Chapter 1

Introduction and Review of Literature

1.1   General

Human health always relies on the environment which surrounds it. The macro environment which includes human lifestyle, the air it breaths, the food it eats, radiation and chemicals human is exposed to, as well as viruses and physical agents it comes in contact with; combine with the microenvironment of human cells (to either prevent or enhance carcinogenesis), deciding the final human health (McMichael, 1994; Daily, 1997). Simple inorganic chemicals (oxygen, carbon dioxide, and water), major organic nutrients (proteins, carbohydrates, fats, vitamins) and inorganic minerals (calcium, zinc, iron, copper, sodium, potassium etc) are essential to life. To maintain health, human beings and all other life forms must ingest, inhale, and otherwise have to absorb these essential environmental chemicals and minerals, but within certain limits. People have continuously been working to recognise these limits out, and as more is learned, more helping it is to reduce the risk of a number of important diseases.

Human health is also affected by the entry of pollutants in the environment including chemical vapors from industry, exhaust gases from vehicles and many types of explosions, radioactive fallout and penetrating radiations and radioactive material-continuously pervading the atmosphere from outer space. In the environment, some of these pollutants exist as gases, some are in the form of a suspension of small liquid or solid particles (aerosols) and some are dissolved in cloud vapor or raindrops. Pollutants can reach our body from the atmosphere by inhalation, skin contact or via ingestion (in food or water). Human exposure to radiation, if occurs by radiation from sources outside the body is called external exposure and through ingestion and inhalation and consequent decay of radionuclides in the body is called as internal exposure, discussed in detail in section 1.5.

1.2 Chemical Elements: Toxicity and Essentiality

The action of an element in an animal system depends on its ability to be absorbed and excreted, its valence and electrochemical behavior. These elements play their respective roles in human body (as catalysts for enzymes), and all of them can be toxic at high concentrations. As more and more research is being carried out on the role of elements, which often work at trace levels in biological systems, the clearer it is becoming that there is no way to make a definite distinction between essential and non-essential elements. Metals like copper, zinc and iron which are essential for the physiological functioning of living organisms, become toxic at high concentrations (Adriano, 2001). Factors influencing trace-element bioavailability often dominate relationships between total element supply and the extent to which this remains within the limits required to maintain health. The effect of any substance on a living system is always dependent on the concentration of it available to cells. The term bioavailability refers to describing the effects of a process, physicochemical or physiological, which influences the fraction of an ingested trace element ultimately presented to tissues in forms that can be used to meet functional demands. Use of the term in this context in fact includes processes which modify the systemic utilization of elements after their absorption and is not confined to digestive or absorptive phenomena. Such an approach takes account of the variables including-physicochemical characteristics of trace-element sources in the diet, biochemical interactions of these elements with synergists or antagonists either in the gut lumen or in tissues and physiological variables which, in response to changing relationships between element supply and demand, influence the efficiency of element absorption, storage or incorporation into functional sites.

The criterion for essentiality of an element is that absence or deficiency of this particular element brings abnormalities that can be connected to specific biochemical changes reversed by intake of this particular element. To evaluate essentiality or toxicity one needs quantitative dose-response relationships. Several metal ions are crucial to the metabolism of cells at low concentrations but are toxic at high concentrations, leading to a bell-shaped type of dose-response relationships (Marschner, 1995). These metals are sometimes called micronutrients (Sherameti and Varma, 2010). The factors constituting toxicity of a metal are metal itself, its total concentration, the

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availability of the metal to the organism, and the organism in particular. Depending on the organism and the metal, different modes of action recognized are: binding to macromolecules, disruption of enzymatic functions and catalysis of radical formation, etc. Zinc for example, is a component found in a variety of enzymes like dehydrogenases, proteinases, peptidases, but it is also involved in the metabolism of carbohydrates, proteins, phosphate, auxins, and in RNA and ribosome formation in plants (Mengel and Kirkby, 1982; Kabata-Pendias and Pendias, 2001).

1.3   Radioactive Chemical Elements

Apart from stable chemical elements, radioactive elements (radionuclides) are also formed due to nuclear reactions taking place in stars. A radionuclide is a nuclide with an unbalanced and unstable nucleus. All chemical elemental atoms except Hydrogen, consist of neutrons and protons in their nuclei. The ratio of neutorns to protons is unity for lighter isotpes on the so callled "stability line" and increases gradually as one approaches the behavior of elements at the end of periodic table or moves off the stabilty line. As this ratio increases, a stage is reached where the nuclide is no longer stable. The heaviest stable nuclide is $^{209}_{83}Bi$. Nuclides heavier than this are unstable because they have excess energy to dissipate. Unstable nuclides are called radionuclides and they dissipate their surplus energy by the emission of radiation. This process is called radioactivity or radioactive decay.

Radioactivity is a phenomenon of spontaneous nuclear transformation of radionuclides (so-called radioactive decay), which is accompanied by the emission of particles (α, β–, β+), electron capture, proton emission, or the emission of fragments (i.e., the most common decay modes), and the emission of γ radiation.

Symbolically the process can be described as follows:

$$(A, Z)X \rightarrow (A1, Z1)Y + (A2, Z2)Particle \qquad (1.1)$$
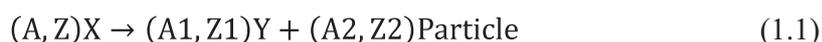
where,

X is the parent nuclide and Y is the daughter nuclide.

Radioactive decay is always exoergic process and the generated products always carry a certain kinetic energy. The basic condition for a radioactive process to take place

is that primary nucleus has more rest energy (mass) than the sum of the rest energies (masses) of the products of the decay:

$$m(X) > m(Y) + m \text{ (Particle)} \tag{1.2}$$

The energy equivalent to this difference in masses is the energy of radioactive decay.

Like stable chemical elements, radionuclides in an environment can be present in comfortable abundances or can be in 'trace' or 'ultra trace' concentrations. Out of 340 nuclides found in nature, about 70 are radioactive and are formed mainly among the heavy elements. More precisely, all elements having atomic number greater than 80 possess radioactive isotopes, and all isotopes of elements heavier than 83 are radioactive. This number of radioactive elements must have been more at the time of formation of universe, but many of them decayed out of existence owing to their short half-lives. Radioactive elements have again essential/beneficial and toxic aspects. Radon exposure, which is considered second leading cause of lung cancer after tobacco smoking (IARC, 1988); has been attracting tourists for Austria and United States; where exposure to natural radioactivity due to high radon concentrations in the air of old mines has been believed to be of curative ability for arthritis, general debility, and certain other diseases (Lewis, 1955). Similarly curative powers of mineral water of spas of Europe, Japan, and South America was a mystery, till the phenomenon of radioactivity was discovered and tests of these mineral waters confirmed to contain abnormally high concentrations of natural radioactive elements. Like stable elements their availability is essential up to a level and hazardous after that. Radioactivity results in radiation in human senses. In most places on earth the radioactivity varies only within narrow limits, but in some localities there are wide deviations from normal levels because of abnormally high rock/soil concentrations of radioactive minerals. These minerals are a natural source of radioactivity. Radionuclides can also be introduced in the environment through human intervention. So, on the basis of origin and formation radionuclides can be categorized into these two categories:

1. Naturally Occurring Radionuclides

2. Artificially Produced Radionuclides

1.3.1   Naturally Occurring Radionuclides

Taking natural radionuclides first, these have been separated into following three types:

(I) Primordial (II) Secondary and (III) Cosmogenic

(I)   Primordial Radionuclides

These are nuclides originated with other (stable) nuclei in the course of cosmic nucleogenesis by thermonuclear reactions in the core of a star, which then exploded as a supernova and enriched the nucleus cloud from which the sun and the solar system originated. They became part of the Earth at the time when the solar system was formed about 4 to 5 billion years ago. To sum up, these are nuclides with half-lives sufficiently long that they have survived since their creation. There are 255 stable primordial nuclides and 33 radioactive primordial nuclides. In addition to primordials that head a decay series like ^{235}U, ^{235}U and ^{232}Th; ^{40}K, ^{50}V, ^{87}Rb, ^{113}Cd, ^{115}In, ^{123}Te, ^{138}La, ^{142}Ce, ^{144}Nd, ^{147}Sm, ^{152}Gd, ^{174}Hf, ^{176}Lu, ^{187}Re, ^{190}Pt, ^{192}Pt, ^{209}Bi are some non-series primordial radionuclides (Eisenbud and Gesell, 1997; Pöschl, and Nollet, 2006). ^{40}K, ^{232}Th and ^{238}U are most important primordial radionuclides, and form the basis of radioactivity analysis of any region. Radioactive elements ^{232}Th and ^{238}U and the long-lived primordial nuclide ^{40}K account for much of the external background radiation dose from radioactivity to which humans are exposed. Primordial nuclides have very long lifetimes, were produced at or before the creation of planet earth and therefore play important role in the Earth's processes. Apart from providing basic natural background to the planet, primordial radionuclides (in particular ^{40}K) have been for example, suggested to be key source of long-term heat in the Earth's core over the past 4.5 billion years (Murthy *et al*., 2003). Some important primordial nuclides are listed in Table 1.1 with their half lives and estimated abundance in earth's crust.

Uranium

Uranium is an important actinide primordial with atomic number 92 and is a ubiquitous radioactive element. Uranium naturally occurs in three isotopic forms ^{238}U (T½= 4.468×10^9 years), ^{235}U (T½ = 7.038×10^8 years), and ^{234}U (T½ = 2.455×10^5 years) (though a large number of synthetic isotopes of uranium have been prepared). The

relative abundances of the three natural isotopes have been measured by various investigators, and 'best values' for the ^{238}U, ^{235}U, and ^{234}U relative abundances have been chosen to be as (Holden, 1977): ^{238}U is present in the amount of 99.28% and is usually in radioactive equilibrium or near-equilibrium with ^{234}U, which is present in the amount of 0.0058%. Uranium-235, the parent isotope of the actinium series, is present in the amount of 0.71%. The isotope ^{238}U is the parent of the natural 4n+2 radioactive series, and the isotope ^{235}U is the parent of the natural 4n+3 radioactive series. ^{234}U arises from ^{238}U by radioactive decay and these two isotopes are thus linked to each other, but ^{235}U is of independent origin.

There are as many as 200 minerals that contain uranium as an essential component (Burns, 1999a; Finch and Murakami, 1999). It is found in almost all types of rocks, soils and natural waters (USDOE, 2001). The uranium concentration is estimated at 1-10 μgg^{-1} (micrograms per gram) in the igneous rocks of the Earth's crust; some sedimentary and granitic rocks can contain much more however. Uranium (+6) form is the most abundant. Uranium (+4) dominates at Eh< 200 mV for water-logged/wet soils whereas U(+6) predominates in sufficiently aerated soil (Abdelouas *et al*., 1998). The greater solubility of U (+6) as the uranyl (UO_2^{++}) compounds, is due to its ability to form stable complexes with various organic and inorganic ligands. Uranium (+4) is rather insoluble and forms complexes by various inorganic ligands such as fluoride, chloride, sulphate, and phosphate. The occurrence and distribution of uranyl species in surface and subsurface water is controlled by the redox condition, pH, and CO_2 partial pressure (Giblin *et al*., 1981). U(VI) is reduced to U(IV) in groundwater systems, if an effective reductant is present, such as H_2S. Among other possible reducing agents are fossil plants, methane, and transported humic material.

Thorium

Thorium is another natural radioactive chemical element with the symbol Th and atomic number 90. In nature, thorium is found solely as thorium-232, and it decays by emitting an alpha particle with a half-life of about 14.05 billion years. Thorium series originates with ^{232}Th. Thorium, like uranium present in the earth's crust in parts per million levels (Evans, 1969). It is estimated to be about three times more abundant

than uranium in the Earth's crust. Thorium occurs in several minerals, the most common being the rare earth-thorium-phosphate mineral, monazite. It contains up to 12% thorium oxide an average of 6-7%. Monazite is found in igneous and other rocks. World monazite resources are estimated to be about 12 million tonnes, two thirds of which are in heavy mineral sands deposits on the south and east coasts of India.

Table 1.1 Some important primordial radionuclides, their half-lives and natural abundances **(**Pöschl and Nollet, 2007)

Radionuclide	Half-life (Years)	Estimated Abundance in Crust (μgg^{-1})
^{40}K	1.38×10^9	2-3
^{87}Rb	4.8×10^{10}	3-9
^{138}La	1.1×10^{11}	$1 \times 10^2 - 2 \times 10^2$
^{147}Sm	1.1×10^{11}	0.5-1
^{187}Re	4.0×10^{10}	$3-5 \times 10^4$
^{232}Th	1.4×10^{10}	1-20
^{235}U	7.0×10^8	$0.3 \times 10^2 - 3 \times 10^2$
^{238}U	4.5×10^9	0.5-5

Potassium

^{40}K with half-life of 1.28×10^9 years, is most widespread primordial radionuclide on the Earth's surface. Potassium is quite an abundant element, composing more than 2% of the Earth's crustal mass. ^{40}K can decay by gamma emission (11% of the decay pathway) to give ^{40}Ar, and this is the basis for the potassium/argon methods used for dating old rocks and meteorites. ^{40}K can also emit a β particle and lead to the formation of ^{40}Ca (89% of the decay processes). ^{40}K is in fact, the most significant natural source of radioactivity ingested by humans, because of its ubiquity and biological uptake.

(II)   Secondary Radionuclides

Secondary radionuclides are derived from radioactive decay of their corresponding primordials. Natural radionuclides ^{232}Th, ^{238}U and ^{235}U decay into nuclei, which are radioactive, themselves. ^{226}Ra, ^{222}Rn are important and widespread secondary radionuclides. ^{210}Pb (half-life 22.3 years), ^{210}Bi (half life 5 days) and ^{210}Po (half-life 138 days) are other secondary radionuclide that are formed from ^{238}U decay via ^{222}Rn. These decay products attach themselves to fine atmospheric aerosols in the respirable size range and have been used to estimate the residence times of submicron aerosols in the environment (Marley *et al*., 2000; Gaffney *et al*., 2004).

Radium

Radium with atomic number 88, exists in 25 isotopic forms, ^{223}Ra, ^{224}Ra, ^{226}Ra and ^{228}Ra being the most common ones. Radium is extremely radioactive, and its decay products are responsible for a major fraction of the dose received by humans from the naturally occurring internal emitters (Singh *et al*., 2005). ^{226}Ra, a member of ^{238}U decay series, is an alpha emitter that decays, with a half-life of 1622 years, to ^{222}Rn, with a half-life of 3.8 days. Radium-226 is present in all rocks and soils in variable amounts. Igneous rocks contain somewhat higher concentrations of radium than sandstones and lime stones. An average concentration of 0.42 pCig^{-1} (~16 Bqkg^{-1}) in limestone and 1.3 pCig^{-1} (~48 Bqkg^{-1}) in igneous rock is found reported (Rankama and Sahama, 1950). Radium-226 is generally in approximate equilibrium with ^{238}U. ^{228}Ra is product of ^{232}Th breakdown, with a half-life of 5.75 years. Although ^{228}Ra frequently occurs in soil and water in an approximate 1:1 ratio to ^{226}Ra, there is little information about its occurrence in foods or in human tissues. Radium, being an α-emitter, does not add directly to the γ activity of the environment but does so indirectly through its γ-emitting decay products. The decay of ^{222}Rn is followed by successive disintegration of a number of short-lived alpha and beta-emitting progeny. After six decay steps, in which isotopes are produced that range in half-lives from 1.6×10^{-4} seconds to 26.8 minutes, ^{210}Pb is produced, which has a half-life of 22 years. This nuclide decays through ^{210}Bi to produce ^{210}Po ($\Gamma_{1/2}$=138 days), which decays by α emission to stable ^{206}Pb.

Radon

Radon gas can be found wherever uranium is present in the environment, occurs naturally as the decay product of radium in the decay chain of uranium. Radon is one of the densest substances that remains a gas under normal conditions and is considered to be a health hazard due to its radioactivity (UNSCEAR, 1993). Among the three commonly occurring isotopes- ^{222}Rn, ^{220}Rn and ^{219}Rn of radon; ^{222}Rn is most stable with a half-life of 3.8 days and principal decay mode again being α decay of energy 5.49 Mev. Similarly, ^{224}Ra, which is a descendant of the ^{232}Th chain, decays by α emission to 54-second ^{220}Rn, historically known as thoron. ^{219} Rn, historically called actinon, is a member of the ^{235}U chain and decays most rapidly, having a half-life of about 3.92 seconds. All radon isotopes are noble gases, occurring as non polar, monatomic molecules, and are inert for practical purposes. ^{222}Rn and its progeny have been of particular concern as environmental hazards, particularly in homes and buildings where air infiltration rates are low. Radon can concentrate significantly in the lower levels of buildings like basements and cellars from ground-source uranium contents and lead to potential inhalation risks in indoor environments (USEPA, 2004).

(III)   Cosmogenic Radionuclides

Cosmogenic radionuclides are continuously produced by bombardment of stable nuclides by cosmic rays, primarily in the atmosphere. These are natural radionuclides that originate by nuclear reactions when high-energy cosmic radiation passes through the Earth's atmosphere. Examples include radiocarbon (^{14}C) and tritium (^{3}H). More than 20 cosmogenic radionuclides have been identified. Apart from (^{14}C) and ^{3}H, ^{24}Na and ^{7}Be are other nuclides of this category which are of any consequence from the perspective of dose to humans. Cosmogenic radionuclides have higher production rates in the stratosphere than in the troposphere, because of higher intensity of cosmic rays in the stratosphere.

1.3.2   Artificially Produced Radionuclides

Radionuclides that do not exist naturally; or occur naturally but with abundance far from sufficiency to meet the demands of present science and technology, industry, and health services are produced artificially. For example, ^{14}C, ^{32}P and ^{24}Na exist in

nature in very low abundance, but are very significant in some biological processes. These are produced artificially. Nuclear reactors, particle accelerators and particle generators are some means to produce radionuclides. This can be achieved by bombardment of the initial nucleus with suitable particles, whereby a radioactive nucleus from a stable nucleus is produced by changing the number of protons or neutrons without disturbing the equilibrium configuration. Neutron is most common bombardment particle. Proton, alpha particle and deuteron are other examples. Among other artificial radionuclides are those used in medicine as tracers for diagnostics or as agents for radiotherapy and including ^{131}I, ^{51}Cr (27.7 d), ^{54}Mn and ^{60}Co, ^{90}Sr, ^{137}Cs, and ^{131}I are among many of the artificial radionuclides that have contaminated soils and plants, and have been studied most thoroughly because they usually result in the highest doses to humans from the release of mixed fission products (Eisenbud and Gesell, 1997).

Artificial radionuclides have also entered the human environment as a consequence of atmospheric nuclear weapon tests apart that produced for peaceful purposes. Global fallout of fission products in the 1950s and 1960s involved deposition of ^{137}Cs (30.17 y), ^{90}Sr (28.5 y), ^{89}Sr (50.5 d), ^{3}H, ^{54}Mn (312 d), ^{65}Zn (244 d), ^{95}Zr (64 d), $^{103/106}$Ru (39.4d/368 d), ^{129}I (1.6×107 y) and ^{144}Ce (284.8 d). At present the most important sources are routine releases from nuclear power (NPP) and reprocessing plants. However, high emissions of these isotopes can take place in developments like - NPP accidents; such as Chernobyl (Ukraine) accident in 1986 (UNSCEAR, 1988). This accident increased $^{134/137}$Cs inventories of many European countries considerably.

1.4   Ionizing Radiation

The term radiation is used to mean ionizing radiation. The term has been in use to refer the radiation exposure (or exposure in short) in a generic sense to mean the process of being exposed to radiation or radionuclides, the significance of exposure being determined by the resulting radiation dose (ICRP, 1991b). Ionizing radiations include electromagnetic waves (photons) of high frequency, and particles of varying mass and charge, e.g., electrons, protons, neutrons, and alpha particles, travelling at initial velocities approaching the speed of light (NCRP, 1989). In contrast to other

forms of radiation, each-ionizing radiation track is able to deposit enough localized energy in an absorbing material to disrupt atoms and molecules in its path. The resulting atomic changes, e.g., the ejection of an electron from its orbit around the nucleus of an atom, or the ejection of a proton from the nucleus of an atom, can produce ion pairs. The ion pairs, in turn, may lead to molecular damage and, ultimately, biological injury. Ionizing radiations are generally characterized by their ability to excite and ionize atoms of matter with which they interact. Radiations are mainly categorised into following four major groups (Pöschl and Nollet, 2006):

- Heavy charged particles, including all particles with a mass greater than or equal to one atomic mass unit (amu), such as alpha particle, proton and fission products.

- Lighter charged particles like electrons, positrons, internal conversion electrons and auger electrons.

- Electromagnetic radiations gamma rays, characteristic x-rays, annihilation radiation and bremsstrahlung. X-ray and γ-ray photon of a given quantum energy have identical properties, differing only in mode of origin.

- Neutrons; including fast neutrons, intermediate neutrons, epithermal neutrons, thermal neutrons, and cold neutrons. Neutrons can be generated from spontaneous fission, radioisotope (alpha-neutron) sources, photo-neutron sources, or reactions from accelerated charged particles.

Along with above categorization of ionizing radiation, there exists another basis, which divides radiation into two broad categories: directly and indirectly ionizing radiation. This categorization stems from the fact that interactions of ionizing radiation within cells are direct or indirect in nature and deals with biological consequences of radiation with matter (ICRU, 1971; Streffer, 2004). The mechanism of interaction of radiation depends actually on both, the physical properties of the radiation and the physical and structural properties of the materials. This may also depend upon the fact that weather the interaction is direct or indirect.

Directly Ionizing Radiation: These are fast charged particles, which deliver their energy to matter directly, through many small Coulomb-force interactions along the particle's track. For alpha particles, which are considered high linear energy transfer (LET) radiations, direct action is the dominant process by which the critical targets are affected.

Indirectly Ionizing Radiation: X- or γ-ray photons and uncharged particles (neutrons etc), first transfer their energy to charged particles in the matter through which they pass. Deposition of energy in matter by indirectly ionizing radiation is thus a two-step process. The resulting fast charged particles then in turn deliver the energy to the matter. Indirectly ionizing such as beta, gamma and X-rays radiation produce free radicals that can then diffuse and damage the critical target. This mode of delivering radiation damage is called indirect action.

Direct effects are produced by the initial action of the radiation itself and indirect effects are caused by the later chemical action of free radicals and other radiation products. For example, whereas it takes about 10^{-3}s, to completely react radicals produced by a charged-particle track in a biological system, cell division is affected in a time of the order of minutes; and so on. Further, including type and energy of radiation, spatial distribution of delivered energy, total dose (energy per mass) delivered and dose rate are other quantities which matter while studying biological interaction of radiation. Radiobiological studies have shown that, in general, cells most sensitive to the effects of ionizing radiation are those that are undifferentiated, well oxygenated, are highly metabolically active, and rapidly reproduce.

Depending upon the consequences of the random nature of radiation, biological effects on the cell can further be divided into two general categories, stochastic and deterministic, or non-stochastic. Stochastic effects are those that occur in a statistical manner. As an example, if a given population is exposed to a significant amount of a carcinogen, such as radiation, then an elevated incidence of cancer can be expected. But, since there is a certain natural incidence of cancer without specific exposure to radiation, it may never be completely certain whether a given case was induced or would have occurred without the exposure. One only be able to predict the magnitude

of the increased incidence, and cannot say which particular individuals in the population will contract the disease and which will not. Stochastic effects (cancer and hereditary effects) may be caused by damage in a single cell. Important advances and judgments are given in ICRP Publication 92 (ICRP, 2003).

Deterministic effects in contrast, are those that show a clear relationship between dose and effect in a given individual. Strand break in DNA caused by ionization, Skin reddening and radiogenic cataract are examples of deterministic or non-stochastic effects. Deterministic (threshold) effects will occur only if the radiation dose is substantial, for example, in an accident. Usually there is a threshold below which no effect is observed, and the severity increases with dose. At low doses, one cannot say with certainty what the risk is to an individual. As a practical hypothesis, one usually assumes that any amount of radiation, no matter how mall, entails some risk. A cataract is an opacification of the lens of the eye. The threshold for ophthalmologically detectable lens opacification, as observed in patients treated with X rays to the eye, ranges from about 2Gy for a single exposure to more than 5Gy for multiple exposures given over several weeks. This level is also consistent with data from Hiroshima and Nagasaki.
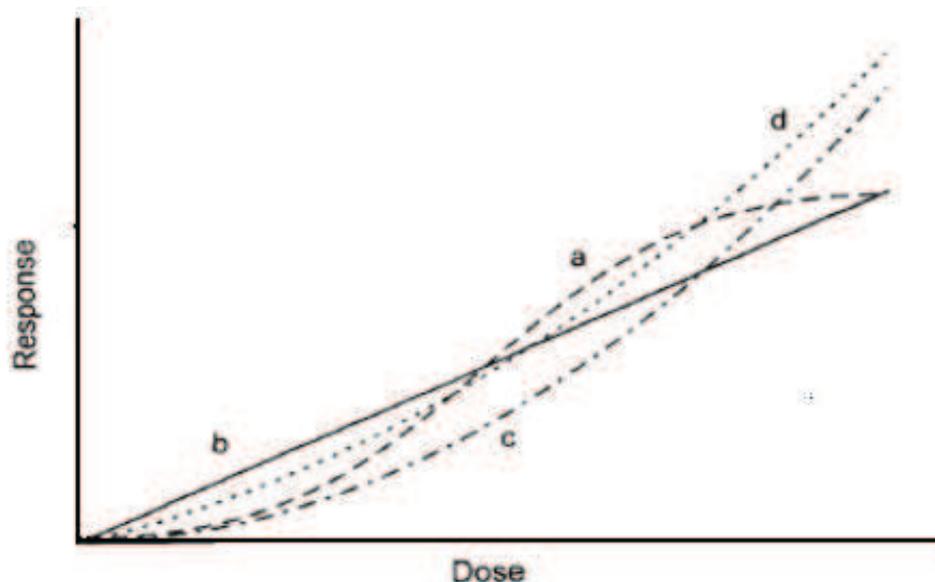

Figure 1.1 Various proposed types of dose-response curves for radiation exposure (Eisenbud and Gesell, 1997)

During the 1950s there was already evidence the dose-response relationship for induction of genetic effect was linear and that it was probably without threshold. This was in contrast to cancer induction, for which a threshold was then believed to exist and for which linearity of response was not generally accepted. It was believed the dose-response relationship for cancer was sigmoidal in form and that a threshold existed below which cancer was not produced (Eisenbud and Gesell, 1997). Sigmoidal dose-response relationship, which is widely applicable in toxicology, is no longer applicable for stochastic effects, such as cancer in radiobiology. The sigmoidal response is shown in curve 'a' of Figure 1.1**.** In this type of relationship, effects per unit of dose gradually increase until a dose level is reached at which the rate of increase begins to diminish and eventually reaches a platue when all susceptible members of the population have been affected. This can be noted form the curve that there may be a threshold below which no effects occur, or the question as to whether a threshold exists may be indeterminate because of statistical limitations in the data. Curve 'b' in this figure illustrates the linear-no threshold response, which, because of statistical limitations, cannot always be distinguished from other dose-response relationships. Another dose-response relationship is the quadratic form KD^2 (curve 'c'), where the number of effects produced per unit dose increases progressively with increasing dose. The linear-quadratic relationship (curve 'd') has got wider acceptance however, and is given by the equation (Upton, 1977):

$$I = aD + bD^2 + C \qquad\qquad (1.3)$$

Here,

I is incidence at dose D, c is the incidence at zero dose, and a and b are constants determined empirically. In this relationship the effects are linear at low doses but become curvilinear at higher doses according to the relationship.

1.5   Terms and Units used in Radiological Risk Assessment

Radiological assessment is the process of estimating dose and risk to humans from radioactive materials in the environment (Till and Helen, 2008). Radioactive materials are generally released from a man-made or natural source may be transported through the environment and appear as concentrations in environmental media. These

concentrations can be converted to dose and risk by making assumptions about exposure to people. Activity of a radioactive material and the radiation dose are two basic quantities in the assessment of radiation levels. Activity of a radioactive material is the number of nuclear disintegrations per unit time with unit Becquerel (Bq). One Becquerel is one disintegration per second. The term radiation dose can serve several terms (absorbed dose, dose equivalent or effective dose equivalent). Table 1.2 summarizes various dose quantities and their units with related conversion factors.

The dose or risk to a person depends upon a number of exposure factors, such as time, location, transport of radionuclides through the environment, and the traits of the individual like physiological parameters (e.g., breathing rate), type of dietary and living habits, use of local resources and recreational activities. In radiological assessment, a specific set of these characteristics is referred to as an exposure scenario. The target of radiological assessment may be real individuals or representative individuals. Real individuals are those who are or were actually exposed. Their characteristics should be defined as closely as possible to those that actually exist. Representative, or hypothetical, individuals are not characterized by specific persons but have characteristics similar to people in the area who are or were exposed in the past or who may be exposed in the future.

1.5.1   Exposure

Exposure is oldest and one of the important fundamental (nonstochastic) quantities with which radiological physics concerns. Exposure is defined for gamma and X-rays in terms of the amount of ionization they produce in air. Exposure is symbolized by X, and is defined as "the quotient of dQ by dm, where the value of dQ is the absolute value of the total charge of the ions of one sign produced in air when all the electrons (negatrons and positrons) liberated by photons in air of mass dm are completely stopped in air." (ICRU, 1980)

$$X = \frac{dQ}{dm} \tag{1.4}$$

The unit of exposure is called the roentgen (R) and was introduced at the Radiological Congress in Stockholm in 1928. It was originally defined as that amount of gamma or X radiation that produces in air 1 esu of charge of either sign per

0.001293g of air. (This mass of air occupies 1 cm^3 at standard temperature and pressure.) The charge involved in the definition of the roentgen includes both the ions produced directly by the incident photons as well as ions produced by all secondary electrons. The unit roentgen is now defined as

$$1R = 2.58 \times 10^{-4} \text{ Ckg}^{-1} \tag{1.5}$$

The concept of exposure applies only to electromagnetic radiation; the charge and mass used in its definition, as well as in the definition of the roentgen, refer only to air.

Exposure can be divided into two main categories:

External exposure: External irradiation is caused by radioactivity present in the soil and in any other material surrounding our bodies, including the air.

Internal exposure: Internal irradiation is caused by radionuclides that are inhaled or ingested.

Radioactivity in air due to cosmic radiation is a source of external irradiation of human beings as well as internal irradiation. Exposure from cosmic rays at ground level is primarily from muons, electrons, photons, and neutrons. This exposure is realized as external exposure. The intensity of cosmic rays and the dose absorbed depends on the altitude and on geographic latitude. Thicker the layer of atmosphere above, lower the absorbed dose. At sea level, the typical annual effective dose due to cosmic rays is about 350 µSvy^{-1}. World average (altitude and latitude averaged) annual effective dose due to cosmic rays is about 460 µSvy^{-1} (UNSCEAR, 2000). Terrestrial radiation is formed mainly by radionuclides from the ^{238}U and ^{232}Th series and from ^{40}K. These radionuclides irradiate our body with γ radiation (externally and internally) and alpha and beta irradiation (mainly internal). Irradiation and effective doses caused by inhalation of terrestrial nuclides results due the presence of dust particles containing radionuclides of the ^{238}U and ^{232}Th series. The most important and dominant contributors to inhalation dose are decay products of radon.

The presence of radioactivity in drinking water is a potential source of internal exposure to radioactivity and comprises of a possible mode of ingestion of radioactivity. Depending on the mineralogy of the bedrock where the water comes from, and the

contamination due to man-made activities, radioactivity content varies. Primordial radionuclides are always present in drinking water to a certain degree, especially in well water or mineral water. The most relevant of these nuclides for radiological protection are ^{226}Ra and ^{222}Rn. A non-negligible radiation dose can be received by the drinking water with a high radioactive content. This is especially relevant in the case of ^{222}Rn and ^{226}Ra. Dose rates can be calculated by using the obtained concentrations and the conversion factors (dose equivalents per unit activity) published by agencies like UNSCEAR and International Commission on Radiological Protection (ICRP).

1.5.2 Absorbed Dose

Realizing that it would no longer be sufficient to express all exposure restrictions in roentgen units, ICRP in 1954 came up with glossary defining absorbed dose and the corresponding unit, rad (=0.01 Gy in modern terms), as described by ICRU (ICRP, 1955). Sub-Committee I also introduced a new RBE-weighted unit, the rem (=0.01 Sv in modern terms). Absorbed dose is relevant to all types of ionizing radiation fields, whether directly or indirectly ionizing, as well as to any ionizing radiation source distributed within the absorbing medium. Absorbed dose D can best be defined in terms of the related stochastic quantity energy imparted ε (ICRU, 1980). The energy imparted by ionizing radiation to matter of mass m in a finite volume V is defined as

$$\varepsilon_{tr} = (R_{in})_u - (R_{out})_u + (R_{in})_c - (R_{out})_c + \sum Q \qquad (1.6)$$

where,

$(R_{in})_u$= radiant energy of uncharged particles entering V,

$\sum Q$ = net energy derived from rest mass in V

$(R_{out})_u$ is the radiant energy of all the uncharged radiation leaving V,

$(R_{in})_c$ is the radiant energy of the charged particles entering V,

$(R_{out})_c$ and is the radiant energy of the charged particles leaving V. We can now define the absorbed dose D at any point P in V thus is:

$$D = \frac{d\varepsilon}{dm} \qquad (1.7)$$

where,

ε is the expectation value of the energy imparted in the finite volume V during some time interval, dε is that for an infinitesimal volume dv at point P, and dm is the mass in dv. The unit of absorbed dose, Jkg^{-1}, is called the gray (Gy). The older unit rad, is defined as 100 $ergg^{-1}$. It follows that

$$1Gy = \frac{1J}{kg} = \frac{10^7 erg}{10^3 g} = \frac{10^4\ erg}{g} = 100\ rad \qquad (1.8)$$

1.5.3 Equivalent Dose

It is established that the absorbed dose needed to achieve a given level of biological damage e.g., 50% of cell killing is different for different kinds of radiation. Radiation with a high linear energy transfer (LET) is generally more damaging to a biological system per unit dose than radiation with a low LET. The term linear energy transfer (LET) (ICRU, 1980) is used to characterize the distribution of ionizing events along the path of impinging radiation. Since the high-LET radiations are capable of causing more damage per unit absorbed dose, equivalent dose- a weighted quantity, or its average over all organs-effective dose, is used for radiation protection purposes. For low-LET radiation, equivalent dose equals absorbed dose. For high-LET radiations, such as neutrons, α-particles or heavier ion particles- equivalent dose or effective dose equals the absorbed dose multiplied by a factor, the quality factor or the radiation weighting factor. The dose equivalent H is defined as the product of the absorbed dose D and a dimensionless quality factor Q, which depends on LET:

$$H = QD \qquad (1.9)$$

It is here to be noted that weighting factor for radiation quality being dimensionless, the unit of equivalent dose is also joules per kilogram. The special unit sievert (Sv) has been introduced for use with equivalent dose and effective dose to avoid confusion between the two dose quantities. For simplicity, all dose units in the Public Summary are reported in sieverts (Sv) (NRC, 2001).

Table 1.2 Summary of various dose quantities and their units （NRC, 2001)

Unit [a]	Symbol	Conversion Factors
Becquerel (SI)	Bq	$1Ci = 3.7 \times 10^{10}\,Bq$
Curie	Ci	
Gray (SI)	Gy	$1\,Jkg^{-1} = 100\,rads$
Rad	Ra	$0.01\,Gy = 100\,ergg^{-1}$
Sievert (SI)	Sv	$1\,Jkg^{-1} = 100\,rem$
Rem	Rem	$1rem = 0.01\,Sv$

[a]International Units are designated SI.

1.5.4 Kerma

Kerma refers to kinetic energy released in material. This again is a dosimetric quantity and stands for the kinetic energy transferred to charged particles per unit mass of irradiated medium when indirectly ionizing (uncharged) particles, such as neutrons, traverse the medium. This non-stochastic quantity is relevant only for fields of indirectly ionizing radiations (photons or neutrons) or for any ionizing radiation source distributed within the absorbing medium. Kerma, K is the quotient of dE_{tr} by dm, where dE_{tr} is the sum of the initial kinetic energies of all the charged particles liberated by uncharged particles in a mass dm of material. The widest application of kerma involves photon beams and air. This is referred to as air kerma. The conventional and SI units for kerma are $ergg^{-1}$ and Jkg^{-1}, respectively. The conventional and SI special names for the units of kerma are the rad and gray.

The kerma K can be defined in terms of the related stochastic quantity energy transferred, ε_{tr} (Attix, 1979, 1983) and the radiant energy R (ICRU, 1980). The energy transferred in a volume V is:

$$\varepsilon_{tr} = (R_{in})_u - (R_{out})_u + \sum Q \qquad (1.10)$$

where

R_{in} and ΣQ are defined the same as for Equation (1.6),

$(R_{out})_u$= radiant energy of uncharged particles leaving V, except that which originated from radiative losses of kinetic energy by charged particles while in V.

Radiative losses, here stand for conversion of charged-particle kinetic energy to photon energy, through either bremsstrahlung x-ray production or in-flight annihilation of positrons. In the latter case only the kinetic energy possessed by the positron at the instant of annihilation (which is carried away by the resulting photons along with 1.022 MeV of rest-mass energy) is classified as radiative energy loss. Radiant energy R is defined as the energy of particles (excluding rest energy) emitted, transferred, or received (ICRU, 1980).

Kerma K at point of interest P in V then is defined as

$$K = \frac{d\varepsilon_{tr}}{dm} \qquad (1.11)$$

where

$d\varepsilon_{tr}$ is the expectation value of the energy transferred in the finite volume V. If all of the kinetic energy is absorbed "locally," the kerma is equal to the absorbed dose.

1.6 General Levels of Radiation Exposures in the Environment

Life on earth has evolved in the presence of naturally occurring ionizing radiation, which is continuous and ubiquitous. However, in addition to natural background radiation exposure, mankind is now also exposed to radiation from various man-made sources such as nuclear power industry (from mining to waste disposal), fallout from nuclear weapons testing, and a number of miscellaneous sources, including the use of radioactive pharmaceuticals in the practice of medicine. The average dose rate from these sources received by a person living at sea level is about 3 mSv (300mrem) effective dose equivalent per year (UNSCEAR, 2008). Natural and artificial sources contribute to the total 3 mSv in the following proportions:

a) Natural Sources (~ 2.4 mSv)
b) Artificial Sources (~ 0.6 mSv)

Thus, of the total effective dose of 3mSv, 80% (2.4 mSv) is due to naturally occurring sources of radiation. The dose may be larger at higher altitudes because of increased cosmic ray intensities, and it may be larger in areas where the content of natural radioactive material in the soil is higher**.** Natural background radiation, which further can be classified into external and internal exposure categories, consists mainly of: (1) Radon and radon daughters, (2) cosmic rays, (3) cosmogenic radiation (4) terrestrial radiation, and (5) internal radiation (potassium-40, carbon-14 and other naturally-occurring radionuclides in the body). Approximately 2 mSv of the 2.4 mSv is due to the inhalation of radon and its daughters.

Table 1.3 Average radiation dose from naturally occurring sources (UNSCEAR, 2008)

Source	Worldwide average annual effective dose (mSv)	Typical annual effective dose range (mSv)
External exposure		
Cosmic rays	0.39	0.3-1[a]
Terrestrial radiation (outdoors and indoors)	0.48	0.3-1[b]
Internal exposure		
Inhalation (mainly radon)	1.26	0.2-10[c]
Ingestion (food and drinking-water)	0.29	0.2-1[d]
Total	**2.4**	**1-13**

[a] Range from sea level to high ground elevation.

[b] Depending on radionuclide composition of soil and building material.

[c] Depending on indoor accumulation of radon gas.

[d] Depending on radionuclide composition of foods and drinking-water.

Of the total 0.6 mSv exposure due to artificial sources per year to members of the general population, 19.6% (almost 0.6 mSv) is due to the use of radiation for medical diagnosis and the remaining 0.4% (around 0.01 mSv) is due to other sources of human-made radiation. There can be large variability in the dose received by individual members of the population, depending on where they live, their dietary preferences and other lifestyle choices. Individual radiation doses can also differ depending on medical treatments and occupational exposures.

1.6.1 Exposure to Uranium

Uranium isotopes are all radioactive, and their decay produces a number of secondary radioactive elements that continue to decay until they reach stable nuclei. Uranium isotopes contribute significantly to terrestrial radiation. Inhalation doses due to uranium isotopes are significant only via their decay products as average annual effective dose from inhalation of uranium and thorium series radionuclides in air without contributions of radon ^{222}Rn and thoron ^{220}Rn is typically about 5 to 10 µSvyear^{-1} (Pöschl and Nollet, 2007). But chronic exposure of this radionuclide in drinking water is a potential health risk (Blantz *et al*., 1985). Although ubiquitous in the environment, uranium has no known metabolic function in animals and is currently regarded as non-essential (Berlin and Rudell, 1986). It enters into human tissues mainly through drinking water, food, air and the other occupational and accidental exposures. The toxicity of uranium is a function of the route of exposure, particle solubility, contact time, and route of elimination (ATSDR, 1999). Uranium accumulated in human results in chemical and radioactive effects.

The principal sites of uranium deposition in the body are the kidneys, the liver and the bones. The health effects of uranium can be divided into carcinogenic and non-carcinogenic effects (WHO, 1998) corresponding to radiological risk (carcinogenic) due to radiation of uranium isotopes and the chemical toxicity risk (non-carcinogenic) due to uranium as heavy metal, respectively (Domingo, 1995; Zamora *et al*., 1998). Uranium is a metal whose biological effects are very similar to other heavy metals Pb, Hg and Cd; lead being the closest analogue. Chemical toxicity of soluble uranium compounds is liable to surpass the potential radiotoxic effects. It is an identified

nephrotoxine. Its nephrotoxic effects are more likely due to its chemical properties rather than its radioactivity; ingested uranium may have a radiological effect on other tissues of deposition though (Zamora *et al*., 1998). Many isolated studies are published for the mechanism for the toxic effects of uranium at moderate to high acute doses on experimental animals (Dygert, 1949). In one of the latest experimental studies, maternal stress has been suspected to enhance the metal-induced adverse effects on embryo/fetal and postnatal development during pregnancy in mammals (Domingo *et al*., 2004). Even if kidney is a critical organ for uranium toxicity, the brain could also be a target organ after uranium exposure (Linares *et al*., 2006).

General population may be exposed to low level of uranium by inhalation or through the diet. Uranium may be also introduced into drinking water supplies through the mining and milling of uranium ores (Domingo *et al*., 2001). The United Nations Scientific Committee on Effects of Atomic Radiation (UNSCEAR, 1982) established the limits for natural uranium in drinking water based on its chemical toxicity for kidney rather than on hypothetical radiological toxicity for skeletal tissues. A level of 100 μgl^{-1} of uranium in drinking water was chosen reasonable, based on consideration of renal toxicity with the application of a safety factor of 50 to 150 (Wrenn *et al*., 1985). In 1991, the U.S. Environmental Protection Agency (USEPA) classified uranium as a confirmed human carcinogen (group A) and has suggested that zero tolerance only is a safe acceptable limit for the carcinogenic risk. At present, a realistic regulation level-maximum contaminant level (MCL) of 30 μgl^{-1} due to USEPA exists (USEPA, 1999b). The World Health Organization recommended a reference level of 15 μgl^{-1}, considered being protective for subclinical renal effects in 2004 (WHO, 2004). In its 4th edition of "Guidelines for Drinking Water Quality" WHO however, has modified the recommended provisional guideline value to 30 μgl^{-1} because of uncertainties in the health database (WHO, 2011). This provisional guideline value for total content of uranium in drinking-water is based on its chemical toxicity, which is predominant compared with its radiological toxicity. Separate guidance levels are provided for individual uranium radioisotopes in terms of radioactivity (i.e. expressed as Bql^{-1}). Guideline value for naturally occurring radioactive isotope ^{238}U that heads the uranium

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decay series, is 10 Bql^{-1}. Maximum acceptable level of uranium in drinking water as per India's Atomic Energy Regulatory Board is 60 µgl^{-1} (AERB, 2004).

1.6.2   Exposure to Radon

Potential inhalation risks of radon isotopes in indoor environments find huge space in the literature (Mayya *et al*., 1998; UNSCEAR, 2000; Amgarou *et al*., 2001; Misdaq *et al*., 2003; UNSCEAR, 2008; Singh *et al*., 2007; Prasad *et al*., 2008; Bi *et al*., 2010; Chen *et al*., 2010). There is general agreement among the experts about the significant role of elevated radon levels in dwellings to lead to lung cancer in the dwellers. In a confined space, like that in a mine/dwelling, radon can accumulate in the inner atmosphere due to contribution from the ground and building materials. This leads "indoor" radon concentration to be higher than "outdoor". The volume activity of radon in the air is thus, classified as indoor activity and outdoor activity. Indoor radon exposure is by far the source of the greatest dose received by the public and has been considered second leading cause of lung cancer after tobacco smoking (IARC, 1988).

Atoms of several radionuclides attach to the atmospheric particles which thus appear as radioactive aerosols (Schery, 2001). One of the possible classifications of these aerosol radionuclides may be on the basis of their origin and is as follows: (i) Cosmogenic: ^{7}Be, ^{22}Na, ^{32}P and ^{35}S; (ii) radon and thoron short-lived decay products; (iii) fission products: ^{89}Sr, ^{90}Sr, ^{103}Ru, ^{131}I, ^{132}Te, ^{137}Cs and ^{140}Ba; (iv) originated from high-energy accelerators: ^{7}Be, ^{22}Na, ^{24}Na and ^{52}Mn; (v) plutonium isotopes from nuclear weapons testing and reactor accidents; and (vi) mine particulate matter containing radionuclides of the uranium, thorium and actinium radioactive decay chains (Papastefanou, 2008). We focus here on radon and short-lived decay products.

Lung is considered to be the most vulnerable organ in human body to receive doses from radon and decay products (Marsh *et al*., 2012). Alpha irradiation of cells in the bronchial epithelium can lead to development of lung cancer. When radon gas is inhaled, densely ionizing alpha particles emitted by deposited short-lived decay products of radon (^{218}Po and ^{214}Po) can interact with biological tissue in the lungs leading to DNA damage. When air that contains ^{222}Rn or ^{220}Rn in partial or total equilibrium with their decay products is inhaled, the inert gases are largely exhaled

immediately. However, a fraction of the dust particles containing attached decay products (radio-active aerosols) are deposited in the lung, with the place of deposition and the manner of clearance from the lung dependent on the factors discussed elsewhere (Falk *et al*., 1992). When breathing air, aerosol particulates are deposited partly on the walls of the respiratory tract, deposition depends strongly on the particle size (Hofmann and Koblinger, 1990; Alföldy *et al*., 2009). Apart from size, aerosol particles however, also differ markedly in their shape, structure and chemical composition. In contrast to general aerosols, the health detrimental effects of radioactive aerosols have been known for a long time and their dosimetry well elaborated. Maximum of the inhaled 1 nm particles are deposited in the nasopharyngeal region and the rest, in the tracheobronchial region with no deposition in the alveolar region (Oberdörster *et al*., 2005). 5 nm particles are almost equally deposited in all three regions. On the other hand, half of the 20 nm particles are deposited in the alveolar region and the remaining half in the other two regions equally.

The development of metastatic cancer induced by α-particles from radon is a multistage process. To this effect, Zhao *et al*. (2001) have put forward a model of neoplastic transformation based on human papillomavirus-immortalized human bronchial epithelial (BEP2D) cells. Cancer is generally thought to require the occurrence of at least one mutation, and proliferation of intermediate cells that have sustained some degree of DNA damage can greatly increase the pool of cells available for the development of cancer (WHO, 2009). Since even a single alpha particle can cause major genetic damage to a cell, it is possible that radon-related DNA damage can occur at any level of exposure. Therefore, it is unlikely that there is a threshold concentration below which radon does not have the potential to cause lung cancer.

1.7 Exposure to Heavy Metals

Heavy metals are chemical elements with a specific gravity that is at least 5 times the specific gravity of water. The specific gravity of water is 1 at 4 °C (39 °F). In one of the definitions, heavy metal refers to a metal or metalloid with a density exceeding 5 gcm^{-3} (Parker, 1989; Lozet and Mathieu, 1991). Apart from defining heavy metals on the basis of density, there exist definitions based on atomic number and

chemical properties, to be discussed in chapter 4. Some important toxic metallic elements with a specific gravity 5 or more times that of water are arsenic (5.7), cadmium (8.65), iron (7.9), lead (11.34) and mercury (13.546) (Lide, 1992). Term heavy metal is usually associated with pollution and toxicity (Reinhold, 1975). Heavy metals become toxic when they exceed permissible levels in the body, are not metabolized and hence accumulate in the soft or hard tissues. Lead for example, can accumulate in bones. Metals, as described above for uranium, enter the human body through food, water, air, or absorption through the skin when they take place in contact with humans-for example, in agriculture and in manufacturing, pharmaceutical, industrial, or residential settings. Industrial exposure accounts for a common route of exposure for adults. Ingestion meanwhile is the most common route of exposure in children (Roberts, 1999).

Contamination generated by heavy metals differs from that of organic contamination in the sense that former is persistent in all parts of the environment because metal don't degrade or get destroyed. Heavy metals can bring numerous adverse health effects, including allergic reactions, neurotoxicity, nephrotoxicity, and cancer, when exceed permissible limits. Heavy metal toxicity can result in damaged or reduced mental and central nervous function, lower energy levels, and damage to blood composition, lungs, kidneys, liver, and other vital organs. Long-term exposure may result in slowly progressing physical, muscular, and neurological degenerative processes that mimic Alzheimer's disease, Parkinson's disease, muscular dystrophy, and multiple sclerosis. Among other health problems are: disease such as black lung, silicosis, and radiation sickness (Siegel, 2002).

Table 1.4 introduces few important heavy metals commonly found in drinking water, their acceptable limit with suggested references and remarks. Metals like antimony, arsenic, bismuth, cadmium, cerium, chromium, cobalt, copper, gallium, gold, iron, lead, manganese, mercury, nickel, platinum, silver, tellurium, thallium, tin, uranium, vanadium, and zinc are some important elements pertaining to occupational or residential exposure (Glanze, 1996). Micro amounts of these elements are common in our environment and diet and are actually necessary for good health, but large amounts of any of them may cause acute or chronic toxicity (poisoning).

Table 1.4 Guideline values and suggested references for some important heavy metals in drinking-water

Heavy metal	Guideline value (μgl^{-1})	Principal reference	Remarks
Aluminum	Value not established	FAO/WHO, 2007; WHO, 2010	A health-based value of 0.9 mgl^{-1} could be derived, but value exceeds practicable levels based on optimization of the coagulation process in drinking-water plants using aluminium based coagulants: 0.1 mgl^{-1} or less in large water treatment facilities and 0.2 mgl^{-1} or less in small facilities.
Arsenic	10 (A, T)	FAO/WHO, 2011 WHO, 2011	Levels in natural waters generally range between 1 and 2μgl^{-1}, although concentrations may be elevated (up to 12mgl^{-1}) in areas containing natural sources.
Barium	700	IPCS, 2001; WHO, 2003	Concentrations in drinking-water generally below 100 μgl^{-1}.
Beryllium	12	WHO, 2011	primary source of beryllium compounds in water is coal burning and other industries using beryllium. It is not likely to be found in natural water above trace levels due to insolubility of beryllium oxides and hydroxides in the normal pH range
Boron	2400	WHO, 2009	Concentrations vary widely and depend on the surrounding geology and wastewater discharges; for most of the world, the concentration of boron in drinking-water is judged to be below 0.5 mgl^{-1}.
Cadmium	3	WHO, 2011	Levels in drinking-water are usually less than 1μgl^{-1}.
Chromium	50 (P)	WHO, 2003	Total chromium concentrations in drinking-water usually less than 2 μgl^{-1}, although concentrations as high as 120 μgl^{-1} have been reported.
Copper	2000	WHO, 2003	Concentrations in drinking-water range from $\leq$0.005 to > 30 μgl^{-1}, primarily as a result of the corrosion of interior copper plumbing.
Lead	10	FAO/ WHO, 2011 WHO, 2011	Concentrations in drinking-water are generally below 5 μgl^{-1}, except where lead fittings are present.
Manganese	Not considered necessary to set a formal guideline value	WHO, 2011	Manganese is an essential element for humans and animals, not of health concern at levels causing acceptability problems in drinking-water; may however, adverse neurological effects following extended exposure to very high levels in drinking-water.

Heavy metal	Guideline value (μgl^{-1})	Principal reference	Remarks
Mercury	6	IPCS, 2003; WHO, 2005	Mercury is found in surface and groundwater at concentrations usually below 0.5 μgl^{-1}, although local mineral deposits may produce higher levels in groundwater.
Molybdenum	Not considered necessary to set a formal guideline value	WHO, 2011	Occurs in drinking-water at concentrations well below those of health Concern.
Nickel	70	WHO, 2005	Normal concentrations in drinking-water less than 0.02 mgl^{-1}, although nickel released from taps and fittings may contribute up to 1 mgl^{-1}; in special cases of release from natural or industrial nickel deposits in the ground, concentrations in drinking-water may be higher.
Iron	Guideline not established	WHO, 2003	Not of health concern at levels found in drinking-water.
Selenium	40 (P)	WHO, 2011	Most drinking-water contains concentrations of selenium that are much lower than 10 μgl^{-1}, except in certain seleniferous areas.
Silver	Available data inadequate to permit establishing a value	WHO, 2003	It is occasionally been found in groundwater, surface water and drinking-water at concentrations above 5 μgl^{-1}.
Zinc	10	WHO, 2003	Occurs in drinking-water at concentrations well below those of health concern.
Uranium	30 (P)	WHO, 2011	Associates with it, radiological risks in addition to that of chemical.

In Table 1.4, A signifies that calculated guideline value is below the achievable analytic quantification level; P, provisional guideline value because of uncertainties in the health database and T for calculated guideline value is below the level that be achieved through practical treatment methods, source protection, etc.

Allergies are common and repeated long-term contact with some metals or their compounds may even cause cancer. For some heavy metals, toxic levels can be just above the background concentrations naturally found in nature. Therefore, it is important for us to inform ourselves about the heavy metals and to take protective measures against excessive exposure.

1.8   Literature of Environmental Studies

1.8.1   Historical Background

In the environmental sciences, ever increasing threat of radioactive and non-radioactive pollution, and the demand for continuous surveillance and monitoring activities (e.g. toxic trace elements in foods), has resulted in extensive investigations worldwide. The problems resulting from environmental changes have posed new challenges for traditional public health science. The health effects of global change are often indirect and difficult to assess, and the quality of evidence for the health-related outcomes of global environmental change varies widely (Last, 1998). The relationships between health and exposure to elements through food, air and water has therefore become a major area of analytical endeavor.

Widespread, low-level contamination of the environment from anthropogenic sources can be traced back at least to Roman times, when lead and silver mining and smelting were carried out at several major sites across Europe (Rosman *et al*., 1997). Experiences with the possible dangers of radioactive materials precede the discovery of the phenomenon of radioactivity in 1896 by Henri Becquerel. One of the examples is radioactive atmospheres of mines in Central Europe that had been mined for heavy metals since medieval times. Unaware of this radioactive atmosphere and associated danger, miners were later diagnosed to have developed lung cancer, caused by the presence of radon (Lorenz, 1944). Some radioactive substances were used even before it was known that they were radioactive. In the middle of twentieth century, Radium was injected intravenously for a variety of ills. But instead of being cured, many of the patients developed bone cancer or other malignant disease. To quote another example, Welsbach gas mantle, which was developed in 1885, utilized the incandescent properties of thorium oxide to increase the luminosity of gaslight, and uranium oxide

had long been used to provide a vivid orange color in ceramic glazes. Other oxides of uranium and thorium have also been used as glazes and for tinting glass. Yet another examples to this category is production of luminous paints by deliberate mixing of radium with the fluorescent material zinc sulfide to produce luminous material during the times of World war I. This later was well recognised for hazards arising out of ignorance about the effects of this radioactive element when inhaled or ingested (Martland, 1925; Martland, 1951; Evans *et al*., 1969).

It was only during construction of large water-cooled plutonium producing reactors at the Hanford Reservation, near Richland, Washington, and the associated operations for extracting the plutonium from the irradiated uranium, that the first possibilities of major contamination of the environment by radioactivity came to fore. Ever since, extensive research has been conducted to understand the physical and chemical properties of radioactive substances, the manner in which they are transported physically through the environment, and the way in which some of them enter into man's food supplies, the water he drinks, and the air (Eisenbud and Gesell, 1997).

Testing of nuclear weapons and want of nuclear energy for producing energy has lead to the beginning of contemporary radiological assessment and the efforts to predict the path of radioactive fallout. The early Hanford studies on the behavior of various radionuclides in the environment are exemplary in the field demonstrating the caution needed before discharging radioactive substances to the environment and importance of understanding the properties of the individual radionuclides and their behavior in the environment, when substantial quantities of radioactivity is to be discharged safely (Perkins *et al*., 1966). The reports of accumulation of radionuclides by waterfowl, fish, and mollusks were published as early as 1946 (Parker and Norwood, 1946). The studies provided the initial opportunities under field conditions to obtain environmental bioconcentration factors of the kind which proved vital for the development of transport models. More recently, research to reconstruct historical releases of radionuclides to the environment from atmospheric nuclear weapons testing and from nuclear weapons facilities resulted in significant improvements in methods to estimate risk (Till, 1990; Till *et al*., 2002).

1.8.2 Epidemiological Studies

Epidemiological investigations have been a relied method in estimating radiation-induced diseases including various type cancers. The Life Span Study (LSS) cohort of survivors of the atomic bombings in Hiroshima and Nagasaki continues to serve as a major source of information for evaluating health risks from exposure to ionizing radiation and particularly for developing quantitative estimates of risk (NRC, 2006). *A cohort study is a type of observational study used in medicine, social science, actuarial science, and ecology. It is an analysis of risk factors and follows a group of people who do not have the disease, and uses correlations to determine the absolute risk of subject contraction* (Porta, 2008). Advantages of LSS included its large size, the inclusion of both sexes and all ages, a wide range of doses that have been estimated for individual subjects, and high-quality mortality and cancer incidence data.

The data gathered from epidemiological studies of Hiroshima and Nagasaki is among the most important sources for manufacturing risk coefficients used to predict number of cancers that can be expected in a population exposed to a given dose (Muirhead, 2003). Numerous epidemiologic studies have been carried out since the Chernobyl accident to investigate the potential late health consequences of exposure to ionizing radiation from the accident (Moysich *et al*., 2002). These studies have focused largely on monitoring delayed effects of radioactive contamination like thyroid cancer in children, occurrence of leukemia and solid tumors other than thyroid cancer among exposed recovery operation workers and residents of contaminated areas.

1.8.2.1 Epidemiological Studies of Uranium Miners

Many epidemiological studies carried out world wide among uranium miners are found in literature in the direction of assessing risk of radiogenic cancers like lung cancer, leukaemia, kidney, liver and stomach cancers due to radon and progeny inhalation (Table 1.5). Lung cancer production among uranium miners is an established phenomenon. Studies of uranium miner groups of Schneeberg in Germny, Joachimsthal in Czechoslovakia, Colorado plateau and metal miners in the United States, Ontario uranium in Canada and fluorspar miners of Newfoundland are some of the famous examples to be quoted (Archer and Lundin, 1967; Muller, 1980; Kusiak *et al*., 1993; Těsínská, 2009). Radon concentrations in Schneeberg mines have attracted attention of many researchers (Evans and Goodman, 1940; Hueper, 1942; Lorenz, 1944; Mitchell,

1945). Brockbank reported an average value for radon measurements in the air of these mines as high as 3000 pCil^{-1} (Brockbank, 1932).

To be specific about radon carcinogenicity, until recently, the main basis for estimating risks from its residential exposure have been epidemiological studies of underground miners and extrapolating the results down to the levels of exposure seen in homes (UNSCEAR, 2006). A good understanding of differences in dosimetry of exposures in mines and homes however, is needed before going ahead with this extrapolation. For example, such an extrapolation must consider the impact of relatively short exposure periods-higher exposure rates seen in mines compared to longer exposures-lower exposure rates in homes.

Epidemiological studies of underground miners and more recently from residential radon studies have established association of inhalation of radon and its decay with lung cancer and potential risk to tissues and organs other than lung. UNSCEAR's 2006 report has dealt with many epidemiological studies of residential exposures and miners across the globe (UNSCEAR, 2006). The evidence for radon-related increases in mortality from cancers other than lung cancer has been examined in some studies of radon-exposed miners. For example, a case-cohort study evaluating the incidence of leukaemia, lymphoma, and multiple myeloma in Czech uranium miners (Rericha *et al*., 2006) found a positive association between radon exposure and leukemia, including chronic lymphocytic leukemia. This is possibly due to the reason that although the part of the body that receives the highest dose of ionizing radiation from radon and its decay products, is the bronchial epithelium, the extra thoracic airways and the skin may also receive appreciable doses. In addition, other organs, including the kidney and the bone marrow, may receive low doses (Kendall and Smith, 2002).

Epidemiological studies in Schneeberg, Saxonia, and in Jáchymov, West Bohemia in the 1920s and 1930s are among the very first Epidemiological studies carried out in uranium miners and proved an existence of the specific occupational disease (lung carcinoma). This was followed by first protective measures like artificial ventilation, water-irrigated pigs instead of pneumatic ones, charcoal-filter respirators introduced in Jáchymov; taken to decrease the dust and radon concentration in mining work places, as well as to improve general hygienic conditions.

Table 1.5 Mortality from lung cancer among miners exposed to radon (Durrani and Ilić, 1996)

Mine(s) (Follow-up period)	Number of men	Mean WLM	Person	Number of lung cancer deaths		Reference
				Observed	**Expected**	
Colorado Plateau, US (1951-82)	3,346	821	73,642	256	59.1	Hornung and Meinhardt,1987
Ontario, Canada (1955-81)	13,469	30		152	67.6	Kusiak *et al*., 1993
Beaverlodge, Canada (1950-80)	8,487	13	114, 170	65	34.2	Howe *et al*., 1986
Port Radium, Canada (1950-80)	2,103	144	52,930	57	24.7	Howe *et al*., 1987
West Bhomia, Czech Republic, (1953-90)	4,320	219		702	138	Tomaasek *et al*., 1994
Malm Berget, Sweden (1951-76)	1,294	94	26,567	51	13.1	Radford and St. Clair Renard, 1984
New Mexico, US (1977-85)	3,469	111	59,000	68	17.0	Samet *et al*., 1991
Newfoundland, Canada (1950-90)	1,744	383	42,417	139	31.1	Morrison and Villeneuve, 1995
Yunnan Province, China (1976-87)	17,143	217	175,143	981	267	Xuan *et al*., 1993
Cornwall, UK (1941-86)	3,010	100		105	66.6	Hodgson and Jones, 1990
Radium Hill, Australia (1952-87)	1,429	7		32	23.1	Woodward *et al*., 1991
France (1946-85)	1,785	70	44,995	45	21.1	Tirmarche *et al*., 1993

In 1950s, the United States Public Health Service (USPHS) began studies of exposures in uranium mines as a result of concern about the possible health hazards of uranium mining. By 1950, medical studies had been initiated (Archer *et al*., 1962), and uranium miners were subject to routine medical examinations. Beginning in 1954, medical examinations were performed every three years on all uranium miners who could be reached and who agreed to undergo examination (Cooper, 1962). These were the times, when the term "working level" came into being and all states were told to adopt a tentative "working level" of 10^{-10} Cil^{-1} of radon in equilibrium with its decay products (Holaday, 1969). This recommended standard was called 1 WL or 12 WLM per year of exposure.

1.8.2.2 Epidemiological Studies in Residential Areas

The domain of epidemiological studies expanded to residential epidemiological studies there on. Case-control study of domestic exposure to RDPs and lung cancer in Port Hope, Ontario, (Lees *et al*., 1987), high concentrations of indoor thoron observed in the Loess Plateau region study of China (Tokonami *et al*., 2004; Shang *et al*., 2006), population-based case-control study in radon-prone area of north-west Spain (Barros-Dios *et al*., 2002) and study concerning risks of lung cancer from residential exposure to radon in Devon and Cornwall in the south-west of the United Kingdom (Darby *et al*., 1998) are some examples. Alpha track radon detectors have been used in a case-control study (1,192 cases and 1,640 controls) of residential radon concentrations and lung cancer in Saxony and Thuringia, which are areas in eastern Germany with naturally elevated radon levels (Kreuzer *et al*., 2003). Mean radon concentration reported was 76 Bqm^{-3} among cases and 74 Bqm^{-3} among controls.

In one such study started in 1990, comprised 12,000 inhabitants of an area with elevated radon concentrations up to 1000 Bqm^{-3} in central Bohemia, Czech Republic, 173 lung cancer were reported in the follow-up period 1961-1995 (Tomasek *et al*., 2001). Exposure estimates were based on measurements of the equilibrium-equivalent concentrations (EECs) of radon made in most of the homes in the study area.

In a pooled analysis of 13 European case-control studies of the risk of lung cancer from residential radon, Darby *et al*. (2006) came out with empirical relation of

the form $(1 + \beta\chi)$, where χ is the measured radon level and β is the proportional risk factor per unit increase in radon. The purposed linear relationship between Relative risk (RR) of lung cancer versus observed residential radon concentration was $RR = 1 + 0.00084\chi$, with 95% confidence limits. The relative risk assumed at 0 Bqm^{-3} is equal to 1. Radon exposures during the previous 5-34 years were considered in the analysis. The study included 7,148 lung cancer cases and 14,208 controls. The mean radon level measured using long-term alpha track-etch detection in the houses of the control group was 97 Bqm^{-3}, and in the houses of lung cancer cases was 104 Bqm^{-3}.

In India, many research groups are active to establish dose rate profiles due to natural radiation and indoor radon levels across the country under the specific leadership of Board of Research and Nuclear Sciences (BRNS), Department of Atomic Energy, India. Significant research has been carried out in the laboratories CSIR, New Delhi and Bhabha Atomic Research Centre (BARC), Mumbai in the direction of developing instrumentation used in environment studies (Eappen *et al.*, 2008; Mishra and Mayya, 2008; Mishra *et al.*, 2009; Mayya *et al.*, 2010; Gaware *et al.*, 2011). Levels of major radionuclides responsible for contributing to the radiation dose in environmental matrices and levels of dose rates in the indoor and outdoor atmospheres from the different corners of India have been are discussed in the coming chapters.

Extensive work has been carried out by various research groups in northern region of India comprising states Punjab, Himachal Pradesh, Rajasthan, Haryana and Delhi relating to environmental radioactivity, heavy metals studies and earthquake precursory studies in seismically active areas of NW Himalayas. Table 1.6 summarizes some of the studies principally of the similar nature as of the present work, along with methodology adopted by the respective workers in the Northern India with special emphasis on areas adjoining to the present study region. Table shows wider type of instrumentation has been utilized in these studies depending on availability and suitability of field conditions. Both passive and active techniques have been used for analysis of radioactive elements uranium, Radium, Thorium, Potassium and Radon in water, soil and air for purposes ranging from dose evaluation to fault delineation and earthquake precursory studies.

Table 1.6 Summary of some recent environmental studies in Northern India

Particulars of the study	Study region	Methodology/Experimental technique	References
Heavy metal contamination in Yamuna Basin	Delhi segment	Atomic Absorption Spectrometer	Sehgal *et al.*, 2012
Heavy metal contamination in drinking water	Faridkot, Punjab (India)	ICP-MS	Blaurock-Busch *et al.*, 2010
Radon monitoring for earthquake percussion	NW Himalaya	RAD-H$_2$O Technique	Singh *et al.*, 2010
TDS and conductivity monitoring in groundwater	NW Himalaya	Water quality probes	Kumar *et al.*, 2010
Natural radioactivity measurements in soil	Upper Siwaliks and Punjab	Gamma Ray Spectrometry	Singh *et al.*, 2009
Uranium and radon in drinking water	Upper Siwaliks of India	fission track and alpha scintillometry techniques respectively	Singh *et al.*, 2009
Radon and thoron levels in soil, water and indoor atmosphere	Budhakedar in Garhwal Himalaya, India	Radon Emanometer for soil and water; LR–115 plastic track detectors for indoor	Prasad *et al.*, 2008
Indoor radon measurements	Upper Siwaliks of India	Using SSNTDs	Singh *et al.*, 2007
Uranium and Heavy metals in water	Bathinda, Punjab (India)	Anode stripping method	Kumar *et al.* 2006
Radon and thoron progeny (outdoors)	Malout, Punajb (India)	Grab aerosol sampling and beta counting	Singh *et al.* 2006
Soil gas radon	Nurpur, Himachal Pradesh	Using SSNTDs	Singh *et al.*, 2006
Seasonal variation of indoor radon	Malwa region, Punajb	Using SSNTDs	Singh *et al.*, 2005
Indoor radon, uranium and radium in soil	Bathinda, Punjab (India)	SSNTDs and can technique	Singh *et al.*, 2005
Natural radioactivity measurements in soil	Hamirpur, Himachal Pradesh	Gamma Ray Spectrometry	Singh *et al.*, 2003
uranium, radium and radon exhalation in soil	Nurpur, Himachal Pradesh	Using SSNTSs	Sharma *et al.*, 2003
Indoor radon measurements in the dwellings	Punjab and Himachal Pradesh, India	LR-115 type-II plastic track detector	Bajwa *et al.*, 2003
Uranium in drinking waters	Bathinda, Punjab (India)	Fission track technique	Singh *et al.*, 1995

1.9   Present Study Region

In the present thesis work, area of south-western of Indian Punjab comprising Faridkot, Mukatsar and Ferozepur has been studied in terms of radioactivity levels in outdoor and indoor environment to estimate radiation dose to the public (Figure 1.2). Study region is located approximately between 29°56' and 31°10' north latitudes and 73°52' and 75°30' east longitudes in terms of geo-positioning coordinates. The study area is a part of Indo-gangetic plain and Sutlej Sub basin of main Indus basin. The area as a whole is almost flat with a gentle slope towards the south westerly direction. The soil of study region is loose and sandy, consists of different layers of clay, sticky clay and fine to coarse grained grey micaceous sandstone. The top layer of soil is dark reddish silty soil rich in $CaCO_3$. Hydrogeology of the region is characterised by unconfined and confined aquifers down to 50 m depth. The land is predominantly used for agriculture purposes.

The guiding force behind carrying present study has been increased occurrence of cancer cases in southern part of Punjab. Apart from many reports in local and national newspapers about the prevalent cancer cases, one is epidemiological study in 36 villages of Southern Punjab (Thakur *et al*., 2008). In their study, authors confirmed 125 cases per 100,000 population and 52 deaths per year during the epidemiology period 1993-2005. Though no concrete cause behind the mass cancer cases was concluded by these workers, higher levels of As, Cd, Cr, Se and Hg in limited drinking water samples and some pesticides in water, vegetable and blood were reported. Another publication seen regarding heavy metals analysis is study of heavy metals Zn, Cd, Pb, Cu along with uranium in drinking water samples from adjacent Bathinda district (Kumar *et al*., 2006). In their work, authors analysed 25 odd water samples and inferred exceeding values of Cd, Pb and U in samples. It can be added to this discussion that not much work has done in this region in the direction of heavy metal analysis until recently, when Blaurock-Busch *et al*. (2010) came out with their publication concluding metal exposure and chronic exposure in the physically and mentally challenged children of Baba Farid Centre at Faridkot in Punjab, India. In this study, authors analysed 149

hair samples and used Hair mineral analysis (HMA) to locate chronic exposure. Out of total 53 elements tested using ICP-MS, the hair of the children showed high values for Ba, Cd, Mn, Pb and U; signifying long-term exposures. In their study, authors also reported contaminant level for uranium (U) in three samples and lead (Pb) in one out of total six water samples were they analysed.

In yet another important and pioneering kind of study to detect high contents of uranium in drinking water samples in this region, high contents of uranium in drinking water samples in the Bathinda district of southern Punjab with a maximum value of 113 μgl^{-1} were reported using fission track technique (Singh *et al*., 1995). With an aim to investigate the possible source of high uranium content, Singh *et al*. (2005) later analyzed uranium concentration in the soil samples from this region, but found no significant higher range (0.83-3.68 $mgkg^{-1}$). Hence, it was anticipated that source of uranium in water is deep uranium mineralization, as region is dependent on underground water sources for drinking and irrigation purposes. In these times however, in this region known as Malwa region of Punjab, reports of number of cancer cases increased sharply. As a result, an intensive survey of drinking water from this region was carried out in a BRNS sponsored project, which only confirmed the high concentration of uranium in water (Kumar *et al*., 2011), discussed in chapter 4. Other components of an environmental radioactivity survey, viz. gamma spectroscopic analysis of soil samples, radon studies and heavy metal analysis as well have come automatically to explore every possible reason behind mass health problems in the region.
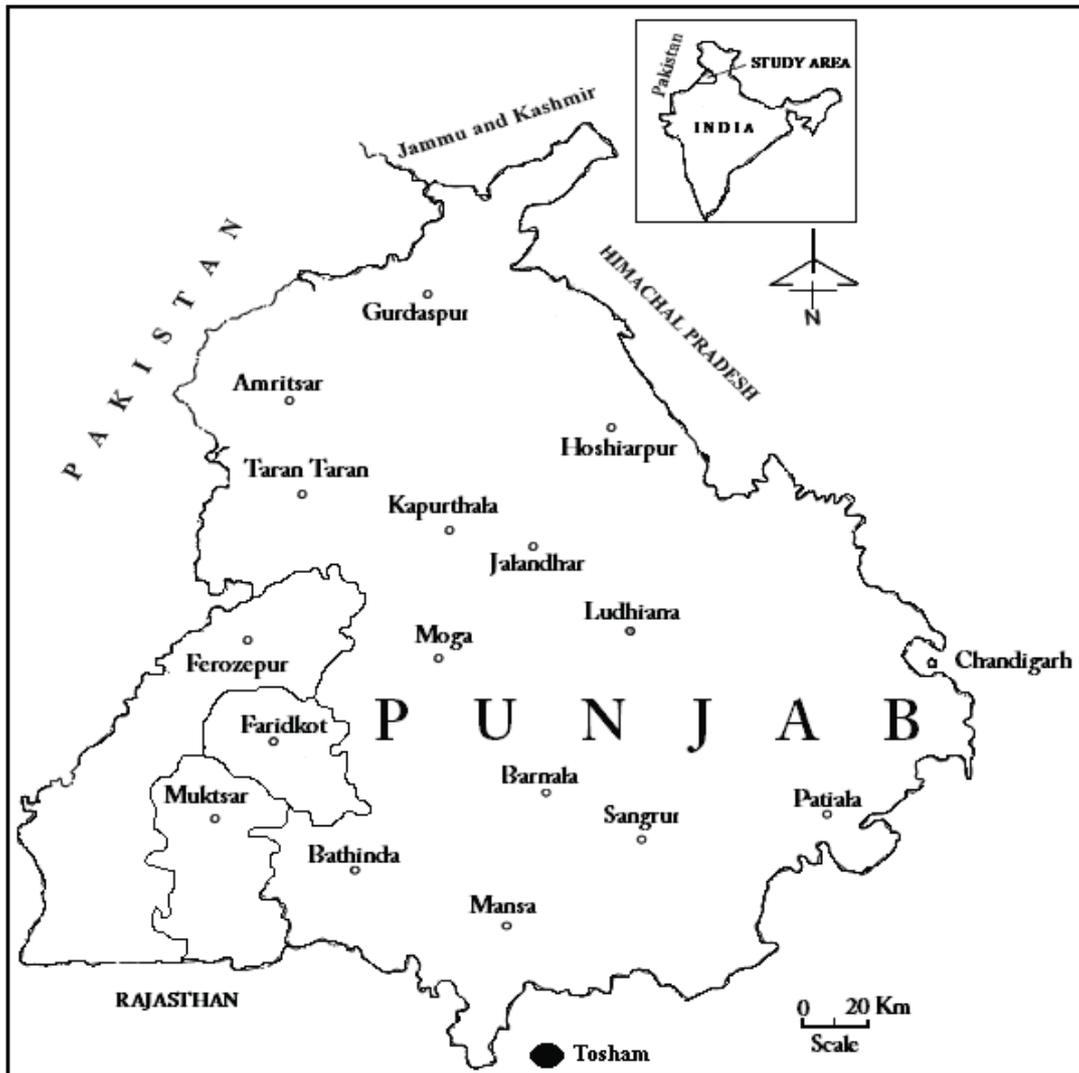
Figure 1.2 Figure showing present region comprising of Faridkot, Ferozepur and Muktsar districts of Punjab, India

1.10   Aim of Present Work

The aim of the present work has been to study radioactivity levels in the environment of the chosen study area of south-western Punjab for health risk assessment. In particular, the study area includes districts of Faridkot, Mukatsar and Ferozepur. The study involves investigating the levels and distribution patterns of radioactive elements viz. U, Ra, Rn, Th and K and other heavy metals of health concern in the environmental matrices of the region to understand levels of chemical and radiation exposure to the residents.

Aims of this study are summarized as follow:

1. Analysis of Uranium in water and soil samples collected from the study region for Radiological and chemical risk assessment,

2. Study of distribution of chemical elements of health concern to calculate corresponding Average Daily Doses and non-carcinogenic hazard coefficients; and physico-chemical water parameters viz. TDS, pH etc in drinking water samples,

3. Radiometric analysis of soil samples using gamma-ray spectrometry to estimate natural radioactive background levels and Effective Dose Rates assessment to the general public and

4. Study of radon levels viz. indoor radon study, soil-gas radon, water- dissolved radon and radon exhalation rates for the study region with emphasis on Inhalation and ingestion doses due to secondary radionuclide.