

## **Chapter two**

### **Nuclear decays**

#### **2-1 Introduction**

Sources of ionizing radiation are inside us and surrounding us all the time and everywhere. This radiation comes from radionuclides which occur naturally as trace elements in rocks and soils of the earth as a consequence of radioactive decay. Radionuclides also exist in different ecosystems such as atmosphere, lithosphere, hydrosphere and biosphere. Since, the middle of the last century, the discovery of nuclear radiation a great interest has been made to study the different sources of ionizing radiation depending on their useful applications and their harmful effects on the human being and his environment. In addition to the naturally occurring radioactive materials (NORM), technological enhanced naturally occurring radioactive materials (TENORM) and man-made (artificially produced) radionuclides have been introduced into the environment due to the proliferation of the different nuclear applications. All of these sources have contributed to the increase in levels of environmental radioactivity and population radiation doses, which require an understanding of the different radionuclides environmental behavior and an estimation of their human risks and how to deal with it. Radioecology is concerned with the behavior of radionuclides in the environment. It deals with the understanding of where radioactive materials originate and, how they migrate, react chemically and affect the ecosphere after their release into the environment [73]. All these aspects are very dynamic processes where the environment greatly affects and is affected by the fate of radioactive substances. So, the main goals of studying the radioactivity in environment and food are providing the scientific basis for the effective utilization of radioactivity such as

geochronology, and prediction of the impacts to man and his environment due to different radionuclides.

## **2.2 Environmental radioactivity:**

Radioactivity is part of nature. All materials alive or dead contain a trace of radioactivity, i.e. radioactive elements (radionuclides). These radionuclides have either a natural or anthropogenic origin, and exist in different levels and chemical status in all environmental compartments (lithosphere, hydrosphere and atmosphere), which is known as environmental radioactivity or radioecology. Radioecology is an area of science that deals with the understanding of where radionuclides originate and how they migrate, react chemically and affect the ecospheres [74]. One of the most important aspects of radio-ecological studies is to provide the scientific basis for predicting the impacts of environmental radioactivity on man and his environment. To assess these impacts quantitatively, it is necessary not only to define the sources of radioactivity and their environmental distribution but also to understand the effect of the environmental parameters and how radionuclides are transferred between the ecological systems [75]. Sources of radiation definition and radioactivity measurements are significant to determine the environmental impacts and to estimate radiation doses. This data can then be linked to epidemiological studies that attempt to relate human health effects to the radiation exposures [76].

Environmental radio activity has been an important area of research throughout the 20<sup>th</sup> century that has been promoted by two powerful stimuli, namely: (i) the remarkable power and sensitivity of radionuclides and their descendants as tracers of the rates and mechanisms of environmental and geological processes; and (ii) the introduction of man-made radionuclides and technologically enhanced natural occurring radioactive material (TENORM) as an important class of environmental

contamination. Research involving natural radionuclides has contributed greatly to developing a quantitative understanding of the functioning of the environment. In order to achieve an improved understanding of the environmental fate of contaminant radionuclides, it is necessary to characterize not only the biogeochemical properties of the radionuclides, but also the biogeochemical processes occurring in the receiving environment [77].

### **2.3 Sources of natural radioactivity:**

Radionuclides, radiation and radioactivity has been an essential constituent of the earth since its creation. Radionuclides are classified according to their origins to Naturally Occurring Radioactive Materials (NORM), Technologically Enhanced Natural Occurring Radioactive Material (TENORM) and man-made or anthropogenic radionuclides. Man-made radionuclides do not exist naturally and are created via human activities that vary with time and location according to domestic and/or world nuclear activities and other factors. Sources of man-made radionuclides include nuclear tests, nuclear power plants and reprocessing facilities, sources used for medical, industrial and agricultural applications, and sources used for research purposes [78]. Both NORM and TENORM have the same natural origin but TENORM exists as a result of human activities, such as tobacco smoking, uranium and phosphate mining and milling, air travel, coal fired power plants, oil exploration and others that enhanced and modified the concentration of NORM, their environmental distribution and radiation exposure dose to human-beings. Generally, some of the non-nuclear industrial processes supplied a considerable contribution to the radio-ecological pollution such as phosphate ore mining and phosphate fertilizers manufacture and agricultural applications [79].

NORM has sources of natural radiation and radioactivity that are classified as extraterrestrial and terrestrial sources. In term of population radiation dose, the sources of natural radiation are the most significant and the main contributor to the population collective doses [80].

Natural radiation sources are classified into three categories; cosmic ray, cosmogenic radionuclides and primordial (terrestrial) radionuclides.

Primordial radionuclides are long-lived species, which have been present on earth since its formation about  $4.5 \times 10^9$  years ago. They are classified into;

- (i) Series radionuclides; groups of radionuclides that are headed by parent radionuclides that decay in sequence to other radionuclides with different half lives and decay modes, and finally end as stable isotopes[81]. There are three natural series headed by uranium-238, uranium-235 and thorium-232, the main members of which are shown in figures 2-1, 2-2 and 2-3.
- (ii) Non-series radionuclides; they decay directly to stable nuclide. The most important radionuclides are the isotopes of potassium<sup>40</sup>K, vanadium<sup>50</sup>V, rubidium<sup>87</sup>R, cadmium113 and indium-115. In term of population dose, the most significant radionuclides are <sup>40</sup>K and <sup>87</sup>R.

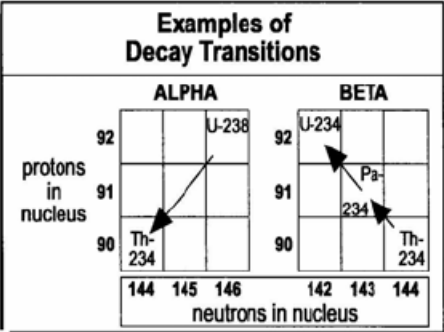
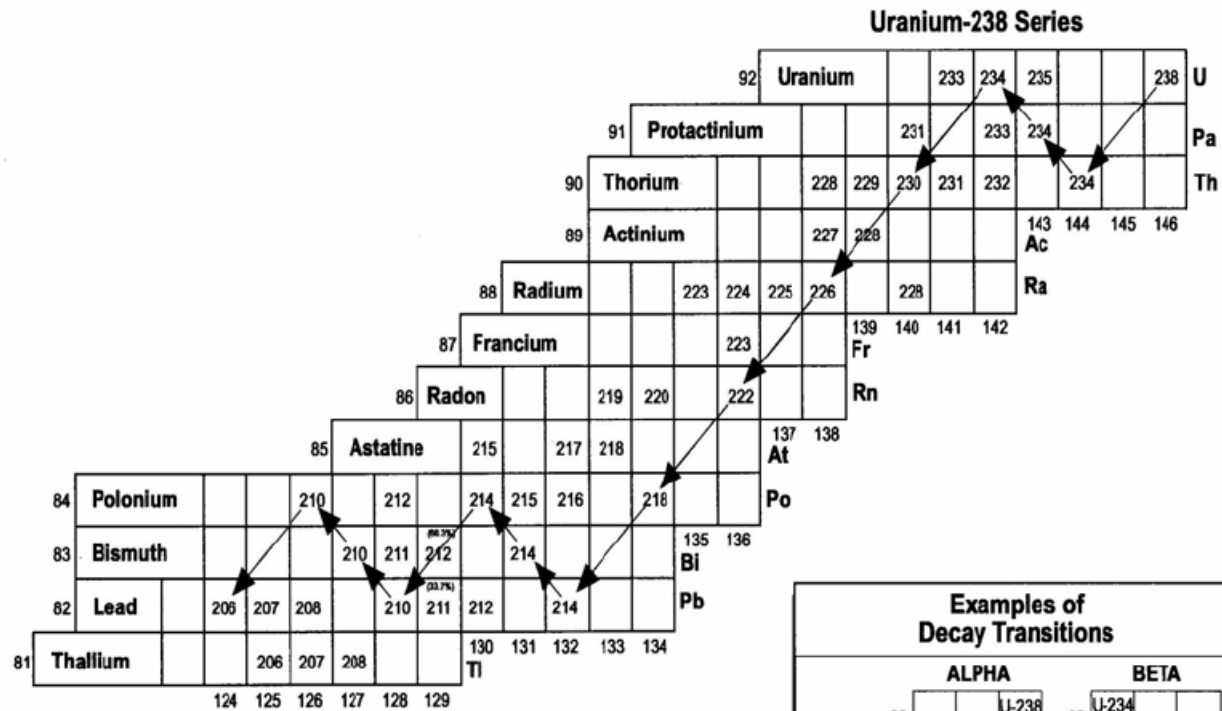


Figure. 2.1: A schematic diagram of the uranium ( $^{238}\text{U}$ ) series [82]





## 2.4 Natural Radioactivity

### 2.4.1 The radioactive decay law

The radioactive atoms in a radioactive substance decay according to a random process. The probability of a nucleus decaying in a time interval is independent of time. It was noted three years after the discovery of radioactivity that decay rate of a pure radioactive substance decreases in time according to an exponential law which is called the Radioactive Decay law [83]. If no new nuclei are introduced into a given radioactive substance, this law predicts how the number of radioactive nuclei which are present at time  $t$  decreases with time. The number,  $dN$ , decaying in a time interval,  $dt$  is proportional to  $N$ , and so

$$\lambda = \frac{(dN/dt)}{N} \quad (2.1)$$

where  $\lambda$  is the decay (disintegration) constant which equals the probability per unit time for the decay of an atom (the right side of equation (2.1)). Regardless of the age of the atoms, this probability is a constant. The exponential law of radioactive decay which is shown below is the solution to the differential equation 2.1 above; i.e.,

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

where  $N_0$  represents the original number of nuclei present at  $t = 0$ . The half-life is the time required for one half of the original nuclei to decay, denoted by the symbol  $t_{\frac{1}{2}}$ . Putting  $N = N_0/2$ , it follows that:

$$t_{\frac{1}{2}} = \frac{\ln 2}{\lambda} = \frac{0.693}{\lambda} \quad 2.3$$

where the mean lifetime is the average time that a nucleus is likely to survive before it decays and equals  $1/\lambda$ . i.e., the reciprocal of the decay constant [83]. The activity,  $A$ , is the rate at which decays occur in a sample



and can be obtained by differentiating Equation 2.4, if the time interval  $dt$  over which the decays take place is much smaller than  $\lambda^{-1}(t_{1/2})$ , i.e.,

$$A = \left| \frac{dN}{dt} \right| = \lambda N_0 e^{-\lambda t} \quad (2.4)$$

$$A(t) \equiv \lambda N(t) = A_0 e^{-\lambda t} \quad (2.5)$$

where  $A_0 = \lambda N_0$ , and is the initial activity at  $t=0$ . The number of decays of the sample per unit time (decays/s) is a convenient unit to measure the radioactivity of a sample. Another unit for measuring the activity is the Curie (Ci) [83]. This unit was originally indicated the activity of one gram of radium but later is defined simply as  $1Ci = 3.7 \times 10^{10}$  decays (becquerel)/s [83,84]. Figure 2.4 shows how schematically the activity decays exponentially with time.

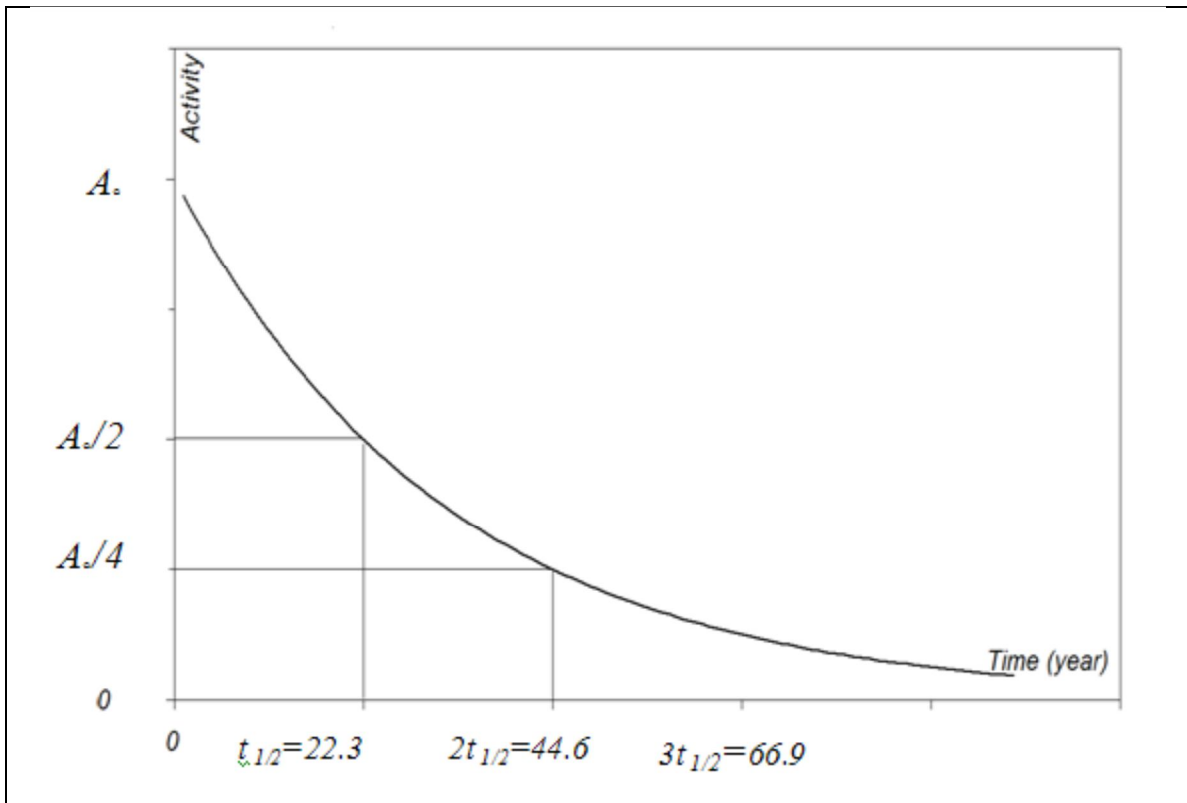


Figure 2.4: Schematic of the exponential decay of activity for  $^{210}\text{Pb}$  ( $t_{1/2} = 22.3$  years)

## 2.4.2 Nuclear Decays Types:

There are three major radioactive decay processes:  $\alpha$ ,  $\beta$  and  $\gamma$  decay.

### 2.4.2.1 $\alpha$ Decay

Alpha ( $\alpha$ ) decay is the spontaneous emission of a helium ( ${}^4\text{He}$ ) nucleus from a heavy nucleus. It is common mode of decay in nuclei with atomic number ( $Z$ )  $\geq 83$ , and mass numbers ( $A$ )  $\geq 209$ . The overall effect of  $\alpha$  decay can be summarized by the following expression [73]:

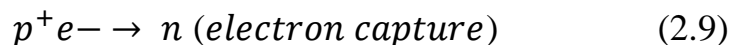
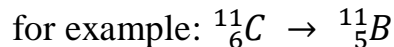
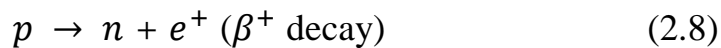
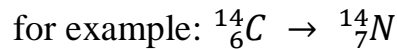


For example,  ${}_{92}^{238}\text{U}$  and  ${}_{88}^{226}\text{Ra}$  decay by  $\alpha$  particle emission, which change them to  ${}_{90}^{234}\text{Th}$  and  ${}_{86}^{222}\text{Rn}$ , respectively.

### 2.4.2.2 $\beta$ Decay

Another common nuclear decay involves the production of a  $\beta$  particle, which is a nuclear particle identical to an electron in mass and charge, in this decay  $Z$  and the neutron number ( $N$ ) of a nucleus each change by one unit, but  $A$  remains constant [80].

There are three processes by which a nuclide may undergo radioactive  $\beta$  Decay:  $\beta$  decay,  $\beta^-$  decay and electron capture. These be simply expressed as follows [81]:



for example:  ${}^{195}_{79}\text{Au} \rightarrow {}^{195}_{78}\text{Pt}$

### 2.4.2.3 $\gamma$ Decay

When  $\alpha$  and/or  $\beta$  decays occur, they will often leave the final nucleus in an excited state. This means that the protons and neutrons in the residual nucleus are not arranged in their lowest energy configuration. The rearrangements of the protons and neutrons in a nucleus can be achieved by  $\gamma$  decay to allow it to regain its lowest energy configuration. For example, when  ${}^{226}\text{Ra}$  undergoes  $\alpha$  decay,  ${}^{222}\text{Rn}$  is produced and this decay is accompanied by  $\gamma$  decay with an energy of 186.21 keV [84,85].

This form of decay has no charge, no mass and no change in the number or type of nucleons in the nucleus (i.e. there are no changes in Z and A numbers).  $\gamma$ -rays are an electromagnetic energy that travel at the speed of light and up to hundred of meters in air before being attenuated [86].

Typical energies for  $\gamma$ -rays emitted from the decay of excited state nuclei range from 0.1 MeV to 10 MeV [85].

The energy of  $\alpha$  and  $\gamma$ - radiation is discrete and well defined, whilst  $\beta$  emission gives  $\beta$ 's with a continuous spectrum due to the varying amount of energy taken away by the neutrino.

In the case of electron capture (EC) the neutron-deficient nuclei must decay by a  $p \rightarrow n$  conversion, but have daughter products whose mass is greater than the maximum acceptable for positron emission. Therefore, these nuclei can only decay by the capture of one of the orbital electrons. The atomic number is then reduced by one unit, due to the negative charge acquired by the nucleus, thus yielding the same daughter that would have been produced by the positron emission.

Figure below presents the decay scheme of  $^{40}\text{K}$  in which the electron capture (EC) is in competition with positron emission; in addition to  $\beta^-$  decay to  $^{40}\text{Ca}$ , the decay to  $^{40}\text{Ar}$  has two competing modes of decay that is  $\beta^+$  and EC. The electron capture is represented by the type of reaction [87]

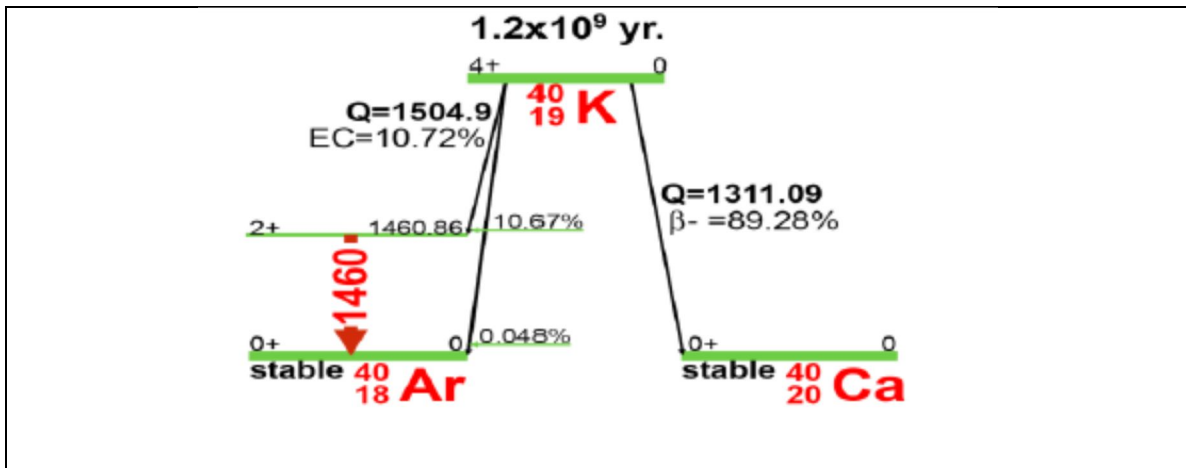


Figure 2.5: Decay scheme of  $^{40}\text{K}$  [86]

### 2.4.3 Interactions of $\gamma$ -rays with Matter

There are three important processes in the attenuation of  $\gamma$ -rays by matter. These are: Photoelectric effect, Compton scattering, and Pair production [88].

Photoelectric interactions are dominant at low energies (up to several hundred keV), and pair production at high energies (more than 5 MeV). Compton scattering being the most important interaction in the mid-energy range as shown in Figure (2.6) [89].

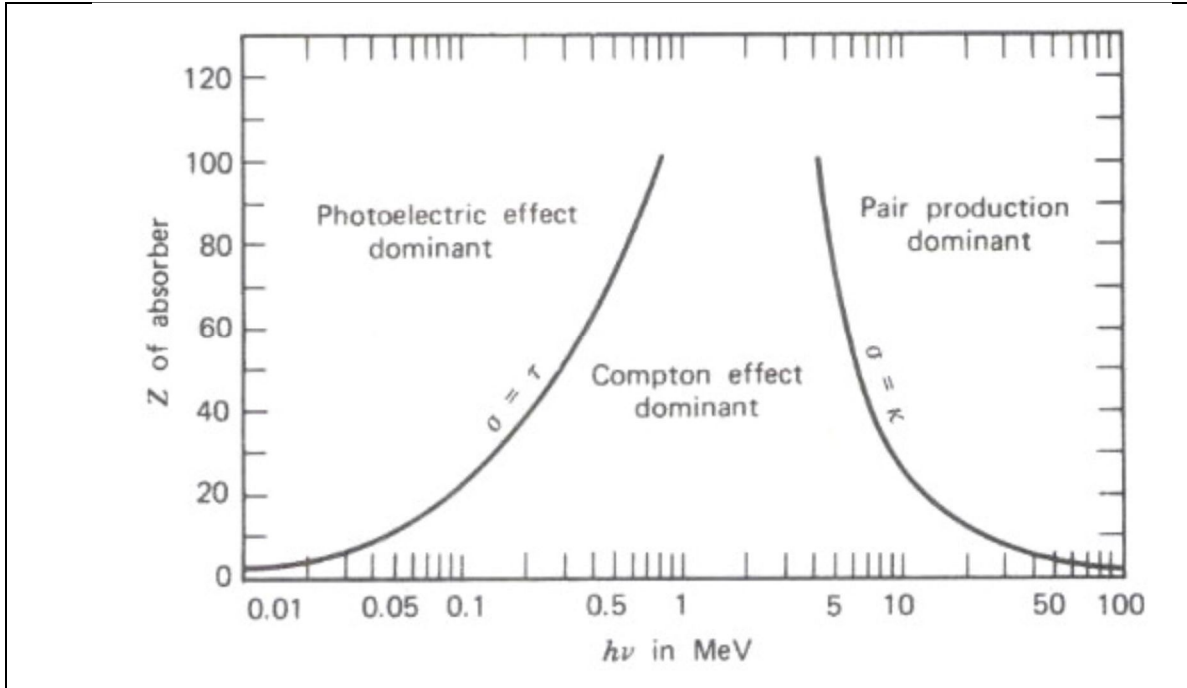


Figure 2.6: Relative importance of the three major types of  $\gamma$ -ray interaction [88].

### 2.4.3.1 Photoelectric Absorption

Photoelectric absorption arises by interaction of the  $\gamma$ -ray photon with one of the bound electrons in an atom. The incident photon transfers all its energy to a bound electron and ejects it from the atom (Figure.2.6) with kinetic energy  $E_e$  given by:

$$E_{e^-} = E_\gamma - E_b \quad (2.10)$$

where  $E_\gamma$  is the  $\gamma$ -ray energy, and  $E_b$  is the energy binding the electron in its shell. The atom now is ionised. Another electron in the atom moves from a higher energy state to fill the vacancy, and during this process a lower energy photon is emitted (called characteristic x-ray photon) in order to carry away the atomic excitation energy. In some cases, an Auger electron may be emitted instead of the emission of characteristic x-ray.

The energy level from which the electron is ejected depends on the energy of the  $\gamma$ -ray. However, the most likely to be ejected is a K-shell electron [88]. Since the energy of  $\gamma$ -ray is transferred to the electron, this allows us to measure directly the energy of the  $\gamma$ -ray and is therefore the most important process for nuclear spectroscopy purposes.

In a spectrum, this process gives rise to well defined peaks called photo peaks. The photoelectric cross section varies approximately as the fourth power of the  $Z$  and inversely as the third power of the photon energy ( $Z^4/E_\gamma^3$ ) [90].

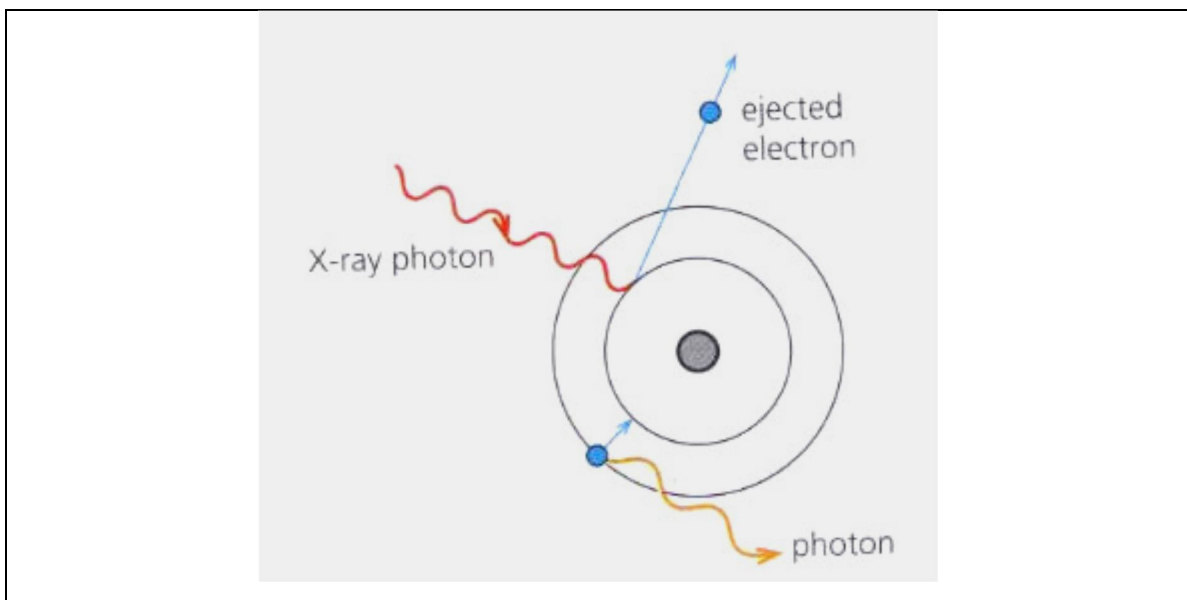


Figure 2.7: The photoelectric effect [90].

### 2.4.3.2 Compton Scattering

Compton scattering is the process in which a photon collides with an electron and is scattered off the electron at an angle. The original energy of the photon is therefore distributed between the  $\gamma$ -ray energy and the recoil

electron as shown in Figure (2.3), the energy of the scattered photon is given by the following equation:

$$E\gamma^- = \frac{E\gamma}{1 + \frac{E\gamma}{m_0c^2}(1 - \cos\theta)} \quad (2.11)$$

where  $E\gamma$  and  $E\gamma'$  are the energies of the  $\gamma$ -ray before and after the collision, respectively,  $m_0$  is the rest mass of an electron,  $c$  is the speed of the light in a vacuum and  $\theta$  is the scatter angle of the  $\gamma$ -ray. While, the energy  $E_{e^-}$  of the electron after the collision is given by the energy Conservation as following:

$$E_{e^-} = E_\gamma - E_b \quad (2.12)$$

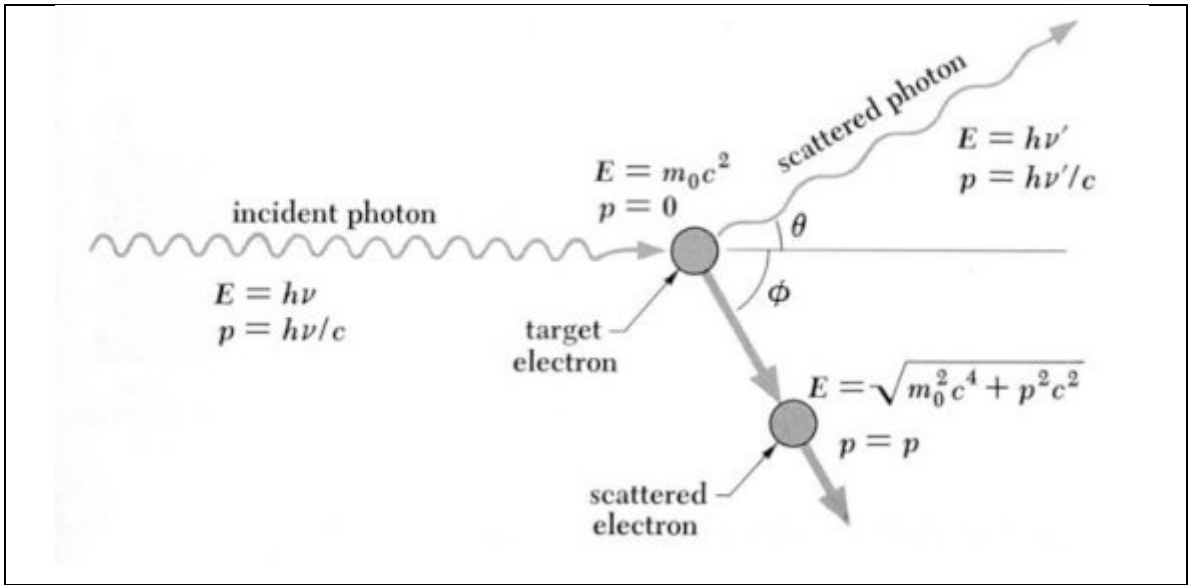


Figure 2.8: Compton scattering [89].

As the above expression depends on the angle  $\theta$ , there is a whole range of energies that the electron can have and the observed continuum in the spectra is known as the Compton continuum. However, one case of interest is when the scattered  $\gamma$ -ray comes out at an angle  $\theta = \pi$ , in other words, it is completely backscattered. By substituting  $\theta = \pi$  into equation 2.11 the energy  $E\gamma'$ BS of the backscattered  $\gamma$ -ray becomes:

$$E'_{\gamma\text{BS}} = \frac{E_{\gamma}}{1 + \frac{2E_{\gamma}}{m_0c^2}} \quad (2.13)$$

The energy of the backscattered  $\gamma$ -ray can then interact in the same way as the original  $\gamma$ -ray and therefore gives rise to an event which known as the backscatter peak. The photon scattering cross-section varies more slowly with energy than does the photoelectric cross-section and is approximately proportional to  $Z$  [88].

#### 2.4.3.4 Pair Production

For incident  $\gamma$  that have energies greater than 1.02 MeV, the most pronounced interaction with matter is pair production. In this process, a photon of sufficient energy gives up all its energy and forms two particles, an electron ( $\beta^-$ ) and positron ( $\beta^+$ ). This process takes place within the Coulomb field of the nucleus [88]. In case if the energy of the incident photon is greater than  $2m_0c^2$ , the excess energy is shared as a kinetic energy between the electron ( $E_{e^-}$ ) and positron ( $E_{e^+}$ ) produced:

$$E_{e^-} + E_{e^+} = E_{\gamma} - 2m_0c^2 \quad (2.14)$$

where,  $m_0$  is the rest mass of an electron or positron. When the energy of the  $\beta^+$  is reduced to near thermal energies, it must unavoidably meet an  $\beta^-$  and the two particles will usually annihilate releasing two 511 keV annihilation  $\gamma$ . The two photons will be emitted at  $180^\circ$  [82]. There is no simple expression exists for the cross-section of the pair production interaction. However, the probability of pair production depends upon  $E_{\gamma}$  and  $Z$  in a complicated way [88,89].



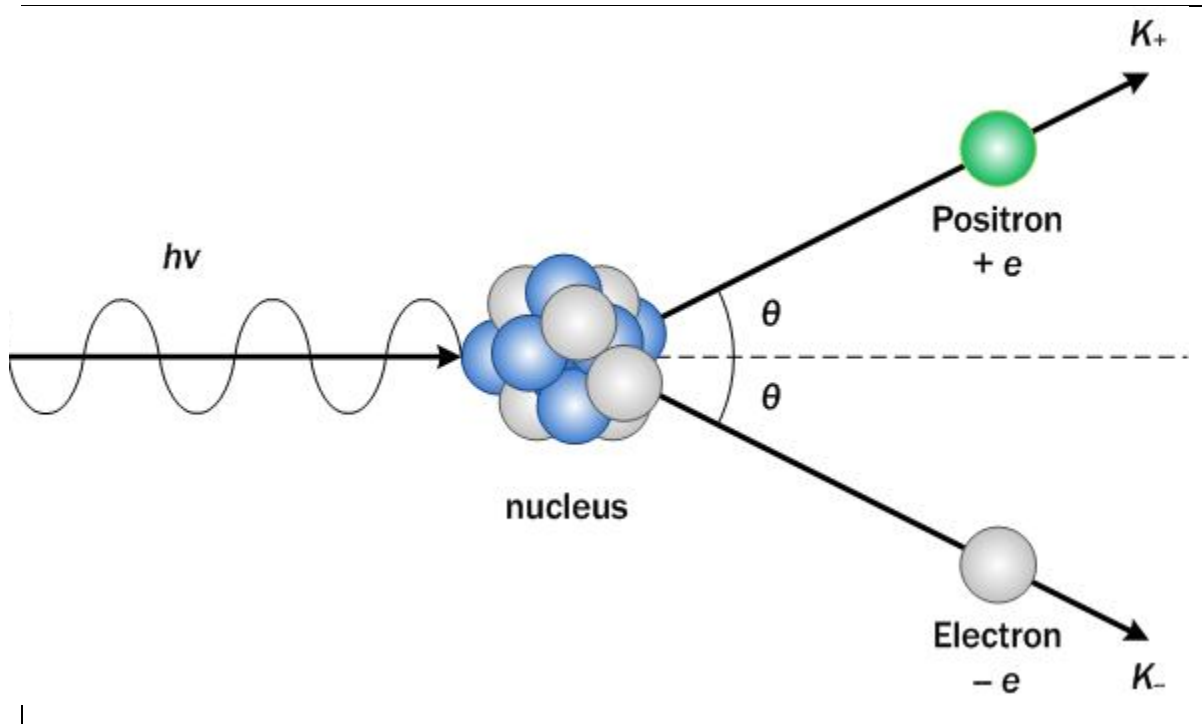


Figure 2.9: Pair production [91].

## 2.5 Natural radioactivity in soil

Soil is the upper part of the earth's crust and is formed as a result of rock deformation by complex physical and chemical processes, which include weathering decomposition, organic matter addition and water movement. Soil consists of minerals, organic matter, water and air; where their percentages vary widely according to soil type, usage, and particle size [92]. Soil has always been important to humans and their health, providing a resource that can be used for shelter and food production. Soils significantly influence a variety of functions (e.g. as a plant growth medium; its importance on the cycling of water; as a foundation for buildings) that sustains the human population. Through ingestion (either deliberate or involuntary), inhalation and dermal absorption, the mineral, chemical and

biological components of soils can either be directly beneficial or detrimental to human health. Specific examples include: cancers caused by the inhalation of fibrous minerals or radon gas derived from the radioactive decay of U and Th in soil minerals. Human health can also be influenced in more indirect ways as soils interact with the atmosphere, biosphere and hydrosphere. Examples include: the volatilization of persistent organic pollutants (POPs) from soils and their subsequent global redistribution that has health implications to the Aboriginal people of the Arctic; the frequent detrimental chemical and biological quality of drinking and recreational waters that are influenced by processes of soil erosion, surface runoff, interflow and leaching; and the transfer of mineral nutrients and PHEs from soils into the plants and animals that constitute the human food chain [93].

Natural radioactivity is wide spread in the earth's environment. It exists in soil, plant, water, air, coal and phosphate. The natural radioactivity in soil comes mainly from  $^{238}\text{U}$  series,  $^{232}\text{Th}$  series and  $^{40}\text{K}$ . Naturally occurring radioactive materials (NORMs) in soil are one of the components of external gamma-ray exposure to which persons are exposed to regularly. Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on geological and geographical conditions and exist at different levels in the soils of each region in the world. The specific levels of terrestrial environmental radiation are related to the geological composition for each lithologically separated area, and to the content of uranium, thorium and potassium in the rock from which the soil originated in each area [94]. Summary of the activity concentration of  $^{238}\text{U}$  ( $^{226}\text{Ra}$ ),  $^{232}\text{Th}$  ( $^{228}\text{Ra}$ ),  $^{40}\text{K}$  and  $^{137}\text{Cs}$  (Bq/kg) in soil samples worldwide is given in table 4.5 in chapter 4 [95].

Elemental distribution in soil is determined by several factors including the parent material, leaching, migration of elements from or within the soil profile, and addition of elements from outside. The parent material is the starting material of the soil and primarily determines its elemental composition. Weathering will modify the elemental composition of the soil. For example, Na, Mg, Ca, K, and Si are apparently lost through leaching. Furthermore, the migration of elements within a profile will occur by processes such as eluviation and eluviations, and by biocycling of plants and animals. These processes change the content of an element in the horizon. Addition of elements will happen by deposition of particles from the air onto the surface of the soil and by applying fertilizer, compost, or sewage sludge [96].

It is expected that the elemental composition of agricultural soils would be affected by agricultural practices such as the application of soil amendments, fertilizers, and pesticides. To compare the compositions of several selected elements in agricultural and non-agricultural soils, the ratios of their contents in the surface horizon to those in the subsurface horizon were calculated. The surface/subsurface ratios for Cr, Ni, Cu, and U in agricultural soils were significantly higher than those in non-agricultural soils, and higher than 1.0 only in agricultural soils, suggesting that the contents of these elements are increased by agricultural activities. These elements are known to be present in considerable amounts in phosphate fertilizers and it is highly probable that their presence in the soil stemmed from phosphate fertilizer application. In addition, sludge, compost and pesticides could be the main sources of these elements in agricultural soils, if these materials have been applied for a long time. The accumulation of U in agricultural soils is also important from the viewpoint of radiation protection. To investigate U accumulation in

agricultural soils in more detail, the concentration ratio of U/Th in soils was examined because the increasing tendency for U to accumulate in agricultural soils becomes clear when the U/Th ratio is used rather than the U content alone. This is because phosphate fertilizers contain 10–200 times more U than that in soils, but the Th content is lower than that in soils. A highly positive correlation is found between the concentrations of Th and U in soils because these elements, which are native to soils, can be expected to behave similarly. Accordingly, the U/Th ratio in the soils was almost constant, although the concentration of U had a wider range. The U/Th ratio and the concentration of U in the surface horizons of agricultural soils were significantly higher than those in non-agricultural soils. Applications of phosphate fertilizer might be the cause of this, thus, the U/Th ratio can be considered a good indicator of the accumulation of U in agricultural soils [97].

## **2.6 Radon gas**

Radon is an inert noble gas. Its most common isotope, and the one that is commonly known as radon, is  $^{222}\text{Rn}$ , which arises in the radioactive decay chain of  $^{238}\text{U}$ . Uranium occurs naturally in varying levels in all rocks and soils. Some fraction of the radon produced in rocks and soils escape to the air; therefore radon is present in the atmosphere. Thus, simply by breathing, people everywhere are exposed to radiation from radon itself and also from short-lived radon decay products (RDPs).<sup>1</sup> Moreover, radon is soluble in water, and groundwater that passes through uranium-bearing soils and rocks contains radon. When radon-rich groundwater is used as drinking water, people are exposed both through water consumption and by radon being released from the water to the air and being inhaled [98].

Radon,<sup>222</sup> Rn-222 ( $t_{\frac{1}{2}}= 3.82$  days), is a daughter product of radium, <sup>226</sup>Ra, which in turn is derived from the longer-lived antecedent, <sup>238</sup>U. Thoron,<sup>220</sup>Rn ( $t_{\frac{1}{2}}=56$  seconds) is a daughter of thorium, <sup>232</sup>Th, which is present in larger amount in the earth's crust than radon. Because of thoron's short half-life, it is essentially all gone before it leaves the ground, and is of no significant radiobiologic consequence. These radionuclide series are present in slowly decreasing amounts in the environment (geologic time scale), due to radioactive decay of their parents, which has been known and understood since the end of the 20<sup>th</sup> century [99].

### 2.6.1 Equations for the radon daughter decay chain

The radioactive daughters of radon follow a decay chain starting with the isotope <sup>218</sup>Po and progressing to <sup>214</sup>Pb, <sup>214</sup>Bi and <sup>214</sup>Po, which decays into the long-lived <sup>210</sup>Pb and effectively ends the chain. The rates of change of the isotopes in the chain are described by the differential equations

$$\frac{dN_1}{dt} = -\lambda_1 N_1 \quad (2.15)$$

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \quad (2.16)$$

$$\frac{dN_3}{dt} = \lambda_2 N_2 - \lambda_3 N_3 \quad (2.17)$$

$$\frac{dN_4}{dt} = \lambda_3 N_3 - \lambda_4 N_4 \quad (2.18)$$

where  $N_1$  is the number of <sup>218</sup>Po atoms,  $N_2$  is the number of <sup>214</sup>Pb atoms,  $N_3$  is the number of <sup>214</sup>Bi atoms,  $N_4$  is the number of <sup>214</sup>Po atoms, and  $\lambda$  is the decay constant, related to the half-life by  $\lambda = \frac{\ln 2}{t_{\frac{1}{2}}}$ . Integrating the differential equations above gives the number of atoms of each isotope as a function of time [100,101]

$$N_1(t) = N_1(0)e^{-\lambda_1 t} \quad (2.19)$$

$$N_2(t) = \frac{\lambda_1 N_1(0)}{(\lambda_2 - \lambda_1)} [e^{-\lambda_1 t} - e^{-\lambda_2 t}] + N_2(0)e^{-\lambda_2 t} \quad (2.20)$$

$$N_3(t) = \frac{\lambda_2 \lambda_1 N_1(0)}{(\lambda_2 - \lambda_1)} \left[ \frac{e^{-\lambda_1 t} - e^{-\lambda_3 t}}{(\lambda_3 - \lambda_1)} - \frac{e^{-\lambda_2 t} - e^{-\lambda_3 t}}{(\lambda_3 - \lambda_2)} \right] + \frac{\lambda_2 N_2(0)}{(\lambda_3 - \lambda_2)} [e^{-\lambda_2 t} - e^{-\lambda_3 t}] + N_3(0)e^{-\lambda_3 t} \quad (2.21)$$

$$N_4(t) = \frac{\lambda_3 \lambda_2 \lambda_1 N_1(0)}{(\lambda_2 - \lambda_1)} \left[ \frac{1}{(\lambda_3 - \lambda_1)} \left( \frac{e^{-\lambda_1 t} - e^{-\lambda_4 t}}{(\lambda_4 - \lambda_1)} - \frac{e^{-\lambda_3 t} - e^{-\lambda_4 t}}{(\lambda_4 - \lambda_3)} \right) - \frac{1}{(\lambda_3 - \lambda_2)} \left( \frac{e^{-\lambda_2 t} - e^{-\lambda_4 t}}{(\lambda_4 - \lambda_2)} - \frac{e^{-\lambda_3 t} - e^{-\lambda_4 t}}{(\lambda_4 - \lambda_3)} \right) \right] + \frac{\lambda_3 \lambda_2 N_2(0)}{(\lambda_3 - \lambda_2)} \left[ \frac{e^{-\lambda_2 t} - e^{-\lambda_4 t}}{(\lambda_4 - \lambda_2)} - \frac{e^{-\lambda_3 t} - e^{-\lambda_4 t}}{(\lambda_4 - \lambda_3)} \right] + \frac{\lambda_3 N_3(0)}{(\lambda_4 - \lambda_3)} [e^{-\lambda_3 t} - e^{-\lambda_4 t}] + N_4(0)e^{-\lambda_4 t} \quad (2.22)$$

## 2.6.2 Numerical Substitution into Decay Equations

Using values for the half-lives of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ , and  $^{214}\text{Po}$  of 185.8s, 26.83 min, 19.9 min, and 164.3  $\mu\text{s}$ , respectively, gives values of the decay constant  $\lambda$  for the isotopes:

$$\lambda_1 = 3.731 \times 10^{-3} \text{ s}^{-1} ( ^{218}\text{Po}), \lambda_2 = 4.306 \times 10^{-4} \text{ s}^{-1} ( ^{214}\text{Pb}),$$

$$\lambda_3 = 5.81 \times 10^{-4} \text{ s}^{-1} ( ^{214}\text{Bi}), \text{ and } \lambda_4 = 4.219 \times 10^3 \text{ s}^{-1} ( ^{214}\text{Po}).$$

Substituting these values of the decay constant into the coefficients in the second set of equations from above gives:

$$N_2(t) = 1.13N_1(0)e^{-\lambda_1 t} + N_2(0) + 1.13N_1(0)e^{-\lambda_2 t} \quad (2.23)$$

$$N_3(t) = 155N_1(0)e^{-\lambda_1 t} + (3.24 N_1(0) + 2.86N_2(0))e^{-\lambda_2 t} + (-158N_1 - 2.86N_2(0) + N_3(0))e^{-\lambda_3 t} \quad (2.24)$$

For  $N_2(t)$ , and  $N_3(t)$ . Equation (2.19) can't be substituted into. Equation (2.25) can be simplified further. Because  $\lambda_4$  has a value at least six orders of magnitude greater than the values of the other decay constants, all terms with the form  $\frac{1}{\lambda_4 - \lambda_n}$  can be approximated as  $\frac{1}{\lambda_4}$ . This allows terms inside (2.25) to be combined, and the formula reduces to

$$N_4(t) = \frac{\lambda_3 \lambda_2 \lambda_1 N_1(0)}{(\lambda_2 - \lambda_1)} \left[ \frac{1}{(\lambda_3 - \lambda_1)(\lambda_4)} (e^{-\lambda_1 t} - e^{-\lambda_3 t}) - \frac{1}{(\lambda_3 - \lambda_2)(\lambda_4)} (e^{-\lambda_2 t} - e^{-\lambda_3 t}) \right] + \frac{\lambda_3 \lambda_2 N_2(0)}{(\lambda_3 - \lambda_2)(\lambda_4)} (e^{-\lambda_2 t} - e^{-\lambda_3 t}) + \frac{\lambda_3 N_3(0)}{\lambda_4} (e^{-\lambda_3 t} - e^{-\lambda_4 t}) + N_4(0) e^{-\lambda_4 t} \quad (2.25)$$

Substituting in numerical values of the decay constants and rearranging, this becomes:

$$N_4(t) = -2.13 \times 10^{-8} N_1(0) e^{-\lambda_1 t} + (4.46 \times 10^{-7} N_1(0) + 3.94 \times 10^{-8} N_2(0) e^{-\lambda_2 t} + (-4.67 - 7N_1(0) - 3.94 \times 10^{-8} N_2(0) + 1.38 \times 10^{-7} N_3(0)) e^{-\lambda_2 t} + (-1.38 \times 10^{-7} N_3 + N_4(0)) e^{-\lambda_4 t} \quad (2.26)$$

### 2.6.3 Concentration of Radon Daughters at Equilibrium

Secular equilibrium is reached when the rates of change of the isotopes in a decay chain are equal and their concentrations are unchanging. At secular equilibrium the rates of decay of all the isotopes in the decay chain are equal. This means that:

$$\lambda_1 N_1 = \lambda_2 N_2 = \lambda_3 N_3 = \lambda_4 N_4 \quad (2.27)$$

Substituting in numerical values shows that at equilibrium among the four daughters:

$$N_2 = 8.66 N_1 \quad (2.28)$$

$$N_3 = 6.42 N_1 \quad (2.29)$$

$$N_4 = 8.8 \cdot 10^{-7} N_1 \quad (2.30)$$

This can be used as a rough approximation for the relative numbers of radon daughters expected on the filter [102].

#### **2.6.4 Units of radon measurement**

One gram of radium was found to decay at a rate of  $3.7 \times 10^{10}$  to the tenth power nuclear disintegrations per second. It was later decided by international agreement that the amount of any radioactive element that will give  $3.7 \times 10^{10}$  to the tenth power disintegrations per second (dps) would be called a Curie, (abbreviated Ci).

A radon-222 gas concentration is commonly measured in pico Curies per liter of air (pCi/L). A picoCurie equates to one-trillionth of a Curie, or a rate of radioactivity indicating 0.037 decays per second (or 2.22 decays per minute). Therefore, one pCi/L represents the concentration of radon 222 atoms per liter of air that will result in 2.22 alpha emissions per minute. Because health effects are primarily due to the radon decay products (RDPs) and not to the radon-222 gas itself, a unique unit of measure exists for quantifying the amount of RDPs in the air[103].

This unit of measure is the working level (WL) and was previously used to measure the occupational exposure of underground miners. The higher the (WL), the higher the risk of adverse health effects. 3-2 A working level is defined as any atmospheric combination of the short-lived radon progeny [polonium-<sup>218</sup>Po , lead<sup>214</sup>Pb, bismuth-<sup>214</sup>Bi , and polonium-<sup>214</sup>Po ] that will deliver  $1.3 \times 10^7$  to the fifth power million electron volts (MeV) of alpha energy per liter of air. If the progeny were in perfect secular equilibrium with the radon gas, that is, if each of the four short lived RDPs were present in the air at the same activity level as the radon, then 1 working level would



be present when there were 100 pCi/L of radon-222 gas (and consequently, 100 pCi/L of each of the progeny). When the maximum possible RDP concentration produced by a given radon concentration is present, the equilibrium ratio (ER) of radon gas to the RDPs would be 1. In practice, the RDPs never reach perfect equilibrium with the radon in homes or other buildings. Due to natural infiltration of outdoor air, all radon atoms do not remain in the house long enough (3 to 4 hours) to reach equilibrium with their progeny. In addition, other environmental factors will not allow all of the RDPs created to remain airborne. RDPs are chemically reactive, are solid particles, and have static electric charges, so they easily attach themselves to breathable particulate matter in the air (e.g., dust, smoke and aerosols). Since a percentage of these RDPs will deposit or plate out on solid objects such as walls, floors, ceilings, furniture and clothing, their airborne concentrations are reduced. The degree to which the progeny approach equilibrium in a specific house can vary significantly. For example, increased air movement will blow more RDPs toward solid surfaces where they will plate out. The concentration of airborne RDPs will be reduced, therefore, and the ER (equilibrium ratio) will decrease. Stagnant air containing an abundance of suspended particles will provide more airborne particles to which the RDPs can attach, creating a higher ER. All of these environmental factors can influence the concentration of airborne progeny without affecting the radon<sup>222</sup>Rn gas concentration, making perfect equilibrium unattainable in home atmospheres. Studies of equilibrium ratios in homes indicate that typically 30% to 70% of the RDPs will be plated out (and therefore not airborne). An ER of 0.50 (or 50%) is commonly assumed to be average. Based on this 0.50 ER assumption, the house with a radon gas concentration of 100 pCi/L would only produce one-half of a working level, meaning it

would take approximately 200 pCi/L to generate one full working level. (However, considering the range of 30% to 70%, 1 WL in any given house could, in fact, correspond to anywhere between 150 and 300 pCi/L.) Based on an average 50% equilibrium ratio assumption, the EPA "action level" of 4 pCi/L is equated to 0.02 WL[104,105].

### **2.6.5 Health Effects of Radon Inhalation**

The adverse health effects of exposure to radon are caused primarily by damage due to Alpha-particles. The possible effects will depend on exposure level. The main danger from high radon exposure is an increased risk of lung cancer. Radon as a noble gas is rapidly exhaled after being breathed in; however, radon progeny combine with other molecules in the air and with particles of dust, aerosols or smoke, and readily deposit in the airways of the lung. While lodged there, the progeny emit ionizing radiation in the form of alpha particles, which can damage the cells lining the airways. Experiments have confirmed that ionizing radiation affecting bronchial epithelial cells could cause cancer. Epidemiological studies on thousands of uranium miners in different countries, including Germany, USA, Canada, Czechoslovakia, and others, also support this fact. Analyses have been undertaken of several miner studies, total including 68,000 men of whom 2,700 died from lung cancer. An increased risk of all histological types of lung cancer, including small cell carcinoma, adenocarcinoma, and squamous cell carcinoma, has been associated with occupational exposure to radon. But exposure to radon in houses can also lead to lung cancer. It is believed, for example, that every year more than 15 000 deaths from lung cancer occur due to radon exposure in the United States and more than 2 500 deaths in the United Kingdom.

The risk of lung cancer from radon in home can be estimated by two ways:

by direct epidemiological studies of residential radon and by projecting occupational risk estimates to lower levels of radon in homes.

The uncertainties associated with residential studies are larger than those with miner studies, primarily because the risk is smaller at the low exposures encountered in most homes [106].