Potatoes	Solanum tuberosum
	6	Cucumbers	Cucumis sativus
	7	Tomatoes	Solanum lycopersium
	8	Cabbage	Brassica oleracea var. capitata
	9	Spinach	Spinacia oleracea
	10	Carrots	Daucus carota
	11	Pumpkin	Cucurbita pepo

The Global Positioning system (GPS) was used to locate the market from which samples were collected. The samples were processed at the South African Nuclear Energy Corporation (NECSA) Radio-Analytical Laboratory (Building P1600).

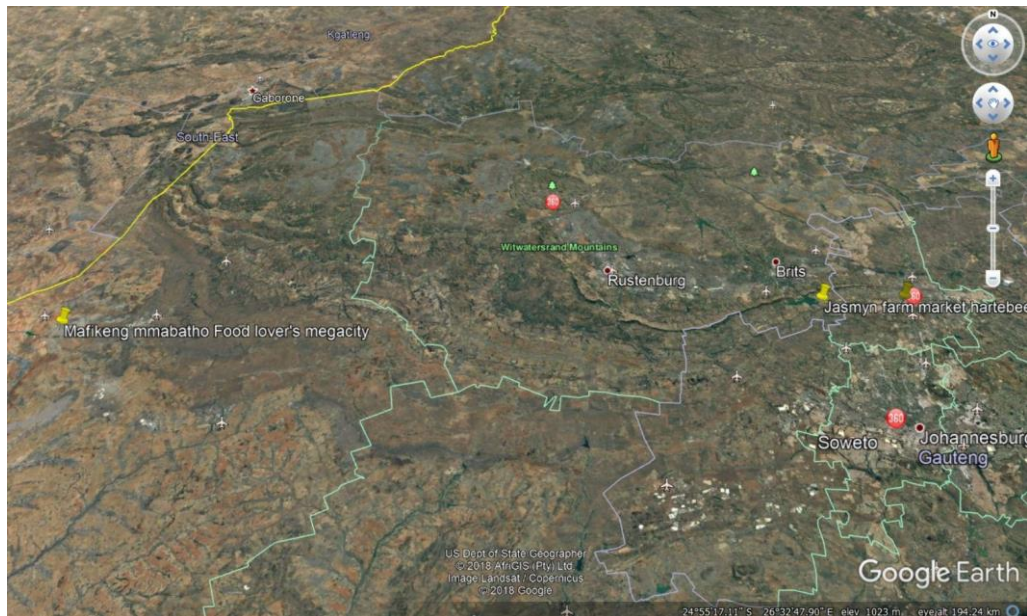


Figure 3.1: A map showing sample collection from local markets in Hartbeespoort, Mahikeng and Pretoria (Google earth maps, 2019).

3.3. Sample Preparation

3.3.1. Determination of the wet and dry mass of samples

An analytical weighing balance was tared to four decimals before each sample was weighed. The measured mass of each sample was recorded on a datasheet.

3.3.2. Drying of plant material

The fruits and vegetables were peeled, and the eatable parts were separated by cutting them into pieces using a knife. The samples were placed in trays and each sample number was written with a marker on the front rim. Each sample was weighed together with the tray as wet plant material and the mass was recorded. All samples were heated in an oven to dry at a temperature of 65°C for 10 hours.



Figure 3.2: Lab preparation of fruits and vegetables from wet to drying.

3.3.3. Determination of dry mass

All the dried samples were removed from the oven and were transferred into marked plastic bags. The dried samples were weighed together with the plastic bags and the masses were recorded.

3.4. Method to ash dry samples for analysis

3.4.1. Ashing of dry material

The mass of the dried sample material was recorded, and the sample transferred into a thin plastic bag. These samples were transferred into ash pans container and were loaded into a furnace at temperature less than 100°C. The temperature of the furnace was ramped up to 200°C. The samples were heated for 2 hours to turn them into charcoal or until fumes were produced. The furnace was left for 30 minutes to cool down and then the ash pans were removed from the furnace.



Figure 3.3: Ash Furnace.



Figure 3.4 Powdered Ash samples.

3.4.2. Determination of the mass of the ash

A mortar and pestle were used to grind the sample into fine powder. An empty sample container was weighed using the weighing balance and the mass was recorded. The ash samples were transferred into the sample container marked using Teflon funnel and clean brush. The container with ash was weighed and the mass was recorded under total.

The actual mass of the sample was calculated as follows:

$$Ash(d) = Total - Tare \quad (3.1)$$

$$\%Ash = (Ash \div Dry) \times 100 \quad (3.2)$$

3.6. Samples preparation for neutron activation analysis

An empty polythene ampoule was placed on the weighing balance to tare. Approximately 0.2 g of each ash finely powdered fruit sample, ash finely powdered vegetable sample, thorium standards and uranium standards were each loaded into polythene ampoule using tweezers, weighed using a weighing balance, sealed with rabbit end cap using a sealer machine and labelled for traceability.

3.6.1. Sample irradiation

The sealed polythene ampoule containing fruits, vegetables, thorium standards and uranium standards samples were placed on the wooden sample tray and were transferred to the channel of the irradiation close to the core of the reactor using a pneumatic rabbit system at the Routine Instrumental Neutron and Gamma Analysis System (RINGAS) laboratory for irradiation at the NECSA (SAFARI-1) research reactor. The irradiations of the samples was for 10 seconds, 15 seconds delayed and counted for 45 seconds using counter of neutron. Containers that were empty and reference materials which were certified and provided by the South African National Standards (SANAS) were irradiated and counted, for quality control under the same conditions as the samples. The isotopes ^{238}U and ^{232}Th capture neutrons during neutron activation in a reactor, and followed by the decay of negative beta into ^{239}U and ^{233}Th , respectively. Each irradiated sample was counted using a Coaxial HPGe gamma detector for one hour.



Figure 3.6: Encapsulated samples.

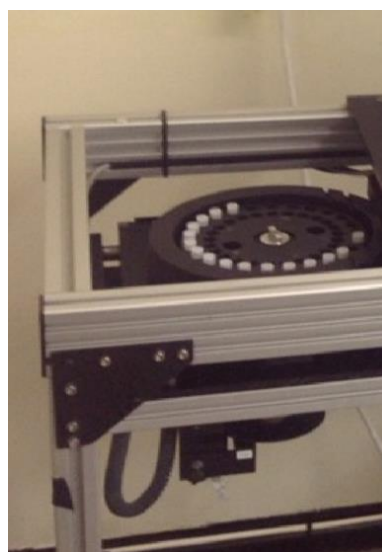


Figure 3.7: Irradiated samples.

3.7. Detector characterization

HPGe-detectors were calibrated for the energy response, peak resolution and counting efficiency using a multi-channel analyser and the spectrophotometry software (Zehringer, 2017). These properties depend on the energy of the incident radiation from $^{241}\text{Am}/^{152}\text{Eu}$ source with different gamma-ray energies (Institute of Metrology, 2018).

Gamma spectroscopy use commercially available calibration sources containing a mixture of gamma-nuclides, which cover the whole energy range (Zehringer, 2017). Gamma-ray detectors are used for detection of natural radiation and their energy properties (Lindsay, 2011). It is better to use a mixture of a low energy nuclide, for example, ^{241}Am with 432.2 years half-life and energy of 59 keV, ^{152}Eu with half-life of 13.5 years which disintegrates slowly with a multiple emission lines from 122 to 1528 keV and a nuclide with a shorter half- life, for example, ^{85}Sr with a half-life of 65 days and energy of 512 keV (Institute of Metrology, 2018). The calibration of detector peak resolution and efficiency were performed with the same calibration sources. Peak resolution and the energy calibration should be tested at the same time on a regular basis (ICRP-60, 2005).

3.7.1. Energy calibration

The detector characterization was performed using Genie 2000 software. Calibration using radiation source in a plastic container of diameter and height of 5.0 x 0.5 mm with the same geometry as those containing the prepared samples was performed. The energy calibration of the HPGe spectrometer system measured the position of known energy gamma-ray peaks (Xhixha, 2012). Gamma-ray energies are allocated to the channel numbers corresponding to the position of the energy peak (Santawamaitre et al., 2010). The gamma-rays cover the energetic range of interest 5–3000 keV from a chosen source. The energy calibration is carried out by using multi-peaked radioactive standard sources emitting gamma rays of known energy to derive channel numbers.

Energy calibration was performed before measuring the sample to eliminate any spectra shift and maintain the quality of the measurements. The aim of energy calibration was to determine the relationship between peak gamma-ray energies and channel number in the MCA spectrum (Faanu et al., 2011). The locations of the channel with known energies on the MCA was determined by counting a standard source, radionuclides which correspond to the gamma-rays of full energy peaks. The calibration curve of the HPGe detector is presented in Figure 3.8.

The machine was calibrated using gamma emitting radionuclides of known energy that allows to determine a linear equation relating gamma energy and number of channels: (Pallavicini, 2011):

$$E = mx + c \quad (3.3)$$

where E is the gamma energy, m and c are constants, and x is the relative channel.

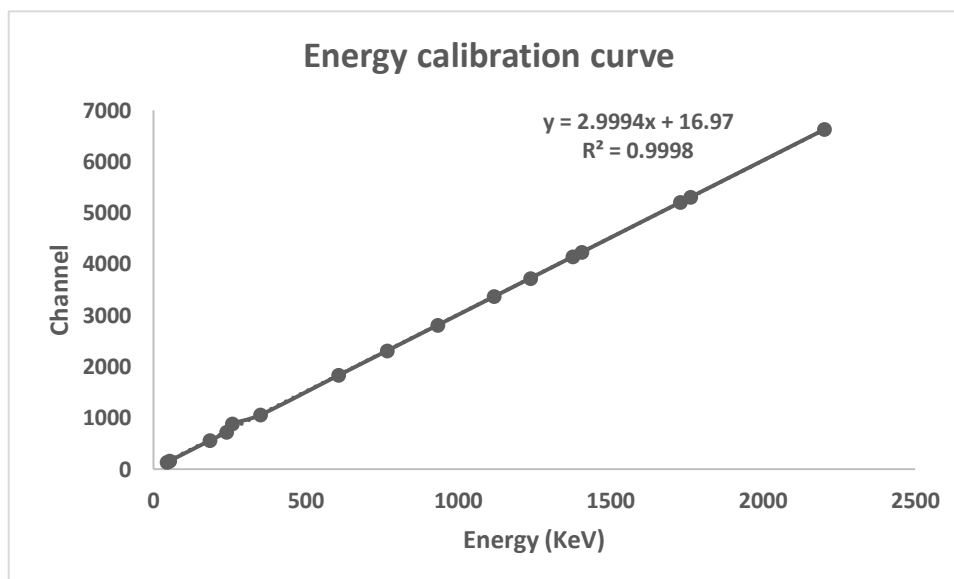


Figure 3.8: Energy calibration curve between gamma energy lines and channel number.

3.7.2. Efficiency calibration

The gamma-ray spectrum of the calibration source (^{152}Eu) was acquired for 600 seconds and the results were used to generate the efficiency of the detector in figure 3.9 below shows the efficiency as function of gamma-ray energy. In order to obtain 10^4 counts, the software was set to count for 100 seconds. The efficiency calibration determines the full energy peak efficiency as a function of energy using two low activity point sources with complex decay schemes (Tsoulfanidis, 2010). The efficiency calibration was obtained by using ^{133}Ba and ^{152}Eu sources. The efficiency was plotted against the energy peak lines, and the peak energy lines were plotted against the generated efficiency values. The efficiency was used to calculate critical energy, minimum detectable activity (MDA) and the activity concentrations of the environmental samples which were dependent on the geometry, density, and composition of the sample (Pallavicini, 2011). The activities of the samples were determined using the total net counts under the selected photo-peaks, the measured photo-peak efficiency, gamma intensity and weight of the samples.

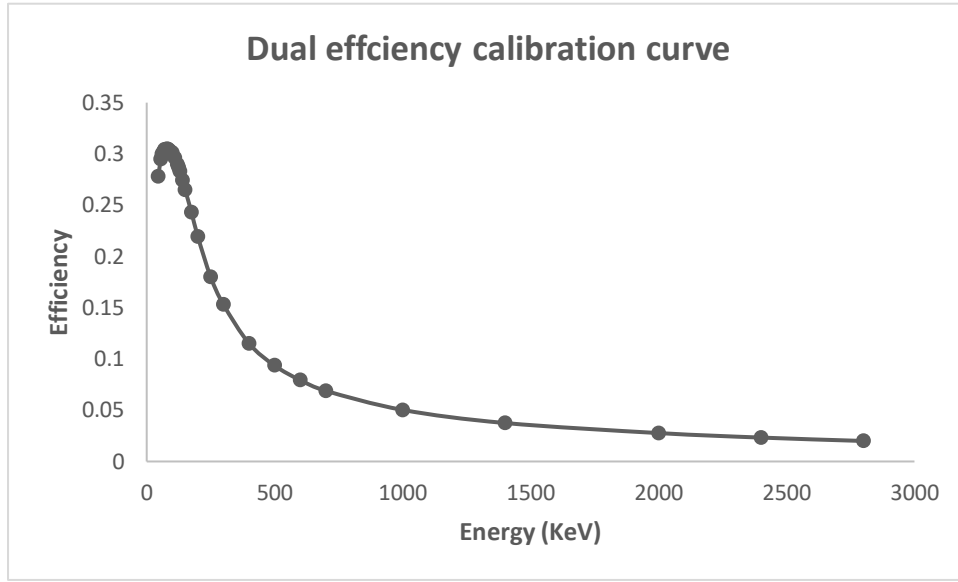


Figure 3.9: Efficiency as function of gamma-ray energy for the HPGe detector.

3.8. Minimum Detectable Activity

Minimum detectable activity (MDA) refers to the smallest net count that can be measured with a certain degree of confidence in gamma spectrometry. The background knowledge of the spectra is required for gamma-ray spectrometry analysis in order to detect any potential residual contaminations and for background corrections (Xhixha, 2012). The determination of the minimum detectable activity of the counting system, were found using the equation:

$$MDA = \frac{L_D}{\varepsilon_f p_\gamma T}, \quad (3.4)$$

where ε_f , p_γ and T are absolute efficiency of the detector, gamma emission probability of specific energy photo peak and live time of spectrum acquisition, respectively. The detection limit L_D , can be found using the following equation:

$$L_D = 2.706 + 4653\sigma_{NB}, \quad (3.5)$$

where σ_{NB} is the standard deviation of the number of counts to determine the background level using an empty container (Thomas, 2013).

3.9. Activity Concentration calculations

The specific activity was calculated after the background was subtracted using the following equation (Al-Sulaiti et al., 2010):

$$A(Bq.kg^{-1}) = \frac{N}{\varepsilon \times I_{\gamma} \times T \times m} \quad (3.6)$$

Such that,

$$N = N_s - N_B \quad (3.7)$$

where;

N - the corrected net peak area of the corresponding full-energy peak.

N_s - the net peak area in the spectrum of the sample.

N_B - the corresponding net peak area in the background spectrum.

A_s - is the activity concentration in $Bq.kg^{-1}$.

ε - the efficiency at peak energy.

I_{γ} - is the gamma-ray emission probability at each energy.

T - is the time for counting (SEC).

m - is the mass of the sample measured in kg .

CHAPTER FOUR: RESULTS AND DISCUSSIONS

4.1. Introduction

Fruits and vegetables collected from Hartbeespoort, Mahikeng and Pretoria local markets were used to determine the activity concentration and to provide a scientific result that would help the regulator to ensure that fruits and vegetables are safe for consumption. For that purpose, agricultural sample types: 4 fruits and 7 vegetables were collected. The agricultural samples were prepared and analysed at the Nuclear Energy Corporation of South Africa (Necsa) RadioAnalysis Laboratory using different techniques. Gross alpha and gross beta Oxford proportional gas-flow series were used to measure the activity concentration in fruits and vegetables from different markets. Neutron activation analysis (NNA) was used to quantify NORM in fruits and vegetables and the gamma spectrometry analysis (HPGe) detector was used to determine the activity concentration of irradiated samples. The results were used to calculate the radiological hazards assessments, Excess Lifetime Cancer Risk (ELCR) and were compared with the international recommended limit values. The statistical analysis of the activity concentrations in fruits and vegetables using a one-way ANOVA stats analysis was carried out to determine the significant difference of the activity concentrations in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria markets. The ANOVA results are presented in appendix A.

4.2. Gross Alpha (α) and Gross Beta (β) Activity Concentrations measurements

Table 4.1 summarises the results in fruits from Hartbeespoort, Mahikeng and Pretoria which are further illustrated in Figures 4.1 and 4.2. Columns 3, 5 and 7 shows gross alpha and columns 4, 6 and 8 show gross beta activity concentrations in fruits from different markets.

Table 4.1: Gross α and gross β activity concentrations in fruits.

Fruits	Gross Alpha and Beta Activity Concentrations (Bq.g ⁻¹)						
	Sample Name	Hartbeespoort		Mahikeng		Pretoria	
		Gross α	Gross β	Gross α	Gross β	Gross α	Gross β
	Oranges	0.753	9.281	0.125	9.566	0.122	6.128
	Apples	0.646	10.482	0.217	8.293	0.220	8.277
	Bananas	0.412	9.630	0.250	8.350	0.275	11.644
	Pineapples	0.294	10.665	0.133	11.871	0.118	11.356
	Mean $\pm$ S.D	0.527 $\pm$ 0.182	10.015 $\pm$ 0.576	0.181 $\pm$ 0.053	9.520 $\pm$ 1.449	0.184 $\pm$ 0.006	9.351 $\pm$ 2.281
WHO (2008) advises 0.1 Bq.g ⁻¹ for gross alpha and 1.0 Bq.g ⁻¹ for gross beta limits for food and drinking water							

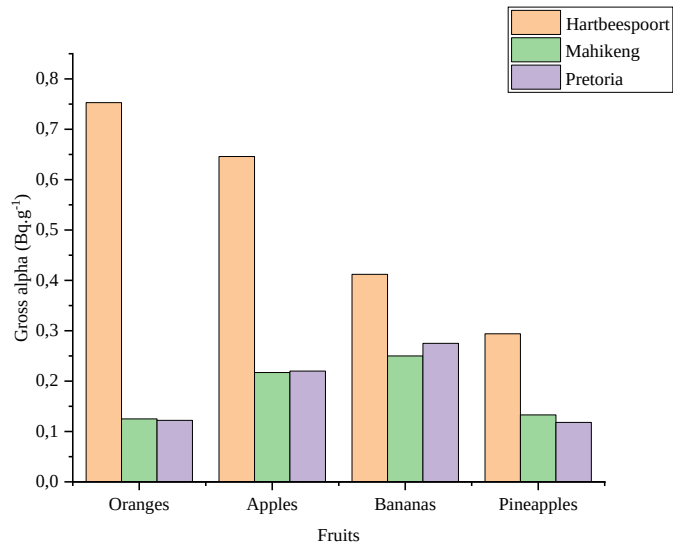


Figure 4.1: Gross α activity in fruits from Hartbeespoort, Mahikeng and Pretoria.

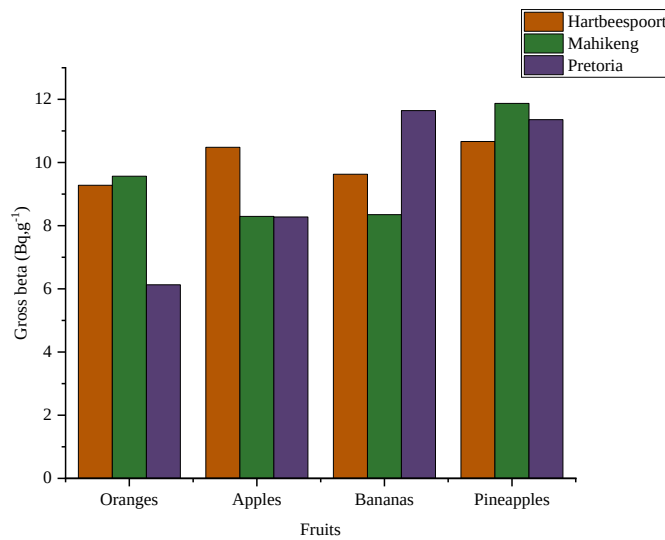


Figure 4.2: Gross β activity in fruits from Hartbeespoort, Mahikeng and Pretoria.

From Table 4.1, results of the samples collected from Hartbeespoort, Mahikeng and Pretoria for gross alpha and gross beta activity concentrations ranged from 0.345 to 0.709 Bq.g⁻¹ and 9.439 to 10.591 Bq.g⁻¹ with mean and standard deviation (Mean $\pm$ S.D) values of 0.527 $\pm$ 0.182 Bq.g⁻¹ and 10.015 $\pm$ 0.576 Bq.g⁻¹ in Haartbeespoort, 0.128 to 0.254 Bq.g⁻¹ and 8.071 to 10.969 Bq.g⁻¹ with mean and standard deviation of (Mean $\pm$ S.D) of 0.181 $\pm$ 0.053 Bq.g⁻¹ and 9.520 $\pm$ 1.449 Bq.g⁻¹ in Mahikeng and gross alpha and gross beta activity concentrations ranged from 0.178 to 0.19 Bq.g⁻¹ and 7.07 to 11.632 Bq.g⁻¹ with mean and standard deviation (Mean $\pm$ S.D) of 0.184 $\pm$ 0.006 Bq.g⁻¹ and 9.351 $\pm$ 2.281 Bq.g⁻¹ in Pretoria, respectively.

The highest gross alpha and gross beta average activity concentrations in fruits were found in Hartbeespoort. From gross alpha and gross beta activity concertations between Hartbeespoort, Mahikeng and Pretoria markets, the highest gross alpha and gross beta

activity concentrations in fruits were found in oranges as shown in Table 4.1, Figure 4.1, and Figure 4.2, followed by Bananas, respectively. As can be seen from Figures 4.1 and 4.2, the highest gross alpha activity concentration was observed in oranges from Hartbeespoort, and the highest gross beta activity concentration were found in pineapples from Mahikeng. The ANOVA analysis for gross alpha in fruits from Hartbeespoort, Mahikeng and Pretoria with p-value of 0.00776 showed there is significant difference ($p < 0.05$). The gross beta analysis in fruit with p-value of 0.87122 showed there is no significant difference ($p > 0.05$).

Table 4.2 provides the results of gross alpha and gross beta activity concentrations measurements in vegetables from Hartbeespoort, Mahikeng and Pretoria. Columns 3, 5 and 7 show gross alpha and columns 4, 6 and 8 show gross beta activity concentrations in vegetables, respectively.

Table 4.2: Gross α and gross β Activity Concentrations (Bq.g^{-1}) in vegetables.

Vegetables	Gross Alpha and Beta Activity Concentrations (Bq.g^{-1})					
	Sample Name	Hartbeespoort		Mahikeng		Pretoria
		Gross α	Gross β	Gross α	Gross β	Gross α
	Potatoes	0.204	11.903	0.363	14.470	0
	Cucumbers	0.255	11.578	0.076	11.939	0
	Tomatoes	0.517	12.602	0.149	13.995	0.482
	Cabbage	0.284	9.238	0.218	10.326	0.341
	Spinach	0.092	6.585	0.285	9.216	0.093
	Carrots	0.165	6.588	0.432	12.067	0.335
	Pumpkin	0.010	12.198	0.028	12.191	0.023
	Mean $\pm$ S.D	0.218 $\pm$ 0.150	10.099 $\pm$ 2.435	0.222 $\pm$ 0.137	12.029 $\pm$ 1.718	0.254 $\pm$ 0.191
	WHO (2008) advises 0.1 Bq.g^{-1} for gross alpha and 1.0 Bq.g^{-1} for gross beta limits for drinking water					

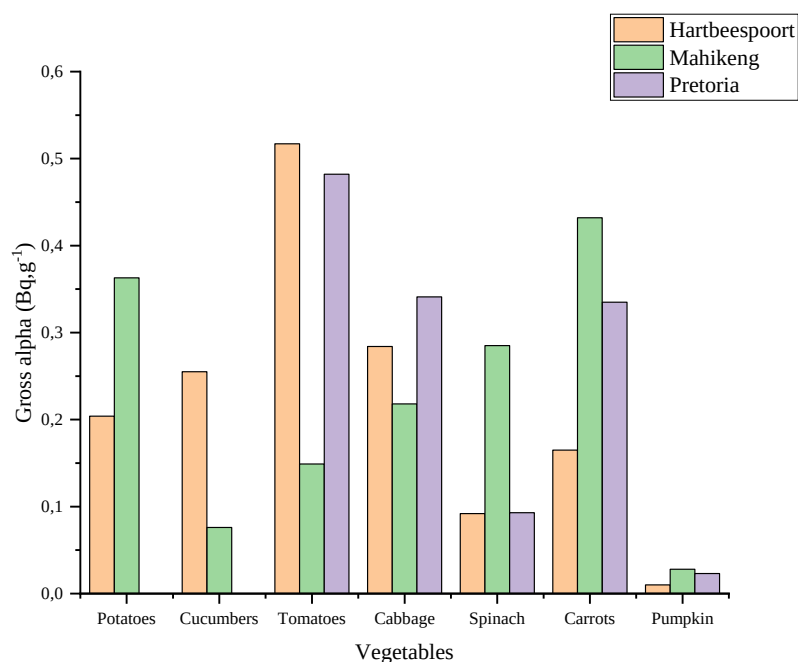


Figure 4.3: Gross α activity in vegetables from Hartbeespoort, Mahikeng and Pretoria.

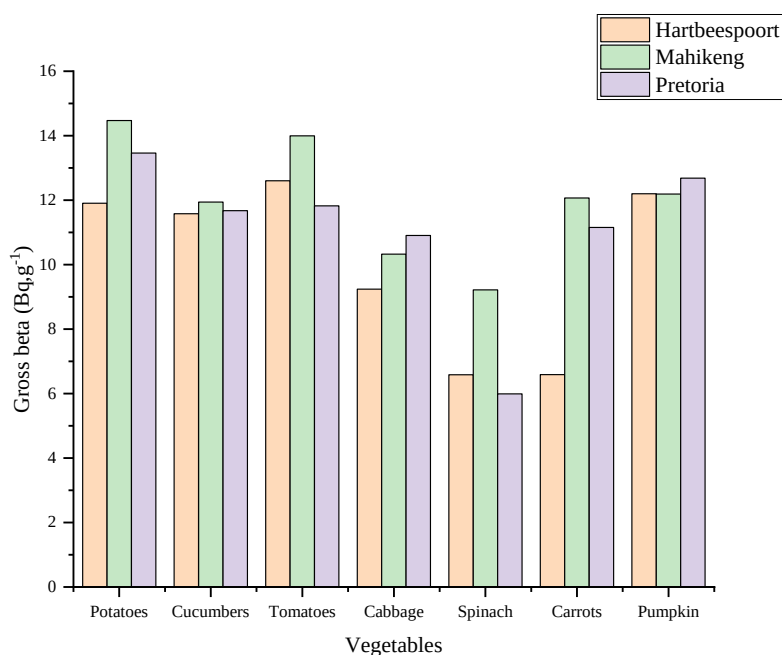


Figure 4.4: Gross β activity in vegetables from Hartbeespoort, Mahikeng and Pretoria.

The gross alpha and gross beta activity concentrations from Hartbeespoort, Mahikeng and Pretoria varied from 0.203 to 0.233 Bq.g⁻¹ and 7.664 to 12.534 Bq.g⁻¹ with the mean and standard deviations (Mean $\pm$ S.D) values of 0.218 ± 0.150 and 10.099 ± 2.435 Bq.g⁻¹, 0.085 to 0.359 Bq.g⁻¹ and 10.311 to 13.747 Bq.g⁻¹ and the mean and standard deviations (Mean $\pm$ S.D) values were 0.222 ± 0.137 and 12.029 ± 1.718 Bq.g⁻¹ and 0.0625 to 0.4455 and 8.861 to 13.337 Bq.g⁻¹ with mean and standard deviation (Mean $\pm$ S.D) of 0.254 ± 0.191 Bq.g⁻¹ and 11.099 ± 2.238 Bq.g⁻¹, respectively. In Table 4.2 gross alpha activity concentrations in vegetables was much lower than the gross beta activity concentrations.

Figure 4.3 shows the highest gross alpha concentration in tomato vegetables from Hartbeespoort. Figure 4.4 shows the highest gross beta concentration in potatoes from Mahikeng. The gross alpha and gross beta results from Mahikeng, Hartbeespoort and Pretoria were higher compared to the study results of (Van et al., 2018) from the city of Ho Chi Minh, Vietnam in food crops and surface soil with a gross alpha activity concentrations of 0.02375 ± 0.00061 Bq.g⁻¹ and Niger Delta Region of Nigeria; 0.303 ± 0.039 Bq.g⁻¹ to 0.482 ± 0.041 Bq.g⁻¹ (Alao, 2016). The gross alpha and gross beta results of the current study were 0.254 ± 0.004 Bq.g⁻¹ and 0.474 ± 0.054 Bq.g⁻¹, respectively and were higher than the (WHO, 2008) references of 0.1 Bq.g⁻¹ and 1.0 Bq.g⁻¹, respectively. The highest gross alpha and gross beta activity concentrations in the current study for both fruits and vegetables might be due to the type of soil, irrigation water, fertiliser application, transportation and manmade radioactivity in the environment (Alao, 2016). The gross alpha and gross beta in vegetables p-values of 0.89223 and 0.32217 which showed no significant

difference in Mahikeng, Pretoria and Hartbeespoort ($p > 0.05$). The gross alpha and beta analysis showed there is no significant difference ($p > 0.05$) in cucumbers from Pretoria and Hartbeespoort, as well as in pumpkin from Mahikeng and Hartbeespoort.

4.3. Neutron Activation Analysis and Radionuclides activity concentrations measurements

Tables 4.3 and 4.4 show the neutron activation results using the (HPGe) coaxial detector to measure the activity concentration of radionuclides ^{238}U , ^{234}U , ^{235}U and ^{232}Th for the irradiated fruits and vegetable samples from Hartbeespoort.

Table 4.3: Activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th (Bq.kg^{-1}) in fruits from Hartbeespoort.

Fruits	Hartbeespoort				
	Sample Name	Activity concentrations (Bq.kg^{-1})			
		^{238}U	^{234}U	^{235}U	^{232}Th
	Oranges	0.573	0.578	0.026	3.800
	Apples	1.370	1.380	0.063	2.700
	Bananas	0.270	0.270	0.023	2.100
	Pineapples	1.850	1.870	0.085	2.200
	Mean $\pm$ S.D	1.015 $\pm$ 0.627	1.024 $\pm$ 0.634	0.049 $\pm$ 0.025	2.700 $\pm$ 0.674
	UNSCEAR (2000)	35 Bq.kg^{-1}	35 Bq.kg^{-1}	-	30 Bq.kg^{-1}

Table 4.4: Activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th (Bq.kg^{-1}) in vegetables from Hartbeespoort.

Vegetables	Hartbeespoort				
	Sample Name	Activity concentrations (Bq.kg^{-1})			
		^{238}U	^{234}U	^{235}U	^{232}Th
	Potatoes	3.790	3.820	0.175	2.000
	Cucumbers	0.742	0.748	0.034	3.800
	Tomatoes	0.910	0.918	0.042	1.800
	Cabbage	0.885	0.893	0.041	3.100
	Spinach	2.090	2.100	0.096	1.700
	Carrots	0.390	0.390	0.018	2.500
	Pumpkin	0.627	0.632	0.029	3.800
	Mean $\pm$ S.D	1.348 $\pm$ 1.116	1.357 $\pm$ 1.125	0.062 $\pm$ 0.051	2.671 $\pm$ 0.838
	UNSCEAR (2000)	35 Bq.kg^{-1}	35 Bq.kg^{-1}	-	30 Bq.kg^{-1}

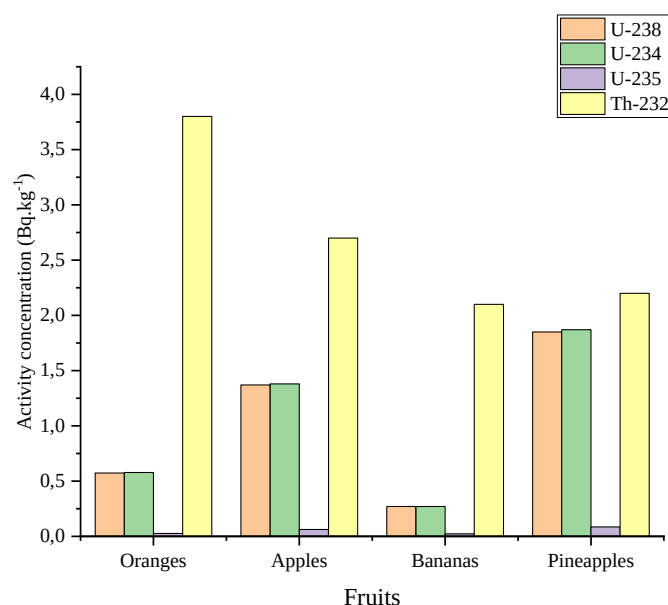


Figure 4.5: Activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits from Hartbeespoort.

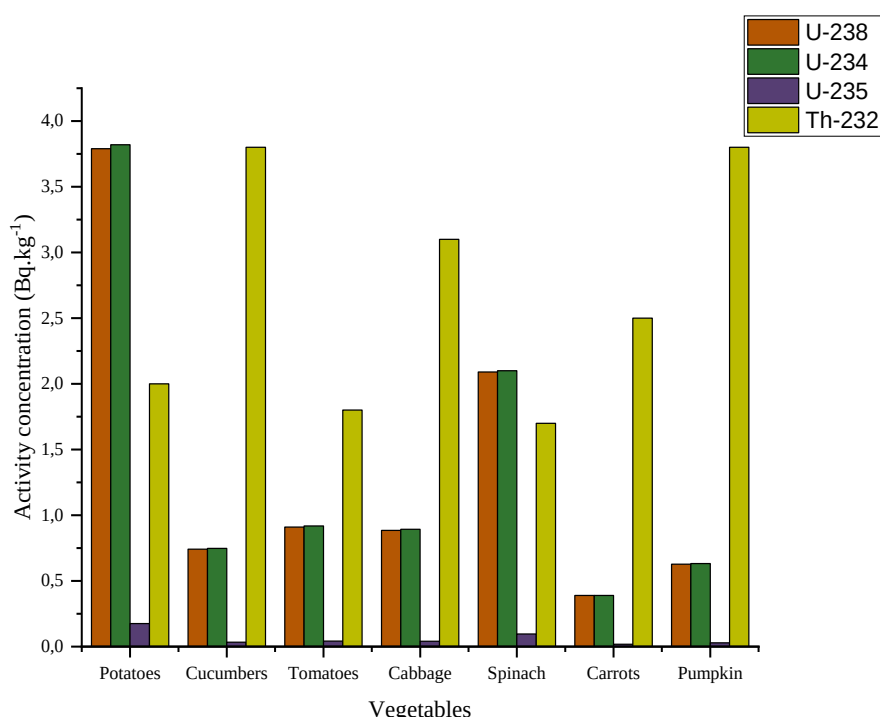


Figure 4.6: Activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in vegetables from Hartbeespoort.

The activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in Table 4.3 in fruits ranged from 0.4245 to 1.6785 Bq.kg⁻¹, 0.3900 to 1.658 Bq.kg⁻¹, 0.029 to 0.069 Bq.kg⁻¹ and 2.2.026 to 3.374 Bq.kg⁻¹, respectively. The mean and standard deviation (Mean $\pm$ S.D) values of the radionuclides ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits were 1.015 ± 0.627 Bq.kg⁻¹, 1.024 ± 0.634 Bq.kg⁻¹, 0.049 ± 0.025 Bq.kg⁻¹ and 2.700 ± 0.674 Bq.kg⁻¹, respectively. The activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in Table 4.4 in vegetables ranged from 0.232 to

2.482 Bq.kg⁻¹, 0.232 to 2.482 Bq.kg⁻¹, 0.011 to 0.113 Bq.kg⁻¹ and 1.871 to 3.471 Bq.kg⁻¹, respectively. The mean and standard deviation (Mean $\pm$ S.D) values of the radionuclides ²³⁸U, ²³⁴U, ²³⁵U and ²³²Th in vegetables were 1.348 $\pm$ 1.116 Bq.kg⁻¹, 1.357 $\pm$ 1.125 Bq.kg⁻¹, 0.062 $\pm$ 0.051 Bq.kg⁻¹ and 2.671 $\pm$ 0.838 Bq.kg⁻¹ respectively. Figures 4.5 and 4.6 show the activity concentrations of ²³⁸U, ²³⁴U, ²³⁵U and ²³²Th in fruits and vegetables from Hartbeespoort.

The NORM with the highest activity concentrations in fruits was ²³²Th measured in oranges and the lowest was ²³⁵U found in bananas (Figure 4.5). The NORM with the highest activity concentrations in vegetables was ²³⁸U and ²³⁴U in potatoes and ²³²Th in cabbages, cucumbers and pumpkins (Figure 4.6). The NORM with the lowest activity concentrations in vegetables was ²³⁵U, in all vegetables, followed by ²³⁴U and ²³⁵U in carrots (Figure 4.6). The activity concentrations of ²³⁸U, ²³⁴U, ²³⁵U and ²³²Th in fruits and vegetables with p-values of 0.00073 and 0.00052 showed that there is a significant difference (p < 0.05) in Hartbeespoort. The current study results in fruits from Hartbeespoort with activity concentrations of ²³⁸U, ²³⁴U, ²³⁵U and ²³²Th were 1.015 $\pm$ 0.627 Bq.kg⁻¹, 1.024 $\pm$ 0.634 Bq.kg⁻¹, 0.049 $\pm$ 0.025 Bq.kg⁻¹ and 2.700 $\pm$ 0.674 Bq.kg⁻¹, respectively and the results in vegetables from Hartbeespoort of ²³⁸U, ²³⁴U, ²³⁵U and ²³²Th were 1.348 $\pm$ 1.116 Bq.kg⁻¹, 1.357 $\pm$ 1.125 Bq.kg⁻¹, 0.062 $\pm$ 0.510 Bq.kg⁻¹ and 2.671 $\pm$ 0.800 Bq.kg⁻¹, respectively. The activity concentrations in fruits and vegetables were lower than the world mean standard of 35 Bq.kg⁻¹, 35 Bq.kg⁻¹, respectively (UNSCEAR, 2000).

Table 4.5: Gross α - β activity concentrations (Bq.g⁻¹) and activity concentrations of ²³⁸U, ²³⁴U, ²³⁵U and ²³²Th (Bq.kg⁻¹) in fruits from Hartbeespoort.

	Hartbeespoort									
	Sample Name	Activity Concentrations (Bq.g ⁻¹)		Activity Concentrations (Bq.kg ⁻¹)			Uranium Isotopic ratios			
		α	β	²³⁸ U	²³⁴ U	²³⁵ U	²³² Th	²³⁴ U/ ²³⁸ U	²³⁵ U/ ²³⁸ U	²³² Th/ ²³⁸ U
	Fruits									
	Oranges	0.754	9.281	0.573	0.578	0.026	3.800	1.009	0.046	6.637
	Apples	0.647	10.482	1.370	1.380	0.063	2.700	1.007	0.046	1.971
	Bananas	0.413	9.631	0.270	0.270	0.023	2.100	1.000	0.085	7.778
	Pineapples	0.295	10.665	1.850	1.870	0.085	2.200	1.011	0.046	1.189
	Mean $\pm$ S.D	0.527 $\pm$ 0.182	10.015 $\pm$ 0.576	1.015 $\pm$ 0.627	1.024 $\pm$ 0.634	0.049 $\pm$ 0.025	2.700 $\pm$ 0.674	1.01 $\pm$ 0.001	0.06 $\pm$ 0.02	4.39 $\pm$ 2.86
	World mean values	0.1 Bq.g ⁻¹	1.0 Bq.g ⁻¹	35 Bq.kg ⁻¹	35 Bq.kg ⁻¹	-	30 Bq.kg ⁻¹			
		WHO (2008)		UNSCEAR (2000)	UNSCEAR (2000)	-	UNSCEAR (2000)			

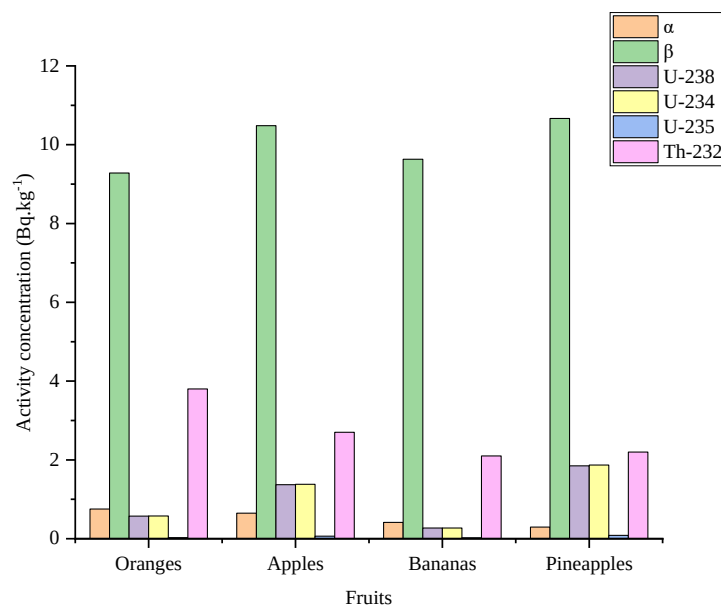


Figure 4.7: Comparison between gross α - β and activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits from Hartbeespoort.

Table 4.6: Gross α - β activity concentrations (Bq.g^{-1}) and activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th (Bq.kg^{-1}) in vegetables from Hartbeespoort.

Vegetables	Hartbeespoort									
	Activity concentrations						Uranium Isotopic ratios			
		Bq.g ⁻¹		(Bq.kg ⁻¹)						
	Sample Name	α	β	²³⁸ U	²³⁴ U	²³⁵ U	²³² Th	²³⁴ U/ ²³⁸ U	²³⁵ U/ ²³⁸ U	²³² Th/ ²³⁸ U
	Potatoes	0.205	11.903	3.790	3.820	0.175	2.000	1.008	0.046	0.528
	Cucumbers	0.255	11.579	0.742	0.748	0.003	3.800	1.008	0.046	5.121
	Tomatoes	0.518	12.603	0.910	0.918	0.042	1.800	1.009	0.046	1.978
	Cabbage	0.284	9.239	0.885	0.893	0.041	3.100	1.009	0.046	3.503
	Spinach	0.093	6.586	2.090	2.100	0.096	1.700	1.005	0.046	0.813
	Carrots	0.165	6.588	0.390	0.390	0.018	2.500	1.000	0.046	6.410
	Pumpkin	0.010	12.199	0.627	0.632	0.029	3.800	1.01±0.000	0.05±0.00	3.49±2.270
	Mean ± S.D	0.219±0.150	10.099±2.436	1.348±1.116	1.357±1.125	0.062±0.051	2.671±0.838			
	World mean values	0.1 Bq.g ⁻¹	1.0 Bq.g ⁻¹	35 Bq.kg ⁻¹	35 Bq.kg ⁻¹	-	30 Bq.kg ⁻¹			
		WHO (2008)		UNSCEAR (2000)	UNSCEAR (2000)		UNSCEAR (2000)			

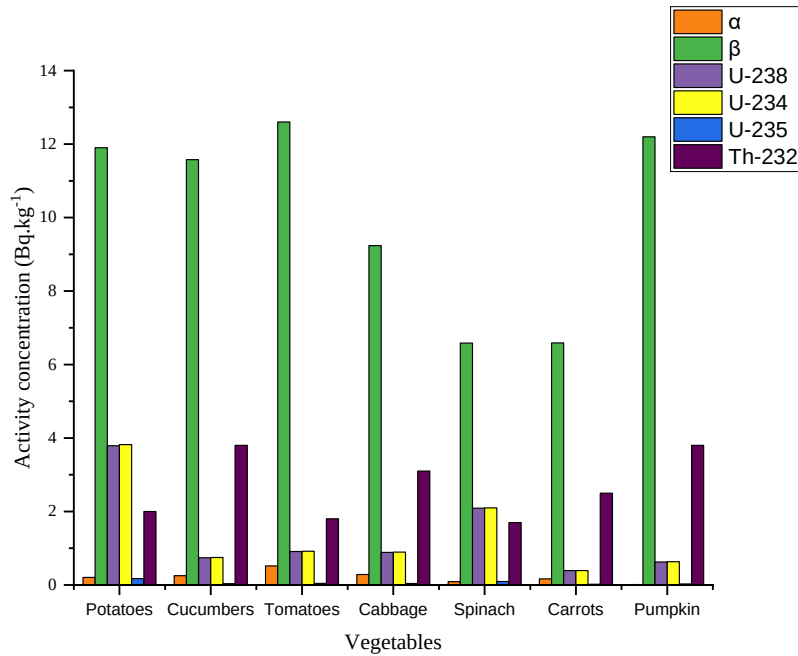


Figure 4.8: Comparison between gross α - β and activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in vegetables from Hartbeespoort.

Tables 4.5 and 4.6 show the results of gross α - β activity concentrations and specific radionuclides activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits and vegetables from Hartbeespoort which are further illustrated in Figures 4.7 and 4.8. The ratio of $^{234}\text{U}/^{238}\text{U}$, $^{235}\text{U}/^{238}\text{U}$ and $^{232}\text{Th}/^{238}\text{U}$ in fruits were 0.01 ± 0.00 , 0.06 ± 0.02 and 4.39 ± 2.86 , respectively. The ratios of $^{234}\text{U}/^{238}\text{U}$, $^{235}\text{U}/^{238}\text{U}$ and $^{232}\text{Th}/^{238}\text{U}$ in vegetables were 1.01 ± 0.00 , 0.05 ± 0.00 and 3.49 ± 2.27 , respectively, which are in good agreement with the natural ratio.

Table 4.7: Activity Concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th (Bq.kg^{-1}) in fruits from Mahikeng.

	Mahikeng				
	Sample Name	Activity concentrations (Bq.kg^{-1})			
		^{238}U	^{234}U	^{235}U	^{232}Th
Fruits	Oranges	0.270	0.270	0.013	3.500
	Apples	2.220	0.230	0.010	2.300
	Bananas	0.220	0.230	0.010	2.000
	Pineapples	0.120	0.120	0.005	2.700
	Mean $\pm$ S.D	0.207 ± 0.054	0.121 ± 0.05	0.009 ± 0.002	2.625 ± 0.563
	UNSCEAR (2000)	35 Bq.kg^{-1}	35 Bq.kg^{-1}	-	30 Bq.kg^{-1}

Table 4.8: Activity Concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th (Bq.kg^{-1}) in vegetables from Mahikeng.

Vegetables	Mahikeng				
	Sample Name	Activity concentrations (Bq.kg ⁻¹)			
		²³⁸ U	²³⁴ U	²³⁵ U	²³² Th
	Potatoes	0.701	0.700	0.032	6.900
	Cucumbers	2.360	2.380	0.109	4.900
	Tomatoes	0.440	0.440	0.020	4.000
	Cabbage	4.000	4.030	0.184	4.300
	Spinach	3.220	3.250	0.148	2.500
	Carrots	0.190	0.190	0.008	3.800
	Pumpkin	0.720	0.730	0.033	3.200
	Mean ± S.D	1.662±1.406	1.674±1.419	0.076±0.064	4.228±1.302
	UNSCEAR (2000)	35 Bq.kg ⁻¹	35 Bq.kg ⁻¹	-	30 Bq.kg ⁻¹

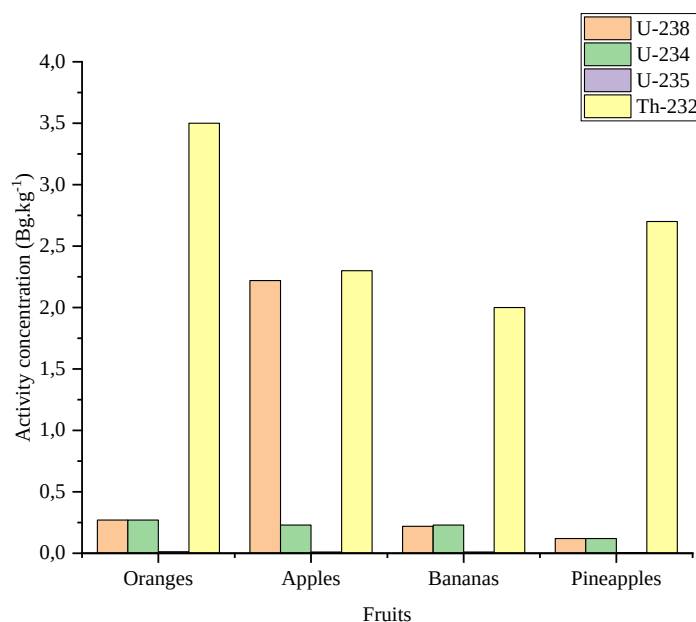


Figure 4.9: Activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits from Mahikeng.

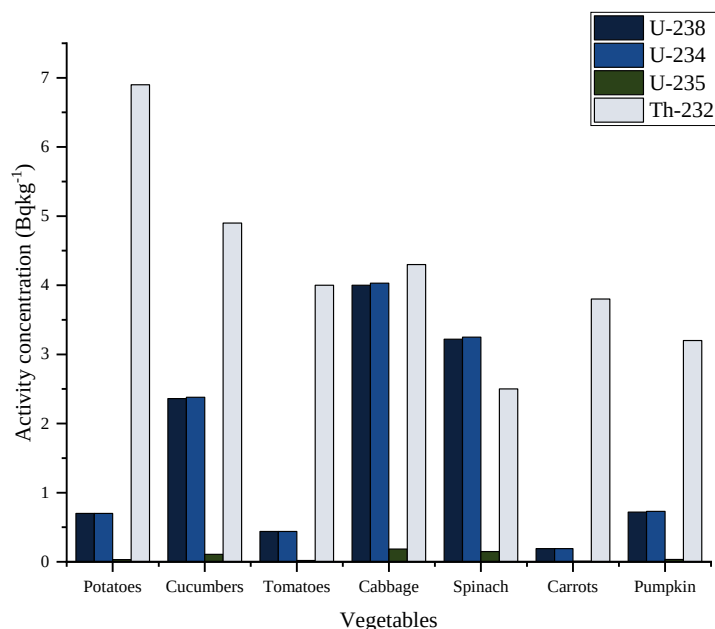


Figure 4.10: Activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in vegetables from Mahikeng.

Tables 4.7 and 4.8 show the measured activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits and vegetables and are illustrated in Figures 4.9 and 4.10, respectively. The activity concentrations of radionuclides ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits in Table 4.7 ranged from 0.153 to 0.261 Bq.kg⁻¹, 0.071 to 0.171 Bq.kg⁻¹, 0.088 to 0.092 Bq.kg⁻¹ and 1.972 to 3.278 Bq.kg⁻¹, respectively. The mean and standard deviations (Mean $\pm$ S.D) values of the radionuclides ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits were 0.207 ± 0.054 Bq.kg⁻¹, 0.121 ± 0.05 Bq.kg⁻¹, 0.009 ± 0.002 Bq.kg⁻¹ and 2.625 ± 0.563 Bq.kg⁻¹, respectively.

The activity concentrations of ^{232}Th was higher in all the fruits, but highest in oranges. ^{238}U was found to be high in apples while ^{234}U and ^{235}U were lowest in all the fruits see, figure 4.9. The activity concentrations of radionuclides ^{238}U , ^{234}U , ^{235}U and ^{232}Th in vegetables ranged from 0.256 to 3.068 Bq.kg⁻¹, 0.255 to 3.093 Bq.kg⁻¹, 0.0128 to 0.140 Bq.kg⁻¹ and 2.926 to 5.520 Bq.kg⁻¹, respectively. The mean and standard deviation (Mean $\pm$ S.D) values of radionuclides ^{238}U , ^{234}U , ^{235}U and ^{232}Th in vegetables were 1.662 ± 1.406 Bq.kg⁻¹, 1.674 ± 1.419 Bq.kg⁻¹, 0.076 ± 0.064 Bq.kg⁻¹ and 4.228 ± 1.302 Bq.kg⁻¹, respectively. Thorium-232 (^{232}Th) was high in all the vegetables, see Figure 4.9, with the highest concentration found in potatoes. Cabbage has the highest activity concentrations of ^{234}U and ^{238}U . The activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits and vegetables with p-values of 0.00018 and 0.00004 showed there is a significant difference ($p < 0.05$) from Mahikeng.

Table 4.9: Gross α - β activity concentrations (Bq.g⁻¹) and activity concentrations of ²³⁸U, ²³⁴U, ²³⁵U and ²³²Th (Bq.kg⁻¹) in fruits from Mahikeng.

Fruits	Mahikeng									
		Activity Concentrations (Bq.g ⁻¹)		Activity concentrations (Bq.kg ⁻¹)				Uranium isotopic ratios		
		α	β	²³⁸ U	²³⁴ U	²³⁵ U	²³² Th	²³⁴ U/ ²³⁸ U	²³⁵ U/ ²³⁸ U	²³² Th/ ²³⁸ U
	Oranges	0.125	9.566	0.270	0.270	0.013	3.500	1.000	0.048	12.963
	Apples	0.217	8.293	2.220	0.230	0.010	2.300	1.045	0.045	10.454
	Bananas	0.250	8.350	0.220	0.230	0.010	2.000	1.045	0.045	9.091
	Pineapples	0.133	11.871	0.120	0.120	0.005	2.700	1.000	0.046	22.500
	Mean ± S.D	0.181±0.053	9.520±1.449	0.207±0.054	0.121±0.05	0.009±0.002	2.625±0.563	1.02±0.02	0.05±0.00	13.75±5.25
	World mean values	0.1 Bq.g ⁻¹	1.0 Bq.g ⁻¹	35 Bq.kg ⁻¹	35 Bq.kg ⁻¹	-	30 Bq.kg ⁻¹			
	WHO (2008)		UNSCEAR (2000)	UNSCEAR (2000)	-	UNSCEAR (2000)				

Table 4.10: Gross α - β activity concentrations (Bq.g⁻¹) and activity concentrations of ²³⁸U, ²³⁴U, ²³⁵U and ²³²Th (Bq.kg⁻¹) in vegetables from Mahikeng.

Vegetables	Mahikeng											
		Activity Concentrations (Bq.g ⁻¹)		Activity concentrations (Bq.kg ⁻¹)							Uranium isotopic ratios	
		α	β	²³⁸ U	²³⁴ U	²³⁵ U	²³² Th	²³⁴ U/ ²³⁸ U	²³⁵ U/ ²³⁸ U	²³² Th/ ²³⁸ U		
	Potatoes	0.363	14.470	0.701	0.700	0.032	6.900	0.999	0.046	9.843		
	Cucumbers	0.076	11.939	2.360	2.380	0.109	4.900	1.008	0.046	2.076		
	Tomatoes	0.149	13.995	0.440	0.440	0.020	4.000	1.000	0.045	9.091		
	Cabbage	0.218	10.326	4.000	4.030	0.184	4.300	1.008	0.046	1.075		
	Spinach	0.285	9.216	3.220	3.250	0.148	2.500	1.009	0.046	0.776		
	Carrots	0.432	12.067	0.190	0.190	0.008	3.800	1.000	0.046	20.000		
	Pumpkin	0.028	12.191	0.720	0.730	0.033	3.200	1.008	0.045	4.419		
	Mean ± S.D	0.222±0.137	12.029±1.718	1.662±1.406	1.674±1.419	0.076±0.064	4.228±1.302	1.00±0.00	0.05±0.00	6.75±6.39		
	World mean values	0.1 Bq.g ⁻¹	1.0 Bq.g ⁻¹	35 Bq.kg ⁻¹	35 Bq.kg ⁻¹	-	30 Bq.kg ⁻¹					
	WHO (2008)		UNSCEAR (2000)	UNSCEAR (2000)	-	UNSCEAR (2000)						

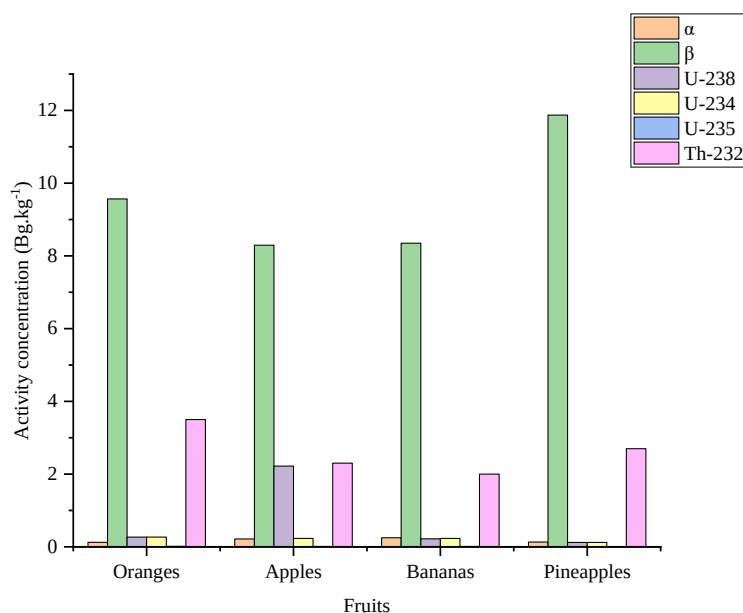


Figure 4.11: Comparison between gross α - β and activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits from Mahikeng.

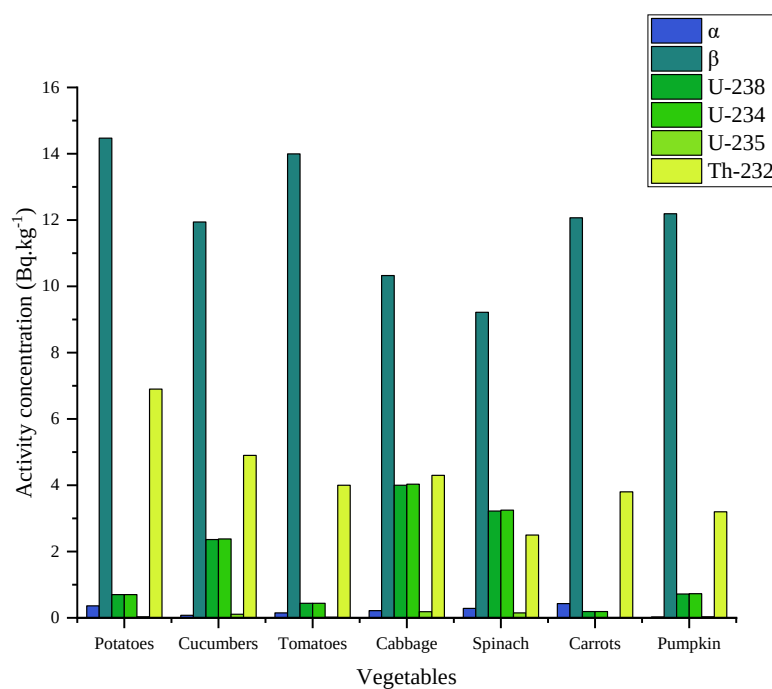


Figure 4.12: Comparison between gross α - β and activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in vegetables from Mahikeng.

Tables 4.9 and 4.10 illustrated in Figures 4.11 and 4.12 show gross α - β activity concentrations and specific radionuclides activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits and vegetables from Mahikeng.

The highest gross beta activity concentrations was observed in fruits and vegetables. The lowest gross beta was from ^{235}U . The ratio of $^{234}\text{U}/^{238}\text{U}$, $^{235}\text{U}/^{238}\text{U}$ and $^{232}\text{Th}/^{238}\text{U}$ in fruits were 1.02 ± 0.02 , 0.05 ± 0.00 and 13.75 ± 5.24 , respectively. The ratio of $^{234}\text{U}/^{238}\text{U}$,

$^{235}\text{U}/^{238}\text{U}$ and $^{232}\text{Th}-^{238}\text{U}$ in vegetables were 1.00 ± 0.00 , 0.05 ± 0.00 and 6.75 ± 6.39 , respectively, which are comparable and in good agreement with the natural ratio.

Table 4.11: Activity Concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th (Bq.kg^{-1}) in fruits from Pretoria.

Fruits	Pretoria			
	Sample Name	Activity concentrations (Bq.kg^{-1})		
		^{238}U	^{234}U	^{235}U
				^{232}Th
	Oranges	0.320	0.320	0.015
	Apples	0.260	0.260	0.012
	Bananas	1.430	1.440	0.066
	Pineapples	0.619	0.624	0.029
	Mean $\pm$ S.D	0.657 ± 0.466	0.661 ± 0.470	0.030 ± 0.021
	UNSCEAR (2000)	35 Bq.kg^{-1}	35 Bq.kg^{-1}	-

Table 4.12: Activity Concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th (Bq.kg^{-1}) in vegetables from Pretoria.

Vegetables	Pretoria			
	Sample Name	Activity concentrations (Bq.kg^{-1})		
		^{238}U	^{234}U	^{235}U
				^{232}Th
	Potatoes	0.350	0.350	0.016
	Cucumbers	0.799	0.805	0.037
	Tomatoes	0.898	0.905	0.041
	Cabbage	0.568	0.573	0.026
	Spinach	3.210	3.240	0.148
	Carrots	0.595	0.600	0.027
	Pumpkin	1.020	1.030	0.047
	Mean $\pm$ S.D	1.060 ± 0.902	1.072 ± 0.909	0.049 ± 0.041
	UNSCEAR (2000)	35 Bq.kg^{-1}	35 Bq.kg^{-1}	-

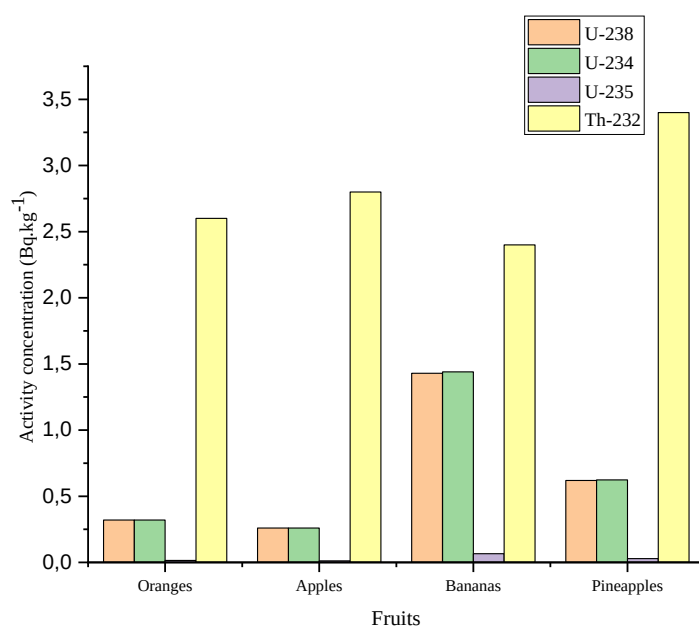


Figure 4.13: Activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits from Pretoria.

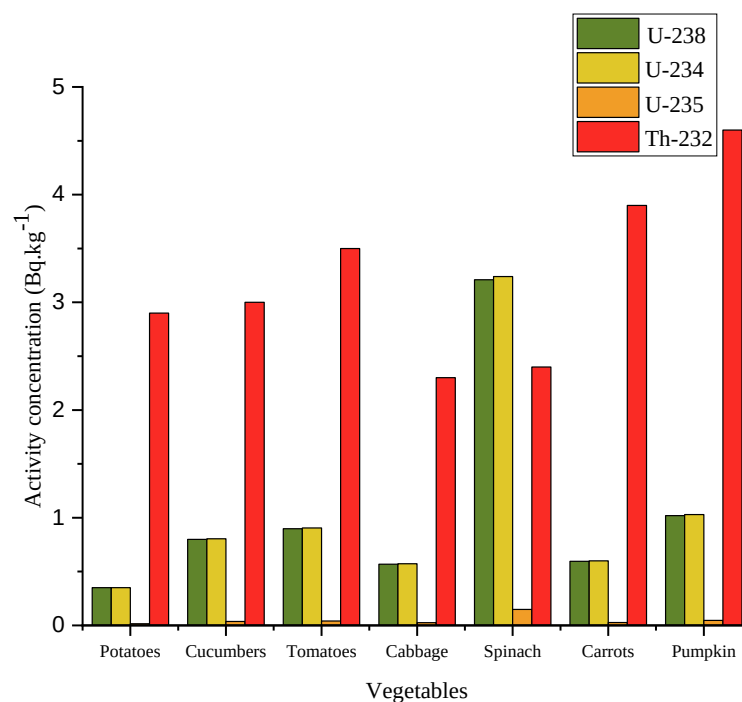


Figure 4.14: Activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in vegetables from Pretoria.

Tables 4.11 and 4.12 illustrated in figures 4.13 and 4.14 show the activity concentrations of radionuclides ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits and vegetables from Pretoria, respectively. The activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits ranged from 0.191 to 1.123 Bq.kg⁻¹, 0.191 to 1.131 Bq.kg⁻¹, 0.009 to 0.051 Bq.kg⁻¹ and 2.426 to 3.174 Bq.kg⁻¹, respectively. The mean and standard deviations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th were 0.657 ± 0.466 Bq.kg⁻¹, 0.661 ± 0.470 Bq.kg⁻¹ and 0.030 ± 0.021 Bq.kg⁻¹ and 2.800 ± 0.374 Bq.kg⁻¹, respectively. The activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in vegetables ranged from 0.158 to 1.063 Bq.kg⁻¹, 0.163 to 1981 Bq.kg⁻¹, 0.008 to 0.090 Bq.kg⁻¹ and 2.461 to 3.9950 Bq.kg⁻¹, respectively. The mean and standard deviation of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in vegetables were found to be 1.060 ± 0.902 Bq.kg⁻¹, 1.072 ± 0.909 Bq.kg⁻¹, 0.049 ± 0.041 Bq.kg⁻¹ and 3.228 ± 0.767 Bq.kg⁻¹, respectively.

Table 4.13: Gross α - β activity concentrations (Bq.g⁻¹) and activity concentrations of ²³⁸U, ²³⁴U, ²³⁵U and ²³²Th (Bq.kg⁻¹) in fruits from Pretoria.

	Pretoria									
		Activity concentrations(Bq.g ⁻¹)		Activity concentrations (Bq.kg ⁻¹)			Uranium isotopic ratios			
		α	β	²³⁸ U	²³⁴ U	²³⁵ U	²³² Th	²³⁴ U/ ²³⁸ U	²³⁵ U/ ²³⁸ U	²³² Th/ ²³⁸ U
	Fruits									
	Oranges	0.112	6.129	0.320	0.320	0.015	2.600	1.000	0.047	8.125
	Apples	0.221	8.278	0.260	0.260	0.012	2.800	1.000	0.046	10.769
	Bananas	0.275	11.644	1.430	1.440	0.066	2.400	1.007	0.046	1.678
	Pineapples	0.119	11.356	0.619	0.624	0.029	3.400	1.008	0.046	5.493
	Mean $\pm$ S.D	0.184 $\pm$ 0.006	9.351 $\pm$ 2.281	0.657 $\pm$ 0.466	0.0661 $\pm$ 0.470	0.030 $\pm$ 0.021	2.800 $\pm$ 0.374	1.00 $\pm$ 0.00	0.05 $\pm$ 0.00	6.52 $\pm$ 3.36
	World mean values	0.1 Bq.g ⁻¹	1.0 Bq.g ⁻¹	35 Bq.kg ⁻¹	35 Bq.kg ⁻¹	-	30 Bq.kg ⁻¹			
		WHO (2008)		UNSCEAR (2000)	UNSCEAR (2000)	-	UNSCEAR (2000)			

Table 4.14: Gross α - β activity concentrations (Bq.g⁻¹) and activity concentrations of ²³⁸U, ²³⁴U, ²³⁵U and ²³²Th (Bq.kg⁻¹) in vegetables from Pretoria.

	Pretoria									
	Sample Name	Activity concentrations (Bq.g ⁻¹)		Activity concentrations (Bq.kg ⁻¹)			Uranium isotopic ratios			
		α	β	²³⁸ U	²³⁴ U	²³⁵ U	²³² Th	²³⁴ U/ ²³⁸ U	²³⁵ U/ ²³⁸ U	²³² Th/ ²³⁸ U
	Potatoes	0	13.463	0.350	0.350	0.016	2.900	1.000	0.045	8.286
	Cucumbers	0	11.674	0.799	0.805	0.037	3.000	1.003	0.047	3.851
	Tomatoes	0.482	11.824	0.898	0.905	0.041	3.500	1.008	0.046	3.897
	Cabbage	0.341	10.907	0.568	0.573	0.026	2.300	1.009	0.046	4.049
	Spinach	0.093	5.993	3.210	3.240	0.148	2.400	1.009	0.046	0.748
	Carrots	0.335	11.156	0.595	0.600	0.027	3.900	1.008	0.046	6.555
	Pumpkin	0.023	12.682	1.020	1.030	0.047	4.600	1.010	0.046	4.509
	Mean $\pm$ S.D	0.175 $\pm$ 0.191	11.099 $\pm$ 2.238	1.060 $\pm$ 0.902	1.072 $\pm$ 0.909	0.049 $\pm$ 0.041	3.228 $\pm$ 0.767	1.01 $\pm$ 0.00	0.05 $\pm$ 0.00	4.56 $\pm$ 2.19
	World mean values	0.1 Bq.g ⁻¹	1.0 Bq.g ⁻¹	35 Bq.kg ⁻¹	35 Bq.kg ⁻¹	-	30 Bq.kg ⁻¹			
		WHO (2008)		UNSCEAR (2000)	UNSCEAR (2000)	-	UNSCEAR (2000)			

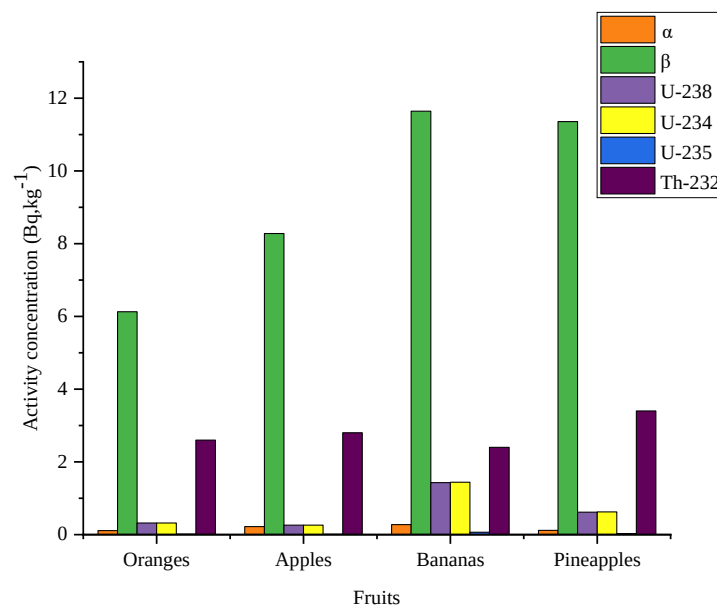


Figure 4.15: Comparison between gross α - β and activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits from Pretoria.

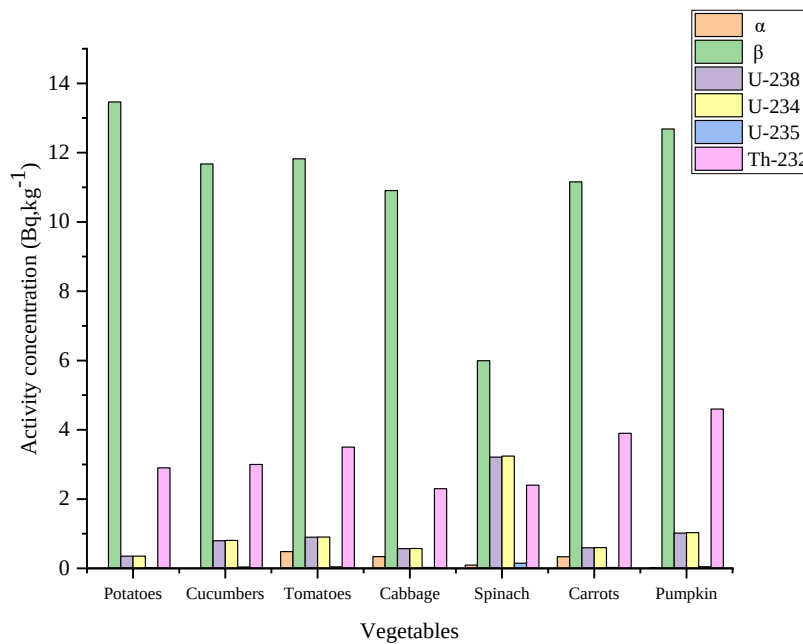


Figure 4.16: Comparison between gross α - β and activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in vegetables from Pretoria.

Tables 4.13 and 4.14 illustrated in Figures 4.15 and 4.16 show the results of gross α - β activity concentrations and specific radionuclide activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits and vegetables from Pretoria. The highest activity concentrations in fruits and vegetables was ^{232}Th in pineapple and pumpkin and ^{235}U was the lowest in apples and potatoes. The highest activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits were observed in bananas. The activity concentrations of ^{238}U were highest in bananas with a

value of $11.04 \pm 3.87 \text{ Bq.kg}^{-1}$ as also observed by Sowole and Olaniyi, (2018). This may be due to fertilisers containing radionuclides such as uranium, thorium and potassium which are used to increase crop yield of fruits and vegetables. The activity concentrations of radionuclides may also have an affect on the internal absorbed dose to peoples consuming them (Harb, 2015). The activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits from Pretoria with p-value of 0.00001 showed significant difference ($p < 0.05$) and the activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in vegetables with p-value of 0.00044 showed there is a significant difference ($p < 0.05$).

The mean ratios of $^{234}\text{U}/^{238}\text{U}$, $^{235}\text{U}/^{238}\text{U}$ and $^{232}\text{Th}/^{238}\text{U}$ were 1.00, 0.05 and 6.52, respectively in fruits. The ratio of $^{234}\text{U}/^{238}\text{U}$, $^{235}\text{U}/^{238}\text{U}$ and $^{232}\text{Th}/^{238}\text{U}$ were 1.01, 0.05 and 4.56, respectively, in vegetables. The mean ratio are close to unity and are comparable to the natural ratio.

Table 4.15: Activity concentrations of ^{238}U (Bq.kg^{-1}) in fruits from Hartbeespoort, Mahikeng and Pretoria.

^{238}U concentrations (Bq.kg^{-1})			
Fruits	Hartbeespoort	Mahikeng	Pretoria
Oranges	0.573	0.270	0.320
Apples	1.370	0.220	0.260
Bananas	0.270	0.220	1.430
Pineapples	1.850	0.120	0.619
Mean $\pm$ SD	1.015 $\pm$ 0.627	0.207 $\pm$ 0.054	0.657 $\pm$ 0.466
UNSCEAR (2000)	35 Bq.kg^{-1}		

Table 4.16: Activity concentrations of ^{238}U (Bq.kg^{-1}) in vegetables from Hartbeespoort, Mahikeng and Pretoria.

^{238}U concentrations (Bq.kg^{-1})			
Vegetables	Hartbeespoort	Mahikeng	Pretoria
Potatoes	3.790	0.701	0.350
Cucumbers	0.742	2.360	0.779
Tomatoes	0.910	0.440	0.898
Cabbage	0.885	4.000	0.568
Spinach	2.090	3.220	3.210
Carrots	3.390	0.190	0.595
Pumpkin	0.627	0.724	1.020
Mean $\pm$ SD	1.348 $\pm$ 1.116	1.662 $\pm$ 1.406	1.060 $\pm$ 0.902
UNSCEAR (2000)	35 Bq.kg^{-1}		

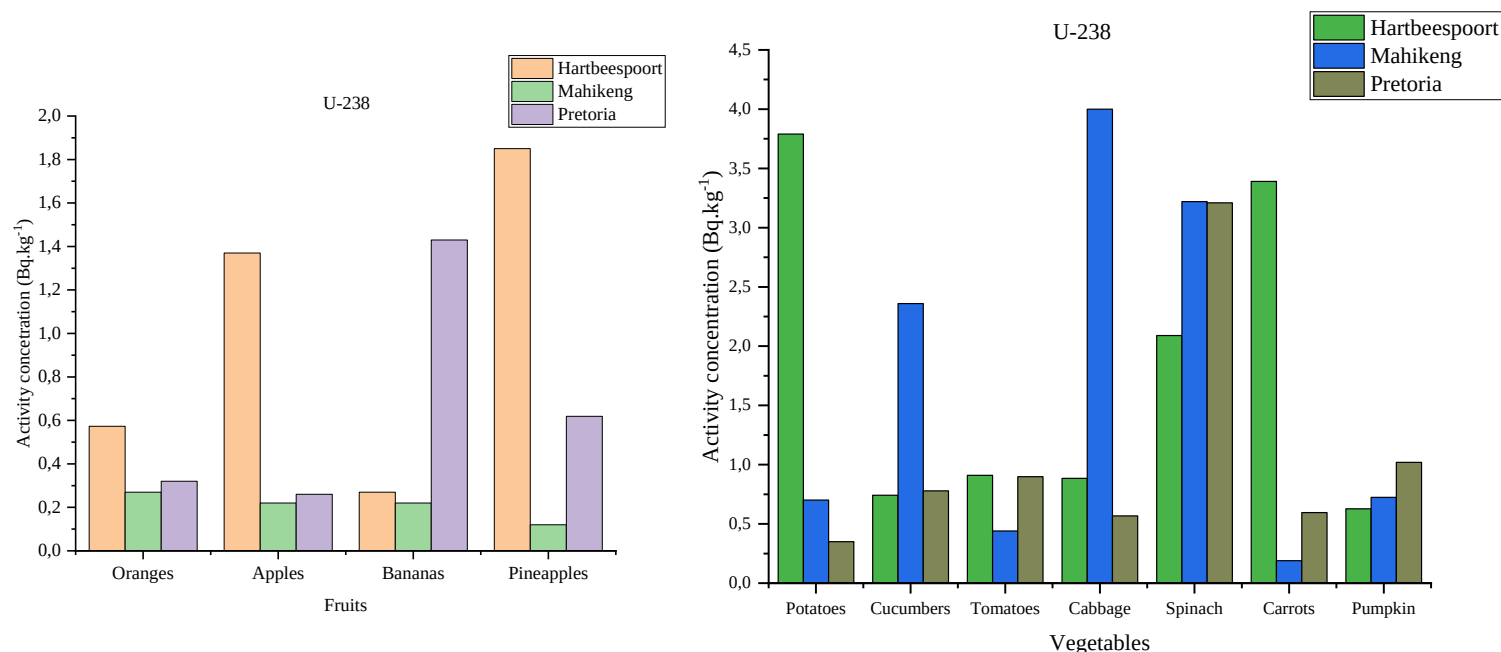


Figure 4.17: ^{238}U Activity Concentrations in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria.

The amount of ^{238}U , ^{234}U , ^{235}U and ^{232}Th were determined in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria. The activity of these radionuclides shown in Tables 4.15, 4.16, 4.17, 4.18, 4.19, 4.20, 4.21 and 4.22 and illustrated in Figure 4.18, 4.19, 4.19 and 4.20. Table 4.15 and Figure 4.17 are the results of ^{238}U activity concentrations in fruits in descending order by areas. In Hartbeespoort, pineapples have the highest ^{238}U concentration followed by apples, oranges, and bananas. Pretoria was second with bananas having the highest concentration of ^{238}U , followed by pineapples, oranges, and apples. Mahikeng had the lowest ^{238}U concentrations in all the fruits measured in the study.

Table 4.16 Figure 4.17 show the results of ^{238}U activity concentrations in vegetables in descending order by market area. Mahikeng had the highest ^{238}U concentrations in cabbage, followed by spinach, cucumber, potatoes and pumpkin, tomatoes and the least ^{238}U concentration was found in carrots. Hartbeespoort was second with potatoes having the highest activity concentration of ^{238}U followed by carrots, spinach, tomatoes, cabbage, cucumber, and pumpkin. Pretoria vegetables had the lowest activity concentrations of ^{238}U with the exception of the spinach. The activity concentrations of ^{238}U in other vegetables were relatively very low. The activity concentrations from ^{238}U with p-values of 0.14568 and 0.55042 showed no significant difference ($p > 0.05$) in fruits and vegetables from Mahikeng, Pretoria and Hartbeespoort.

Table 4.17: Activity concentrations of ^{234}U in fruits (Bq.kg^{-1}) from Hartbeespoort, Mahikeng and Pretoria.

^{234}U concentrations (Bq.kg^{-1})			
Fruits	Hartbeespoort	Mahikeng	Pretoria
Oranges	0.578	0.270	0.320
Apples	1.380	0.230	0.260
Bananas	0.270	0.230	1.440
Pineapples	1.870	0.120	0.624
Mean $\pm$ SD	1.024 $\pm$ 0.634	0.121 $\pm$ 0.05	0.661 $\pm$ 0.470
UNSCEAR (2000)	35 Bq.kg^{-1}		

Table 4.18: Activity concentrations of ^{234}U in vegetables (Bq.kg^{-1}) from Hartbeespoort, Mahikeng and Pretoria.

^{234}U concentrations (Bq.kg^{-1})			
Vegetables	Hartbeespoort	Mahikeng	Pretoria
Potatoes	3.820	0.700	0.350
Cucumbers	0.748	2.380	0.805
Tomatoes	0.918	0.440	0.905
Cabbage	0.893	4.030	0.573
Spinach	2.100	3.250	3.240
Carrots	0.390	0.190	0.600
Pumpkin	0.632	0.730	1.030
Mean $\pm$ SD	1.357 $\pm$ 1.125	1.674 $\pm$ 1.419	1.072 $\pm$ 0.909
UNSCEAR (2000)	35 Bq.kg^{-1}		

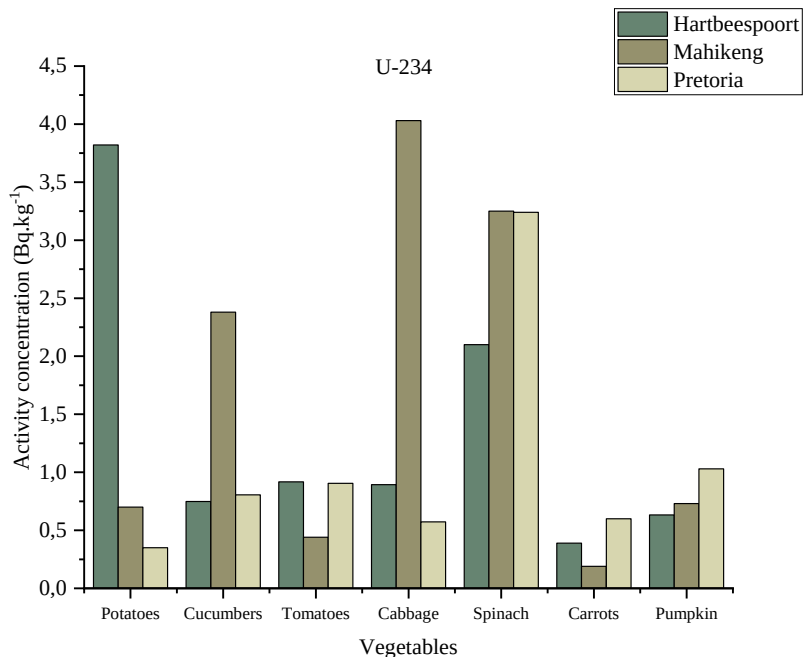
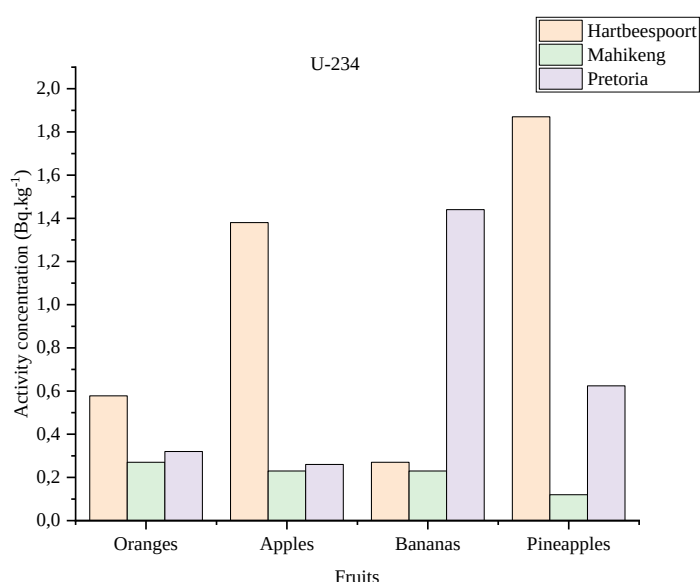


Figure 4.18: ^{234}U Activity Concentrations in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria.

Table 4.17 and Figure 4.18 are the results of ^{234}U activity concentrations in fruits in descending order by market areas. Fruits from Hartbeespoort had the highest ^{234}U activity concentrations in pineapples followed by apples, oranges, and bananas. Bananas having the lowest concentrations of ^{234}U . Pretoria was second with bananas having the highest ^{234}U activity concentrations and the other fruits had low concentrations of ^{234}U . Mahikeng had the lowest ^{234}U activity concentrations in all the fruits measured in the study.

Table 4.18 and Figure 4.18 show the results of ^{234}U activity concentrations in vegetables in descending order by market area. Mahikeng showed the highest ^{234}U activity concentrations in vegetables. Cabbage had the highest concentration followed by spinach, cucumber, pumpkin, potatoes, tomatoes, and carrots had lowest activity concentrations. Pretoria was second where potatoes had the highest ^{234}U activity concentrations followed by spinach, tomatoes and cabbage, cucumber, pumpkin, and carrots the least. Pretoria had the lowest ^{234}U activity concentrations. Spinach had the highest concentration of ^{234}U followed by pumpkin, tomatoes, cucumber, cabbage, and carrots. Potatoes had the least activity concentration of ^{234}U . The activity concentration of ^{234}U with p-values of 0.14847 and 0.67744 showed no significant difference ($p > 0.05$) in fruits and vegetables from Mahikeng, Pretoria and Hartbeespoort.

Table 4.19: Activity concentrations of ^{235}U (Bq.kg^{-1}) in fruits from Hartbeespoort, Mahikeng and Pretoria.

^{235}U concentrations (Bq.kg^{-1})			
Fruits	Hartbeespoort	Mahikeng	Pretoria
Oranges	0.026	0.013	0.015
Apples	0.063	0.010	0.012
Bananas	0.023	0.010	0.066
Pineapples	0.085	0.005	0.029
mean $\pm$ SD	0.049 $\pm$ 0.025	0.009 $\pm$ 0.002	0.030 $\pm$ 0.021
UNSCEAR (2000)	-		

Table 4.20: Activity concentrations of ^{235}U (Bq.kg^{-1}) in vegetables Hartbeespoort, Mahikeng and Pretoria.

^{235}U concentrations (Bq.kg^{-1})			
Vegetables	Hartbeespoort	Mahikeng	Pretoria
Potatoes	0.175	0.032	0.016
Cucumbers	0.034	0.109	0.037
Tomatoes	0.042	0.020	0.041
Cabbage	0.041	0.184	0.026
Spinach	0.096	0.148	0.148
Carrots	0.018	0.008	0.027
Pumpkin	0.029	0.033	0.047
Mean $\pm$ SD	0.062 $\pm$ 0.051	0.076 $\pm$ 0.064	0.049 $\pm$ 0.041
UNSCEAR (2000)	-		

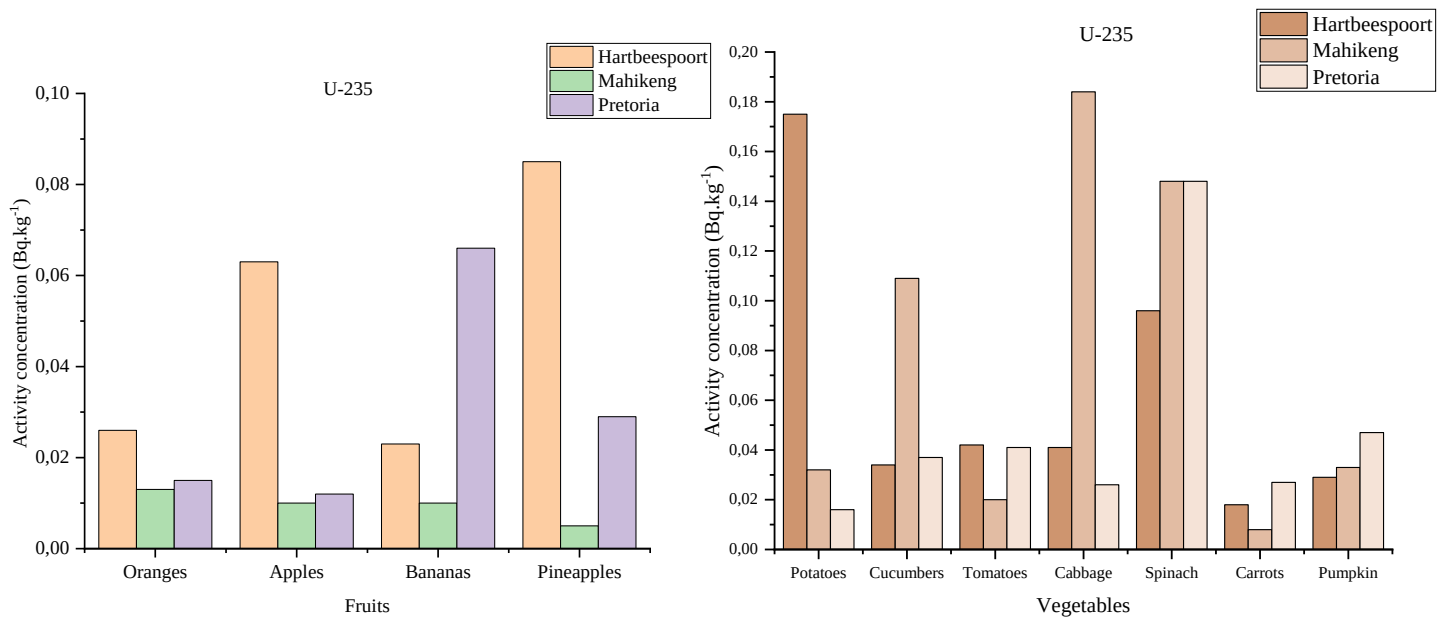


Figure 4.19: ^{235}U Activity Concentrations in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria.

Table 4.19 and Figure 4.19 are the results of ^{235}U activity concentrations in fruits in descending order by market areas. Fruits from Hartbeespoort show the highest ^{235}U activity concentrations with pineapples having the highest ^{235}U followed by apples, oranges, and bananas. The second highest ^{235}U activity concentrations in fruits were found in Pretoria. Bananas had the highest activity concentration followed by pineapples, oranges, and apples having the lowest ^{235}U activity concentrations. Mahikeng had the lowest ^{235}U activity concentration with oranges having the highest ^{235}U concentration followed by apples and bananas. The lowest ^{235}U activity concentration was found in pineapples.

Table 4.20 and Figure 4.19 show the results of ^{235}U activity concentrations in vegetables in a descending order by market areas. Vegetables from the Pretoria market had the highest ^{235}U with cabbages having the highest followed by spinach, cucumbers, potatoes and pumpkins, tomatoes and the least was in carrots. The second vegetable market with ^{235}U activity concentrations in vegetables was Pretoria with cabbages having the highest ^{235}U concentration followed by spinach, cucumbers, potatoes and pumpkins, tomatoes and the least found in carrots. The activity concentrations of ^{235}U with p-values of 0.09361 and 0.68037 showed no significant difference ($p > 0.05$) in fruits and vegetables from Mahikeng, Pretoria and Hartbeespoort.

Table 4.21: Activity concentrations of ^{232}Th (Bq.kg^{-1}) in fruits from Hartbeespoort, Mahikeng and Pretoria.

^{232}Th concentrations (Bq.kg^{-1})			
Fruits	Hartbeespoort	Mahikeng	Pretoria
Oranges	3.800	3.500	2.600
Apples	2.700	2.300	2.800
Bananas	2.100	2.000	2.400
Pineapples	2.200	2.700	3.400
Mean $\pm$ SD	2.700 $\pm$ 0.674	2.625 $\pm$ 0.563	2.800 $\pm$ 0.374
UNSCEAR (2000)	30 Bq.kg^{-1}		

Table 4.22: Activity concentrations of ^{232}Th (Bq.kg^{-1}) in vegetables from Hartbeespoort, Mahikeng and Pretoria.

^{232}Th concentrations (Bq.kg^{-1})			
Vegetables	Hartbeespoort	Mahikeng	Pretoria
Potatoes	2.000	6.900	2.900
Cucumbers	3.800	4.900	3.000
Tomatoes	1.800	4.000	3.500
Cabbage	3.100	4.300	2.300
Spinach	1.700	2.500	2.400
Carrots	2.500	3.800	3.900
Pumpkin	3.800	3.200	4.600
Mean $\pm$ SD	2.671 $\pm$ 0.838	4.228 $\pm$ 1.302	3.228 $\pm$ 0.767
UNSCEAR (2000)	30 Bq.kg^{-1}		

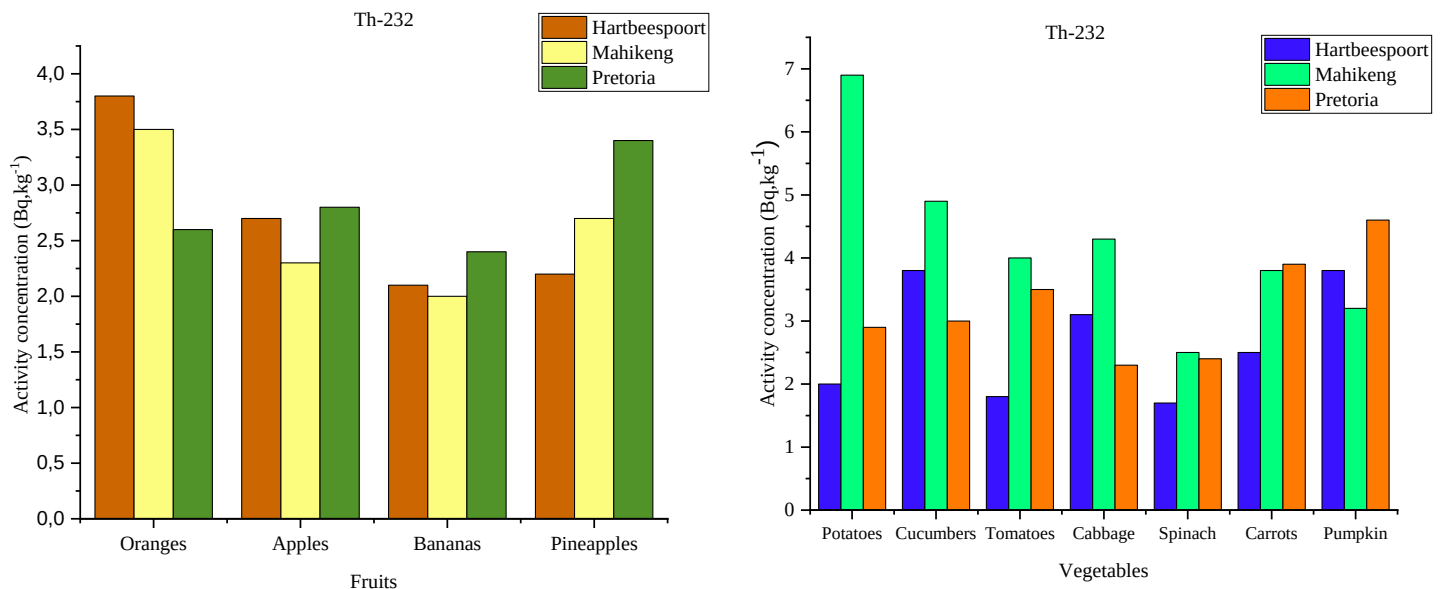


Figure 4.20: ²³²Th activity concentrations in fruits and vegetables from Hartbeespoort and Mahikeng.

The activity concentrations of ²³²Th is the biggest contributor to the total activity concentration in all samples collected from Hartbeespoort, Mahikeng and Pretoria. The maximum radioactivity concentration of ²³²Th (6.900 Bq.kg⁻¹) was detected in potatoes from Mahikeng and the lowest ²³²Th was found in spinach from Hartbeespoort. The activity concentrations of ²³⁸U, ²³⁴U, ²³⁵U and ²³²Th in fruits from Hartbeespoort, Mahikeng and Pretoria showed no significant difference ($p < 0.05$). The activity concentrations of ²³⁸U, ²³⁴U, ²³⁵U in vegetables with p-value of 0.92733 showed no significant difference ($p > 0.05$) however, with a p-value of 0.04343 showed there was a significant difference of ²³²Th ($p < 0.05$) noticed in vegetables.

The differences in activity concentrations of these radionuclides may depend on the range of fertilizers used, irrigation water and the type of soil. The uptake of dissolved NORM in water from the soil by plants depends on various properties such as texture, pH, and organic matter components. On the other hand, the absorption of radionuclides is affected by parameters such as the chemical and physical of the soil and radionuclides, plant species, stages of growth, temperature, rainfall, and sunlight. The different activity concentrations of these radionuclides as the results of the natural presence of NORM in the soil before fertilizers were applied to the agricultural farms and the effect of fertilizers to the agricultural farm which may change the levels of natural occurring radionuclide, but can also affect the chemical forms, physical forms and biological uptake (Khan et al., 2010). The activity concentrations of ²³⁸U, ²³⁴U, ²³⁵U and ²³²Th in fruits and vegetables were lower than the world mean values of 35 Bq.kg⁻¹ and 30 Bq.kg⁻¹ (UNSCEAR, 2000), respectively.

4.4. Radiological Dose Assessment

4.4.1 Radiological indices

The activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th were used to calculate the external hazard index (H_{ext}) and internal hazard index (H_{int}) due to fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria using equation (2.41) and (2.42).

Table 4.23: External hazard index (H_{ext}) and Internal hazard index (H_{int}) due to fruits from Hartbeespoort, Mahikeng and Pretoria.

	External hazard index (H_{ext}) and Internal hazard index (H_{int}) in fruits					
	Hartbeespoort		Mahikeng		Pretoria	
Fruits	(H_{ext})	(H_{int})	(H_{ext})	(H_{int})	(H_{ext})	(H_{int})
Oranges	0.016	0.018	0.014	0.015	0.001	0.002
Apples	0.014	0.018	0.009	0.010	0.001	0.001
Bananas	0.009	0.009	0.008	0.009	0.004	0.001
Pineapples	0.018	0.014	0.011	0.011	0.002	0.003
SD	0.004	0.004	0.003	0.003	0.001	0.003
Median	0.014	0.018	0.010	0.011	0.001	0.002
Min	0.009	0.009	0.008	0.009	0.001	0.001
Max	0.016	0.018	0.014	0.015	0.004	0.001
Mean $\pm$ SD	0.013 $\pm$ 0.004	0.016 $\pm$ 0.004	0.011 $\pm$ 0.003	0.011 $\pm$ 0.003	0.002 $\pm$ 0.001	0.004 $\pm$ 0.003
UNSCEAR (2000)	<1					

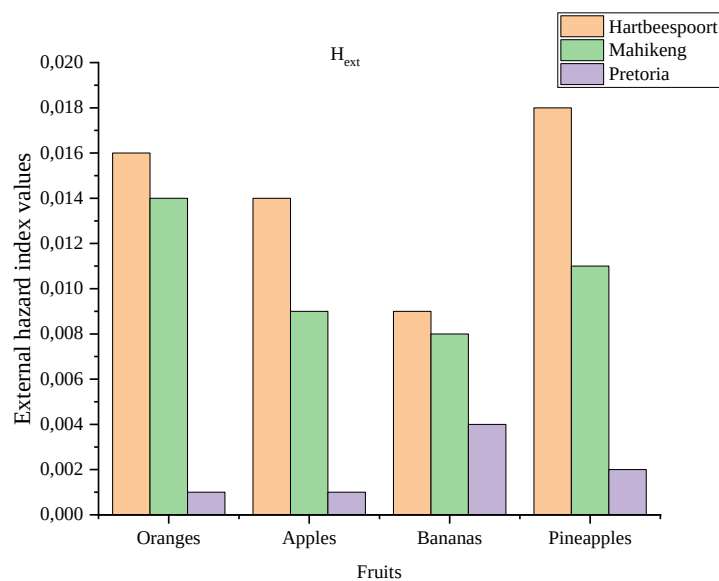


Figure 4.21: The external hazard index in fruits from Hartbeespoort, Mahikeng and Pretoria.

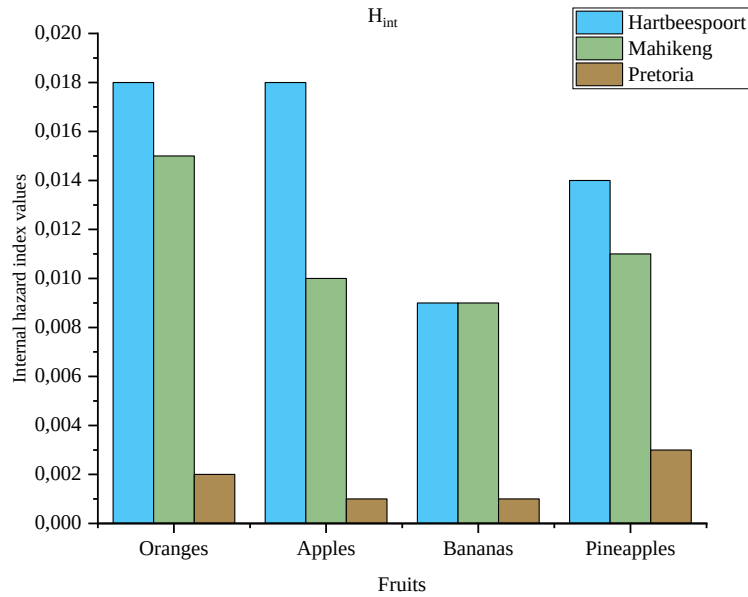


Figure 4.22: The internal hazard index in fruits from Hartbeespoort, Mahikeng and Pretoria.

The results in Table 4.23 and plotted Figure 4.21 are the H_{ext} values in fruits from Hartbeespoort, Mahikeng and Pretoria ranging from 0.009 to 0.018, 0.008 to 0.014 and 0.001 to 0.004 with average values and standard deviations of 0.013 ± 0.004 , 0.011 ± 0.003 and 0.002 ± 0.001 , respectively. The highest indexes were from Hartbeespoort followed by Mahikeng and the least were from Pretoria. Table 4.23 illustrated in Figure 4.22 are H_{int} obtained from Hartbeespoort, Mahikeng and Pretoria ranging from 0.014 to 0.018, 0.009 to 0.015 and 0.001 to 0.003 with the mean and standard deviations of 0.016 ± 0.004 , 0.011 ± 0.003 and 0.004 ± 0.003 , respectively. The internal indexes follow the same pattern as those of the external indexes, Hartbeespoort showed the highest and Pretoria the lowest. The external and internal hazard indices in fruits from Mahikeng, Pretoria and Hartbeespoort with p-values of 0.0005 and 0.00043 showed there is significant difference ($p < 0.05$).

Table 4.24: External hazard index (H_{ext}) and Internal hazard index (H_{int}) in vegetables from Hartbeespoort, Mahikeng and Pretoria.

	Internal hazard index (H_{int}) and external hazard index in fruits					
	Hartbeespoort		Mahikeng		Pretoria	
Vegetables	(H_{ext})	(H_{int})	(H_{ext})	(H_{int})	(H_{ext})	(H_{int})
Potatoes	0.018	0.028	0.029	0.030	0.012	0.013
Cucumbers	0.017	0.018	0.007	0.032	0.014	0.016
Tomatoes	0.009	0.012	0.017	0.018	0.016	0.018
Cabbage	0.014	0.016	0.012	0.038	0.010	0.012
Spinach	0.012	0.018	0.018	0.027	0.018	0.027
Carrots	0.011	0.012	0.001	0.016	0.017	0.018
Pumpkin	0.003	0.018	0.014	0.016	0.021	0.023
SD	0.003	0.006	0.009	0.009	0.003	0.005
Median	0.014	0.018	0.014	0.027	0.016	0.018
Min	0.009	0.012	0.001	0.016	0.010	0.012
Max	0.018	0.028	0.029	0.038	0.021	0.027
Mean $\pm$ SD	0.014 $\pm$ 0.003	0.018 $\pm$ 0.006	0.014 $\pm$ 0.009	0.025 $\pm$ 0.009	0.015 $\pm$ 0.003	0.018 $\pm$ 0.005
UNSCEAR (2000)	<1					

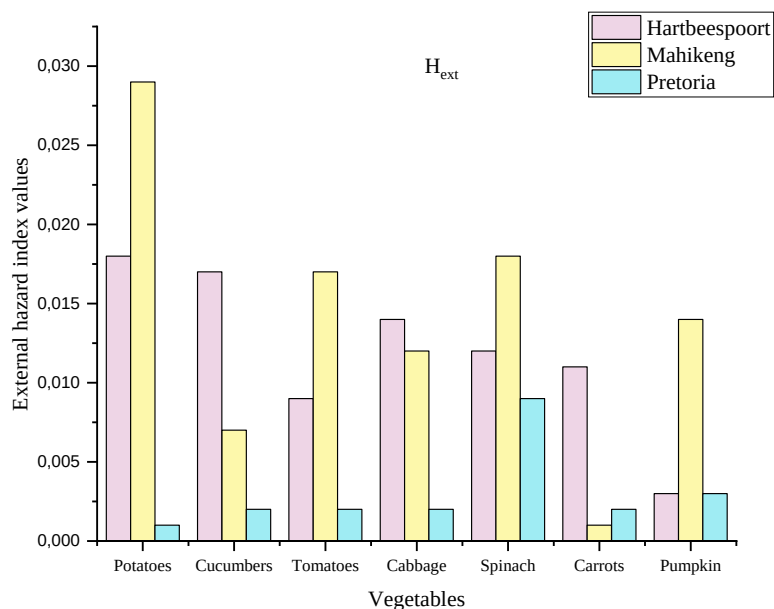


Figure 4.23: The external hazard index in vegetables from Hartbeespoort, Mahikeng and Pretoria.

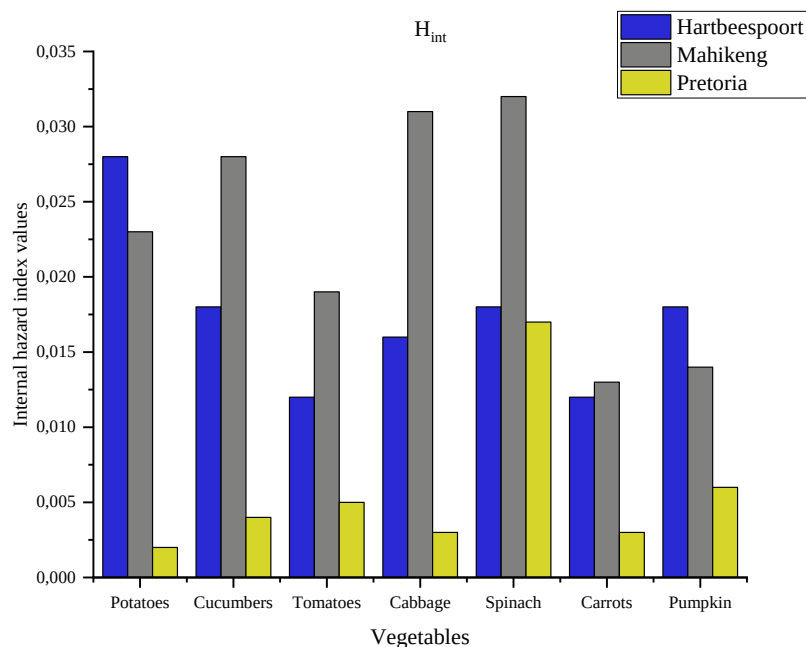


Figure 4.24: The internal hazard index in vegetables from Hartbeespoort, Mahikeng and Pretoria.

Table 4.24 illustrated in Figure 4.23 are the H_{ext} values in fruits ranging from 0.003 to 0.018; 0.001 to 0.029 and 0.010 to 0.021 with the average values of 0.014 ± 0.003 ; 0.014 ± 0.009 and 0.015 ± 0.003 from Hartbeespoort; Mahikeng and Pretoria, respectively.

Table 4.24 illustrated in Figure 4.24 are the H_{int} value in vegetables ranging from 0.012 to 0.028; 0.016 to 0.038 and 0.012 to 0.023 with mean and standard deviations of 0.018 ± 0.006 ; 0.025 ± 0.009 and 0.018 ± 0.005 for Hartbeespoort, Mahikeng and Pretoria, respectively. The highest indexes were observed in fruits from Hartbeespoort and the lowest indexes were in vegetables from Pretoria. The external and internal hazard indexes in vegetables from Hartbeespoort, Mahikeng and Pretoria with p-values of 0.60565 and 0.07392 showed no significant difference ($p > 0.05$).

4.4.2. Annual ingestion radiation dose rate

Tables 4.25 and 4.26 illustrated in Figures 4.25 and 4.26 are the results of the annual ingestion radiation dose ($\mu\text{Sv.y}^{-1}$) of ^{238}U and ^{232}Th (Bq.kg^{-1}) in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria.

Table 4.25: The annual ingestion dose ($\mu\text{Sv.y}^{-1}$) in fruits from Hartbeespoort, Mahikeng and Pretoria.

Fruits	Annual ingestion dose ($\mu\text{Sv.y}^{-1}$)					
	Hartbeespoort		Mahikeng		Pretoria	
	^{238}U	^{232}Th	^{238}U	^{232}Th	^{238}U	^{232}Th
Oranges	3.021	17.480	1.512	16.100	1.792	8.970
Apples	7.672	12.42	1.232	10.580	1.456	9.660
Bananas	1.512	9.660	1.232	9.200	8.088	8.280
Pineapples	10.360	10.120	0.672	12.420	3.466	11.730
Min	1.512	9.660	0.672	9.200	1.456	8.280
Max	10.360	17.480	1.512	16.100	8.008	11.730
Mean $\pm$ SD	5.688 $\pm$ 4.056	12.420 $\pm$ 3.582	1.162 $\pm$ 0.352	12.075 $\pm$ 2.990	3.681 $\pm$ 3.016	9.660 $\pm$ 1.491
UNSCEAR (2000)	290					

Table 4.26: The annual ingestion dose ($\mu\text{Sv.y}^{-1}$) in vegetables from Hartbeespoort, Mahikeng and Pretoria.

Vegetables	Annual ingestion dose ($\mu\text{Sv.y}^{-1}$)					
	Hartbeespoort		Mahikeng		Pretoria	
	^{238}U	^{232}Th	^{238}U	^{232}Th	^{238}U	^{232}Th
Potatoes	15.918	6.900	2.944	23.805	1.470	10.005
Cucumbers	3.116	13.110	9.912	16.905	3.272	10.350
Tomatoes	3.822	6.210	1.848	13.800	3.772	12.075
Cabbage	3.717	10.695	16.800	14.835	2.386	7.935
Spinach	8.778	5.865	13.524	8.625	13.482	8.280
Carrots	1.638	8.625	0.798	13.110	2.499	13.455
Pumpkin	2.633	13.110	3.041	11.040	4.284	15.870
Min	1.638	5.865	0.798	8.625	1.470	7.935
Max	15.918	13.110	16.800	23.805	13.482	15.870
Mean $\pm$ SD	5.660 $\pm$ 5.064	9.216 $\pm$ 3.123	6.981 $\pm$ 6.380	14.589 $\pm$ 4.853	4.452 $\pm$ 4.091	11.139 $\pm$ 10.350
UNSCEAR (2000)	290					

The mean and standard deviation (Mean $\pm$ SD) of the annual ingestion dose from natural radionuclides ^{238}U and ^{232}Th in fruits from Hartbeespoort; Mahikeng and Pretoria were 5.688 $\pm$ 4.056 $\mu\text{Sv.y}^{-1}$ and 12.420 $\pm$ 3.582 $\mu\text{Sv.y}^{-1}$; 1.162 $\pm$ 0.352 $\mu\text{Sv.y}^{-1}$ and 12.075 $\pm$ 2.990 $\mu\text{Sv.y}^{-1}$ and 3.681 $\pm$ 3.016 $\mu\text{Sv.y}^{-1}$ and 9.660 $\pm$ 1.491 $\mu\text{Sv.y}^{-1}$, respectively. The mean and standard deviations (Mean $\pm$ SD) of the annual ingestion dose from natural radionuclide of ^{238}U and ^{232}Th in vegetables from Hartbeespoort, Mahikeng, and Pretoria were 5.660 $\pm$ 5.064 $\mu\text{Sv.y}^{-1}$ and 9.216 $\pm$ 3.123 $\mu\text{Sv.y}^{-1}$; 6.981 $\pm$ 6.380 $\mu\text{Sv.y}^{-1}$ and 14.589 $\pm$ 4.853 $\mu\text{Sv.y}^{-1}$ and 4.452 $\pm$ 4.091 $\mu\text{Sv.y}^{-1}$ and 11.139 $\pm$ 10.350 $\mu\text{Sv.y}^{-1}$, respectively. The highest ingestion dose of ^{232}Th was observed in Figure 4.26

found in potatoes from Mahikeng followed by oranges from Hartbeespoort, Figure 4.25. The annual ingestion dose in fruits and vegetables from the current study was lower than the world annual ingestion dose of ^{238}U and ^{232}Th of $290 \mu\text{Sv.y}^{-1}$ (UNSCEAR, 2000) and the recommended values of $250\text{-}400 \mu\text{Sv.y}^{-1}$ (WHO, 2011). The annual ingestion dose of ^{238}U and ^{232}Th with p-value of 0.15508 and 0.36467 showed no significant difference ($p > 0.05$) in fruits. The annual ingestion dose of ^{238}U with p-value of 0.67331 showed no significant difference ($p > 0.05$) while ^{232}Th with p-value of 0.04343 showed a significant difference in vegetables ($p < 0.05$).

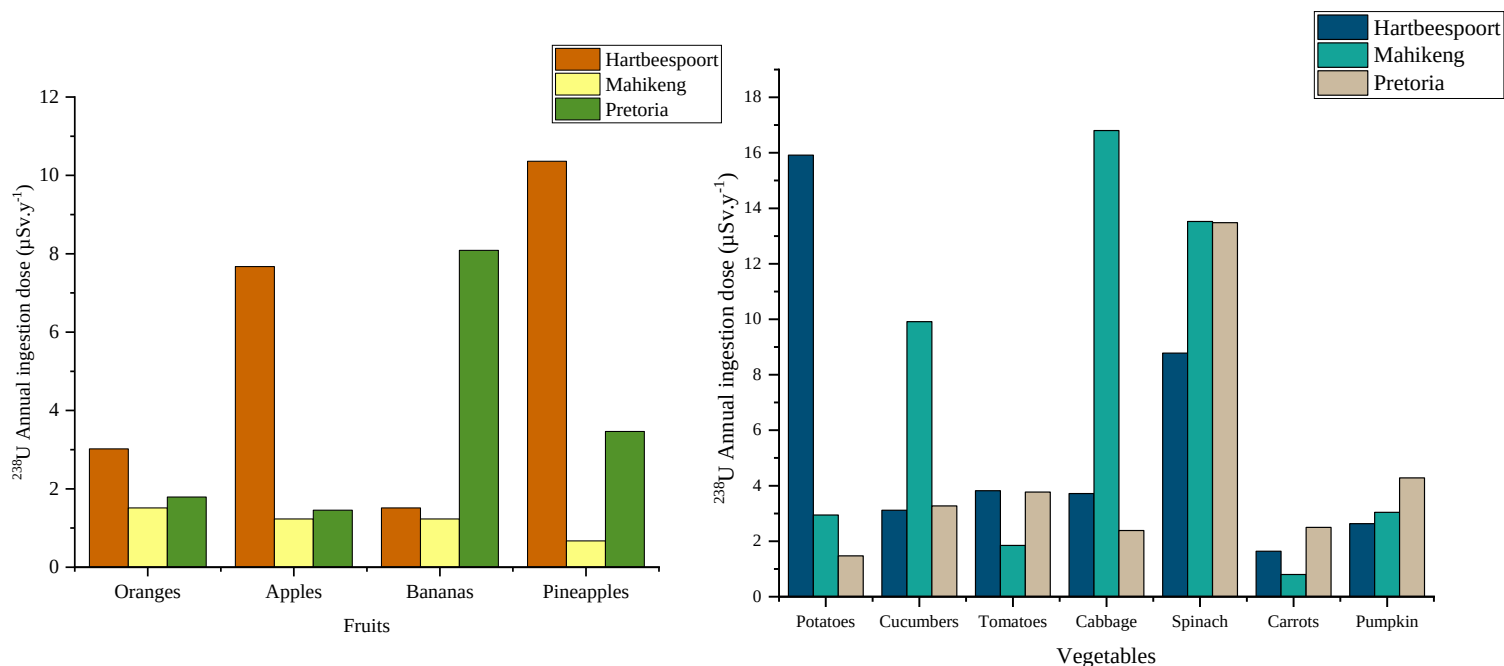


Figure 4.25: Contribution of ^{238}U in various types of fruits and vegetables to annual ingestion dose.

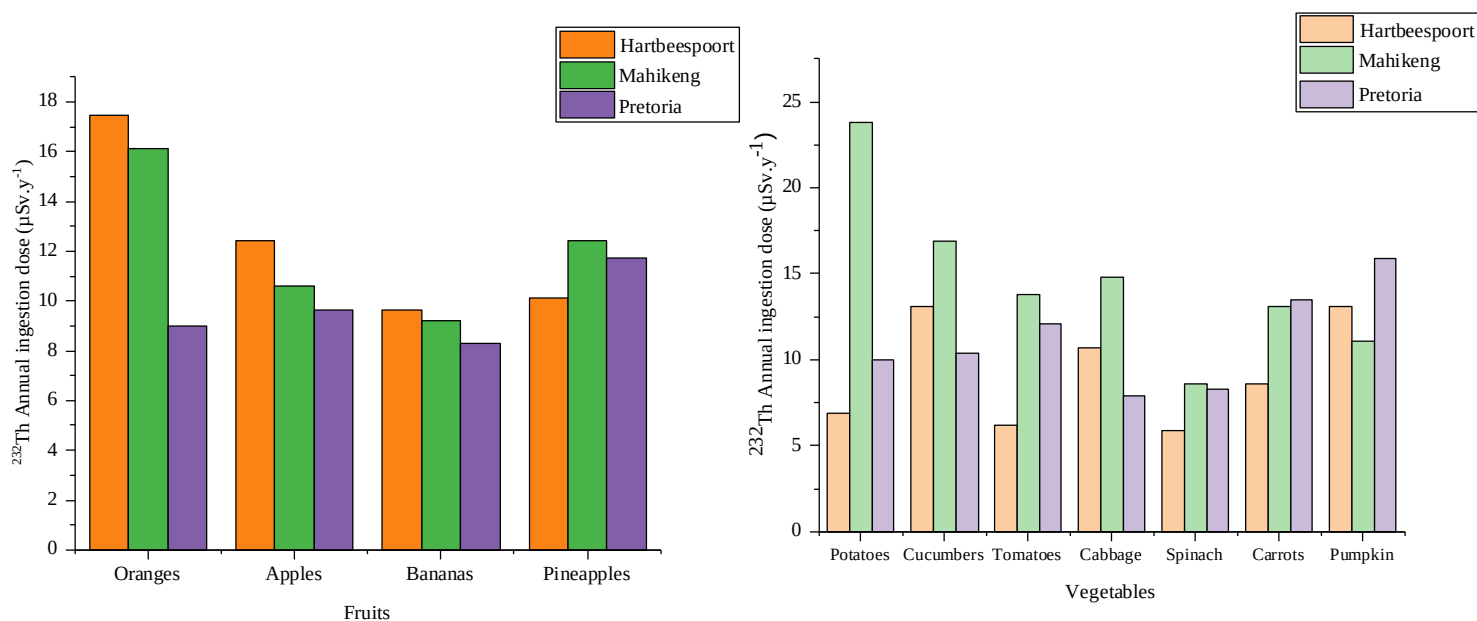


Figure 4.26: Contribution of ^{232}Th in various types of fruits and vegetables to annual ingestion dose.

4.4.3. Absorbed dose rate, Annual effective dose equivalent (AEDE) and Excess lifetime cancer risk (ELCR).

Tables 4.27 and 4.28 are the results of absorbed dose rate $D(\text{nGyh}^{-1})$, outdoor annual effective dose equivalent (AEDE) (mSv.y^{-1}) and excess lifetime cancer risk (ELCR) (mSv.y^{-1}) in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria. The $D(\text{nGyh}^{-1})$; (AEDE) (mSv.y^{-1}) and (ELCR) (mSv.y^{-1}) were calculated using equation 2.44, 2.46 and 2.47, respectively. The results are further illustrated in Figures 4.27, 4.28, 4.29.

Table 4.27: Absorbed dose rate $D(\text{nGyh}^{-1})$, Annual effective dose equivalent (AEDE) (mSv.y^{-1}) and Excess lifetime cancer risk (ELCR) (mSv.y^{-1}) in fruits.

Fruits	Hartbeespoort			Mahikeng			Pretoria		
	Absorbed Dose $D(\text{nGyh}^{-1})$	AEDE (mSv.y^{-1})	ELCR (mSv.y^{-1})	Absorbed Dose $D(\text{nGyh}^{-1})$	AEDE (mSv.y^{-1})	ELCR (mSv.y^{-1})	Absorbed Dose $D(\text{nGyh}^{-1})$	AEDE (mSv.y^{-1})	ELCR (mSv.y^{-1})
Oranges	2.625	0.003	0.011	2.298	0.003	0.010	1.762	0.002	0.007
Apples	2.310	0.003	0.010	1.530	0.002	0.007	1.859	0.002	0.008
Bananas	1.429	0.002	0.006	1.344	0.002	0.006	2.151	0.003	0.009
Pineapples	2.221	0.003	0.010	1.731	0.002	0.007	2.397	0.003	0.010
Min	1.423	0.002	0.006	1.344	0.002	0.006	1.762	0.002	0.008
Max	2.625	0.003	0.011	2.298	0.003	0.010	2.397	0.003	0.010
Mean± SD	2.146±0.508	0.003±0.001	0.009±0.002	1.726±0.413	0.002±0.001	0.007±0.002	2.042±0.289	0.003±0.000	0.008±0.001
World safety limits	59 ICRP (1993)	0.48 UNSCEAR (2000)	2.9×10^{-4} UNSCEAR (2000)	59 ICRP (1993)	0.48 UNSCEAR (2000)	2.9×10^{-4} UNSCEAR (2000)	59 UNSCEAR (2000)	0.48 UNSCEAR (2000)	2.9×10^{-4} UNSCEAR (2000)

Table 4.28: Absorbed dose rate D(nGyh⁻¹), Annual effective dose equivalent (AEDE) (mSv.y⁻¹) and Excess lifetime cancer risk (ELCR) (mSv.y⁻¹) in vegetables.

Vegetables	Hartbeespoort			Mahikeng			Pretoria		
	Absorbed Dose D(nGyh ⁻¹)	AEDE (mSv.y ⁻¹)	ELCR (mSv.y ⁻¹)	Absorbed Dose D(nGyh ⁻¹)	AEDE (mSv.y ⁻¹)	ELCR (mSv.y ⁻¹)	Absorbed Dose D(nGyh ⁻¹)	AEDE (mSv.y ⁻¹)	ELCR (mSv.y ⁻¹)
Potatoes	2.993	0.004	0.013	4.609	0.006	0.020	1.963	0.002	0.008
Cucumbers	2.703	0.003	0.012	4.133	0.005	0.018	2.223	0.003	0.010
Tomatoes	1.538	0.002	0.007	2.687	0.003	0.012	2.588	0.003	0.011
Cabbage	2.334	0.003	0.010	4.518	0.006	0.019	1.691	0.002	0.007
Spinach	2.021	0.002	0.010	3.040	0.004	0.013	2.973	0.004	0.012
Carrots	1.733	0.002	0.007	2.448	0.003	0.011	2.697	0.003	0.012
Pumpkin	2.649	0.003	0.011	2.322	0.003	0.010	3.327	0.004	0.014
Min	1.538	0.002	0.007	2.322	0.003	0.010	1.691	0.002	0.007
Max	2.993	0.004	0.013	4.609	0.006	0.020	3.328	0.004	0.014
Mean ± S.D	2.282±0.539	0.003±0.001	0.010±0.002	3.394±0.996	0.004±0.001	0.015±0.004	2.495±0.574	0.003±0.001	0.011±0.002
World safety limits	59 ICRP (1993)	0.48 UNSCEAR (2000)	2.9 x10 ⁻⁴ UNSCEAR (2000)	59 ICRP (1993)	0.48 UNSCEAR (2000)	2.9 x10 ⁻⁴ UNSCEAR (2000)	59 ICRP (1993)	0.48 UNSCEAR (2000)	2.9 x10 ⁻⁴ UNSCEAR (2000)

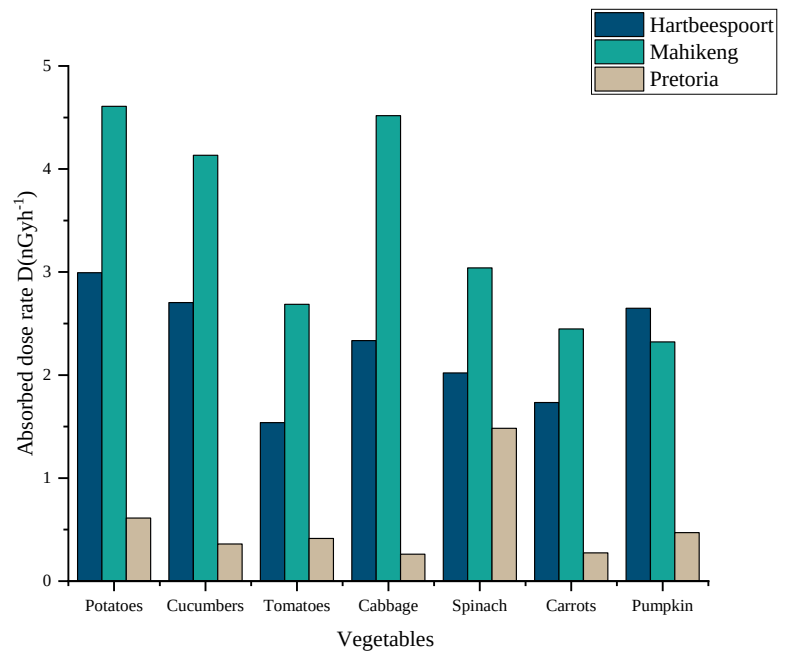
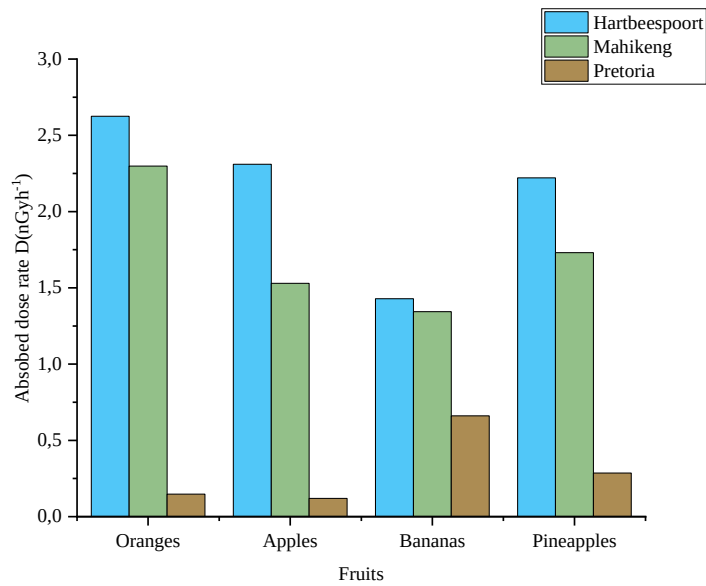


Figure 4.27: Absorbed dose rate in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria.

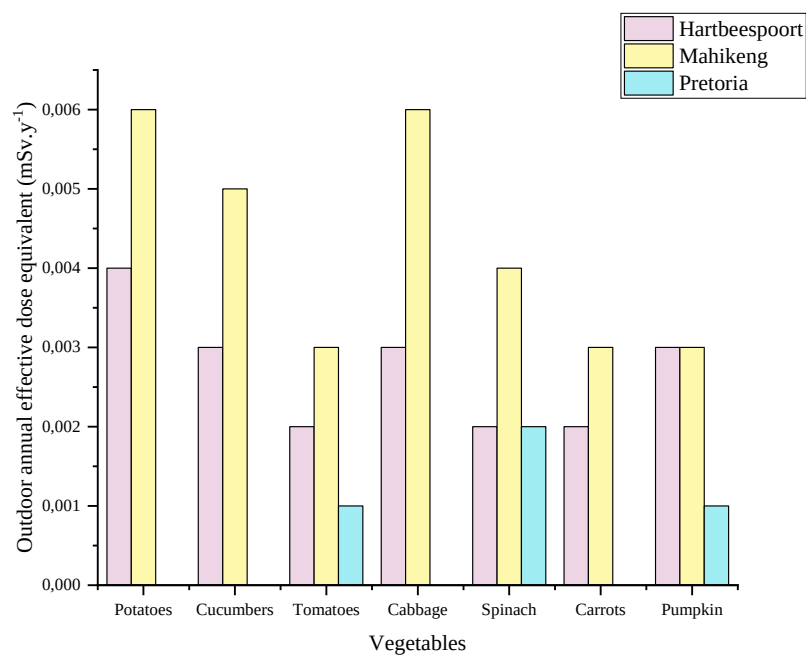
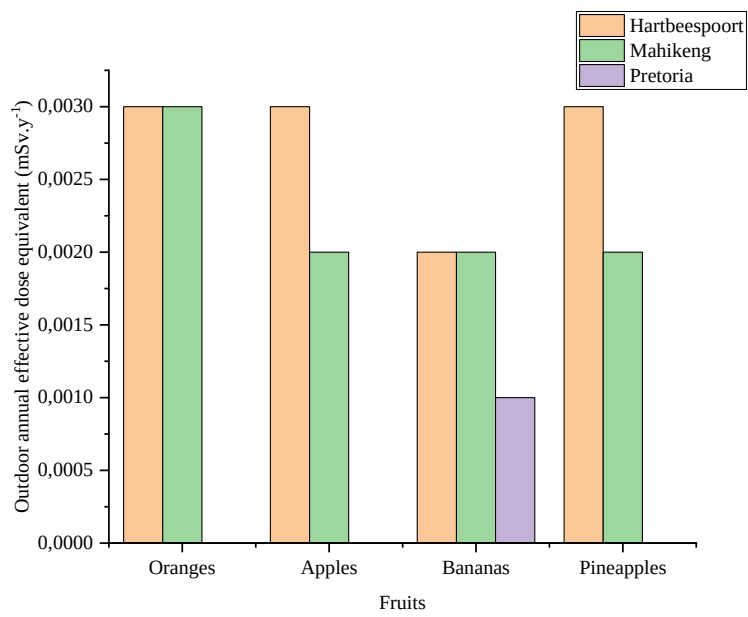


Figure 4.28: Annual effective dose equivalent in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria.

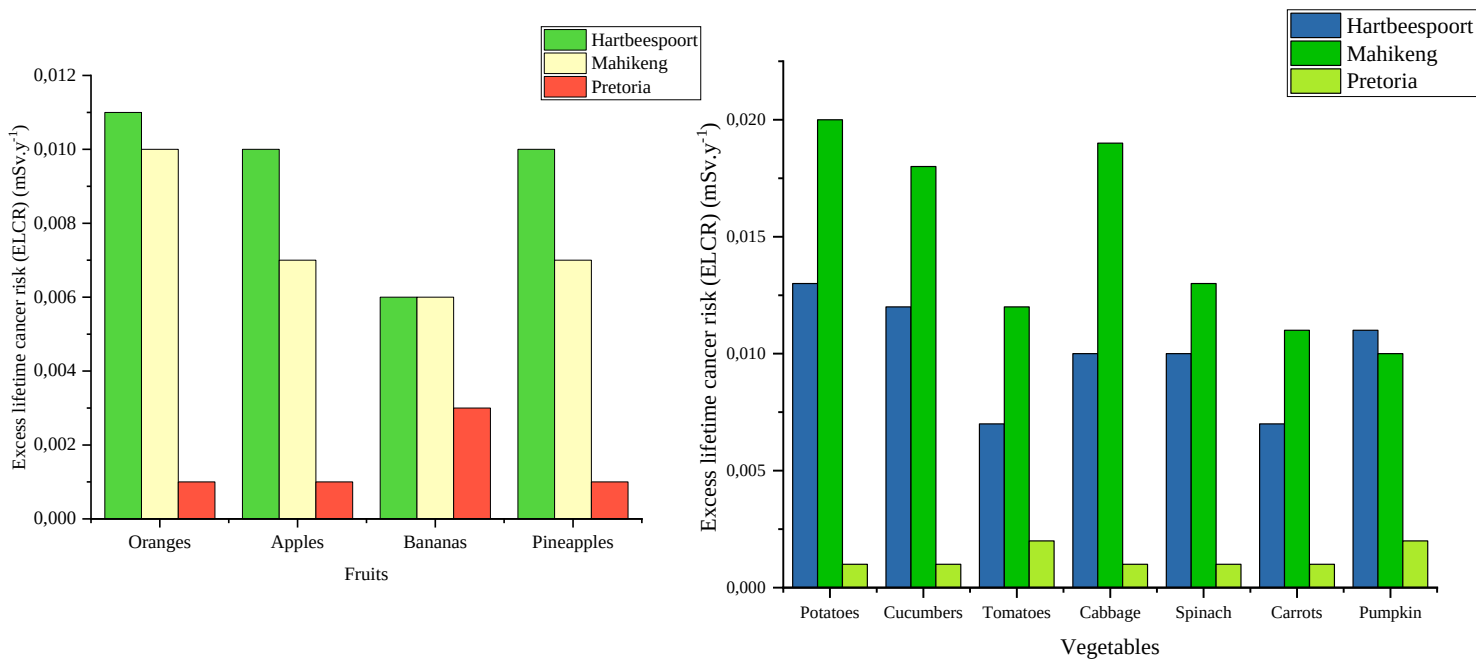


Figure 4.29: Excess lifetime cancer risk in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria.

Table 4.27, illustrated in Figure 4.27 are the absorbed dose rates of NORM in fruits from Hartbeespoort, Mahikeng and Pretoria. The absorbed dose of NORM from Hartbeespoort, Mahikeng and Pretoria ranged from 1.429 to 2.625 nGyh⁻¹; 1.344 to 2.298 nGyh⁻¹; 1.762 to 2.397 nGyh⁻¹ with the mean and standard (mean $\pm$ S.D) values of 2.146 $\pm$ 0.508 nGyh⁻¹; 1.726 $\pm$ 0.413 nGyh⁻¹ and 2.042 $\pm$ 0.289 nGyh⁻¹, respectively. Table 4.28 illustrated in Figure 4.28 are the absorbed dose rates of ²³⁸U and ²³²Th in vegetables from Hartbeespoort, Mahikeng and Pretoria. The absorbed dose rates of ²³⁸U and ²³²Th from Hartbeespoort, Mahikeng and Pretoria ranged from 1.538 to 2.993 nGyh⁻¹; 2.322 to 4.609 nGyh⁻¹ and 1.691 to 2.973 nGyh⁻¹ with the mean and standard deviations (mean $\pm$ S.D) of 2.282 $\pm$ 0.539 nGyh⁻¹; 3.394 $\pm$ 0.996 nGyh⁻¹ and 2.495 $\pm$ 0.574 nGyh⁻¹, respectively. The absorbed dose rates were lower than the world safety limit of 59 nGyh⁻¹ (ICRP, 1993). The Absorbed dose rate (D) analysis with p-value of 0.36687 and obtained in fruits from Hartbeespoort, Mahikeng and Pretoria showed no significant difference ($p > 0.05$) while showed a significant difference ($p < 0.05$) with a p-value of 0.02531 in vegetables.

Table 4.27 illustrated in Figure 4.27 show the results of AEDE from Hartbeespoort, Mahikeng and Pretoria in fruits. The AEDE from Hartbeespoort, Mahikeng and Pretoria ranged from 0.002 to 0.003 mSv.y⁻¹; 0.002 to 0.003 mSv.y⁻¹ and 0.002 to 0.003 mSv.y⁻¹ with the mean and standard deviations (mean $\pm$ S.D) of 0.003 $\pm$ 0.001 mSv.y⁻¹; 0.002 $\pm$ 0.001 mSv.y⁻¹ and 0.003 $\pm$ 0.000 mSv.y⁻¹, respectively. Table 4.28 illustrated in Figure 4.28 show the results of AEDE from Hartbeespoort, Mahikeng and Pretoria in vegetables. The AEDE

from Hartbeespoort, Mahikeng and Pretoria ranged from 0.002 to 0.004 mSv.y⁻¹; 0.003 to 0.006 mSv.y⁻¹ and 0.002 to 0.004 mSv.y⁻¹ with the mean and standard deviations (mean ± S.D) of 0.003 ± 0.001 mSv.y⁻¹; 0.015 ± 0.004 mSv.y⁻¹ and 0.003 ± 0.001 mSv.y⁻¹, respectively. The AEDE results from the current study for both fruits and vegetables were lower than the world safety limit of 0.48 mSv.y⁻¹ (UNSCEAR, 2000). The annual effective dose equivalent (AEDE) analysis with p-value of 0.44626 obtained in fruits from Hartbeespoort, Mahikeng and Pretoria showed there is no significant difference (p > 0.05) and the analysis in vegetables with p-value of 0.02296 showed is significant difference (p < 0.05).

The calculated excess lifetime cancer risk (ELCR) in fruits from Hartbeespoort; Mahikeng and Pretoria are shown in Table 4.27 and illustrated in Figure 4.27. The results of ELCR from Hartbeespoort, Mahikeng and Pretoria in fruits ranged from 0.006 to 0.011 mSv.y⁻¹; 0.006 to 0.010 mSv.y⁻¹ and 0.007 to 0.010 mSv.y⁻¹ with the mean and standard deviation (mean ± S.D) of 0.009 ± 0.002 mSv.y⁻¹; 0.007 ± 0.002 mSv.y⁻¹ and 0.008 ± 0.001 mSv.y⁻¹, respectively.

The excess lifetime cancer risk in vegetables in Table 4.28 and Figure 4.29 from Hartbeespoort, Mahikeng and Pretoria ranged from 0.007 to 0.013 mSv.y⁻¹; 0.010 to 0.020 mSv.y⁻¹ and 0.007 to 0.014 mSv.y⁻¹ with the mean and standard deviation (mean ± S.D) of 0.010 ± 0.002 mSv.y⁻¹; 2.495 ± 0.574 mSv.y⁻¹, and 0.011 ± 0.002 mSv.y⁻¹, respectively. The highest ELCR was observed in potatoes from Mahikeng and the lowest in Pretoria. The excess lifetime cancer risk value in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria are lower than the world safety limit of 2.9×10^{-4} mSv.y⁻¹ (UNSCEAR, 2000). The analysis of Excess lifetime cancer risk (ELCR) with p-value of 0.39359 obtained in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria showed there is no significant difference (p > 0.05) and the analysis in vegetables with p-value of 0.02149 showed significant difference (p < 0.05).

CHAPTER FIVE: CONCLUSIONS AND FUTURE RECOMMENDATIONS

5.1 Conclusions

The study of gross alpha and gross beta activities, the activity concentrations of NORM, estimation of absorbed dose rate, annual effective dose equivalent; internal (H_{int}) and external hazard indexes (H_{ext}), annual ingestion dose rate and the excess lifetime cancer risk (ELCR) was conducted in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria markets in South Africa. The gross alpha analysed in fruits from Hartbeespoort, Mahikeng and Pretoria showed no significant difference ($p > 0.05$). The gross beta analysed in fruits from Hartbeespoort, Mahikeng and Pretoria showed a significant difference. The gross alpha and gross beta in vegetables showed no significant difference from Mahikeng, Pretoria and Hartbeespoort ($p > 0.05$). The study also showed that the gross alpha and gross beta activity concentrations obtained were higher than WHO reference of 0.1 Bq.g^{-1} and 1.0 Bq.g^{-1} , respectively (WHO, 2008).

The activity concentrations of ^{238}U , ^{234}U , ^{235}U and ^{232}Th in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria showed a significant difference ($p < 0.05$), however, there was significant difference noted in ^{232}Th ($p < 0.05$). The activity concentrations of ^{238}U , ^{234}U and ^{232}Th in fruits and vegetables were lower than the world values of 35 Bq.kg^{-1} and 30 Bq.kg^{-1} (UNSCEAR, 2000), respectively. The internal indices followed the same trend as those of the external indexes. Hartbeespoort showed the highest and Pretoria the lowest indexes. The external and internal hazard indices in fruits showed significant differences ($p < 0.05$) from Mahikeng, Pretoria and Hartbeespoort ($p < 0.05$) market products and the external and internal hazard indexes in vegetables showed no significant difference ($p > 0.05$) and were lower than unity (UNSCEAR, 2000). The value of unity corresponds to the maximum radium activity of 370 Bq.kg^{-1} for all terrestrial material. The annual ingestion dose of ^{238}U and ^{232}Th showed no significant difference ($p > 0.05$) in fruits. The annual ingestion dose of ^{238}U in vegetables showed no significant difference ($p > 0.05$) and ^{232}Th showed a significant difference in fruits and vegetables ($p < 0.05$).

The current study showed lower annual ingestion doses of ^{238}U and ^{232}Th which are lower than the world annual ingestion dose of $290 \mu\text{Sv.y}^{-1}$ (UNSCEAR, 2000) and the recommended values of $250\text{--}400 \mu\text{Sv.y}^{-1}$ (WHO, 2011). The AEDE results from the current

study in both fruits and vegetables were lower than the world safety limit of 0.48 mSv.y^{-1} (UNSCEAR, 2000). The absorbed dose rates were lower than the world safety limit of 59 nGyh^{-1} (ICRP, 1993). The excess lifetime cancer risk in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria were lower than the world safety limit of $2.9 \times 10^{-4} \text{ mSv.y}^{-1}$ (UNSCEAR, 2000) and showed no significant difference ($p > 0.05$) in fruits and there is significant difference ($p < 0.05$) in vegetables in excess lifetime cancer risk. From these results obtained in the current study, it can be concluded that the consumption of fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria markets in South Africa poses no health threats to human beings.

5.2 Recommendations for future studies

Future study is recommended which will include soil samples collected from the agricultural farms ; further analysis which should include testing of irrigation water for its pH, conductivity, total dissolved substances, and the level of NORM; the type of fertilizer used to determine the concentration of NORM and the absorption rate of NORM by fruits and vegetables (transfer factors). The outcome will ultimately provide evidence-based information, to the policy makers, about the radiological contribution of NORM found in fruits and vegetables from Hartbeespoort, Mahikeng and Pretoria local markets.

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