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Measurement of Radioactive Contamination of Plant Fertilizers Used in Palestine

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This Thesis is Submitted in Partial Fulfillment of the requirements for the Master's Degree in Chemistry, College of Graduate Studies and Academic Research, Hebron University, Palestine.

2023

Hebron University

Faculty of Science and Technology

Graduate Studies in Chemical Sciences

For the Degree of Master of Science in Chemistry

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DEDICATION

I dedicate my success in this project to God, thanks to whom I was able to overcome this stage. To my parents, brothers and sisters who supported me in order to complete this research. To my dear husband, I dedicate this success to him as an expression of my thanks to him for standing by my side so that I can achieve my ambitions and goals. To those loved ones who are always happy with our success. And to my child, the light of my life, Adam.

Dedication to my dear university, Hebron University. To all of my teachers. To my wonderful, beloved country Palestine.

DECLARATION

I certify that this master's degree project is only a result My own work that was done at Hebron University under the supervision of Prof. Dr. Khalil Thabayneh , and this is a higher degree thesis has not been submitted to any other university or institution.

Name: Heba Musbah Hreaz

Signed

Date:

ACKNOWLEDGMENT

In achieving success in this research, there are many people who have supported me and deserve all my thanks.

First of all, I would like to thank God for being able to successfully complete this thesis. Then I would like to thank Professor Khalil Thabayneh for his expert advice and encouragement throughout this project, my friend Lina, who was the first sponsor to register for the master's degree

It was not an easy stage, and without the support of loved ones, it would not have been completed, so all thanks to them.

My dear supportive husband, all thanks to him, and all thanks to the family, relatives and friends for their continuous support to complete the research. Thanks to all the doctors in the college for their help, my colleagues in developing the thesis and people who have willingly helped me out with my abilities.

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LIST OF ABBREVIATIONS

Bq: Becquerel

α: Alpha

B: Beta

γ: Gamma

mSv: Millisievert

CNSC: Certified Nutrition Support Clinician

TNT: Trinitrotoluene

H: Hydrogen

Pb: Lead

Bi: Bismuth

Z: Atomic Number

A: Activity

λ: Decay constant

N: Number of atoms

$t_{1/2}$**:** Half life

U: Uranium

E_{α}**:** Alpha particle Kinetic Energy

MeV: MelionelectronVolt

E_{γ}**:** Gamma Kinetic Energy

Th: Thorium

N: Neptunium

G_y**:** Gray

W_t**:** Tissue weighting factor

W_R**:** Radiation weighting factor

DNA: Deoxyribonucleic Acid

Rn: Radon

Sec: Second

Ra:Radium

NORM: Naturally Occurring Radioactive Material

NPK: Nitrogen, Phosphorous, Potassium

CR-85: Columbia Resins No.85 Laser Radar Detector

CR-115: Columbia Resins No.115 Plastic track detector

CR-39: Columbia Resin No. 39 Plastic Nuclear Track Detector

SSNTD: Soiled State Nuclear Detector

NaOH: Sodium Hydroxide

$\mathbf{E_A}$: Surface Radon Exhalation Rate

$\mathbf{E_M}$: Mass Radon Exhalation Rate

ICRP: International Commission on Radiological Protection

Abstract

Intensive agriculture requires extensive use of fertilizers as a supportive and fortifying source for the soil to increase its productivity. Among these fertilizers used are industrial fertilizers, which are an essential source of nitrogen, phosphorus and potassium. With the increased use of such types of fertilizers, the percentage of environmental pollution with radon gas increases.

Radon is a naturally occurring radioactive element produced from the decay series of uranium. This radioactive gas rises from Earth, enters homes and human lungs.

The research aims to study the level of radioactive contamination in selected samples of common vegetable fertilizers use, due to what these fertilizers cause of an increase in the level of natural radiation in the soil, and the extent of its impact to increase the level of environmental pollution by radiation.

The analysis of radon and radium concentrations for different types of fertilizer samples is important, because of its effects on correcting the consumption of these products and reducing their side effects.

Both radon and radium concentrations, radon exhalation rate in terms of area and mass, as well as the annual effective doses were calculated for 34 types of plant fertilizers. The study was conducted using the solid state nuclear trace detector technology (CR-39) This technique has become an important tool in calculating radon levels in the surrounding environment by using test tube with a diameter of (6.5 cm). The chemical etching process was carried out by NaOH at a concentration of 6.25M for 4 hours at a temperature of 105 °C.

After study it is found, The average values of radon concentration in all collected samples vary from 119.0 Bqm^{-3} in Super phosphate to 570.4 Bqm^{-3} in Agro-Grow with total average values of 270.6 Bqm^{-3} .

The values of average radium concentration are ranged between (13.5-72.1) $BqKg^{-1}$ with total average values of 31.9 $BqKg^{-1}$.

The total average value of the annual effective dose is 9.4 $mSvy^{-1}$.

The surface exhalation rate in these collected samples varies from 7.0 $Bqm^{-2}h^{-1}$ (Super Phosphate) to 33.7 $Bqm^{-2}h^{-1}$ (Agro-Grow) with a total average value of 16.1 $Bqm^{-2}h^{-1}$

The mass exhalation rate has been found to vary from 0.08 $BqKg^{-1}h^{-1}$ to 0.40 $BqKg^{-1}h^{-1}$ with an a total average value of 0.18 $BqKg^{-1}h^{-1}$

The measurements taken in this study represent a baseline database of activity levels that can serve as a reference point for future studies to indicate impacts from future events.

الملخص
قياس التلوث الإشعاعي لأسمدة النبات المستخدمة في فلسطين

تتطلب الزراعة الحديثة استخداما واسعا للأسمدة باعتبارها مصدر غذاء وداعم ومقوي للتربة لزيادة انتاجياتها لتلبية الحاجة المتزايدة للغذاء. ومن ضمن هذه الأسمدة المستخدمة هي الأسمدة الصناعية التي تكون مصدر اساسي للنيتروجين والفسفور والبوتاسيوم . ومع زياده استخدام مثل هذه الانواع من الاسمدة تزداد نسب التلوث البيئي بغاز الرادون.

الرادون هو عنصر مشع ينتج من سلسلة اضمحلال اليورانيوم والتي تحدث بشكل طبيعي. يرتفع هذا الغاز المشع من الأرض ، يدخل المنازل ورئتي الإنسان. يهدف البحث إلى دراسة مستوى التلوث الإشعاعي في عينات مختارة من الأسمدة النباتية الشائع استخدامها في فلسطين ، لما تسببه هذه الأسمدة من زيادة الاضرار في التربة والبيئة وتأثيرها على صحة الانسان . يعد تحليل تركيزات الرادون والراديوم لأنواع مختلفة من عينات الأسمدة أمرًا مهما, لما له من تأثيرات على تصحيح استهلاك هذه المنتجات والتقليل من الاضرار الجانبية لها

تم حساب كل من تركيزات الرادون والراديوم ، ومعدل زفير غاز الرادون من حيث المساحة والكتلة ، وكذلك الجرعات الفعالة السنوية لـ 34 نوعاً من الأسمدة النباتية. أجريت الدراسة باستخدام تقنية كاشف التتبع النووي (CR-39) حيث اصبحت هذه التقنية أداة مهمة في حساب مستويات الرادون في البيئة المحيطة وتمت باستخدام أنبوب اختبار بقطر (6.5 سم). تم إجراء عملية الحفر الكيميائي بواسطة هيدروكسيد الصوديوم بتركيز 6.25 م لمدة 4 ساعات عند درجة حرارة 90 درجة مئوية.

بعد الدراسة وجد أن متوسط قيم تركيز الرادون في جميع العينات التي تم جمعها تتراوح من 119.0 بيكريل/ ^{3}م في سوبر فوسفات إلى 570.4 بيكريل/ ^{3}م في Agro-Grow بمتوسط قيم إجمالية قدرها 270.6 بيكريل/ ^{3}م .

ولقد تراوحت قيم متوسط تركيز الراديوم بين (13.5 - 72.1) بيكريل/كغم بمتوسط إجمالي قيم 31.9 بيكريل/كغم .وتبلغ متوسط القيمة الإجمالية للجرعة الفعالة السنوية 9.4 ميلي سفيري/ سنه.

يتراوح معدل الزفير السطحي في هذه العينات المجمعة من 7.0 بيكريل/ ^{2}م ساعه في السوبر فوسفات إلى 33.7 بيكريل/ ^{2}م ساعه في Agro-Grow بمتوسط إجمالي بقيمة 16.1 بيكريل/ ^{2}م ساعه. وتم

الحصول على معدل الزفير الكتلي يتراوح من (0.08 – 0.40) بيكريل/كغم ساعه بمتوسط إجمالي يبلغ 0.18 بيكريل/كغم ساعه .

تمثل القياسات المأخوذة في هذه الدراسة قاعدة بيانات أساسية لمستويات النشاط يمكن أن تكون بمثابة نقطة مرجعية للدراسات المستقبلية للإشارة إلى التأثيرات من الأحداث المستقبلية.

CHAPTER ONE

INTRODUCTION

Chapter 1: Introduction

1.1 Introduction

All life has evolved in an environment filled with radiation. Radioactive materials and radiations have become a part of our daily life. This radiation either comes from cosmic radiation that occurs in space or from radioactive materials that are used naturally on Earth. Together, these are known as background radiation. The forces at work in radiation are revealed upon examining the structure of atoms. Atoms are a million times thinner than a single strand of human hair, and are composed of even smaller particles – some of which are electrically charged. Radiation is high-speed energy that travels through space or through materials. This energy is transmitted either in the form of particles such as alpha and beta particles, or in the form of waves such as light, x-rays and gamma rays. Radioactive substances are called radionuclides, which are atoms that do not reach a stable state, and until they reach a stable state, they emit radiation [1].

Radiation is divided according to energy into two types: ionizing radiation that has sufficient energy to liberate electrons and thus the atom becomes ionized, and the type that does not have the ability to leave the atom charged such as ultraviolet rays and radio waves. Both radioactivity and the radiation it produces existed on earth long before life emerged. In fact, they have been present in space since the beginning of the universe and radioactive material was part of the Earth at its very formation. But humanity first discovered this elemental, universal phenomenon only in the last years of the nineteenth century [2] . Through many human activities, radioactive materials and radiations are produced, and among these activities is the use of x-ray equipment and also through particle accelerators that are used in medicine. Today, radiation is one of the things that are used in several fields, including the medical field, where it is used to diagnose diseases and can be used in high doses to treat cancer, as well as in high doses to kill and sterilize food from harmful bacteria. It is also possible to use radiation in the field of industry through the use of heat resulting from radiation in generating electricity in nuclear power reactors, and for many other industrial purposes [1] . In 1895, Wilhelm Conrad Roentgen, a German physicist, discovered Radiation which he called X-rays that could be used to look into the human body.

This discovery heralded the medical uses of radiation, which have been expanding ever since. Roentgen was awarded the first Nobel Prize in physics in 1901 in recognition of the extraordinary services he had rendered to humanity. One year after Roentgen's discovery, Henri Becquerel, a French scientist, put some photographic plates away in a drawer with fragments of mineral containing uranium [3]

When he developed them, he found to his surprise that they had been affected by radiation. This phenomenon is called radioactivity and occurs when energy is released from an atom spontaneously and is measured today in units called Becquerel's (Bq) after Henri Becquerel. Soon afterwards, a young chemist, Marie Skłodowska-Curie, took the research further and was the first to coin the word radioactivity. In 1898, she and her husband Pierre Curie discovered that as uranium gave off radiation, it mysteriously turned into other elements, one of which they called polonium, after her homeland, and another they called radium, the "shining" element. Marie Curie shared the Nobel Prize in physics in 1903 with Pierre Curie and Henri Becquerel. She was the first woman to win the Nobel Prize a second time in 1911 for her discoveries in radiation chemistry [2].

Figure 1.1 Some scientists who are credited with discovering different types of radiation.

1.2 Ionizing Radiation

Ionization is the process in which atoms become positively or negatively charged by losing or gaining electrons [2]. Ionizing radiation has the ability to remove electrons from their orbits around atoms and creates a problem in the electron and proton balance and gives the atom a positive charge. This type of radiation includes both

natural radiation and man-made radiation from radioactive materials. Particulate radiations include electrons, positrons, protons, neutrons, α-particles, and other ions. With the exception of neutrons, all of these particles are charged and are classified as directly ionizing (if they have sufficient energy) because they directly ionize the medium they are traversing, producing chemical and biological damage [4].

1.2.1 Alpha particles (α)

Alpha particle (α) is the nucleus of helium, it was Ernest Rutherford (considered as the father of nuclear physics) that finally could conclude that the α -particle was the nucleus of the helium atom. A radioactive source that emits α-particles is called an α-emitter. Many of the natural sources consisting of heavy elements, like radium and thorium are α-emitters. alpha particles that are made up of two protons and two neutrons each and that carry a double positive charge. Because these particles have a relatively large mass and charge, they have a limited ability to penetrate materials, as they can be stopped by a piece of paper or a layer of dead skin.

Thus, alpha radiation from nuclear material outside the body does not pose a radiation hazard. However, when materials that emit alpha rays are introduced into the body through the mouth or inhalation, the full alpha energy is absorbed, so alpha radiation is considered as internal danger. An example of a nuclear material undergoing alpha decay to radon-222, which decays to polonium-218 [4].

1.2.2 Beta particles (β)

An unstable nucleus may attain a more stable configuration by emitting a β-particle. In this process a neutron in the nucleus is transformed into a proton and an electron. Beta particles generally have a negative charge; beta particles are very small and can penetrate more deeply than alpha particles. Both natural sources and artificial radioactive sources may be β-particle emitters. The energy of the emitted β-particle is usually much smaller than that of the α-particles. However, most beta radiation can be stopped by small amounts of shielding, such as sheets of plastic, glass or metal. In fact β-particle emission involves a whole spectrum of energies. The reason for this situation is that together with the β-particle, a tiny neutral particle is emitted. This particle was named the "*neutrino*" by the Italian physicist Enrico Fermi. (The term neutrino means a "small neutral particle"). When the source of radiation is outside the

body, beta radiation with sufficient energy can penetrate the body's dead outer layer of skin and deposit its energy within active skin cells. However, beta radiation is very limited in its ability to penetrate to deeper tissues and organs in the body. Beta-radiation-emitting nuclear substances can also be hazardous if taken into the body. An example of a nuclear substance that undergoes beta emission is tritium (hydrogen-3), which decays to helium-3 [4].

1.2.3 Gamma rays (γ)

Gamma rays are high-energy electromagnetic radiation emitted in the excitation of the atomic nucleus. Gamma radiation consists of photons that originate from within the nucleus. Gamma rays often accompany the spontaneous alpha or beta decay of unstable nuclei. X-rays are identical to gamma rays except that they are emitted during rearrangement of the atomic electron structure rather than changes in nuclear structure and X-ray radiation consists of photons that originate from outside the nucleus, and are typically lower in energy than gamma radiation. Gamma rays are characterized by the high ability to penetrate materials deeply, and the radiation intensity can be reduced through the use of very dense materials such as lead and steel. In general, gamma radiation can travel much greater distances than alpha or beta radiation, and it can penetrate bodily tissues and organs when the radiation source is outside the body. Gama radiation can also be hazardous if gamma -emitting nuclear substances are taken into the body. An example of a nuclear substance that undergoes gamma emission is cobalt-60, which decays to nickel-60 [4].

1.3 Sources of Radioactive Isotopes in Environment

1.3.1 Naturally occurring radioactive nuclides

Life has evolved and has a high level of ionizing radiation, and many radioactive isotopes occur and our bodies are accustomed to it. There is no effective way to avoid exposure to these radiations, which cause most human health problems [4].

1.3.1.1 Cosmic radiation

Cosmic rays are a major natural source of external exposure to radiation. Cosmic rays consist of neutrons, electrons, and atomic nuclei that have been accelerated to a very high level. Through the basic composition, the chemical fraction and the nature of the

source area are identified. Cosmic ray isotopes probe more deeply the nature of the source region and the timescales of the injection and initial acceleration [3]. This type of radiation radiates directly to the earth and interacts with the atmosphere and produces other types of radiation. It is the most widespread type of radiation in outer space. But the Earth's atmosphere and the magnetic fusion that takes place greatly reduce cosmic radiation [2]. When particles of cosmic rays reach us, they arrive equally from all directions of the sky, but this does not mean that their sources are distributed evenly around us, most likely that they are dispersed and deflected by magnetic fields until their original motion is lost [3].

1.3.1.2 Terrestrial radiation

The composition of the earth's crust is a major source of natural radiation. And natural sediments are the main sources of this radiation, including potassium-40; uranium-238 and thorium-232 in addition to the radionuclides into which they decay such as radium-226 and radon- 222 have been emitting radiation since before the Earth took its current shape [4].

Outdoor exposure varies greatly from place to place. Studies in France, Germany, Italy, Japan and the United States, for example, indicate that about 95 per cent of their populations live in areas where the average annual dose outdoors varies from 0.3 to 0.6 mSv [2]. However, in some places in these countries people can receive doses higher than 1 mSv annually. There are other places in the world where radiation exposure from terrestrial sources is higher still. For example, on the southwest coast of Kerala, India, a densely populated 55-kilometre long strip of land contains thorium-rich sands, where people receive, on average, 3.8 mSv annually. Other regions with high levels of natural terrestrial sources of radiation are known to exist in Brazil, China, the Islamic Republic of Iran, Madagascar, and Nigeria [2].

1.3.1.3 Internal radiation

"Radiation exposure" refers to the situation where the body is in the presence of radiation. Radiation exposure is classified into internal and external. External exposure is by taking the radiation that comes from radioactive materials on the ground or attached to the surfaces of objects. Either internal exposure is when eating food contaminated with radioactive materials (ingestion), or through inhalation,

absorption through the skin, or entry of radioactive materials through wounds. When these radioactive materials enter the body, their effect will continue on the body until they are eliminated through urine or feces [2].

Trace amounts of radioactive minerals are naturally found in the contents of food and drinking water. For instance, vegetables are typically cultivated in soil and ground water which contains radioactive minerals. Once ingested, these minerals result in internal exposure to natural radiation. Naturally occurring radioactive isotopes, such as potassium-40 and carbon-14, have the same chemical and biological properties as their non-radioactive isotopes. These radioactive and non-radioactive elements are used in building and maintaining our bodies. Natural radioisotopes continually expose us to radiation and are commonly found in many foods, such as Brazil nuts [4].

1.3.2 Artificially occurring radioactive nuclides

In the past decades, there has been a significant increase in the use of radiation, as scientists have begun to use atomic energy in several fields, including military and medical (such as cancer treatment), and from electricity production to local applications (such as smoke detectors) [2]. Humans are also exposed to radiation from medical treatments. Radioisotopes are produced as a by-product of the operation of nuclear reactors, and by radioisotope generators like cyclotrons. Many man-made radioisotopes are used in the fields of nuclear medicine, biochemistry, the manufacturing industry and agriculture. Individual doses from artificial sources of radiation vary greatly. Most people receive a relatively small dose from such sources but a few receive many times the average. Artificial sources of radiation are generally well controlled by radiation protection measures [4].

1.3.2.1 Non-nuclear supplies

Radiation sources are used in a broad spectrum of industrial applications. These include industrial irradiation used for sterilizing medical and pharmaceutical products, preserving foodstuffs or eradicating insect infestation; industrial radiography used for examining welded metal joints for defects; alpha or beta emitters used in luminizing compounds in gun sights and as low-level light sources for exit signs and map illuminators; radioactive sources or miniature X-ray machines used in well logging to measure geological characteristics in boreholes drilled for mineral, oil or gas

exploration; radioactive sources used in devices to measure thickness, moisture, density and levels of material; and other sealed radioactive sources used in research.

Radiation also has industrial uses, ranging from nuclear meters used in road construction to density gauges that measure the flow of material through pipes in factories. Radioactive materials are used in smoke detectors, as well as to estimate reserves in oil fields. Other applications include sterilization, which is performed using large, heavily shielded irradiators. Industrial activities are licensed by the CNSC [4]

1.3.2.2 Medicine supplies

Radiation has many uses in medicine. And the most common place where it is used is x-ray machines, which are used to diagnose diseases or diagnose bone fractures. Another example is nuclear medicine, which uses radioactive isotopes to diagnose and treat cancer. A gamma camera is one piece of medical equipment commonly used in diagnosis .The CNSC regulates these applications of nuclear medicine, as well as related equipment. It also licenses reactors and particle accelerators that produce isotopes destined for medical and industrial applications [4]. There are substantial differences between medical exposure and most other types of exposure. Medical exposure typically involves only a portion of the body, whereas other exposure often involves the whole body. The main general categories of medical practice involving radiation are radiology (including interventional procedures), nuclear medicine and radiotherapy [2].

1.3.2.3 Nuclear weapons and nuclear tests

In 1945, during Second World War , atomic bombs were dropped on Hiroshima and Nagasaki. As a result of the detonation of these bombs, approximately 130,000 people were killed. This incident is considered the only use of nuclear weapons in wars. However, after 1945, many nuclear weapons were tested in the atmosphere, mostly in the northern hemisphere. The period between 1952 and 1962 is considered the most active for testing. More than 500 tests have been carried out, with a total yield of 430 megatons of trinitrotoluene (TNT) equivalent, the last in 1980. In that period, people in the world were exposed to radiation resulting from these tests. The estimated average annual effective dose due to the global fallout from nuclear weapons testing

was 0.11 mSv, and has subsequently decreased to its current level of 0.005 mSv. This exposure will decline only very slowly in the future because most of it is now due to the long-lived radionuclide carbon-14 [2].

Nuclear weapons can be grouped into three categories:

- Pure fission weapons: the kind of weapon used to destroy Hiroshima and Nagasaki.
- Boosted fission weapons: a fission explosion causes a small fusion reaction that reacts back onto the fissioning material with fusion neutrons to "boost" the fission explosion yield.
- Thermonuclear weapons: the X-ray energy from a primary fission stage implodes a separate (secondary) thermonuclear (fusion) stage. This is the "H-bomb" [2].

1.4 Radioactivity

Radioactivity is a natural part of our environment. Present-day Earth contains all the stable chemical elements from the lowest mass (H) to the highest (Pb and Bi). Every element with higher Z than Bi is radioactive. The earth also contains several primordial long-lived radioisotopes that have survived to the present in significant amounts [5].

1.4.1 Radioactive decay law

When radioactive processes occur, either particles or electromagnetic radiation are emitted. The most common forms of radiation emitted are alpha (α) particles, beta (β) particles and gamma (γ) radiation. Nuclear radiation occurs in other forms, including the emission of protons or neutrons or spontaneous fission of a massive nucleus. The activity of a radioactive source (A), i.e. the number of disintegrations per second (Becquerel), is given as [5]

$$A = -\frac{dN}{dt} = \lambda . N \quad (1.1)$$

Where λ is the decay constant and it varies from one isotope to another. N is the number of atoms that, in time, will disintegrate and dN is the change in N during the time interval dt. The negative sign shows that the number remaining is decreasing.

In order to determine how the number of atoms (*N*) decreases with time, the change in *N* must be summed over time. This is done mathematically by integrating, giving:

$$N = N_0 e^{-\lambda t} \quad (1.2)$$

Where N_0 is the number of radioactive atoms at time zero (i.e., when the first measurement was made). By substituting a later time (day, year) for *t* in the equation (1.2) we can solve the equation and determine the radioactivity at the new time. The two equations (1.1) and (1.2) are very important in order to evaluate risks and radiation doses .

1.4.2 Half- life and Mean life time

The period it takes half of any amount of an element to decay is known as its *half-life.* It is usually denoted as $T_{1/2}$. After one half-life the intensity of the radiation has decreased to 50%. After two half-lives only 25 % remains, and so on. Each half-life reduces the remaining amount by one half. The Earth still contains large amounts of naturally occurring radioactive isotopes, such as U-238. For this to occur, the half-lives must be very long. It was noted above that there is a relation between the half-life $T_{1/2}$ and the disintegration constant λ. The relationship can be found from equation (1.2) by setting $N = ½N_0$. This gives [2] :

$$T_{1/2} = \frac{ln2}{\lambda} \quad (1.3)$$

Where *ln 2* (the natural log of 2) equals 0.693. If the disintegration constant (λ) is given, it is easy to arrive at the half-life, and vice-versa. In calculations using radioactive compounds one of these two constants must be known.

The expectation value of the time needed for an initial population of N_0 radioactive nuclei to decay to *1/e* of their original number is called *the mean life.*

$$\tau = \frac{1}{\lambda} \quad (1.4)$$

Where τ is the mean lifetime of the parent nuclei? From this relation, it can be shown that:

$$T_{1/2} = ln2\ \tau = 0.693\ \tau \quad (1.5)$$

Where τ is also the time that would be needed for all the nuclei to disintegrate if the initial activity of the group, $\lambda.N_0$, were maintained constant instead of decreasing exponentially

1.4.3 Decay rates

A rate of decay of the sample, which is also called its activity (*A*) as the decrease in the number of the radioisotope's nuclei per unit time or number of disintegrations per unit time [5]

$$A = \frac{\Delta N}{\Delta t} \quad (1.6)$$

As shown above, the activity of a radioactive source (*A*) is given by equation (1.1)

$$A = -\frac{dN}{dt} = \lambda . N \quad (1.1)$$

1.5 Transformation Mechanism

The paragraphs below show how a variety of decay mechanisms transform an initial (parent) nucleus ${}^{A}_{Z}X$ into different final (daughter) nuclei.

1.5.1 Alpha emission

The parent nucleus ${}^{A}_{Z}X$ emits a particle. The daughter product is 4 mass units lighter and with 2 fewer units of electric charge: $\boldsymbol{{}^{A-4}_{Z-2}X}$. Alpha decay occurs in large nuclei, in which the ratio of proton to neutron is very high. An alpha particle consists of two protons and two neutrons and is a stable particle. Alpha radiation brings the nucleus to a stable state by reducing the ratio of protons to neutrons. Often the nuclei that have a mass greater than the mass of lead are decomposed in this way [5].

The atomic number in alpha decay changes and results in atoms of the decay product that are different from the original atoms, and therefore have different chemical properties. Alpha decay occurs almost exclusively in heavy nuclei, when A is large the daughter product of decay is more stable than the parent. Mass numbers of nearly all emitters > 209 and typical α -particle kinetic energies $E_{\propto}$ = 4 to 6 MeV. When alpha decay occurs, there is a change in the binding energy, and this change appears as the kinetic energy of the alpha particle and the daughter nucleus. Because this energy must be shared between these two particles, and because the alpha particle and

daughter nucleus must have equal and opposite momenta, the emitted alpha particle and recoiling nucleus will each have a well-defined energy after the decay. Because of its smaller mass, most of the kinetic energy goes to the alpha particle [5].

1.5.2 Isobaric Transitions

Nuclear decay to nuclides with the same mass number, can have different atomic number and neutrons number by converting proton to neutron or vice versa.

1.5.2.1 Beta emission

Beta particles are electrons or positrons (electrons with positive electric charge, or antielectrons). Beta decay occurs when, in a nucleus with too many protons or too many neutrons, one of the protons or neutrons is transformed into the other. Nuclei with either too many protons or too many neutrons will undergo β decay via the weak nuclear force.

- A neutron-rich nucleus will undergo β^- emission.
- A proton-rich nucleus will either emit a β^+ particle or be transformed by capturing an atomic electron in a process called electron capture.

Proton decay, neutron decay, and electron capture are three ways in which protons can be changed into neutrons or vice-versa; in each decay there is a change in the atomic number, so that the parent and daughter atoms are different elements. In all three processes, the number A of nucleons remains the same, while both proton number, Z, and neutron number, N, increase or decrease by 1 [5]

In beta minus decay, a neutron decays into a proton, an electron, and an antineutrino:

$$n \rightarrow p + B^- + \bar{v}$$

The β^-particle is an electron (mass 0.0005486 atomic mass unit, 0.511 MeV). It is considered to be created at the moment of decay. A second light particle, -antineutrino ($\overline{\boldsymbol{v}}$) is also created and emitted.

A neutrino, which is a nearly zero-mass, zero-charge particle that is emitted along with each B^-particle, thus conserving energy and momentum in the disintegration process [6] .

It interacts extremely weakly with matter. In beta decay the change in binding energy appears as the mass energy and kinetic energy of the beta particle, the energy of the neutrino, and the kinetic energy of the recoiling daughter nucleus. The energy of an

emitted beta particle from a particular decay can take on a range of values because the energy can be shared in many ways among the three particles while still obeying energy and momentum conservation [5].

1.5.2.2 Positron emission

Positron decay or beta plus emission β^+ change a proton rich nucleus into a more stable isobar. The nuclear charge is decreased by one unit. A β^+ particle is an antielectron, called a positron. It is identical to an ordinary electron except that it is positively charged. Effectively, β^+ decay converts (via the weak nuclear force) a proton into a neutron and a positron. A neutrino is also created in the process [5] .

$$p \rightarrow n + \beta^+ + v$$

1.5.2.3 Electron capture

in the electron capture process the parent nucleus instead of emitting beta particle, capture one of its own atomic electron and emit amonoenergetic neutrino [6]

$$p \rightarrow n + v + E_b + E_\gamma$$

In electron capture, the mass of an atomic electron is converted into energy, whereas, in β^+ decay, energy is required to create a positron. This means that EC can occur when β^+ decay cannot. No particle is emitted in electron capture and so, except for a very small amount of recoil energy of the daughter nucleus, the energy released escapes undetected [5].

1.5.3 Isomeric Transitions

Nuclear decay from nuclei in metastable state to the same nuclei in the ground state by emitting energy in two process: gamma decay or internal conversion process.

1.5.3.1 Gamma ray emission

In gamma decay a nucleus changes from a higher energy state to a lower energy state through the emission of electromagnetic radiation (photons). The number of protons (and neutrons) in the nucleus does not change in this process, so the parent and daughter atoms are the same chemical element. In the gamma decay of a nucleus, the emitted photon and recoiling nucleus each have a well-defined energy after the decay

[5]. The characteristic energy is divided between only two particles. Gamma-ray energies E_γ typically, are in the range 0.1 to 10 MeV and can be determined very accurately with a modern detector. E_γ is characteristic of the emitting nucleus and are widely used to identify radioactive nuclei. Gamma rays of about an MeV do not interact strongly in matter and will penetrate many cm of moderate-density material

1.5.3.2 Internal conversion

This is an alternative to emission whereby an excited nucleus de-excites by ejecting an electron from an atomic orbit. Both emission and internal conversion are due to the action of the electromagnetic force.

1.6 Elements of the Radioactive Series

The radioactive atom sooner or later will emit radiation in order to attain a more stable state. In some cases it turns out that the daughter nucleus is also unstable – and consequently will emit another particle. Thus, we have a whole series of radioactive atoms. Uranium is a typical example. Three very massive elements, Th-232 (14.1 billion year half-life), U-235 (700 million year half-life), and U-238 (4.5 billion year half-life) decay through complex "chains" of alpha and beta decays ending at the stable Pb-208, Pb-207, and Pb-206 respectively [7].

1.6.1 The Uranium-238 decay series

The U-238 series of radionuclides is of relevance in a variety of environmental contexts ranging from the remediation of former uranium mining and milling facilities to the deep geological disposal of solid radioactive wastes. In particular, low and intermediate level radioactive wastes, as defined in some countries, can contain substantial amounts of uranium-series radionuclides [7]. The Uranium decay chain consists of 15 main steps; The U-238 decay chain comprises nine radionuclides with half-lives longer than one day, a succession of shorter-lived radionuclides, and one stable isotope at the end of the chain, Pb-206.

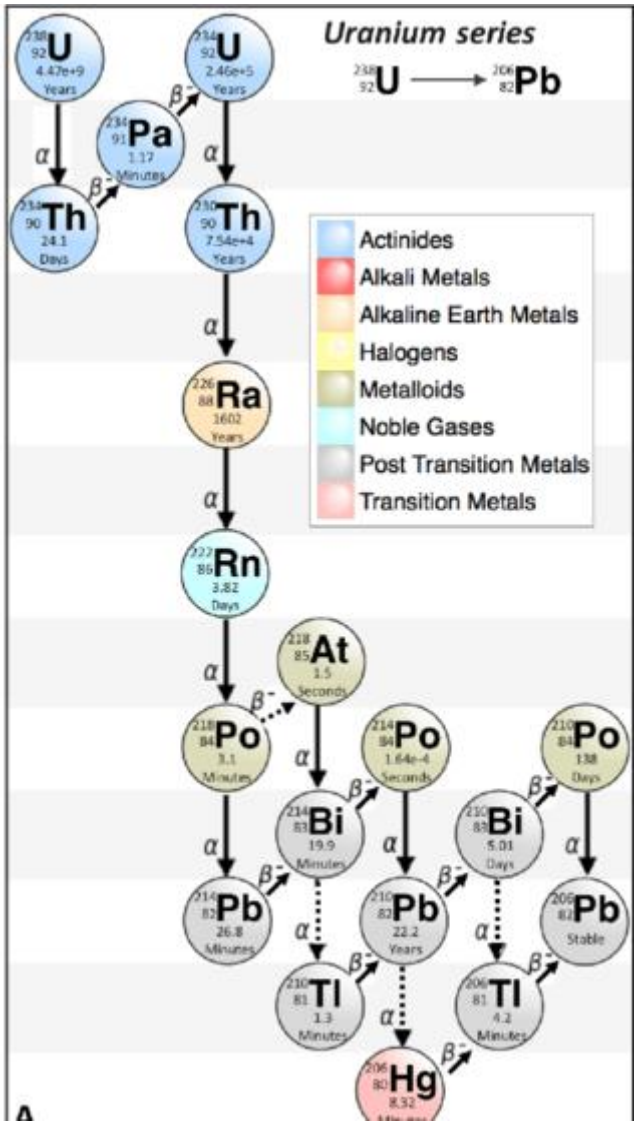

Figure 1.2 The U-238 decay chains [8]

1.6.2 The Thorium-232 decay series

Th-232 is an element that occurs naturally in the earth's crust. The only naturally occurring isotope of thorium is ^{232}Th and it is unstable and radioactive. Thorium in its pure form is a silvery-white heavy metal that is about as dense as lead. In nature, almost all thorium is thorium-232, although several additional isotopes can be present in small amounts. The half-life of thorium-232 is very long. Its low specific activity means that tis isotopes are not highly radioactive. Thorium-232 presents in soil and ores in secular equilibrium with radium-228. The radioactive decay series of ^{232}Th is complex and produces alpha, beta, and gamma radiation. The figure below shows the

important isotopes in the decay series indicates whether the primary decay mode is via alpha or beta emission, and gives the half-life [8]

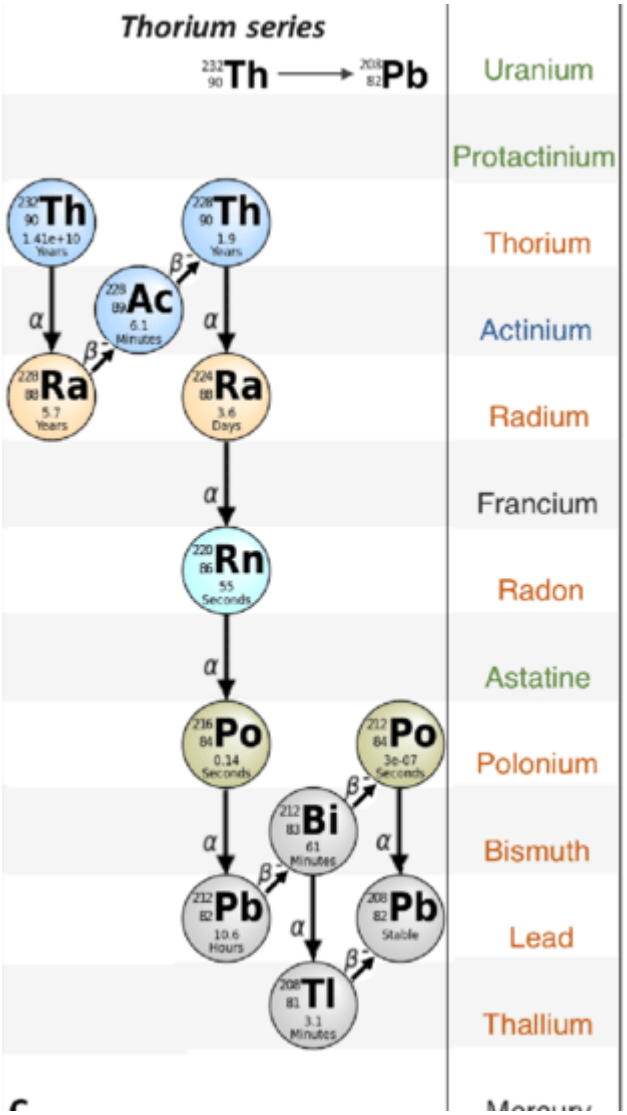

Figure 1.3 The Th-232 decay chains [8]

1.6.3 The Uranium-235 decay series

U-235 is relatively abundant in nature. Typical concentrations of uranium in soil: U-238 1 pCi/g of soil,U-234 1 pCi/g of soil, U-235 0.05 pCi/g of soil. Uranium-235 occurs naturally in small quantities; useful because it fissions, U-235 used in fission bombs because it splits apart more readily than U-232. The U-235 decay chain comprises ten radionuclides with a half-life of 703.8 million years, and one stable isotope at the end of the chain, Pb-207 [8]

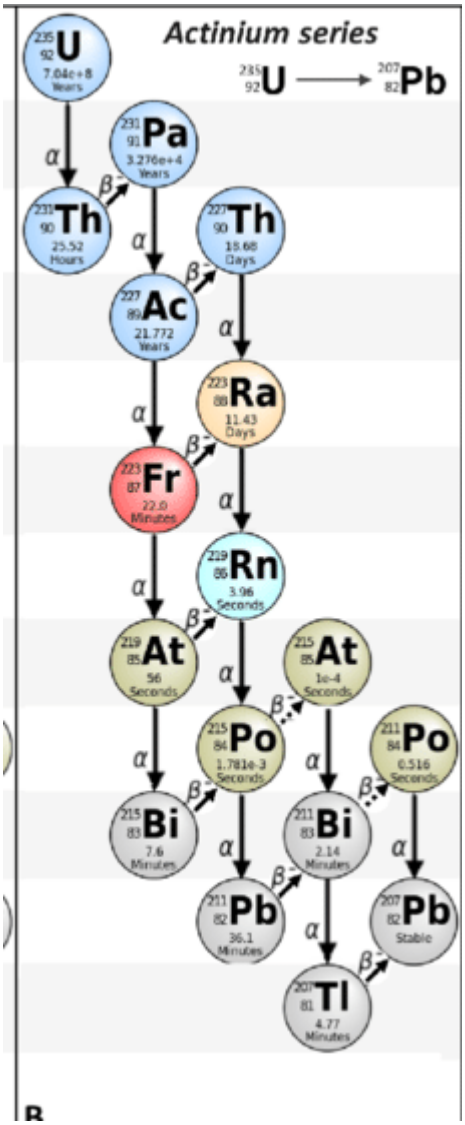

Figure 1.4 The U-235 decay chains [8]

1.6.4 The Neptunium-237 decay series

The radioactive actinide element *Np*-237 is formed by neutron bombardment of uranium (*U*), with more than 50,000 kg of *Np* produced annually as a by-product of nuclear-power generation. Smaller quantities of *Np* can be found as a decay product of americium-241 (*Am-241*) used in ionizing smoke detectors, with trace quantities being produced from nuclear bomb testing, and naturally (from natural neutron capture) in U ores. 237Np sits at the top of the "neptunium decay series" which in turn produces a series of other radioactive elements with medium to very short half-lives, eventually forming *B*i and *Tl*. *Np-237* is extremely mobile in the environment, as it readily forms aqueous solutions (more so than any other actinide), attaches to

particles and colloids, and does not readily become trapped in humus substrates such as soil. Np also has a high affinity for calcium-rich materials including certain clays and concrete, so care is needed with its storage. Although Np is not readily absorbed in the human gut, once in the body, it will pre-concentrate in the bones. The relatively long half-life of Np-237 (~2.14 million years) means that it is persistent in the environment, so it requires suitable containment, and needs to be monitored at low levels [9].

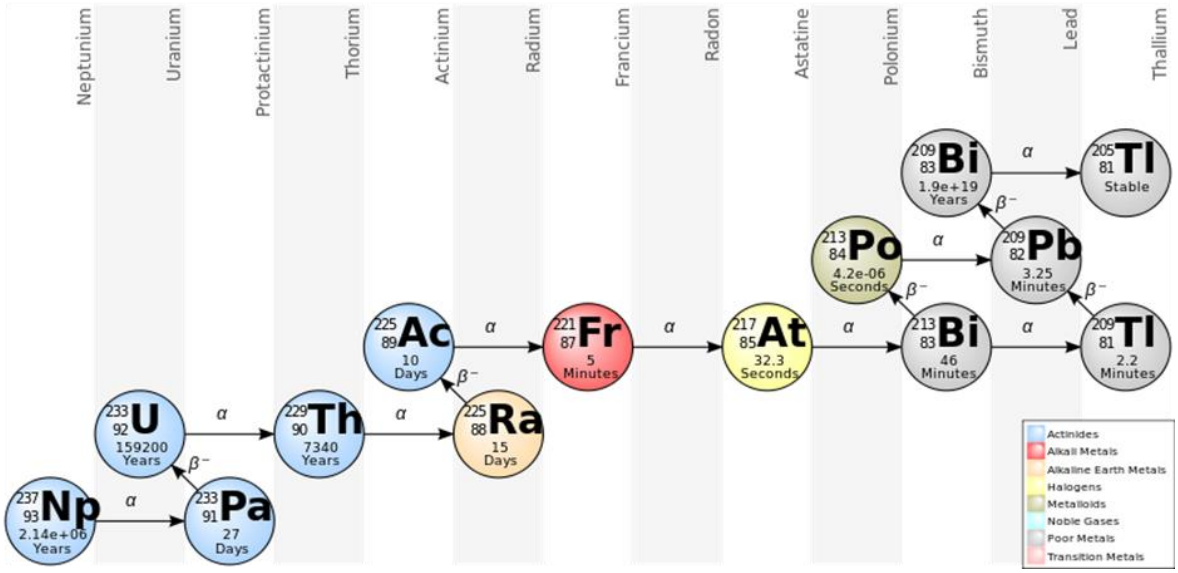

Figure 1.5 The Np-237 decay chains [9]

1.7 The Radiological Effects

For the purpose of radiation protection, dose quantities are expressed in three ways: absorbed, equivalent, and effective.

1.7.1 Absorbed Dose

The effect of radiation depends on the amount you have received. Therefore, amounts of radiation received are referred to as doses; the unit of **ABSORBED DOSE** is specified in terms of the amount of energy deposited by radiation in 1 kg of material. This unit is the gray, abbreviated *Gy*. (It was named in honor of Louis Gray, who was a very big name in the early days of radiation measurement). The gray is a measure of energy absorbed by 1 *kg* of any material, be it air, water, tissue or whatever. A person

who has absorbed a whole body dose of 1 *Gy* has absorbed one *joule* of energy in each *kg* of body tissue [2].

1.7.2 Effective Dose

Tissues and organs differ in their degree of sensitivity to radiation. For example, bone marrow is more affected by radiation than muscle or nervous tissue. To obtain an indication of how exposure can affect overall health, the equivalent dose is multiplied by a tissue weighting factor (W_T) related to the risk for a particular tissue or organ. The sum of the multiplication provides us with the effective dose that the body absorbs. The effective dose is measured in Sieverts. For example, if someone's stomach and bladder are exposed separately to radiation, and the equivalent doses to the organs are 100 and 70 mSv respectively, the effective dose is: (100 mSv x 0.12) + (70 x 0.05) = 15.5 mSv. The risk of harmful effects from this radiation would be equal to a 15.5 mSv dose delivered uniformly throughout the whole body [4]

1.7.3 Dose Equivalent

When radiation is absorbed into living matter, some biological effects will be observed. However, it is not necessary that equal amounts of absorbed radiation have the same biological effects. The effect depends on the type of radiation (e.g., alpha, beta or gamma). For example, 1 Gy of alpha radiation is more effective in tissues than 1 Gy of beta radiation. It is symbolized by the symbol H. The unit of H is the Sievert (Sv). It was named after the Swedish scientist Rolf Sievert, who did a lot of the early work on dosimetry in radiation therapy, rather than Hans Sievert, a different Swede who was once a big name in the Decathlon. In practice, we use the millisievert (mSv) and microsievert (µSv). The absorbed dose is multiplied by a specified radiation weighting factor (W_R). A radiation weighting factor (W_R) is used to equate different types of radiation with different biological effectiveness [4]. Equivalent dose, radiation weighting factor and absorbed dose are related by the expression:

$$H = D \times W_R \qquad (1.7)$$

1.8 Health Effects of Radiation

Since the discovery of radiation, a lot of radiation research has been conducted, which gave a lot of information about the biological mechanisms by which radiation affects

the human body and health. When radiation is absorbed, it causes changes at the level of the cell, either through its death or causing a defect in the DNA in the chromosome [2]

Studies have been conducted on survivors of the atomic bombing of the cities of Hiroshima and Nagasaki, and it has been found that the main effect of radiation exposure is an increase in the incidence of cancer, especially leukemia. As well as obtaining groups of people who are also at high risk of developing cancer, including: workers in uranium mines, workers who manufactured atomic weapons, and people who were exposed to radiation for a therapeutic purpose or to diagnose a specific disease [4]

Studies have shown that exposure to radiation will increase the incidence of naturally occurring cancer [2]. And this increase is higher the higher the radiation dose. Generally, acute doses higher than 50 Gy damage the central nervous system so badly that death occurs within a few days. Even at doses lower than 8 Gy, people show symptoms of radiation sickness also known as *acute radiation syndrome*, which could include nausea, vomiting, diarrhea, intestinal cramps, salivation, dehydration, fatigue, apathy, listlessness, sweating, fever, headache and low blood pressure. And the term acute means effects that occur immediately after exposure to radiation rather than problems that appear after a long period of time [2]. However, victims may survive at first only to die from gastrointestinal damage one to two weeks later. Lower doses may not inflict gastrointestinal injury but still cause death after a few months, mainly from damage to the red bone marrow. Exposure to lower doses of radiation will delay the onset of the disease and also produce less severe symptoms. About half of those who receive doses of 2 Gy suffer from vomiting after about three hours, but this is rare for doses below 1 Gy. High radiation doses to the heart increase the probability of cardiovascular diseases (e.g. heart attacks). Such exposure may happen during radiotherapy, although treatment techniques nowadays result in lower cardiac doses. However, there is no existing scientific evidence to conclude that exposure to low dose of radiation causes cardiovascular disease [2].

CHAPTER TWO

RADON AND RADIUM

Chapter Two: Radon and Radium

2.1 Characteristics of Radon and Radium

2.1.1 What is Radon?

Radon is a noble gas, radioactive in nature, produced naturally from the decomposition of uranium, thorium, or radium, and these metals are radioactive and decompose in rocks, soil, and groundwater. Most of the time people are exposed to radon gas through inhaling it from the air that comes from cracks in buildings and homes. And because this gas comes naturally from the earth, most people are exposed to it continuously, and it is also considered the second leading cause of lung cancer after smoking. Ernest Rutherford is the scientist who discovers radon. He actually discovered the alpha particle radiation given off by radon [10].

2.1.2 Properties of Radon

Radon is one of the noble gases without smell, visible, tasteless and soluble in water. It is one of the heaviest noble gases. Its atomic number is 86 and it has an atomic mass equal to 222g.mol^{-1} . Its half-life is 3.82 days . Radon has a melting point of -71°C, boiling point of -61.8 °C, gas density of 9.73 g/ℓ, specific gravity of the liquid state of 4.4 at -62°C, specific gravity of the solid state of 4, usually with a valence of 0 (it does form some compounds, however, such as radon fluoride). When exposed to cooling below its freezing point, it displays a phosphorescent color. When the temperature drops, the color changes from phosphorous to yellow, and becomes reddish-orange at the air-liquid temperature. Radon buildup is a health consideration when working with radium or thorium, as well as in uranium mines [10].

2.1.3 Radon isotopes and daughters

Of all radon isotopes only two, radon-222 and radon-220 occur in significant amounts indoors. Radon-222 is one of the decay products of uranium-238 (half-life 4.5×10^9 years) and radon-220, nuclide thorium-232 decay (half-life 1.4×10^{10} years). The mother nuclides are radium-226 and radium-224, respectively. Radon is the heaviest of the noble gases and has 12 short-lived isotopes. Rn-222 is the most abundant, consisting of 99 percent of the radon present, and it's the most stable

isotope and called thoron and emanates naturally from thorium. Thoron is an alpha-emitter. Radon-219 is called Actinon and emanates from actinium. It is an alpha-emitter with a half-life of 3.96 sec [10].

2.1.4 What is Radium?

Radium (Ra) is a radioactive element of the heavy alkaline earth metals, located in group two of the periodic table. It is a silvery white metal. Radium is found in all ores that contain uranium, and it can be produced from uranium refining. Radium was discovered (1898) by Pierre Curie, Marie Curie, and an assistant, G. Bémont. However, it was used in many daily products such as toothpaste and in the wrist watch, and they were believed in the past to have therapeutic properties until its radioactivity was discovered, which causes health damage [11]. One of the most important uses of radium was in medicine, mainly in the treatment of cancer by exposing tumors to gamma rays resulting from its radioactivity [12].

2.1.5 Properties of Radium

The chemical properties of radium can be expected because it is an alkaline element, but what distinguishes it most is its intense radioactivity. Compounds containing radium give off a bluish glow in the dark, a result of their radioactivity in which emitted alpha particles excite electrons in the other elements in the compound and the electrons release their energy as light when they are de-excited. Radium is a highly reactive metal and always exhibits its group oxidation state. The Atomic number of Radium is 88 and it has an Atomic mass of 226.0254 g.mol^{-1}. Radium has a melting point of 700 °C, boiling point of 1140 °C, gas density of 5 g.cm^{-3} at 20°C. Radium is a bright, silver-colored, soft, and intensely radioactive element. When exposed to air, it quickly oxidizes, and turns from white to black. Radium corrodes in water to form radium hydroxide [11].

2.1.6 Radium isotopes and daughters

Radium isotopes are used in several fields, including: tracking the movement of groundwater and also to know the masses of ocean waters, but these radionuclides (and their daughters) are also useful chronometers for the determination of the time scales of other Earth and environmental processes [13]. Radium has 34 isotopes, all of

which are radioactive, and their half-lives are less than a few weeks, with the exception of radium-226 (1600) years, and radium-228 (5.75) years. Radium-226 is found in nature as a result of the decay of uranium-238. Thus radium is found in all uranium ores, but it is spread more widely because it forms water-soluble compounds; Earth's surface contains an estimated 1.8×10^{13} grams of radium. Because all isotopes of radium are radioactive and are short-lived, any primordial radium would have vanished long ago. So radium is naturally produced only from the three natural radioactive decay series (thorium, uranium, and actinium series). Radium-226 is a member of the uranium-decay series. Its parent is thorium-230 and its daughter radon-222 [11].

2.2 The Radon Sources in Environment

Radon comes from the natural (radioactive) breakdown of uranium in soil, rock and water and gets into the air you breathe. It can get into any type of building—homes, offices, and schools—and result in a high indoor radon level. Radon ultimate source is uranium.

2.2.1 Sources of radon in rocks

Radon and radium are found in all rocks and soils, because they are daughter products of uranium, which is already present in rocks. In 2019, about a quarter of a million Americans were diagnosed with lung cancer, and 143,000 people died from the disease. The use of tobacco was the cause of these deaths, but the presence of rocks that contain natural uranium, which decomposes with time to produce radon gas, was also a partial cause. Radon is the second leading cause of lung cancer in the United States. Genetic carcinogens are formed such as uranium, which is present in the rock, and then begins to decompose into radioactive decomposition products, and enters the houses from the holes that are present in the foundations. The Environmental Protection Agency estimates that 1 in 15 homes in the United States has elevated radon levels [14]. Mylonite is a kind of rock that is formed by ductile shear where the temperature is higher than 250°C. When shearing elastic, the rock does not break, but behaves as if it were plastic. The development of Mylonite changes the microstructure, porosity, and chemical properties of the parent rock. These changes can influence the mobility and concentration of uranium and may promote the

emanation of radon. One of the properties of Mylonite is to reduce the size of the minerals in the grains, and this is achieved through plastic processes such as the crystals undergoing ductile deformation that reduces their size. Granulation and fracturing play a minor role, especially with more resistant minerals such as uraniferous titanite and zircon. When the grain size of these minerals is reduced, the uranium is released, and becomes available for chemical reaction and redistribution. Mylonite rocks have strong anisotropy, due to the expansion and alignment of minerals during gradual deformation. This anisotropy may channelize fluids, which dissolve or replace minerals and thus change both the volume and composition of the deformed rock. The liquids may also react with the liberated elements while reducing the particle size. As a result, the bulk composition of Mylonite often differs from that of the parent rock [15].

2.2.2 Sources of radon in soil

Radium is produced through the radioactive decomposition of uranium-238 in the earth, and radium decomposes into radon. Part of the radon is released to rocks and soil. The higher the amount of uranium in the soil, the greater the amount of radon. The concentration of radon in the soil varies to a depth of less than a meter, and that depends on the weather condition, for example when there is snow or rain, the soil pores are filled with a larger amount of freezing or water, respectively. Thus, the air laden with radon gas becomes difficult to get out of the soil. As a result, the radon concentration increases in the upper layers of the soil. Air pressure also plays a role in the concentration of radon, as the increase in air pressure increases the concentration of radon in the soil. An increase in atmospheric pressure increases the air pressure into the soil pores, which leads to the difficulty of the exit of air containing radon from the soil and remains inside it. When the air pressure decreases, the concentration of radon in the soil decreases due to the increase in leakage. The process of transporting radon from deep in the soil to the surface is called is determined by the gas permeability of the soil and locally occurring flow paths. The more cracks and gaps exist in the underground, the more easily radon propagates [15].

2.2.3 Sources of radon in air

Radon gas is one of the gases present in all places, for example, it is found in soil and rocks. It also results from the burning of coal and fuel. When radon escapes from the soil or from exhaust stacks into the outdoor air, it is diluted to lower levels than in the indoor air [15].

2.3 Characteristics of Fertilizers

2.3.1 Types and classifications

All plants need a certain type of nutrient to keep them alive. In the natural state, these nutrients are found in the soil and are absorbed through the roots of the plant. The right soil contains sufficient quantities of these nutrients. However, some of these nutrients are reduced in amounts because they are used up by plants, or are leached from the soil. So we replace them to maintain perfect growth. Fertilizers can be defined as materials used to increase nutrients in the soil to improve plant growth, and are divided into natural and artificial fertilizers [16].

Naturally Occurring Radioactive Material (NORM) is usually composed of materials that are not normally considered radioactive, but contain naturally occurring radionuclides with activity concentrations above average in the Earth's crust. NORM It can be raw materials, products or waste, and in these cases its radiological damage is accidental and is less than the chemical one. Fertilizers are included among NORM, because they can increase the exposure of the population radioactivity [16].

2.3.1.1 Natural fertilizers

Here, the word natural, which refers to fertilizers, means that all the nutrients in the fertilizer are derived from the remains of a living organism. If these products are registered with the Ministry of Agriculture as fertilizers, they will have the grade of fertilizer mentioned on the package labels. These labels may or may not contain the total nutrient value, but what is guaranteed. Some organic materials are often registered and sold instead of fertilizers as soil conditioners. Soil conditioners do not have a nutrient guarantee, even though various amounts of plant available nutrients are usually present. Soil conditioners are materials having properties that may improve the soil’s physical condition. Soil conditioners of good quality and high

nutritional value can be used in a cost-effective manner. Nutrients are released from organic fertilizers over a long period of time. This can cause a number of drawbacks, including high costs, odors, and the uncertainty of being able to release the right amount of nutrients at the right time. Some of them may attract animals. Examples include cottonseed meal, fish meal, blood meal, fish emulsion and all animal fertilizers [17]. Fertilizers are generally classified into ‘straight’ or ‘complex/mixed’, based on the presence of one or more primary plant nutrients (nitrogen, phosphorus, and potassium), respectively [16].

2.3.1.2 Artificial fertilizers

Artificial fertilizers are made from petroleum or natural gas. It contains several nutrients, including magnesium, potassium, phosphorus and nitrates. Fertilizers that contain nitrogen, phosphorous and potassium are called NPK fertilizers. Synthetic fertilizers contain a high amount of nutrients, so we need to use small amounts of them, and their manufacture is usually less expensive than natural fertilizers. Also, unlike natural fertilizers, they release nutrients very quickly into the soil [17].

2.3.2 Fertilizers uses

With the huge increase in the world's population, it is important to provide enough crops each year to provide food for all people around the world. Plants such as wheat and maize obtain the nutrients they need for their growth from the soil, and different crops deplete soil nutrients in different ways. Some crops deplete soil nutrients after a few growing seasons. Therefore, fertilizers play an important role in compensating for the lack of nutrients in the soil to obtain good-growing crops. The use of fertilizers has several goals, including providing additional nutrients to plants, as well as improving crop yields. Fertilizers rich in nitrogen are used to green the lawn. Farmers use fertilizers treated with the specific needs of plants to make up for the missing nutrients [17].

2.4 The Production of Radon and Radium in Fertilizers

2.4.1 Radon in fertilizers

Radon-222 is the most important naturally occurring isotope of the radioactive element. It is a decay product of U-238 and has a half-life of 3.82 days. U-238 has a

strong affinity for phosphate ore, which is the main raw material for the production of phosphate fertilizers. High U-238 has been reported in phosphate ores of sedimentary origin. The latter also has some amount of natural radioactivity for Th-232 and K-40 due to a geological reason. The fertilizers derived from the phosphate rocks further contain naturally occurring radioactive nuclides, which are members of the thorium and uranium decay series. These radionuclides will reach the crop through the absorption of these fertilizers, and through the consumption of these agricultural products, they will reach the human being and pose risks to his health. The average of the worldwide annual effective dose from the natural sources of radiation in areas of normal background is reported to be 2.4 mSv. An amount of 1.275 mSv is estimated to be contributed by radon [18].

2.4.2 Radium in fertilizers

Commercial fertilizers are primarily composed of materials containing biologically available nitrogen, phosphorus and potassium. Radium, uranium and thorium are among the most important radioactive elements found in these fertilizers. Phosphorus is a mineral that contains U-238 series radionuclides in quantities that cannot be ignored; In the production of phosphoric acid by the wet process, the radioactive balance of phosphate rock is disrupted, which leads to a redistribution of radionuclides. Through previous studies, it was found that these nuclides migrate based on their solubility in phosphoric acid or phosphogypsum. Accordingly, Ra-226 is incorporated in the phosphogypsum, as its chemical behavior is similar to calcium; Pb-210 and Po-210 also concentrate into phosphogypsum. The behavior of Thorium depends on the chemical reaction and it is distributed in the two reaction products. Uranium isotopes form highly soluble compounds which are mostly combined into phosphoric acid [16].

2.5 Health Effect of Radon

2.5.1 Health risks of radon

Radon is a major cause of lung cancer. It is estimated that radon causes between 3% to 14% of all lung cancers in a countries. The higher incidence of lung cancer was first observed in uranium miners exposed to very high concentrations of radon. In addition, studies in Europe, North America and China have confirmed that even low

concentrations of radon expose people to health risks and increase the incidence of lung cancer [19]. When radon gas is inhaled, it breaks down into radioactive particles and sticks to the lungs. As they decay further, these particles release tiny bursts of energy. This can lead to lung tissue damage and lung cancer over the course of your life. Not everyone exposed to radon gas in large quantities will develop lung cancer. And the amount of time between exposure and the onset of the disease may be many years. Like other environmental pollutants, there is some uncertainty about the magnitude of radon health risks. However, we know more about the dangers of radon than any other carcinogen. This is because the dangers of radon are based on studies of cancer in humans (underground miners). Smoking with radon is a particularly serious health risk. Stop smoking and lower your radon level to reduce your lung cancer risk. Children have been reported to be at greater risk than adults for certain types of cancer from radiation, but there is currently no conclusive data on whether children are at greater risk of radon than adults [20].

2.5.2 Mechanism of lung cancer induction

Cancer is one of the most common causes of death after cardiovascular diseases, accounting for 20 percent of all deaths. The incidence of cancer in the general population is about four out of every ten people, even if they were not exposed to radiation. In recent years, lung, prostate, colon and stomach cancer were among the most common cancers among men. And breast, uterine, lung, stomach and colon cancer among women [4]. Lung cancers are characterized by abundant genetic diversity with relatively few recurrent mutations occurring at high frequency. However, the genetic alterations often affect a common group of oncogenic signaling pathways. There have been vast improvements in our understanding of the molecular biology that underpins lung cancer in recent years and this has led to a revolution in the diagnosis and treatment of lung adenocarcinomas based on the genotype of an individual's tumor.

Lung cancer is known to spread rapidly in its early stages, as well as the possibility of spreading to a group of organs. Because lung cancer patients are diagnosed at an advanced stage, multiple metastases would have already developed, making targeted therapy extremely difficult and systemic therapy less effective [21].

2.6 Radionuclides in Fertilizers

During the past decades, agricultural activities have increased on a large scale, and this has helped to increase the use of chemical fertilizers, which provide the chemical and nutritional elements that the plant needs. Fertilizers have become essential to the agricultural field all over the world. Phosphate rock with potassium and nitrogenous compounds are the basic raw materials used for fertilizers. More than 30 million tons of phosphate fertilizers per year are consumed worldwide, which increases crop production. However, a possible negative effect of fertilizers is the contamination of cultivated lands by some naturally occurring radioactive materials. The natural radionuclide of fertilizers consists mainly of uranium and thorium series radioisotopes and natural K-40. Rocks that contain phosphate are the raw material for the production of all products consisting of phosphate and the main source of phosphorous used in fertilizers. Phosphate rocks consist of sediments, which constitute about 85 percent of these rocks, and the rest of the phosphate rocks are of volcanic origin. The concentration of U-238 and its decay products tends to be elevated in phosphate deposits of sedimentary origin, where typical ^{238}U series concentration is about 1500 Bq. Therefore, when this rock is processed into phosphates' fertilizers, most radionuclides come into the fertilizers. Thus, fertilizers redistribute naturally occurring radionuclides at trace levels throughout the environment and become a source of radioactivity [22].

CHAPTER THREE

SAMPLING, METHODOLOGY AND EXPERIMENTAL TECHNIQUES

Chapter Three: Sampling, Methodology and Experimental Techniques

3.1 Fertilizer samples

In order to study the radioactive contamination in plant fertilizers, thirty four samples of natural and artificial fertilizers, widely used in the cultivation of crop in Palestine, were collected. These samples were placed under the same conditions to achieve the principle of homogeneity in the distribution of radioactive material

Methodology 3.2

3.2.1 Samples sources

A total of 34 plant fertilizers samples were collected from different sites in the Hebron province in Palestine. Sixteen of these samples were brought from the Faculty of Agriculture at Hebron University, ten samples from the Red Rose Nursery in Hebron, and eight samples from animal farms.

3.2.2 Samples collection

The samples collected were formed between natural animal fertilizers and synthetic fertilizers that contain nitrogen, phosphorous and potassium

Natural samples were collected from animal farms, and there were 8 samples, including horse, chicken, sheep and pigeon droppings. As for artificial samples, they were collected from agricultural nurseries.

3.2.3 Samples preparation

After the samples were collected, the samples containing large objects were crushed and then placed inside a plastic cylindrical container, all bottles were marked with the name of sample; date and time were written upon them before closing the containers, then the detector was stuck under a cover . The container was then sealed for 80 days; during that time, α particles emitted by radon and their daughters bombarded the CR-39 track detectors, The container's height was 12 cm and its diameter was 6.5 cm, with the distance between the surface of the sample and detector being constant at 1.5 cm [23].

3.3 Measurement technique- passive technique

3.3.1 Solid State Nuclear Track Detectors SSNTDs (CR 39 detectors)

Solid state detectors are used to accurately measure radiation energy. These devices are sensitive to alpha particles more than any other particles emitted from radon, which is largely unaffected by beta and gamma rays. It is also distinguished that it is not affected by humidity or low temperatures.

There are three types of SSNTDs:

- Polyallyl diglycol carbonate, known as CR-39
- Cellulose nitrate, known as CR-85
- Plastic track detector known as CR-115

CR-39 is a better detector when compared to other radon detectors concentration measurement [24].

3.3.2 Preparation of dosimeters

The study will be carried out by adopting the long-term measurement method, as the test tube technique is considered one of the preferred techniques in determining the concentrations of radon, radium and uranium.

We install the detector with dimensions of 1 cm x 1 cm in the center of the bottom surface of a rubber stopper and then put the other cover quickly after lifting the previous seal that does not contain the detector in order to avoid a defect in the state of equilibrium that has been reached, after that I left the system for (80) days, based on the long-term measurement method [24].

3.3.3 Collecting detectors and chemical etching

After the mentioned period, the detectors were collected and exposed to chemical etching in NaOH solution at a concentration of (6.25 N) for 4 hours at a temperature of 70 °C , after which they were washed with distilled water to get rid of suspended impurities, and then dried. A optical microscope with 400 times magnification was used to count the number of tracks per field of view [24]

3.3.4 Detectors scanning counting tracks

The main objective of this process is to count the tracks engraved on the detector. We will find differences in the size and shape of these tracks, where vertically falling alpha particles will form circular pits, and alpha particles falling at an angle will be an elliptical pit. Any small hole or any scratch will be ignored. The real crater can be

identified by moving the microscope slowly up or down and looking for the bright spot.

Using a light microscope at 400 times magnification, the number of tracks per field of view was calculated. About ten fields of view were randomly scanned for each detector, and then the average number of tracks per field of view was calculated [24].

3.4 Theoretical Calculations

3.4.1 Calculations of Radon Concentration in Fertilizers

In order to measurement of radon, radium and uranium concentrations, the calibration constant (*K*) of the system used must be determined. The calibration constant varies from system to system depending on the geometric dimensions of the tube.

The calibration constant (*K*) of the tube used in this study was determined according to the following relationships [26]:

$$C_{Rn} = K\left(\frac{\rho}{t}\right) \qquad (3.1)$$

The calibration process for detectors in this survey was carried out at a nuclear lab in Ain Shams University at the physics department. Since then, the average *K* was calculated:

$$K = \frac{C_0 t_0}{\rho_0} \qquad (3.2)$$

Where C_0 is the activity concentration of ^{226}Ra (solid radon source) equal 800 Bqm^{-3} , ρ_0 is the track density (number of tracks cm^{-2}) in detectors exposed to ^{226}Ra, t_0 is the exposure time (in days) of detectors are exposed to ^{226}Ra, equal 70 days.

$$K = 24.2\left(\frac{day.Bq.cm^2}{track.m^3}\right) \qquad (3.3)$$

The concentration of ^{222}Ra in Fertilizers samples will be calculated in $\frac{Bq}{m^3}$ unit from this equation:

$$C_{Rn} = 24.2\left(\frac{\rho}{t}\right) \qquad (3.4)$$

Where C_{Rn}: the activity concentration of ^{222}Rn in fertilizer sample, ρ: track density, t:exposure time of detectors and equal 80 days, K: is the calibration factor of CR-39 nuclear track detector which equal 24.2 [27].

3.4.2 Calculations of Radium Concentration in Fertilizers

The concentration of Ra (C_{Ra}) in fertilizers samples will be calculated from the following relationship [28]:

$$C_{Ra} = \frac{\rho h A}{k M T_e} \qquad (3.5)$$

Where ρ: is the track density, h: is the distance between the top of the sample and the detector which equal 1.5 cm, A: is the surface area from which radon is exhaled (m^2), M: is the mass of the sample (Kg) and T_{eff}: is the effective exposure time in (hour), which is related to the actual exposure time t, by the equation [24]:

$$T_{eff} = t - \frac{1}{\lambda(1 - e^{-\lambda t})} \qquad (3.6)$$

λ: is the decay constant for radon which equal ($7.56 \times 10^{-3} h^{-1}$)

3.4.3 Calculation of Radon Exhalation rates

The study of exhaled radon is important to understand the relative contribution of the substance to the total radon concentration found in dwelling. The equation used for surface expiratory rate is written in the form [28]:

$$E_A = \frac{CV\lambda}{AT_{eff}} \qquad (3.7)$$

The unit is $Bqm^{-2}h^{-1}$.

$$E_M = \frac{CV\lambda}{MT_{eff}} \qquad (3.8)$$

The unit is $BqKg^{-1}h^{-1}$.

WhereE_A: is the surface radon exhalation rate E_M: is the mass radon exhalation rate, C: is the integrated radon exposure in ($Bqm^{-3}h$), V: is the void volume of the container (m^3), A: is the area of the sample (m^2), M: is the mass of the sample (kg), λ: is the decay constant of radon ($7.56 \times 10^{-3} h^{-1}$).

3.4.4 Calculation of Annual Effective Dose

To calculate the average annual effective dose to which people are exposed to and absorbed from radon, there must be a conversion factor to relate the absorbed dose to the internal occupancy factor. The following equation can be used to predict the dose here for 1 year of radon exposure [29]:

$$AED = C_{Rn} \times F \times T \times Q \qquad (3.9)$$

Where C_{Rn} : radon concentration , F: conversion factor and equal 9 nSv, T: time and equal 8760 hours of a year (Assuming an indoor occupancy factor is about 80% of 8760 hours, which equals 7008 hours and 20% for outdoors, which equals 1752 hours), and Q is the equilibrium fraction (0.6) for outdoors and (0.4) for indoor (30).

CHAPTER FOUR
RESULTS AND DISCUSSIONS

Chapter four: Results and Discussions

4.1 Introduction

In this chapter, the nature of alpha radioactive particles emitted from the decomposition of radium into radon gas in some types of fertilizers is studied using nuclear solid state detectors (CR 39 detectors). Also, the results will be presented and discussed for radon and radium concentration, radon exhalation rate, and the annual effective dose using the laws mentioned in the previous chapter.

Equations 3.1 through 3.9, respectively, were used for calculating Radon concentrations, Radium concentrations, Radon Exhalation Rates and The Annual Effective Doses. All the results are summarized in tables 4.1 to 4.3. The correlation between radon concentration and radium concentration is represented in figure 4.2.

4.2 Results of Radon and Radium Concentrations

Table 4.1, shows the number of samples, the average values of radon concentrations and the average values of radium concentrations in the collected fertilizer samples which are used as a fertilizer for plants in Palestine. The average values of radon concentration in all collected samples vary from $119.0\ Bqm^{-3}$ in Super phosphate to $570.4\ Bqm^{-3}$ in Agro-Grow with total average values of $270.6\ Bqm^{-3}$. The values of average radium concentration are ranged between $13.5 - 72.1\ BqKg^{-1}$ with total average values of $31.9\ BqKg^{-1}$.

Table 4.1: Radon concentration (C_{Rn} in Bqm^{-3}) and Radium concentration (C_{Ra} in $Bqkg^{-1}$) in fertilizer samples

Sample name	No of samples	Mass (kg)	Av. C_{Rn}	Av. C_{Ra}
ACiDAX	2	0.605	317.2	30.0
ARES Ts	2	0.701	231.8	18.9
Potassium sulfate	2	0.714	186.1	14.9
11-8-20	2	0.553	183.0	18.8
Magnesium sulfate	2	0.450	363.0	46.0
Ammonium sulfate	2	0.498	195.2	22.4
13-13-13	2	0.571	234.9	23.5
Mono Ammonium phosphate	2	0.510	125.1	14.0
Super phosphate	2	0.502	119.0	13.5
Aluminum Sulfate	2	0.406	488.0	68.6
12-12-12	2	0.627	231.8	21.1
Agro-Grow	2	0.605	570.4	53.9
Flower BOMB	2	0.753	509.4	38.7
Horse Manure	2	0.422	228.8	30.9
Chickens Manure	2	0.337	131.2	22.2
Sheep Manure	2	0.390	225.7	33.1
Pigeons Manure	2	0.205	259.3	72.1
Total Average			**270.6**	**31.9**

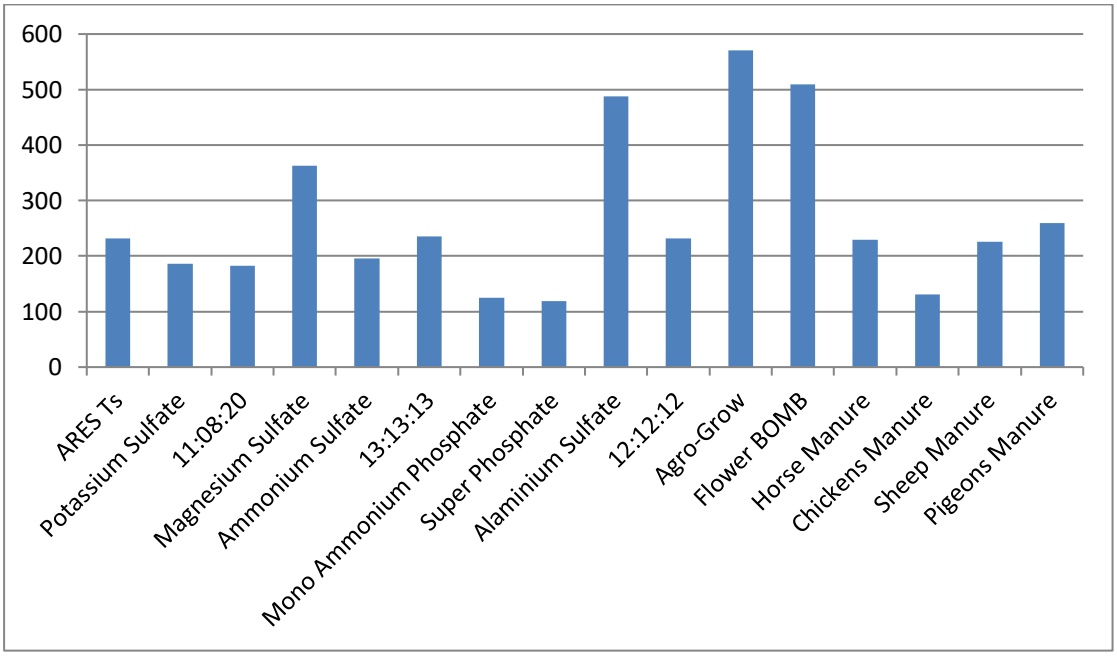

Fig 4.1: The Radon concentration in fertilizer samples.

4.3 The Radon Exhalation Rate

Table 4.2, shows the values of the surface and mass exhalation rates of radon for fertilizer samples which used as fertilizer for plants collected from different sites in Palestine. The surface exhalation rate in these collected samples varies from 7.0 $Bqm^{-2}h^{-1}$ (Super Phosphate) to 33.7 $Bqm^{-2}h^{-1}$ (Agro-Grow) with a total average value of 16.1 $Bqm^{-2}h^{-1}$.

The mass exhalation rate has been found to vary from 0.08 $BqKg^{-1}h^{-1}$ to 0.40 $BqKg^{-1}h^{-1}$ with an a total average value of 0.18 $BqKg^{-1}h^{-1}$.

Table 2: The surface Exhalation Rate (E_A) (in $Bqm^{-2}h^{-1}$) and the Mass Exhalation Rate (E_M) (in $BqKg^{-1}h^{-1}$) in fertilizer samples

Sample name	**Av. E_A**	**Av. E_M**
ACiDAX	20.1	0.18
ARES Ts	13.7	0.11
Potassium sulfate	11.0	0.08
11-8-20	10.8	0.11
Magnesium sulfate	21.4	0.26
Ammonium sulfate	11.5	0.13
13-13-13	13.9	0.13
Mono Ammonium phosphate	7.4	0.08
Super phosphate	7.0	0.08
Aluminum Sulfate	28.8	0.38
12-12-12	13.7	0.12
Agro-Grow	33.7	0.30
Flower BOMB	30.1	0.22
Horse Manure	13.5	0.17
Chickens Manure	7.8	0.12
Sheep Manure	13.3	0.18
Pigeons Manure	15.3	0.40
Total Average	**16.1**	**0.18**

4.4 The Annual Effective Dose

Table 4.3, shows the values of the average annual effective dose, AED_{in} ,AED_{out} and AED_{tot} in fertilizer samples. The average annual effective dose AED_{in} in these samples varies from 3.0 $mSvy^{-1}$ in Super Phosphate to 14.4 $mSvy^{-1}$ in Agro-Grow, with an a total average value of 6.8 $mSvy^{-1}$; The average annual effective dose

AED_{out} in these samples varies from 1.1 $mSvy^{-1}$ in Super Phosphate to 5.4 $mSvy^{-1}$ in Agro-Grow, with an a total average value of 2.6 $mSvy^{-1}$;and the total average annual effective dose in these samples varies from 4.1 $mSvy^{-1}$ in Super Phosphate to 19.8 $mSvy^{-1}$ in Agro-Grow, with an a total average value of 9.4 $mSvy^{-1}$.

Table 4.3: The annual effective dose for indoors and outdoors and the total annual effective dose (in $mSvy^{-1}$) in fertilizer samples

Sample name	**Av. AED_{in}**	**Av. AED_{out}**	**Av. AED_{tot}**
ACiDAX	8.0	3.0	11.0
ARES Ts	5.9	2.2	8.0
Potassium sulfate	4.7	1.8	6.5
11-8-20	4.6	1.7	6.4
Magnesium sulfate	9.2	3.4	12.6
Ammonium sulfate	4.9	1.9	6.8
13-13-13	5.9	2.2	8.2
Mono Ammonium phosphate	3.2	1.2	4.3
Super phosphate	3.0	1.1	4.1
Aluminum Sulfate	12.3	4.6	16.9
12-12-12	5.9	2.2	8.0
Agro-Grow	14.4	5.4	19.8
Flower BOMB	12.9	4.8	17.7
Horse Manure	5.8	2.2	7.9
Chickens Manure	3.3	1.2	4.6
Sheep Manure	5.7	2.1	7.8
Pigeons Manure	6.5	2.5	9.0
Total Average	**6.8**	**2.6**	**9.4**

4.5 Discussions

Alpha particles and radon decay products have negative health effects on humans. Radon and its products can enter the human body through two ways: inhalation and digestion, and then they work to destroy tissues.

Radium is a radioactive element that is widely distributed in water, soil and air. Chemically, it is similar to calcium, it is deposited on the surfaces of bones. When radium is ingested, because the chemical behavior of radium is similar to that of calcium, it is absorbed into the blood through the gastrointestinal tract and followed by the behavior of calcium, it is deposited primarily in the bones. Because radium is a highly radioactive chemical element , there has been an increasing interest in studying

its radioactivity in various plants and food over the last decade. If it consumed or exposed to the body, it can cause major health problems such as sores, anemia, bone cancer, and other problems [2].

Fertilizers are among the important components used in agriculture in Palestine. Thus, it is very important to find the radon activity concentration it has an estimate of radiation risks to the population. The aim of this study is to determine the level of radioactive contamination in the studied fertilizer samples, so the radon concentration inside the samples, as well as the radium concentration as a measure of this pollution. In table 1, the names of the used samples were mentioned, which were classified as synthetic and natural, and their weight were mentioned, as well as the concentration of radon and radium within the samples. It is noted that radon has high concentrations in Agro-Grow and Flower BOMB . The reason for the variation in radon concentration in plant fertilizers is that phosphate fertilizers it contains high concentrations of uranium (where uranium is present in association with phosphate) greater than what is It is found in non-phosphate fertilizers, and phosphate fertilizers contain varying proportions of phosphate compounds, and therefore they vary in uranium and radon concentrations [31].

By comparing the results with the permissible limits for exposure to radon gas published by (ICRP) and amounting to (200-600 Bqm^{-3}) [32] , We find that the results are within the permissible limits.

Referring to previous soil studies in different regions, we find that the average concentration of radon in the Gaza soil is 207.24 Bqm^{-3} [33] , in another study of the soil of Bethlehem, the average concentration of radon was found 145.0 Bqm^{-3} [28], and in a study in Hebron, an average concentrations of radon was found 294, 357, 433 and 512 Bqm^{-3} at different depths [26], while in our study is equal 270.6 Bqm^{-3} .

And by referring to previous fertilizers studies in different places, we find radon concentration 1.39 Bqm^{-3} to 593.06 Bqm^{-3} in fertilizers that used in Iraq [34]. And the activity concentration of naturally occurring radionuclides in fertilizer samples for Radium were 87 $BqKg^{-1}$ in Serbia [35].

The radon exhalation study is important for understanding the relative contribution of the material to the total radon concentration found in the environment. The values of

radon exhalation rate were found well low the world average value of 57.600 $Bqm^{-2}h^{-1}$ (UNSCEAR 2000).

In all studied samples, the annual effective dose is 9.4 $mSvy^{-1}$ It was found that the results are within the normal range (3-10 $mSvy^{-1}$) recommended by ICRP [32]. And the maximum permissible is the upper limit of allowed radiation dose that one may receive without the risk of significant side effects. The annual whole-body dose limit for adult is 50 mSv. For the fetus, the annual maximum permissible dose is 5 mSv [36].

CHAPTER FIVE

CONCLUSION AND RECOMMENDATIONS

Chapter 5: Conclusion and Recommendations

5.1 Conclusion

Natural radioactive isotopes have existed in rocks, air and soil since the formation of the Earth, and because some of these isotopes have a long half-life, large quantities of them are still present on our planet today.
Adding fertilizers to treat crop soils and increase crop yields has become a necessity and a tool used around the world. These fertilizers contain varying amounts of uranium and thorium. Therefore, tons of naturally occurring radioactive uranium and thorium series radionuclides are scattered in the environment due to the high use of fertilizers in agriculture.

Radon concentration levels of the fertilizer samples collected from different sites in Hebron region were measured using the sealed can technique. The radon exhalation rates (both the surface and mass exhalation rates); the radium concentrations, in these samples were determined to assess the radiological hazards. The values of Radon concentrations ranged from (119.0) to (570.4) $Bqm^{-3}$and this variation in the values is due to the difference in the chemical composition and the total average values of Radon concentration is $270.6\ Bqm^{-3}$. The values of radon concentrations are in the range of action levels from 200 to 600 recommended by ICRP (1993).

The total average value of the annual effective dose was $9.4\ mSvy^{-1}$. The value is found in the range of the action levels (3-10 $mSvy^{-1}$) recommended by ICRP- 1993.

By observing the results, we can conclude the following: Plant fertilizers contribute to an increase in radioactive contamination of the environment by increasing the radioactivity of the soil. Plant fertilizers contain varying concentrations of radioactive substances, and these concentrations depend on the substances included in the composition of these fertilizers. Phosphate fertilizers contain high concentrations of uranium compared to other types of Fertilizers, because uranium is associated with phosphates.

As the long-term average concentration of radon increases, so the risk of lung Cancer increase. Smokers are more likely to get cancer caused by radon. The percentage of other types of cancer or other health problems is low, although inhaling radon gas can deliver radiation to other organs, but at a much lower level than the lungs

5.2 Recommendations

Based on the results of this research, it is clear that the excessive or wrong use of fertilizers endangers human health and increases the incidence of various diseases It such as cancer, also has a negative impact on the environment, as it affects the ecosystem, causing an imbalance in the natural balance.

Some recommendations for the safe use of fertilizers:

1- The competent authorities must collect the available facts about the misuse of fertilizers and their negative effects, and indicate the method of proper use of them.

2- Use substitutes for fertilizers if available.

3- Putting supervision of the competent authorities on farmers.

4- Use the least amount of fertilizers possible without affecting the ecological balance.

5- Place masks when using these fertilizers.

6- Increasing researches that study fertilizers, which also study the soil in which these fertilizers are used and the plants grown in this soil.

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