CHAPTER 1

INTRODUCTION

1.1 RADIATIONS AND RADIOACTIVITY: GENERAL INFORMATION

Radiations exist in the environment due to cosmic rays and presence of radioactive material in the earth's crust since the creation of universe. Radiations were referred to as 'act or process of emitting light' or 'beam shining' in the middle of the 15th century. Since the 1560s, these have been referred to as rays or beams released. In the 1650s, its definition changed to 'divergence from a centre' and from that point on, it has been referred to as 'radiations' where 'radi' stands for radius. Nowadays, radiations are defined as form of energy that travels through space or matter in the form of particle or wave. These radiations were thought to come from a variety of natural sources like cosmic rays, terrestrial sources, visible light, the sun and many more.

The modern scientists first discovered this elemental, universal phenomenon only in the late nineteenth century. In 1895, W.C. Roentgen, a German Physicist, photographed his wife's hand with revealing the unmistakable image of her skeleton complete with wedding ring. His wife's had placed her hand in the path of X- rays where beaming an electron ray energy source onto a cathode tube. He was awarded the first Nobel Prize in 1901 for the discovery of these unknown X-rays capable of producing an image on a photographic plate.

After one year, in 1896 Henri Becquerel, a French scientist, put some photographic plates away in a drawer with fragments of mineral containing uranium. When he developed them, he found they had been affected by radiation. This phenomenon later termed as radioactivity by Marie Curie, occurs when energy released from an atom spontaneously and is measured in becquerel (Bq). In 1898, Marie Curie and her husband Pierre Curie discovered that as uranium gave off some radiations and mysteriously turned into another shinning elements, that is polonium and radium. In 1903, Heneri Becquerel, Marie Curie and Pierre Curie were jointly awarded the Nobel Prize in physics for their work on radioactivity. Later Marie Curie was also awarded in 1911 Nobel Prize in chemistry for her discovery of radium. On the basis of simple experiment on penetration power in materials, Rutherford discovered two elements alpha and beta particles in 1899. Gamma rays were discovered in 1903 by Rutherford. In 1907 Rutherford and Soddy has identified several separate series of naturally occurring radionuclides which transform into another radioactive nuclides,

until they eventually ended up as non-active lead. Some kinds of radioactive gases were found emanating from thorium and radium which were later identified as isotopes of thoron and radon. The radioactive process was related to the transition of one nucleus into another by the emission of radiations such as alpha, beta, and gamma. The cosmic rays were discovered by Millica in 1925 (Blaauboer and Smetsers, 1996), which enter the atmosphere consists mainly of nucleonic components comprising of 88% protons, 11% alpha particles and remaining 2% electrons.

1.2 TYPES OF RADIATIONS

Radiations that exist in the environment are categorised into different types based on their energy, ionizing ability, penetration power and sources of emission etc. Based upon the presence of mass and charge, radiations can be either be electromagnetic or particulate in nature. Electromagnetic radiations have no mass and charge but only energy e.g., photons, x-rays, and gamma rays whereas particulate radiations are made up of particles having mass and energy and may or may not have charge e.g., alpha particles, beta particles and neutrons. On the basis of energy, radiations are basically classified into two categories: non-ionizing and ionizing radiations (Figure 1.1) as explained below.

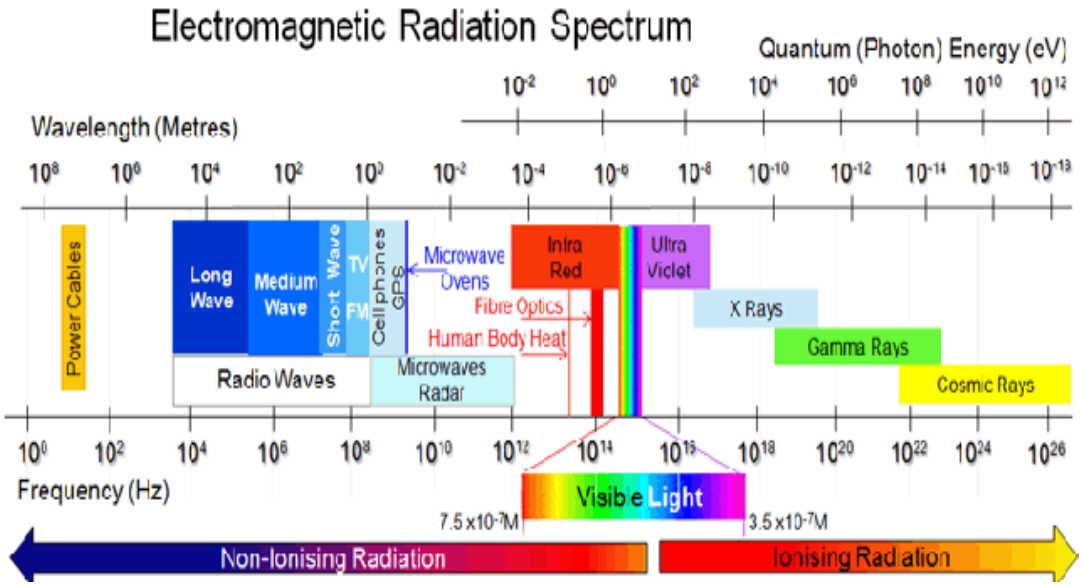

Figure 1.1: Ionizing and non-ionizing radiations

1.2.1 Non- ionizing Radiations

The non-ionizing radiations do not have enough energy to ionize the material with which they interact and these dissipates their energy in the form of heat. Microwave, laser, visible light, radio waves, TV waves are the examples of non-ionizing radiations. There is no serious health effect produced by these types of radiations. These are mainly used for communication purposes and other useful purposes.

1.2.2 Ionizing Radiations

Ionizing radiation possess enough energy so as to cause ionisation of the matter with which these interact by stripping out the electrons from the outer shell of the atom and leaving behind the positive ions. These radiations have energy in the range of 4-25 eV, which is the usual order of valence energy for most of the atoms. As, ionizing radiation can affect the atoms in living things, so these may pose serious health risk by damaging tissue and DNA in genes. Ionizing radiations are emitted from x-ray machines, cosmic particles from outer space and radioactive elements. Radioactive elements emit ionizing radiation whenever their atoms undergo radioactive decay. Based upon relative biological effectiveness, ionization radiations are categorized into High Linear Energy Transfer (LET) and Low Linear Energy Transfer radiations. Alpha, beta, gamma, neutrons and other particles of similar or greater mass (Radiation weighting factor greater than 1) are included in high LET radiations whereas photons (X-rays, gamma rays), electrons, muons, and positrons (Radiation weighting factor equal to 1) are described as low LET radiations. The ionizing radiations emitted from natural radionuclides are explained below:

a) Alpha Particles. An alpha particle is identical to helium nucleus having 2 protons and 2 neutrons with two unit positive charge and is emitted from unstable nuclei due to their large size. It is emitted from a radioactive element with a speed of 5% of light i.e. 15000 kms^{-1} and with energy in the range of 4 to 9 MeV. While passing through the matter, these particles are capable of producing many ion pairs due to relatively large mass, high electrical charge and low velocity and therefore possess very high specific ionisation. The range in air is only several centimetres even for the most energetic alpha particles. The penetration power of these particles is very low, even

can't pass from the skin. However, once inside the body, surrounded by living tissue, damage will be inflicted to the local area in which the alpha emitter is deposited. Thus, alpha emitters cause internal hazard and therefore the intake to the body must be prevented. In this present work, the detectors have been used to detect alpha particles emitted by radioactive elements like radon, thoron and their progeny. The schematic representation of alpha decay of parent nuclei (X) to daughter nuclei (Y) is given as:

$$ {}^{A}_{Z}X \rightarrow {}^{A-4}_{Z-2}Y + {}^{4}_{2}He^{2+} $$

b) Beta Particles. These particles are emitted from the nucleus having too large or too small neutron to proton ratio. These are like electrons with a wide range of energies upto some maximum value which is the characteristic of each radionuclide. Beta particles, like alpha particles, lose their energy by ionization and excitation. Due to their small mass and less charge, their interaction is less frequent, hence these particles do not produce as many ion pairs per centimetre of path of an alpha particle and have greater range in passing through matter. The particles with energy greater than 70 keV penetrate the protective layer of skin, thus are somewhat external hazard. These can be also constituting internal hazard if present with in the body. The schematic representation of their decay formation is as follows:

$$ {}^{A}_{Z}X \rightarrow {}^{A}_{Z+1}Y + \beta^{-} + \vartheta $$

c) Gamma Rays. Gamma rays are emitted with discrete energies from excited nuclei. These are massless, chargeless radiations and have sufficient energy to release secondary particles from matter through basic interactions that is photoelectric effect, compton effect and pair production depending upon energy. These can be blocked by dense materials like lead or thick concrete. These rays can easily passes through human body, causing ionizations to damage tissue or DNA along their path. The generalized atomic equation for nuclear transition is:

$$ {}^{A}_{Z}X \rightarrow {}^{A}_{Z}X + \gamma $$

d) Neutrons. Neutrons are neutral particles which cause indirect ionization when interact with matter and produce several types of the radiations which leads to further ionization. The penetration power of neutrons is much higher than all other type of particles like alpha, beta, gamma as shown in Figure 1.2. Only concrete wall and water are used to stop the neutron.

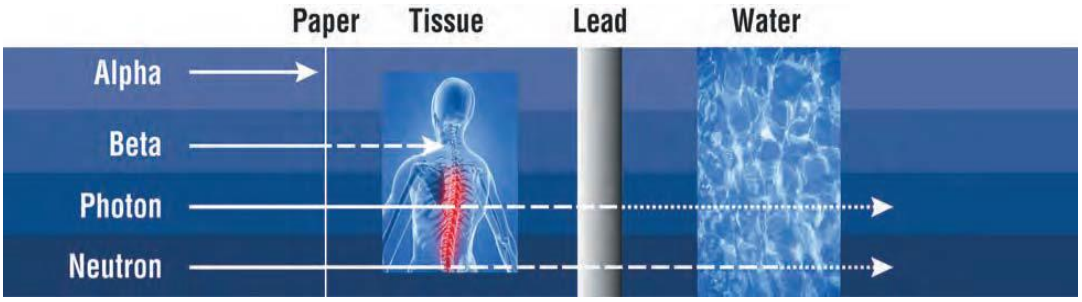

Figure 1.2: Penetration power of ionizing radiations

(UNEP, Radiations Effects and Sources, 2016)

1.3 RADIATION SOURCES

Radiations have always been present all around us and living beings are continuously exposed to these radiations emitted from different type of sources. The sources of radiations can be natural or man-made. All species on earth have existed and evolved in environment, have been exposed to radiation from natural background and artificial sources, the detail for which have been discussed below. Figure 1.3 shows the distribution of exposure from all sources of ionizing radiations.

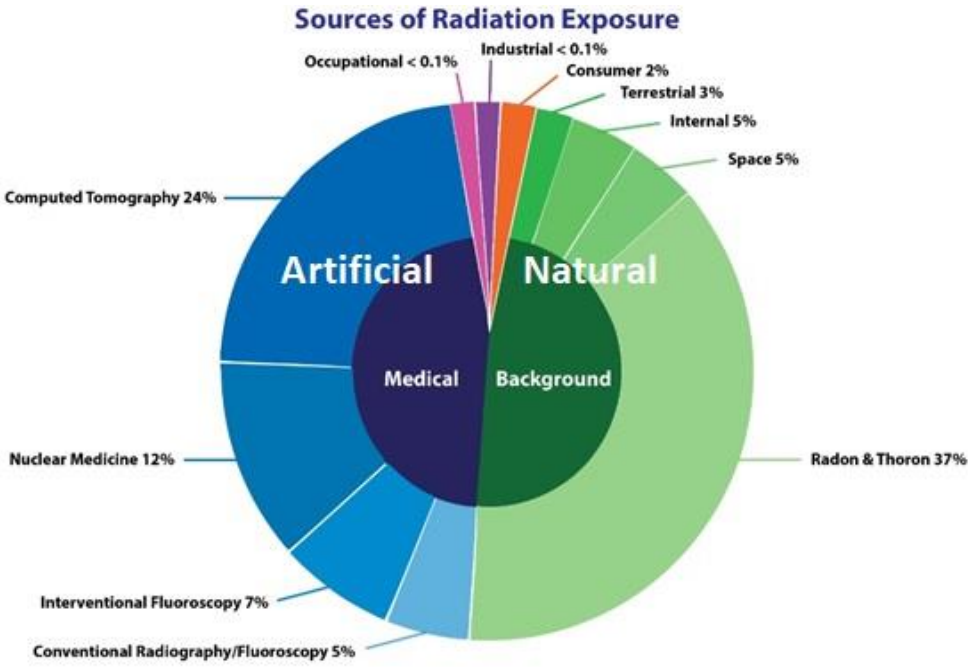

Figure 1.3: Sources of distribution of all radiation dose

(NCRP Report no. 160, 2009)

1.3.1 Natural Sources

Since the creation of earth, its environment has been exposed to radiation from both outer space and from radioactive material in its crust and core. Natural background radiation is the largest source of radiation exposure to humans (about 50%). The global average effective dose per person is about 2.4 mSv and ranges from about 1 to more than 10 mSv depending upon where people live (UNEP, 2016). According to UNSCEAR annual effective dose from radon is 1.3 mSv about half of the dose received by public from natural sources (UNSCEAR, 2000). Depending upon weather the radiations are originated from earth or outside the earth, these sources have been classified as terrestrial and extra-terrestrial and are discussed below:

a) Terrestrial Sources. Terrestrial radiations are those which are emitted because of radioactive decay of radionuclides present in the earth itself. The radioactive atoms like uranium, thorium and potassium, are present in varying amounts in all soils and rocks, in the atmosphere and in the hydrosphere. These radiations are emitted by decay of natural radioisotopes in rocks, soil, vegetation and groundwater. These sources can be classified further as outdoor sources (like rocks, soil, air, natural gases) and indoor sources (like building materials, tap water).

b) Extra-terrestrial Sources. These radiations have origin outside earth in the deep interstellar space or inside the stars. Cosmic rays are the major source of external source outside solar system. These high energy radiations are mainly protons (89%), nuclei of light to heavy elements including hydrogen, helium upto uranium. When these radiations enter earth's atmosphere they collide and create more particles like pions, muons to which living beings on the earth's surface are exposed.

1.3.2 Artificial Sources

Accidental discovery of x-rays and phenomenon of radioactivity led to the development of man-made radiations as well as applications of these ionising radiations. Over the last few decades, man has "artificially" produced several hundred radionuclides. Out of the total radiation exposure, artificial sources contribute half of the exposure which is mainly from medical sources of radiation exposure (about 48%) and remaining 2% comes from consumer products, occupational exposure, industrial exposure and nuclear

power plants (UNEP, 2016) (Figure 1.3 shown above). Consumer products mainly comprise of building and road materials, x rays security system, televisions, fluorescent lamp starters, smoke detectors, luminous watches etc. Main sources of artificial radiations are the use of medical facilities, pharmaceutical facilities, in research and teaching institutions, nuclear reactors and their supporting facilities such as uranium mills and fuel preparation plants, and facilities involved in nuclear weapons. Many of these facilities generate radioactive waste, and some release a controlled amount of radiation into the environment. Consequences of nuclear tests and radiological accidents are also sources of man-made radiation. The exposure to these radiations contributes towards the dose received by individuals. Figure 1.4 shows the annual effective dose per person received from various sources.

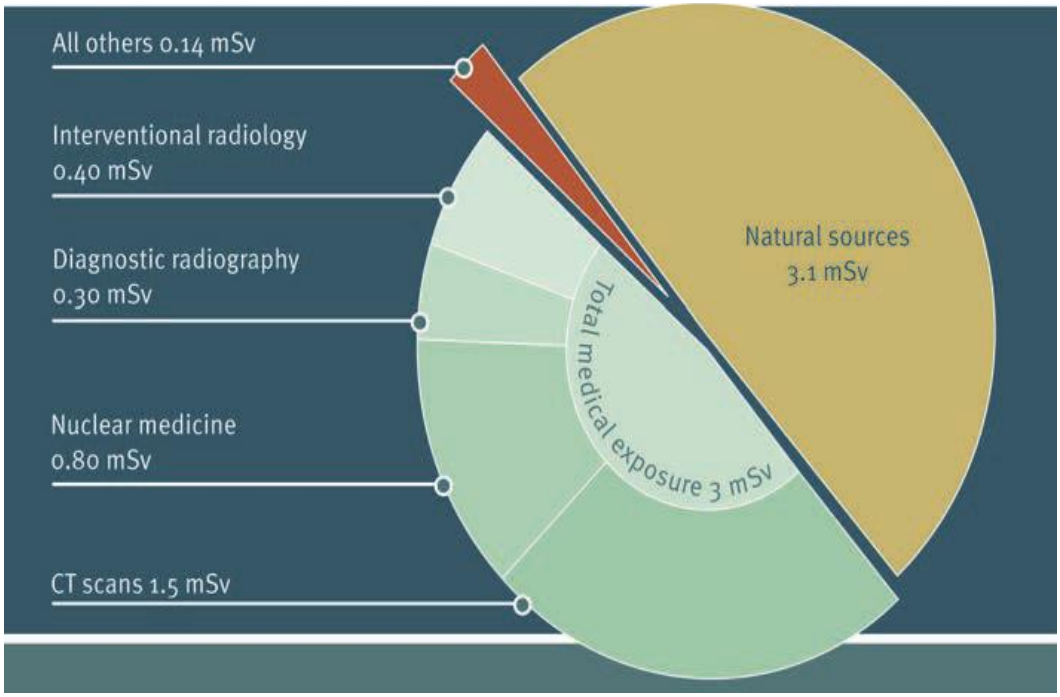

Figure 1.4: Annual effective dose per person in United States (2007)
(UNEP, Radiations Effects and Sources, 2016)

1.4 TERMINOLOGY

For the assessment of risk due to ionizing radiation, the few characteristics terms related to radiations like activity, exposure, units, energy, dose has been explained below:

1.4.1 Activity

As hazardous substances are usually measured in the units of mass, similarly the decay rate of radionuclides is measured in terms of activity. Mass and activity are related by the decay constant of the radionuclide. The activity of radionuclide is defined as the number of disintegrations per unit time. Becquerel (Bq) is the standard SI unit of radioactivity and is the activity of radionuclide decaying at an average rate of one spontaneous nuclear transition per second. One Becquerel equals one nuclear transformation per second. Curie (Ci) is the older unit of activity is which corresponds to 3.7×10^{10} nuclear transformations per second.

1.4.2 Energy and Exposure

The energy of particle emitted during the nuclear transformation of a radionuclide is expressed in electron-volts (eV). One electron volt is the energy of an electron submitted to a potential difference of 1 V ($1\ eV = 1.6 \times 10^{-19}$ J). Radiation exposure is measured in terms of the amount of radiations received per unit time. SI unit of radiation exposure is Roentgen (R) and is also expressed in coulomb per kilogram.

1.4.3 Dose and Units

Radiation dose is related to the damage inflicted on the body by the exposure to radiations. Depending upon the energy absorbed and the damage caused by the radiations and the dose can be expressed as the absorbed dose, equivalent dose and effective dose as discussed below:

a) Absorbed Dose is the primary physical quantity of radiation dosimetry. It is defined as the radiation energy absorbed per unit mass of organ or tissue and is used to study the damage to particular organ or tissue. Gray (Gy) is the unit of absorbed dose and defined as the absorbed dose when the energy per unit mass imparted to matter by ionizing radiation is 1 joule per kilogram. The former unit of absorbed dose is rad (rd) = 10^{-2} Gy.

b) Equivalent Dose (H) to an organ or tissue is the primary dosimetric quantity of the radiation protection, which is concerned with inferring the biological effects associated with irradiation of tissues with rays of various characteristics. The equivalent dose is obtained by weighting the absorbed dose in an organ or tissue by radiation weighing factor which reflects the biological effectiveness of the charged particles that produce ionization within the tissue. It is measured in Sievert (Sv), millisievert (mSv) and micro sievert (μSv). One sievert is a large quantity, radiation doses normally encountered are expressed in millisievert (mSv) or microsievert (μSv) which is one-thousandth or one millionth of a sievert.

c) Effective Dose (E) is a biological dose that is often used in radioprotection to estimate how damaging a person's radiation exposure can be. It considers not only the nature of the incoming radiation, but also the sensitivity of the afflicted bodily parts. The units of the effective dose are same as that of equivalent dose that is Sievert (Sv), millisievert (mSv) and micro sievert (μSv). The effective dose is considered in the subject of radioprotection whereas the equivalent dose is matters in medicine.

1.5 HEALTH EFFECTS DUE TO RADIATION EXPOSURE

The exposure of human beings to the ionizing radiations is a cause of concern for health effects and has been widely studied by the researchers. The effects and variability of radiation exposure to a person are influenced by a variety of variables, including the type of radiation, the total dose of radiation, the time period over which the dose is received, the radiosensitivity of the part of the body exposed, the person's age, gender, and health, as well as the type of exposure (internal or external). When these radiation traverses through the body, it may deposit its energy and damage biomolecules in the surrounding region directly or indirectly. These may stick to alveoli and could expose cells of bronchial and pulmonary epithelium in the lungs, thereby damaging the cells, DNA and may cause lung cancer (Seve et al., 1976; El-Hussein et al., 1998; Butterweck et al., 2002; Sannappa et al., 2014; Alberta et al., 2017). Indoor radon exposure has been linked to the development of leukaemia and other malignancies such as melanoma, kidney and prostate cancers (Henshaw et al., 1990; Quindos et al., 1994), as well there is 16 percent increase in relative lifetime lung cancer risk per 100 Bqm^{-3} due to chronic radon inhalation (Fintan et al., 2019). A radon concentration of 50 Bqm^{-3} can cause

25% of leukaemia cases in children and adults of all ages (Richardson et al., 1991; Reddy et al., 2017). Uranium exhibits chemical and radiological toxicity which effects mostly to the two vulnerable organs kidneys and lungs (Wrenn et al., 1985; WHO, 2000; Nagaraju et al., 2013; CGWB, 2020). Higher uranium intake can cause uranium to accumulate in organs like the kidney, bones, GI Track, soft tissues which can be cancerous (ICRP, 1976; Byju et al., 2012). In addition to being toxic heavy metal, it is also a radioactive element and can cause several adverse health effects ranging from renal failure, diminished bone growth and damage to DNA when consumed in high quantities (ICRP, 1993; ATDSR, 2013). Cancer of lungs, bladder, skin, stomach, and breast is the most notable uranium-related disease (Brugge et al., 2011; Sahoo et al., 2021). Health effects mainly depend upon the amount of energy deposited per unit mass of the tissue. These effects can be categorized in two types (Grover and Kumar, 2002): a) Non-Stochastic Effects (deterministic effects) b) Stochastic Effects (non-threshold function of the dose). The details about these effects have been discussed below:

a) Non-Stochastic Effects. Non-Stochastic (deterministic) effects appears when an organism is exposed to high radiation dose above some threshold value for a short period of time and shows a clear connection between exposure (dose) and effect (ICRP, 2007) (Figure 1.5). It may cause skin reddishness, loss of visual acuity (cataract), burning of skin and tissues and radiation sickness. These after effects of radiation exposure can be experienced within few days to few weeks of exposure (Hamda and Fujimichi, 2014).

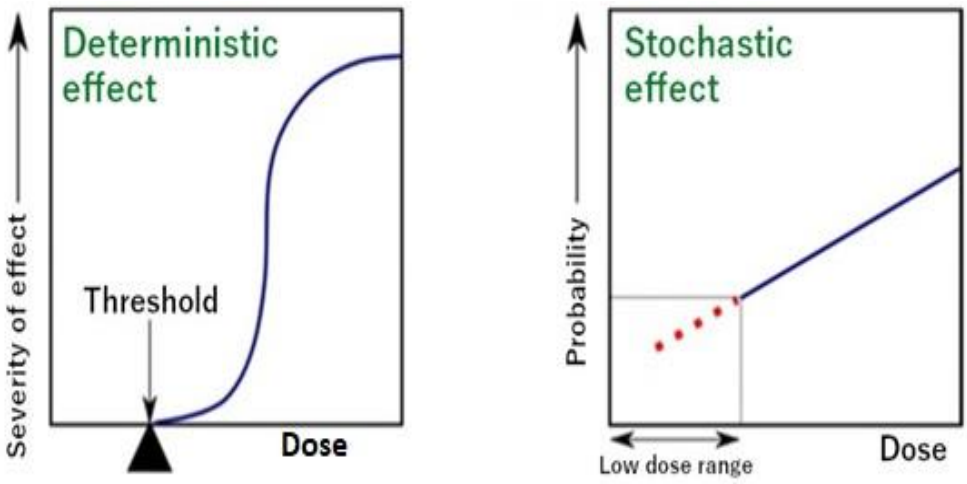

Figure 1.5: Deterministic and stochastic health effects

b) Stochastic Effects. Stochastic effects occur by chance and there in no threshold limit below which the effect will not occur (Figure 1.5), which means exposure to any amount of dose can also initiate the pathological changes to the exposed organism (UNSCEAR, 2012). Several diseases like sickness, internal bleeding and hair loss appear after the exposure but cancer appears after a number of years.

1.6 RADIATION EXPOSURE PATHWAYS

Living beings are constantly exposed to ionizing radiations and about 50% of the radiation exposure comes from naturally occurring sources. Radionuclides found in dietary food items (particularly seafood) and drinking water cause intake doses. At great heights, cosmic radiation dosages are noticeable. Frequent travellers and air crew should be cautious with such doses. Additionally, some persons are exposed to ionising radiation created intentionally while performing their professions (occupational exposure) or when undergoing medical procedures like radiotherapy or X-ray diagnosis (medical exposure). Workplace exposure occurs where ionising radiations are used, such as in industries (sterilisation), scientific research labs, and the medical industry (radiation medicine diagnosis and treatment). There are two ways of a person may be exposed to ionizing radiation - internal and external exposure as discussed below:

1.6.1 Internal Exposure

Internal exposure to human beings is mainly due to ingestion and inhalation. The person may inhale radioactive material in the atmosphere, ingesting contaminated food and drinks, inadvertent ingestion such as from eating with contaminated hands and drinking, eating or smoking in a contaminated environment where radioactive material is present in air or on ground and other surfaces and absorption through the skin or open wound.

1.6.2 External Exposure

External exposure to human beings is mainly due to radioactive materials present in the atmosphere, materials deposited on the ground and contamination on skin and clothes.

These substances may then travel into the body by physical, biological, and chemical processes, exposing other organs and tissues.

1.7 RADIATION MONITORING

It has long been recognized that large doses of ionizing radiation can damage human tissues. Over the years, as more was learned, scientists became increasingly concerned about the potentially damaging effects of exposure to large doses of radiation. The need to regulate exposure to radiation prompted the formation of a number of expert bodies to consider what is needed to be done. In 1928, an independent non-governmental body of experts in the field, the International X-ray and Radium Protection Committee was established. It later was renamed the International Commission on Radiological Protection (ICRP). Its purpose is to establish basic principles for, and issue recommendations on, radiation protection.

An intergovernmental body was formed in 1955 by the General Assembly of the United Nations as the UN Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). UNSCEAR is directed to assemble study and disseminate information on observed levels of ionizing radiation and radioactivity (natural and man-made) in the environment, and on the effects of such radiation on man and the environment.

However, for sources that can be controlled, International Atomic Energy Agency (IAEA) works in close collaboration with United Nations Scientific Committee on Effects of Atomic Radiation (UNSCEAR) and other international organizations (World Health Organization, International Labour Organization, Organization of Economic Cooperation and Development/ Nuclear Energy Agency) to research and address safety measures for use of ionizing radiation.

International organizations and governments can use the International Basic Safety Standards (BSS), developed by the IAEA, as a guide to protect people and the environment from the harmful effects of ionizing radiation. All activities and nuclear facilities, old and new, used for peaceful purposes must adhere to these safety requirements. They also apply to protective measures meant to lower radiation risks. International agreement is represented by the IAEA safety recommendations, which promote world trade and peace. However, they do not apply to exposures that are

unamenable. For example, it is not feasible to control 40K in the body and cosmic radiation at the surface of the earth.

Different radiation protection agencies were set up in India with the mission of protection of human body and environment from the exposure of radiation and to establish nuclear power systems with the use of atomic energy. Development of radioactive monitor, Environmental monitoring and radiological impact assessment at different sites and monitoring the background levels of gamma radiations at different sites are the main features of (Bhabha Atomic Research Center) BARC and Department of Atomic Energy (DAE). BARC and DAE give the various research projects to the professors for the assessment of seasonal variation of indoor radon and thoron and their progenies, concentration of uranium in drinking water, radon concentration in water and soil, radon exhalation rate from soil, measurement of terrestrial radionuclides (226Ra, 232Th and 40K) in soil etc. in different locations of India. According to the protection policies of Atomic Energy Regulatory Board (AERB, 2004) to make sure that use of ionizing radiation sources and nuclear energy in India does not make inappropriate risk to the health of people and environment. The main objective of AERB is that the effect for radiation exposure from nuclear and radiation facilities remains lower than the permissible limit and use of these facilities are allowed for the benefits of society. All the nodal centers should be arranged appropriate radiation protection programmes time to time for the safety of public from radiation exposure. The radioactive waste which is produced from the nuclear and radiation generating factories shall be maintained according to the human health point of view.

1.8 NATURAL RADIONUCLIDES AND DECAY SERIES

Radionuclide is an atom (element) having an unstable nucleus which have excess energy that is released by different types of radioactive decay processes. Radionuclides occurs naturally in numerous rocks and minerals. Some radionuclides like uranium (238U), thorium (232Th) and potassium (40K), radium (226Ra), Plutonium (239Pu) etc. have been found in rocks since the earth's formation. These radionuclides decay continuously into other daughter nuclides which are also unstable and decay further with the emission of alpha (α), beta (β) and gamma (γ) particles. These radionuclides will terminate at the stable element. The concentrations of radionuclides are influenced

by the geological and geographical conditions of each place across the world (Radhkrishna et al., 1993; Quindos et al., 1994; UNSCEAR, 2000), are causes of variation of doses.

The radioactive elements uranium, thorium, potassium etc. are widely distributed in the earth's crust all over the world. Detailed study of naturally occurring radioactive elements show that these nuclei decays to form naturally radioactive decay series out of which U-238 decay series and Th-232 decay series has been explained below.

U-238 undergoes radioactive decay into a series of 13 radionuclides by emitting alpha, beta, gamma particles before reaching the stable Pb-206 (Figure 1.6). Main elements studied for natural radioactivity in this series are Ra-226, Rn-222, Po-218, Po-214, Bi-214.

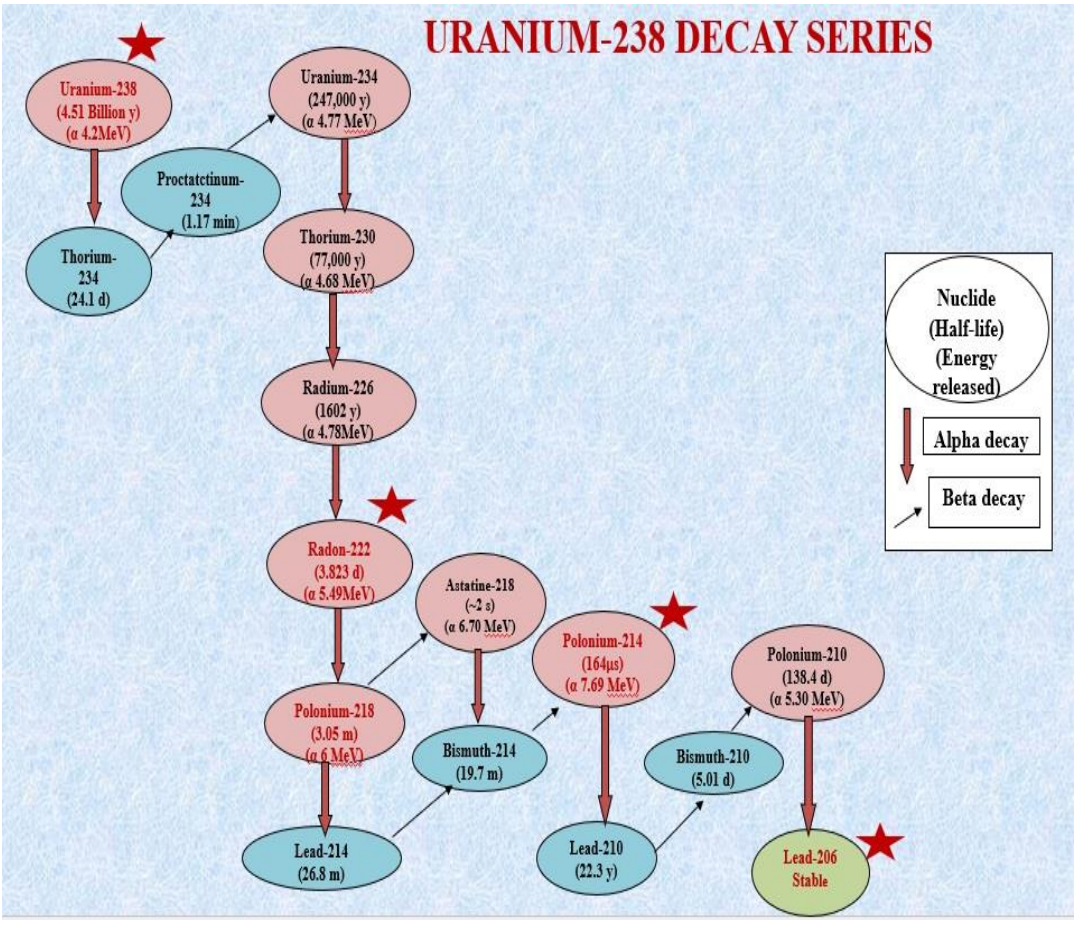

Figure 1.6: Uranium decay series

In Th-232 decay series, Th-232 finally decays to stable element Pb-208 by emission of alpha, beta, gamma particles. The main radioactive elements are Rn-220, Po-216, Pb-212 in this decay series (Figure 1.7).

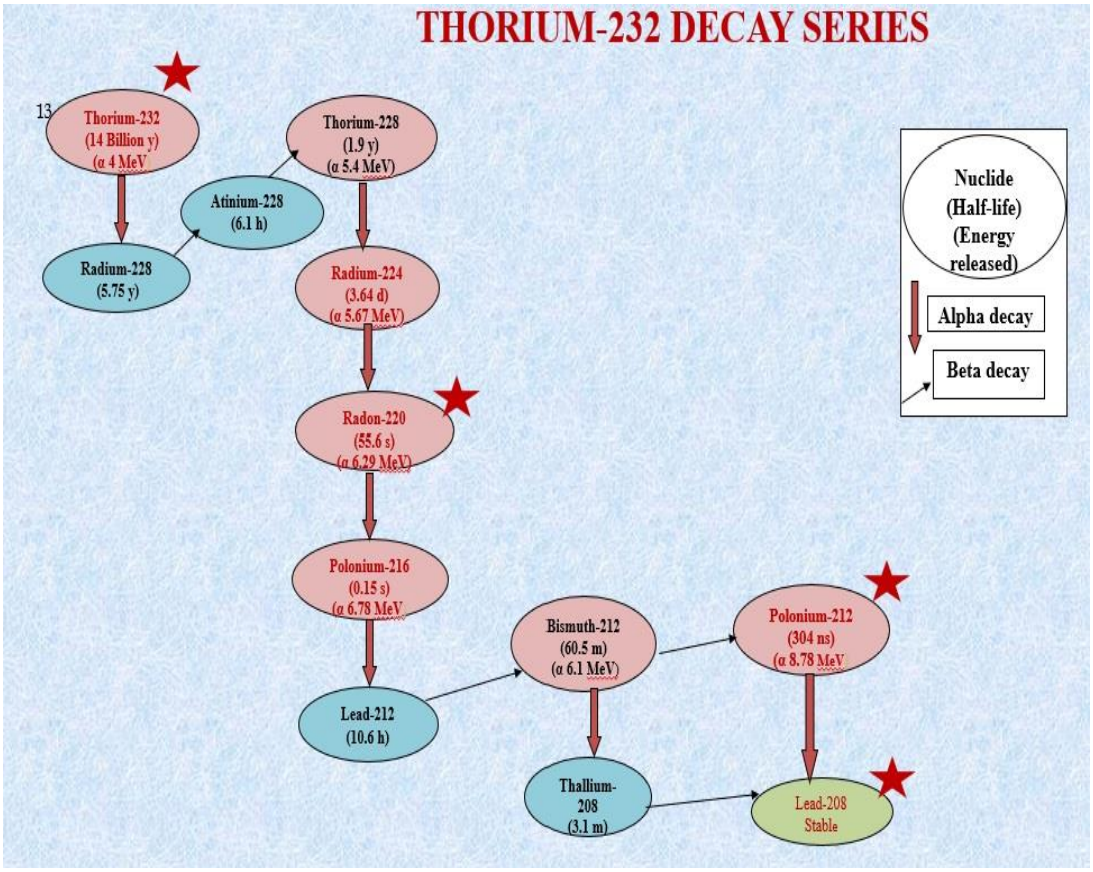

Figure 1.7: Thorium decay series

1.9 URANIUM

1.9.1 General Information

Although existing in the earth's crust billion years ago, the discovery of uranium in 1789 in the mineral pitchblende is credited to Martin Heinrich Klaproth, who named the new element after the recently discovered planet Uranus. Eugène-Melchior Péligot was the first person to isolate the metal and its radioactive properties were discovered in 1896 by Henri Becquerel. Research by Otto Hahn, Lise Meitner, Enrico Fermi and others, such as J. Robert Oppenheimer starting in 1934 led to its use as a fuel in the nuclear power industry and the first nuclear weapon used in war.

Uranium is a ubiquitous, ductile, weakly radioactive, one of the densest metals and the second massive naturally occurring element, that prevails in the earth's crust having a mean abundance of 1.8 ppm. There are three naturally occurring radio-isotopes of uranium that exists in environment: 99.27% of 238U (half-life: 4.468 × 109 y), 0.72% of 235U (half-life: 7.04 × 108 y), and 0.005% of 234U (half-life: 2.33 × 105 y), out of which 238U is most stable. Earth's crust constitutes 48.9% of 238U, 2.2% of 235U and 48.9% of 234U isotopes which has been obtained from their respective mass percentage and half-lives (UNSCEAR, 2016). Uranium gets oxidized easily when it comes in contact with air, forming a layer of oxide. Uraninite, pitchblende and davidite are the three basic uranium ores that exists in the nature (Stalder et al., 2012).

1.9.2 Sources

Uranium is abundant, occurring in granites, rocks and soils and various other naturally occurring mineral deposits. Uranium is present in the environment as a result of leaching from natural deposits, release in mill tailings, emissions from nuclear industry, the combustion of coal and other fuels and the use of phosphate fertilisers that contain uranium. Uranium can be enriched in soil via natural paedogenesis processes with uranium being released from parent materials to soil and soil solution, and via human activities. Mining phosphate can carry uranium to soil and river water sources surrounding the mines. The average concentration of uranium in U.S. soils is about 3 ppm.

The various factors affecting the distribution of uranium in groundwater are lithology, geo-morphology, geochemical factors such as dissolution of uranium bearing minerals and its persistence in groundwater (Sahu et al., 2020). The predominant species of uranium existing in primary mineral ores is in the tetravalent U(IV) state (Smedley et al., 2006), existing as hydroxides, phosphates and fluorides (Keith et al., 2007) and is comparatively insoluble. By weathering and chemical transformation in aquifers, primary uranium minerals are transformed to secondary minerals and get dissolved or mobilized in groundwater which result in change of oxidation state from tetravalent U(IV) to hexavalent U(VI). U(IV) is easily oxidised to U(VI) as both surface water and groundwater behaves as oxidising media. For example, insoluble U(IV) (e.g. uraninite) is readily oxidized through rock-water interactions to soluble U(VI)

(e.g. UO_2^{2+}), which gets leached and then transported with groundwater by forming highly soluble and stable uranyl complexes with HCO^{3-}, PO_4^{3-}, F^- and SO_4^{2-} ions (Shin et al., 2016). Uranium generally exists as complexes of uranyl cation, UO_2^{2+} and commonly found in groundwater at larger concentrations than is supposed from the underlying rocks. The various methods which control the uranium distribution in groundwater are weathering of uranium bearing rocks/minerals, sorption/desorption reactions and reductive precipitation (Alam and Cheng, 2014). Geological processes are a significant source of uranium in groundwater. These processes include water-rock interactions that extract uranium from various rocks, oxidising conditions that increase the uranium's solubility in water, and interactions between the extracted uranium and other chemicals in the groundwater, such as carbonate, that can further increase its solubility. Granite regions are where abnormally high uranium concentrations in groundwater are most frequently seen. For instance, higher uranium concentrations in the granite and gneiss rocks of the northern half of the Karnataka state may be the cause of higher uranium concentrations in the Kolar district (Figure 1.8).

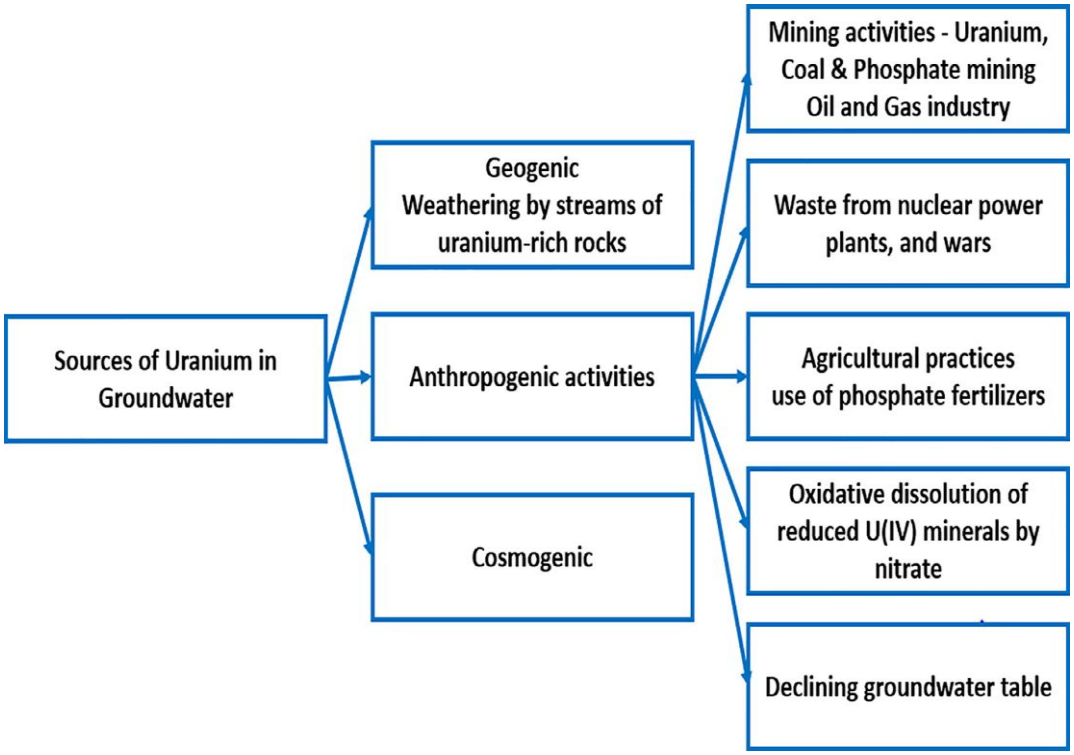

Figure 1.8: Various sources of uranium in groundwater

The anthropogenic activities such as uranium mining and milling activities, uranium transformation and enrichment for uranium fuel fabrication or nuclear weapon

production and the production and utilization of fertilizers/pesticides on agricultural land can't be ruled out as they are predominant in causing uranium contamination in groundwater. Cosmic rays produce radionuclides, such as uranium, yet some of them do reach the surface of the earth and interact with water and soil. Additionally, naturally occurring radionuclides that are found in the hydrosphere, crust of the earth, and atmosphere circulate throughout the environment. 3H, 14C, 32P, 40K, 222Rn, 226Ra, 232Th, 235U, and 238U are some of the radionuclides in this group. Some of them are known as cosmogenic isotopes because some of them are produced under the influence of cosmic radiation. According to recent study, some uranium is created when neutron stars and meteorites collide, bringing some uranium with them that later becomes enriched in the continental crust.

1.9.3 Exposure Mechanism

Exposure of uranium in human beings through inhalation and ingestion. As uranium is available in soil, water therefore its exposure through ingestion can be by eating contaminated food or drinking water that consists of natural uranium in high amount (Brugge et al., 2011). Vegetables such as potatoes, turnips, and sweet potatoes etc. which are generally root crops, contribute to highest uranium content in the diet and the amount of uranium present in these foodstuffs is directly proportional to uranium concentration in the agricultural land in which they are cultivated.

Exposure due to uranium may also occur by inhalation which is the primary pathways for radiation workers but for general population this is very minor source of exposure. The areas where mining activities are done, there is a great possibility of increase in the air uranium concentration. The possibility of uranium exposure through dermal contact is also very low that may occur with uranium workers due to skin contact with uranium powders, metals or uranium wastes (ATSDR, 2013). According to a survey, the daily uranium intake through ingestion varies from 0.9 to 1.5 μg, while through inhalation, it varies from 0.001 to 0.01 μg (ATSDR, 2013). Inhaled uranium compounds are absorbed in the respiratory tract via transmission across the cell membranes as dust particles and get deposited in the lungs depending on their size and respective absorption is also affected by their solubility in biological fluids (Zavodska et al., 2008). Hardly, 0.76 - 5% of the inhaled uranium gets into the bloodstream by the

way of respiratory tract. However, 0.1 - 6% of the ingested uranium gets absorbed into the blood flow through gastrointestinal tract (ATSDR, 2013). Most of the uranium intake is not captivated by the body as it gets eliminated via faeces and urine. About 67% of the uranium absorbed in blood is sieved through kidneys and then leaves the body within 24 hours and 33% of uranium is found in kidneys, liver and bones (Kitahara et al., 2017). Figure 1.9 shows the exposure of uranium via various pathways like ingestion, inhalation, dermal contact through air, water and soil.

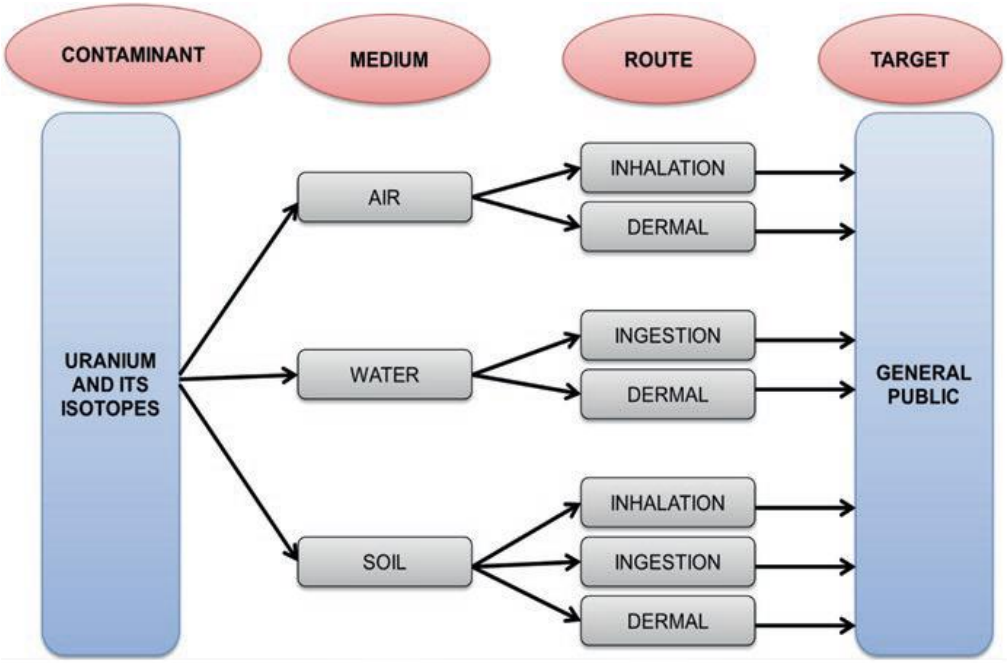

Figure 1.9: Pathways of uranium exposure (Mehra and Kaur, 2019)

1.9.4 Significance of Study

Being present everywhere, uranium shows no definite metabolic functions and therefore, it is regarded as redundant component, but when accumulated inside human body it causes chemical and radioactive effects causing various health hazards. In nature, only dissolved uranium is found to be chemically harmful which occurs as a uranyl ion in groundwater (Zavodska et al., 2008). Uranium absorption in the gastrointestinal tract rely on the solubility of the uranium compound (WHO, 2011). The various uranium isotopes are naturally radioactive, but chemically it is six times more hazardous than its radioactivity (Atkins et al., 2016). It is very harmful to inhale or ingest the uranium with higher dose or quantity because these nuclides emit α- rays of

intense ionization power. The chemical and radiological toxicity has been discussed below:

a) Chemical Toxicity. Chemical toxicity is the side reactions of ingested molecules with DNA and the interference with enzymatic system. The main chemical effect associated with exposure to uranium and its compounds is kidney toxicity. This toxicity can be caused by breathing air containing uranium dusts or by eating substances containing uranium, which then enters the bloodstream. Once in the bloodstream, the uranium compounds are filtered by the kidneys, where they can cause damage to the kidney cells (ICRP, 1993; ATDSR, 2013). Very high uranium intakes (ranging from about 50 to 150 mg depending on the individual) can cause acute kidney failure and death (ICRP, 1975). At lower intake levels (around 25 to 40 mg), damage can be detected by the presence of protein and dead cells in the urine, but there are no other symptoms. Also, at lower intake levels, the kidney repairs itself over a period of several weeks after the uranium exposure has stopped. Insoluble inhaled uranium particles having size 1-10 μm, tend to be retained in the lung and may lead to irradiation damage of lung and even lung cancer if a high enough radiation dose results over a prolonged period. A study of Uranium on its chemical toxicity revealed that body burden of 0.1 mg kg^{-1} of body weight produce nephrotoxic effect (Waseem et al., 2015).

It is recognised as a nephrotoxine as its nephrotoxic effects are known due to its chemical properties instead of its radioactivity, even though ingested uranium may produce radiological effect on other tissues due to deposition (Llobet et al., 1991). The primary sites for uranium deposition inside the human body are liver, bones and kidneys. 95% of uranium received by the body is accumulated in bones and causes high risk of bone cancer (ICRP, 1975). It can also injure the capillary membrane and induce minor injury to muscle tissues and liver. Its impact on nervous system is same as of heavy elements.

b) Radiological Toxicity. Radiological toxicity deals with the effects of radiations emitted by radioactive elements when it accumulates in the body. Several possible health effects are associated with human exposure to radiation from uranium. Because all uranium isotopes mainly emit alpha particles that have little penetrating ability but have more ionizing power which can damage the tissues, cells or organs. The main radiation hazard from uranium occurs when uranium compounds are ingested or inhaled. However, workers in the vicinity of large quantities of uranium in storage or in a processing facility also are exposed to low levels of external radiation from uranium

decay products. At the exposure levels typically associated with the handling and processing of uranium, the primary radiation health effect of concern is an increased probability of the exposed individual developing cancer during their lifetime. Cancer cases induced by radiation are generally indistinguishable from other "naturally occurring" cancers and occur years after the exposure takes place. The probability of developing a radiation-induced cancer increases with increasing uranium intakes. Direct contact of uranium metal with the skin, even for several weeks, is unlikely to produce radiation- induced erythema (superficial inflammation of the skin) or other short term effects.

1.9.5 Biokinetic Model

Biokinetic models are complex tools for calculating tissue and organ dosages after ingestion as well as for prospective and retrospective assessment of radio element retention. They use mathematics to represent the translocation, retention, and removal of chemicals within the body while taking into account their solubility, particle size distribution, uptake and residence times in different tissues and organs, and clearance kinetics from the body. As a result, they acquire utmost significance in determining the toxicity and forecasting the biological effects of intakes. Numerous variables, including physiological traits of the individual, age, dietary habits, exercise, and medicine, as well as environmental elements like temperature, moisture, sweating, etc., have an impact on the biokinetics of uranium. Various models namely Bernard and Struxness Model (1957), Lipsztein's Model (1981), Wrenn's Model(1985), ICRP's Uranium Systemic Model (1959; 1979) evolved with time and availability of data.

Li et al. (2009) Hair Compartment Model, which incorporates a hair compartment for excretion of uranium in addition to faeces (GI tract) and urine which was proposed in ICRP's Biokinetic Model for Uranium (urinary bladder). All uranium absorption after drinking water takes place in the small intestine (SI), with an alimentary tract transfer factor (f_1) of 0.6 percent. Uranium is released and reabsorbed from soft tissue, the liver, the bones, and the kidneys after entering plasma from SI. There are two ways that uranium enters the hair compartment: directly through the blood or indirectly through the skin's subcutaneous tissue. The assumption used to calculate the transfer rate to the hair compartment is that only a portion of the uranium

that passes from the plasma to the urine is redistributed, keeping alternative removal pathways from plasma intact.

Figure 1.10 (below) shows the block diagram of the compartmental model of uranium excretion pathways. The numbers (in percentage) between compartments represent fractional removal from first compartment to other. k_i, i=1,…,5 are the transfer rates exclusively used in Hair Compartment Model and the rest are taken from ICRP's Biokinetic Model for uranium. Uranium excretion via hair is controlled by k_3 and k_1. First-order differential equations represent movement of uranium among the various compartments: where dq_i is change in amount of uranium (in units of mass) in a particular compartment, dt is a small time interval, λ_{ji} is transfer rate between two specificcompartments (from compartment j to i), λ_i is the total transfer rate from the i^{th} compartment, λ_r is rate of radioactive decay and n is the number of compartments. Hair growth rate is normalized at 0.1g/day taking into account a number of factors like type, color, length and density of hair.

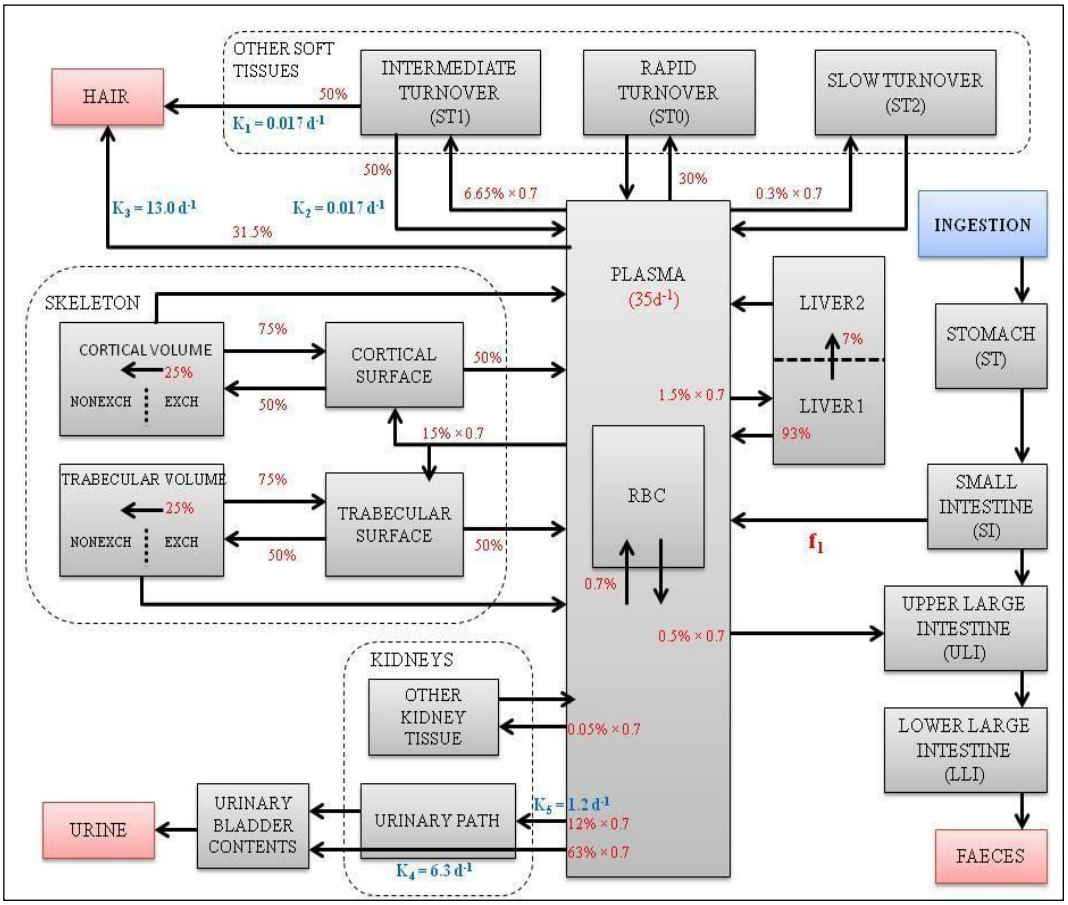

Figure 1.10: Compartmental model of uranium with hair excretion pathway (Kaur and Mehra, 2018)

1.10 RADON, THORON AND THEIR PROGENY

1.10.1 General Information

Radon belongs to the noble gas series in the periodic table. There are three natural isotopes of radon namely, radon (222Rn; $t_{1/2}$ = 3.8 days), thoron (220Rn; $t_{1/2}$ = 55.6 sec.) and actinon (219Rn; $t_{1/2}$ = 3.92 sec.) resulting from the radioactive decay of the uranium, thorium and the actinium series. 222Rn is formed from the decay of 226Ra, the immediate parent from the 238U series, while its isotope 220Rn decays from 224Ra, a member of the 232Th series. Actinon results from the decay of 223Ra from 235U series and is normally neglected because its presence is negligible in atmosphere due to its shorter half life.

The discovery of radium in 1898 followed the discovery of radon by Curies (Kolthoff and Philip, 1966). The Curies noted that air in contact with radium compounds becomes a conductor of electricity. In 1900, Dorn showed that this phenomenon was due to an emanation from radium and Rutherford established that thorium compounds too gave off a similar emanation. Rutherford called the gaseous element from radium "emanation" and gave a symbol "*Em*". The International Committee on Chemical Elements officially adopted the name "Radon" for this element in 1923. Investigations for its chemical properties and the emanating spectrum from radium were carried out by Ramsay and Rutherford. Soddy and other scientists showed that chemically radon is kin to the inert gases and it was included in the argon group.

The presence of radon in the free atmosphere was first noted by Elster and Geitael (Elster, 1901) around 1901. The study of conduction of electricity in air and other gases led Elster and Geitel to the conclusion that gaseous ions are the carriers of electricity. They later found that radiations from radioactive substances form ions in the surrounding air. Even though, both radon and thoron are present in the atmosphere, most studies generally ignore the presence of the thoron because of its short half-life. However, thoron may not be negligible under all circumstances. In several places in the world, high amount of thorium bearing regions exist, which are known as High Background Radiation Areas (HBRAs). Examples of such places are monazite-bearing sands in Kerala, India, and regions of Yangjiang in China.

Thoron along with its decay products is found in excess quantities in the atmosphere in these areas. As general population resides in these areas, they constitute a natural study group for examining the consequences of exposure to thoron. It all indicates that there is a growing concern due to the presence of thoron that may not be negligible in the global context especially in situations like India, where thorium presence is higher.

Radon thoron are the gases which decays by emitting alpha particles into the progenies Polonium, bismuth, lead which are considered as important elements in finding out the concentration of radon and thoron.

1.10.2 Sources

Presence of radon/thoron radionuclides in the surrounding environment may be because of naturally occurring (NORM) and man-made sources of radiations. The distribution of radon/thoron in ground depends upon the number of factors like soil temperature, humidity. porosity, soil moisture content and surface winds etc. Basically, there are three types of sources for radon/thoron contribute to the indoor and outdoor environment such as soil and rocks, building material (natural and technically enhanced) used for construction and ground water (Ramola et al., 2005; Parshad et al., 2008) has been discussed below:

a) Soil and Rocks Under Beneath. Soil and rocks rich of uranium (238U) and thorium (232Th) are the main source of indoor and outdoor radon/thoron levels. When radium isotope (intermediate product of uranium and thorium series) decays, it releases the radon/thoron atoms to the soil pore. After the emanation they migrate to the surface of the soil and enter in the outdoor as well as indoor environment via micro-fissures and foundations. In the outdoor environment it mixes with other gases and dispersed but in the indoor environment its concentration is high especially at those levels which are close to the soil such as basements and crawl spaces.

b) Building Materials. The building materials mainly composed of bricks, cements, mud, granite etc. which are mainly obtained from earth's crust. These are used for the construction and decorative purpose are considered as second main cause of indoor radon/thoron exposure by inhalation (Kumar and Chauhan, 2015). Radon because of its long half-life (3.82 days) was constantly a matter of concern whereas

thoron because of its short half-life (55.6 sec.) was not to be counted for the indoor radiological assessment. But now days it is well known that building materials are a significant source of indoor thoron levels (Hafeza et al., 2001). Some building materials like sand stone, bricks, granite, marble and concrete etc. emanates radon/thoron just after the radium during the decay of uranium and thorium from building materials and contribute a significant role to the indoor exposure to radiations.

c) Ground Water. Radon is dissolved in water and hence ground water is also a potential source of radon. Frequent activities of the inhabitants like showering, cooking, washing dishes and clothes etc. released radon from the water supply where it mixes in the indoor air and can be inhaled at whatever time of their use (Mjonas and Falk, 2005). Radon can be ingested through drinking water also.

d) Entry Routes of Radon/Thoron in Indoor Air. There are a number of entry routes of indoor radon/thoron from top few cm of soil like cracks in the floor, joints between floor and wall, construction joints, pores in concrete blocks, cracks in walls below the ground level, loose fitting pipe, fractured bedrock, building materials and water from wells as shown in Figure 1.11.

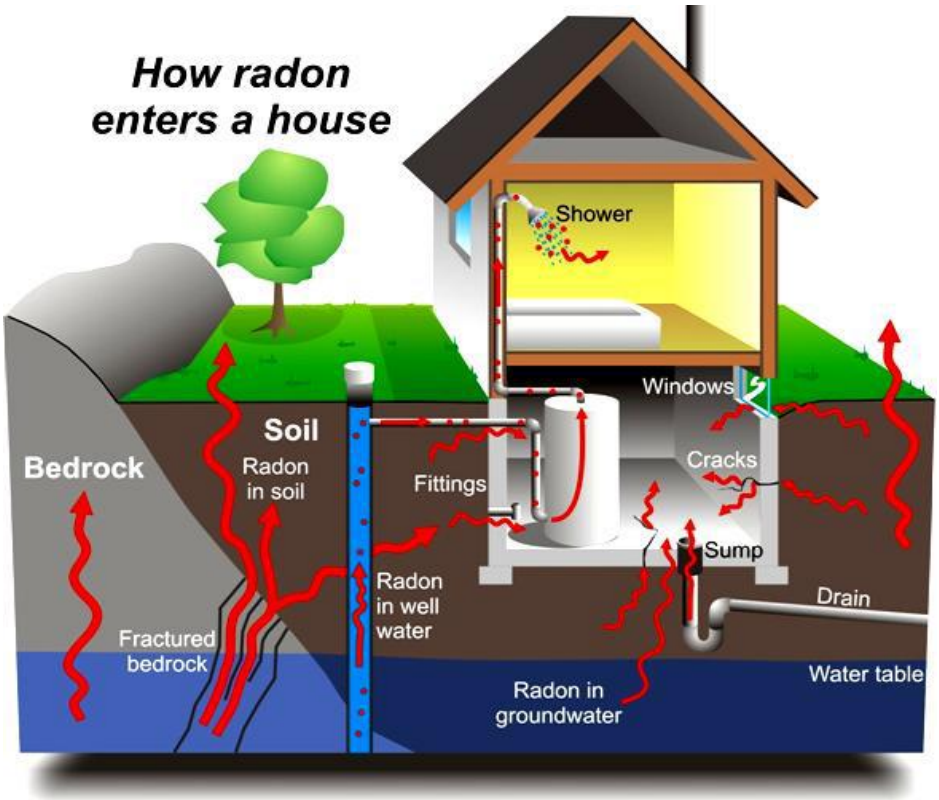

Figure 1.11: Entry routes of radon and thoron in dwellings

1.10.3Transport of Radon from Soil to Atmosphere

Radon/thoron atoms to be found in solid grain release to the interstitial space between the solid grains as a recoil effect of alpha particle produced from the decay of radium isotopes. Now, these atoms in the interstitial space can easily diffuse to the atmosphere (Hassan et al., 2013). Release mechanism of radon/thoron from the radium isotopes to the open surface as shown in Figure 1.12 can be described in three steps as discussed below:

a) Emanation. It is a process in which radon atoms emanates from the soil grain to the interstitial space between the soil grains due to the recoil effect of alpha particle produced during the decay of radium isotope.

b) Transport: A process in which radon atoms placed in the soil pores migrate (move) to the ground surface by the process of diffusion or advective flow.

c) Exhalation: In this process the radon atoms transported to the surface of ground are free to release in the open atmosphere.

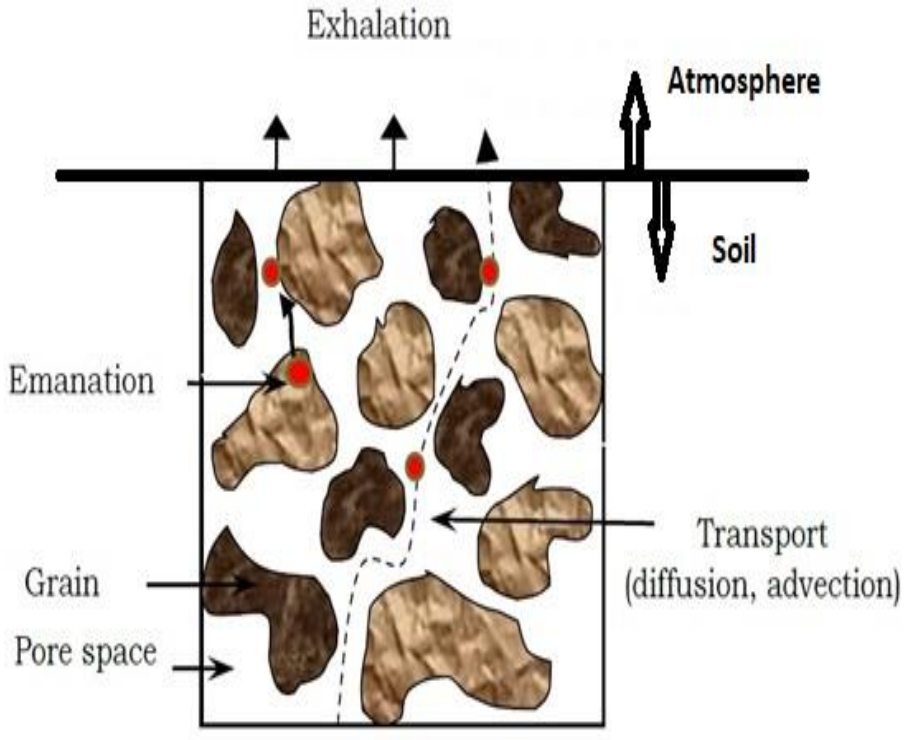

Figure 1.12: Release mechanism of radon into atmosphere (Nazaroff and Nero, 1988)

1.10.4 Significance of Study

There are shreds of evidence of adverse health effects due to radon even before its discovery. In the sixteenth century a large number of workers' deaths in mines of Eastern Europe attracted the attention towards the existence of some gases (Agricola, 1556). In 1879, high lung cancer was diagnosed by F.H. Harting and W Hesse among the miners and known as "Schneeberger Berkrankheit" and after 45 years it was recognized that this high lung cancer may be because of high radon activity (Harting and Hesse, 1879; Ludewig and Lorenzer, 1924; ICRP, 1987; BEIR IV 1988). Actually, radon and thoron itself are not primary source of health hazards but their short lived progenies play a dominant role when they inhaled (NRC, 1999). Most of the inhaled gas is exhaled during the process of breathing but their progenies attach themselves to the lung tissues and causes lung cancer (Cohen, 1987; Brookins, 1990). A recent study of European case-control studies (Darby et al., 2005) suggests an increased risk of lung cancer of about 8% (3–16%) per 100 Bqm^{-3} of 222Rn concentration, which is consistent with an estimate of 11 percent (0–28%) obtained in a combined review of North American studies (Krewski et al., 2005).

Radon/ thoron progenies may get attached to aerosols present in ambient air and are inhaled during breathing (Lubin et al., 1995, 1997; ATDSR, 2012, 2019; Uskalu et al., 2020; Chauhan et al., 2016). These may stick to alveoli and could expose cells of bronchial and pulmonary epithelium in the lungs, thereby damaging the cells, DNA and may cause lung cancer (Seve et al., 1976).Indoor radon exposure has been linked to the development of leukaemia and other malignancies such as melanoma, kidney and prostate cancers (Henshaw et al., 1990; Quindos et al., 1994), as well there is 16 percent increase in relative lifetime lung cancer risk per 100 Bqm^{-3}due to chronic radon inhalation (Fintan et al., 2019). A radon concentration of 50 Bqm^{-3} can cause 25% of leukaemia cases in children and adults of all ages (Richardson et al., 1991). Inhalation of short-lived decay product of radon (Po-214), responsible for 40% of the total radiation dose received by the population, is a major contributor to problems of respiratory system, lung cancer, and damage to sensitive cells in the skin and causes skin cancer (NRC, 1991). Radon and its progeny leads to about 69% of the total annual effective dose in contrast to another natural sources (Khan 1993; Mehra et al., 2015). For non-smokers radon is the primary cause of lung cancer while for smokers it is

secondary, encompassing an approximated 3-20% of deaths due to lung cancer globally (WHO, 2009; Chunxio et al., 2020). Indoor radon causes 21,000 (13.4%) lung cancer deaths in the United States each year, with 2900 of these being non-smokers (USEPA, 2003).

Health hazards posed by radon and thoron gases are acknowledged worldwide by many agencies such as WHO (World Health Organization), UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation), USEPA (United States Environmental Protection Agency) and ICRP (International Commission on Radiological Protection). On behalf of above stated facts, it is clear that the study of naturally occurring uranium, thorium and their subsequent daughter products is necessary.

1.11 STUDIED AREA

The studied area (Figure 1.13) comprises two districts Barnala and Moga in the Malwa region of Punjab, India with a longitude from 30º22’24” N to 30º48’44.19” N and latitude from 75º32’55” E to 75º10’15.71” E. The geographical area of studied area is spread over the area of 3717 sq km with population density of 432 per sq km and population is 1,592,040 as per census of 2011.

According to the Central Ground Water Board report (2017), it is located in the Ghaggar Sub Basin and is occupied by the Indo Gangetic alluvial plain of quaternary age. Soil is rich in nutrients, suitable for crops like wheat, cereals, vegetables etc. Most of the land of the district with loamy sand and sandy loam kaller sand is supported in few parts of the land. Mainly two types of soils Siozerem and Desert soil are present. The siozorem soils are found in major parts of the district and desert soils are comparatively found in a relatively smaller area towards the western part of the district. The alluvial formations are overlain by Aeolian sands, except in the area falling in the proximity of the Satluj River. The soil is also used for the construction of the building and in agriculture for growing crops.

Groundwater is found in alluvium deposits, which are made up of fine to coarse sand and create a possible aquifer. The majority of the district's terrain is covered in loamy sand and sandy loam kaller sand, which is only supported in a few places. The geological formations comprised of the thick sequence of Quaternary deposits of

mid-pleistocene to recent age. The alluvium is the principal aquifer of the studied area, whereas the older alluvium and Aeolian alluvium are the major aquifers. The granite rocks present in the Tosham hill region in the nearby state of Haryana affect the geology of district Bathinda and Barnala. The major rock types present in the area are silicate, granite, igneous and sedimentary rocks. The major minerals reported in the region are quartz, muscovite, plagioclase, feldspars, amphibole, biotite, anhydrite (gypsum), chlorite, fluorite, kaolinite, etc. (Kochar et al., 2015). The climate of these districts for the year can be classified into three seasons that is summer, rainy, and winter. The summer season starts from mid-March to the last week of June and the rainy season starts from July to October and the winter season from November to the first week of March.

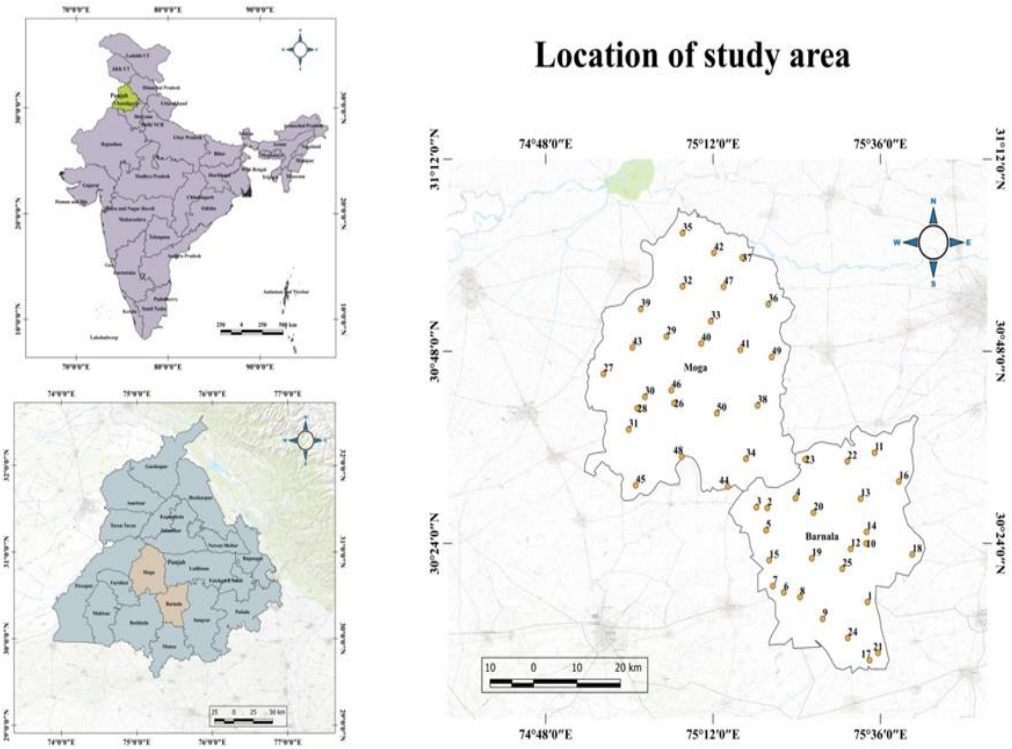

Figure 1.13: Map of studied area (Barnala and Moga districts of Punjab)

The land of Punjab is highly fertile and useful for agriculture. Over the past few decades the land has been used for crop production with excessive use of fertilisers and pesticides. As a result, the land and water has been contaminated with phosphate fertilisers which are the major source of uranium. Over use of water leads to depletion

of groundwater level. The study area has many thermal power plants in its surrounding and the fly ash from these plants is used in the formation of cement which is the major component of building materials. So, these factors make the residents of the area vulnerable to the exposure of natural radiation through ingestion and inhalation. Also, the studied area falls in the Malwa region of Punjab which is known for having the highest cancer rate in the country (Godara et al., 2015).

Although plenty of literature related to radiological health risk assessment is available for the Malwa region but there is scarcity of data for the studied area. Hence the present study has been carried out for the first time to find out the activity concentration of these radiations and their associated carcinogenic health effects for the residents of the area. The fieldwork has been done for various seasons throughout the year in the dwellings under different ventilation conditions. It would also help to identify the areas where concentration is higher and provide the data for the national pool.

1.12 OBJECTIVES

Risk Assessment studies in the environs of our study region (Moga and Barnala districts of Punjab) were carried out with the following objectives has been finalized by the Departmental Doctoral Research Committee (DDRC) during synopsis presentation:

1. To measure the indoor radon, thoron and their progeny concentrations in different types of dwellings and to study their variation with different seasons, ventilation conditions and the metrological parameters based on grid pattern by using DTPS, DRPS and Pin-Hole cup Dosimeters in the Moga and Barnala Districts of Punjab.
2. To study the equilibrium factor (EF) variation in different dwellings of the study region based on ventilation conditions and the metrological parameter.
3. To study the Radon mass exhalation and Thoron surface exhalation in soil samples of study region on the grid pattern basis, using the Smart RnDuo and also study their correlation with indoor radon/thoron concentration levels.
4. To study the spatial and depth-wise distribution of radon concentration in groundwater samples on the grid pattern basis using Smart RnDuo.

5. To study the spatial and depth-wise distribution of Uranium concentration in groundwater samples on the grid pattern basis using LED Fluorimeter.

6. To study the various Radiological health risk parameters for the inhabitants of the study region.

The physicochemical parameters of water has also been studied as suggested by the experts during the pre-thesis presentation in the DDRC meeting.