STUDY OF RADIOACTIVITY IN NATURALLY OCCURRING RADIOACTIVE MATERIALS (NORMs)

THESIS

submitted in fulfillment of the requirement of the degree of

DOCTOR OF PHILOSOPHY

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by

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FEBRUARY, 2019

DECLARATION

I hereby declare that this thesis entitled **"STUDY OF RADIOACTIVITY IN NATURALLY OCCURRING RADIOACTIVE MATERIALS (NORMs)"** being submitted in fulfillment of the requirements for the Degree of Doctor of Philosophy in **Physics** under Faculty of **Sciences**, is a bonafide record of my original work carried out under joint guidance and supervision of **Dr. Maneesha Garg, Assistant Professor (Physics), J.C. Bose University of Science and Technology, YMCA, Faridabad and Dr. Krishan Kant Gupta, Principal, Aggarwal PG College Ballabgarh, Faridabad** and has not been presented elsewhere.

I further declare that the thesis does not contain any part of any work which has been submitted for the award of any degree, either in this university or in any other university.

(Raj Kumari)
YMCAUST /PH08/2011

CERTIFICATE

This is to certify that the thesis entitled, **"STUDY OF RADIOACTIVITY IN NATURALLY OCCURRING RADIOACTIVE MATERIALS (NORMs)"** by **Raj Kumari,** being submitted in fulfillment of the requirement for the Degree of Doctor of Philosophy in Physics under the Faculty of Sciences, J. C. Bose University of Science & Technology, YMCA, Faridabad, during the academic year 2019-20, is a bonafide record of the work carried out under our joint guidance and supervision.

We further declare that to the best of our knowledge, the thesis does not contain any part of any work which has been submitted for the award of any degree either in this university or in any other university.

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ABSTRACT

Radon is non-reactive naturally occurring radioactive gas. It is invisible, tasteless, odorless and colorless noble gas and it contributes significantly to the total dose received by the general population from ionizing radiation. Radon and radium are the daughter products of uranium decay series.

In view of these facts, organized investigations on naturally occurring radioactive materials are needed to evaluate and to understand the distribution of radionuclides. Many research scientists from all over the world and India are involved in this research work because of their deleterious effects on human health. This was the inspiration behind the present research work with the topic **"STUDY OF RADIOACTIVITY IN NATURALLY OCCURRING RADIOACTIVE MATERIALS (NORMs)".** The present study is an attempt in this direction in which systematic investigation on the distribution and behavior of naturally occurring radionuclides have been carried out in the southern Haryana, which is an industrial area situated in north India. Faridabad, in southern Haryana, is the most populated city in northern India. Stone mining in the Aravali range is the dominant activity in the region. This area is at the verge of becoming a major industrial hub with the setting up of many production industries. The results of the current research work not only provide the identification of radionuclide but also throw light on basic aspects of the enhanced mechanism of radionuclides and their transportation.

In the present investigations, radium content, radon exhalation rates, and gamma dose rates in Naturally Occurring Radioactive Materials (NORMs) viz. vegetables, fruits, cereals and building materials are measured by CAN technique and gamma-ray spectroscopy. LR-115 type-II SSNTDs (Solid State Nuclear Track Detectors) have been used. It is a long term technique which is widely used all over the world for long-term integrated measurements of radon and their progeny levels. The methodology is depending on the track registration in SSNTDs. This method is reliable and sensitive for the measurements of radon and their progeny. This study will be significant in the understanding of the main sources of indoor radon and its health risks, and various factors that are influencing it.

Keeping in view of the detrimental effects of radon and its progeny, measurements of underground radon, indoor radon and its progeny levels besides radium content in the environment have been carried so as to find out the health risks.

Gamma-ray spectroscopy is one of the most accepted technique for the detection and analysis of low-level radioactivity. High purity germanium (HPGe) detector is the most sensitive detector and has excellent energy resolution, hence it is widely used for determining radioactivity of radionuclides.

Inhalation risk is associated primarily with the alpha particles from radon and its progeny that are emitted within the lungs, causing damage to the cells lining. This ultimately leads to lung cancer in many cases. Some studies indicate that chronic exposure to radium can also induce bone sarcomas. Higher values of ^{226}Ra in solid samples contribute significantly to the enhancement of environmental radon.

The effective environmental monitoring and assessment of the health effects and risks indicate the impact of all activities involving radionuclides. The present measurements have been carried out to study various aspects viz. radon and their progeny concentrations in different samples, effective dose, and occurrence of lung cancer risk due to its inhalation.

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List of Symbols and Abbreviations

S.No.		
1	**UNSCAER**	United Nations Scientific Committee on the Effects of Atomic Radiation
2	**IARP**	Indian Association for Radiation Protection
3	**OECD**	Organization for Economic Cooperation and Development
4	**ICRP**	International Commission on Radiological Protection
5	**IAEA**	International Atomic Energy Agency
6	**BEIR**	Biological effects of Ionizing Radiation
7	**EPA**	Environmental Protection Agency
8	**ICRP**	International Commission on Radiological Protection
9	**WHO**	World Health Organisation
10	**DNA**	Deoxyribo-Nucleic Acid
11	**SSNTDs**	Solid State Nuclear Track Detectors
12	**LET**	Linear Energy Transfer
13	**CR-39**	Columbia Resin-1939
14	**$C_6H_8O_9N_2$ (LR-115)**	Cellulose Nitrate
15	**NaOH**	Sodium Hydroxide
16	**NaI**	Sodium Iodide
17	**HPGe**	High Purity Germanium
18	**mBq**	milli Becquerel (1 mBq = 10^{-3} Bq)
19	**ADC**	Analog Digital Converter
20	**MCA**	Multi Channel Analyzer
21	**ρ**	track density
22	**C_{Rn}**	Radon concentration
23	**C_{Ra}**	Effective Radium content
24	**T**	exposure time

25	**T_e**	Effective time of exposure
26	**U**	Uranium
27	**Th**	Thorium
28	**K**	Potassium
29	**Rn**	Radon
30	**Po**	Polonium
31	**Pb**	Lead
32	**Ra**	Radium
33	**He**	Helium
34	**Ge**	Germanium
35	**s**	Second
36	**µs**	micro second

CHAPTER-I

INTRODUCTION

1.1 INTRODUCTION

Radiation exposure to the living organism from birth is inescapable. One of the causes of radiation exposure is natural radioactivity generated by natural radioactive elements present naturally in the environment and radiation coming from cosmic sources like stars and sun. Radioactive materials like radium, thorium, and uranium are present on the earth. Humans can easily come in contact with these radioactive elements [1].

All living organisms that inhabit the earth, including humans have adapted up to a certain level of radiations. The incorporation of manmade activities like household and industrial activities has dramatically increased the environmental radioactivity by redistributing the natural radioactive isotopes [2]. The invention of new radioactive isotopes, which do not exist naturally in the environment, has added to complications of exposure of radiation to the human race [3]. Knowledge of radioactive materials concentration and their distribution in the environment around a human is important as the exposure to humans causes various diseases. This fact forms the basis of different researches in the field of science which are related to well being of the human population.

Radiations can be classified into two categories

I. Ionizing Radiations
II. Non-ionizing radiations

1.1.1 Ionizing Radiations

The spontaneous disintegration of atoms to reach a stable state is called radioactivity, and its excess energy is released in terms of ionizing radiations. The radioactive elements release ionizing radiation from their nuclei [4].

Types of ionizing radiation:

a. Gamma rays and x-ray
b. Alpha particles
c. Beta particles
d. Neutrons

During the disintegration of a radioactive element, usually an alpha (α) particle is emitted from the nuclei of the element. If the newly formed element is again a radioactive element it decays further till the formation of nonradioactive elements [5].

1.1.2 Non-Ionizing Radiation

The radiations which have comparatively less energy than ionizing radiations and excite the molecules.

These radiations can be classified into two categories on the basis of their sources:

a) Natural background radiations
b) Artificial radiations.

1.1.2.1 Natural Background Radiations

Low-level radiation which comes from the environmental sources like earth's crust, sun, radioisotopes, and atmosphere are known as natural background radiation. The natural background radiations present in nature can be further classified into three subcategories depending upon the source from which they emerge.

a) Cosmic Radiation

High energy radiation arrives from the outside environment of the earth is called cosmic ray. All the living and non-living creatures are exposed to these radiations from space constantly.

b) Terrestrial Radiation

Electromagnetic wave emitting from radioactive element present in the earth's crust is known as terrestrial radiation. Radioactive materials like uranium, thorium and their decay products are ubiquitous. It is found in the earth's crust, vegetation and water.

Radiation emitted by these materials which exist in various form reaches the human via inhalation of air, ingestion of food and water. Radiation dose varies with geographical location and is mainly dependent on the presence of radioactive material present in the environment.

c) Internal Radiation

The living beings have radioactive carbon-14, lead-210, potassium-40 and their isotopes inside their bodies since birth [5] and hence themselves act as sources of radiations. These types of radiations are called internal radiation.

1.1.2.2 Artificial Radiations

Radiations produced by bombardment of very high energy particles are known as artificial radiation. Although natural radiations majorly affect the living organisms but the man-made sources also significantly contribute to radiation exposure of living beings.

Mainly following groups of people are affected by artificial radiation sources.

I. Humans who have interaction with

a) Medical facilities like diagnostic, nuclear medicine procedures, x-rays.
b) Radiation causing consumer products like fluorescent lamp starters, luminous watches, televisions, smoke detectors, lantern mantles, tobacco, building materials, and road construction materials.

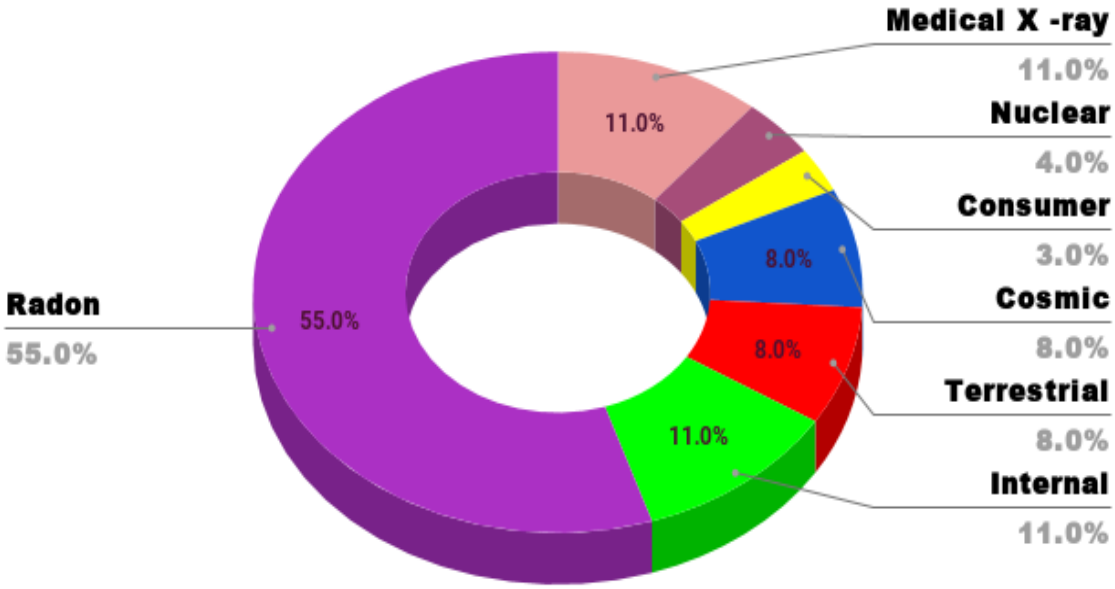

Figure 1.1: Radiation exposure to the public from different sources of Radiations [6]

Humankind also encounters radiation during mining of uranium, extraction of nuclear fuel and disposal of used fuel. The unlikely events such as reactor accidents and the fallout of nuclear weapons testing also cause radiation exposure to human.

II. The other class of human which are exposed to radiations are people working in a radiation-exposed environment like nuclear power plant workers.

1.2 RADIUM

Uranium and thorium decay series produce radioactive elements like radium (^{226}Ra) and radon (^{222}Rn) as the intermediate elements. Radium is found in almost all geological formations on earth like water, soil, plants, and rock etc. Radium is present in high concentrations in the deep mines viz. uranium ores mines, coal mines, metal mines [7].

Radium decays to produce radon (^{222}Rn), which when inhaled by humans emits alpha particles and may cause lung cancer by damaging the red blood cells (RBCs) and bones sarcomas [8].

Radium, not only through inhalation, may also enter the body through the food chain. The crops may contain a high amount of radium concentration. Many studies [9-11] have reported a direct relationship between concentrations of radium present in the soil to radium concentration in plants.

1.3 RADON

Radon (^{222}Rn) gas is a radioactive inert gas. It is a naturally occurring, colourless, odourless and tasteless gas which cannot be sensed by humans but can be measured by special instruments. Radon was discovered by Friedrich E. Dorn, a German scientist in 1898 [6]. Niton (Nt) name was given to it by R. Whylaw Gray and W. Ramsay in 1908 [8]. In 1923, new name Radon was given to it by scientists as it was emitted from the radium. It is represented as ^{222}Rn [12]. Several studies have confirmed that it is a significant cause of lung cancer after smoking [13].

The radioactive decay chain resulting in the generation of radon and its progeny are shown in figure 1.2 and decay series and half-life is depicted in table 1.1 [14].

Figure 1.2: Radon and its daughter products generation from uranium

Table 1.1: The decay series and half-life of naturally occurring radioactive elements

S. no.	Radionuclide	Decay particle	Half-life	Energy
1.	$_{92}U^{238} \rightarrow {}_{90}Th^{234}$	Alpha	4.51×10^9 years	4.20 MeV
2.	$_{90}Th^{234} \rightarrow {}_{91}Pa^{234}$	Beta	24.1 days	0.19 MeV
3.	$_{88}Ra^{226} \rightarrow {}_{86}Rn^{222}$	Alpha	1620 years	4.77 MeV
4.	$_{86}Rn^{222} \rightarrow {}_{84}Po^{218}$	Alpha	3.825 days	5.49 MeV
5.	$_{88}Ra^{224} \rightarrow {}_{86}Rn^{220}$	Alpha	3.62 days	5.69 MeV
6.	$_{86}Rn^{220} \rightarrow {}_{84}Po^{216}$	Alpha	55.6 sec	6.29 MeV
7.	$_{83}Bi^{212} \rightarrow {}_{81}Tl^{208}$	Alpha	60.6 min	6.05 MeV
8.	$_{84}Po^{218} \rightarrow {}_{82}Pb^{214}$	Alpha	3.05 min	6.0 MeV
9.	$_{84}Po^{216} \rightarrow {}_{82}Pb^{212}$	Alpha	0.15 sec	6.78 MeV
10.	$_{84}Po^{214} \rightarrow {}_{82}Pb^{210}$	Alpha	164 µsec	7.69 MeV
11.	$_{83}Bi^{212} \rightarrow {}_{84}Po^{212}$	Beta	60.6 min	2.25 MeV
12.	$_{84}Po^{212} \rightarrow {}_{82}Pb^{208}$ (stable)	Alpha	3.04×10^{-7} sec	8.78 MeV

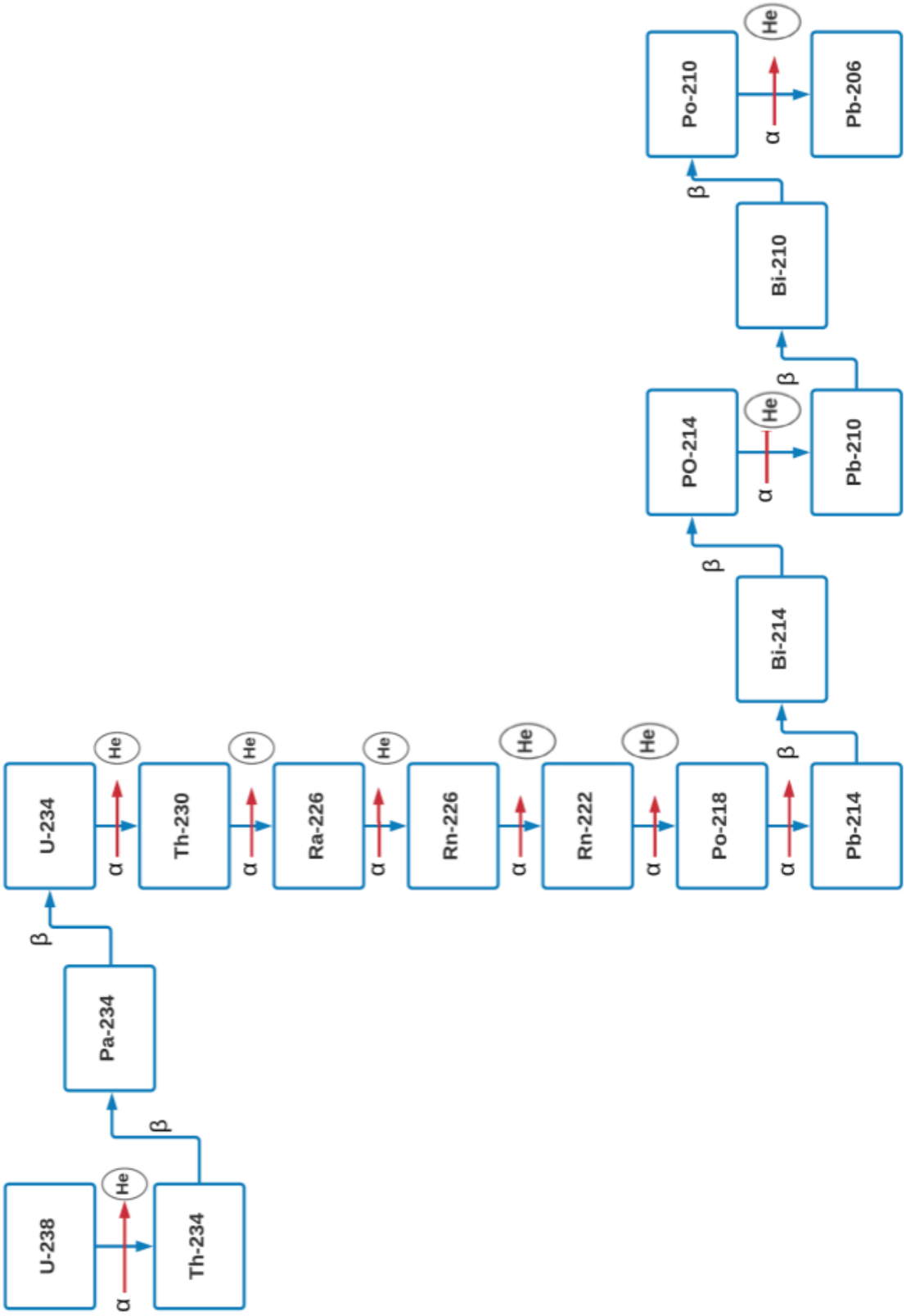

Figure 1.3: Uranium - 238 decay series

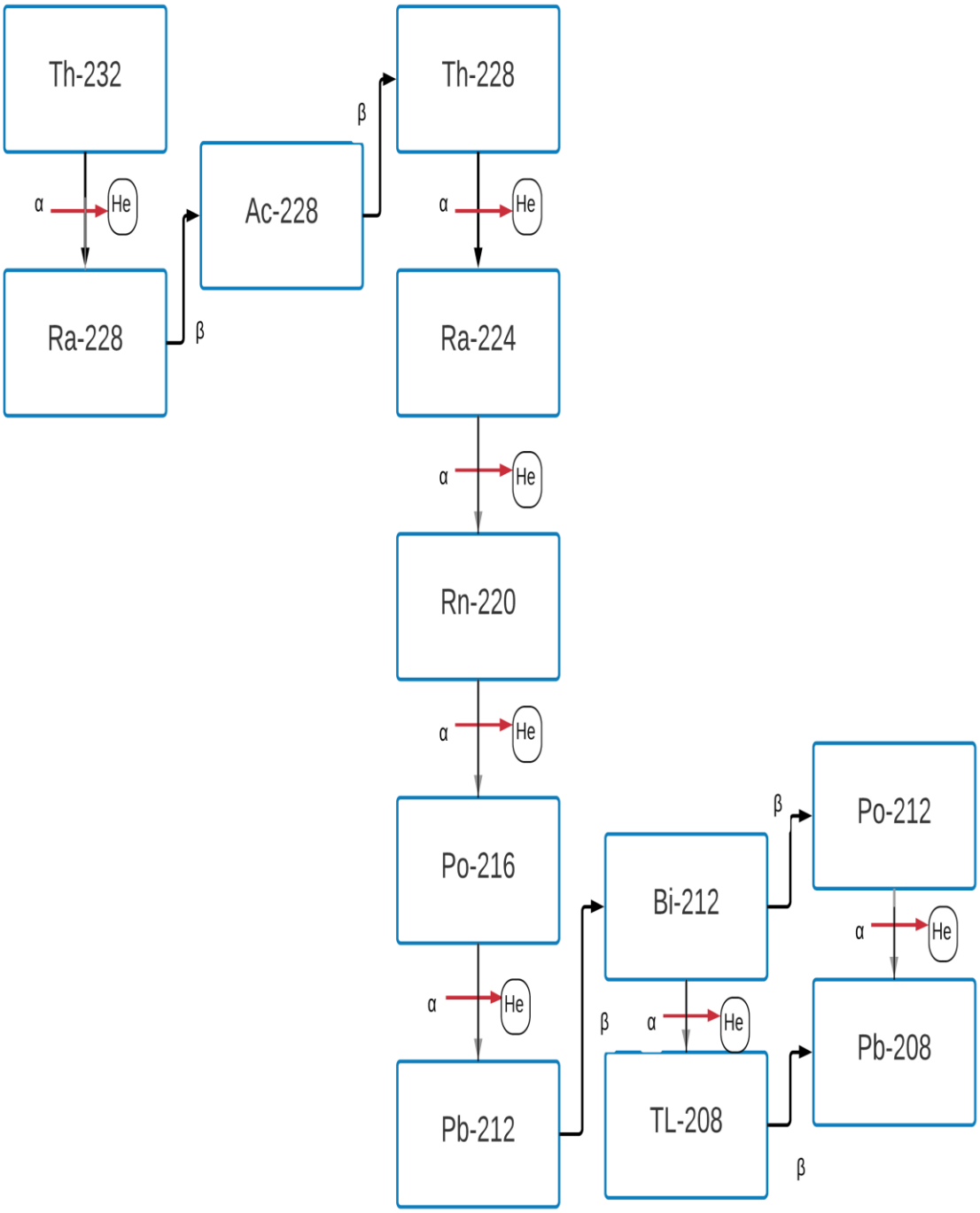

Figure 1.4: Thorium-232 decay series

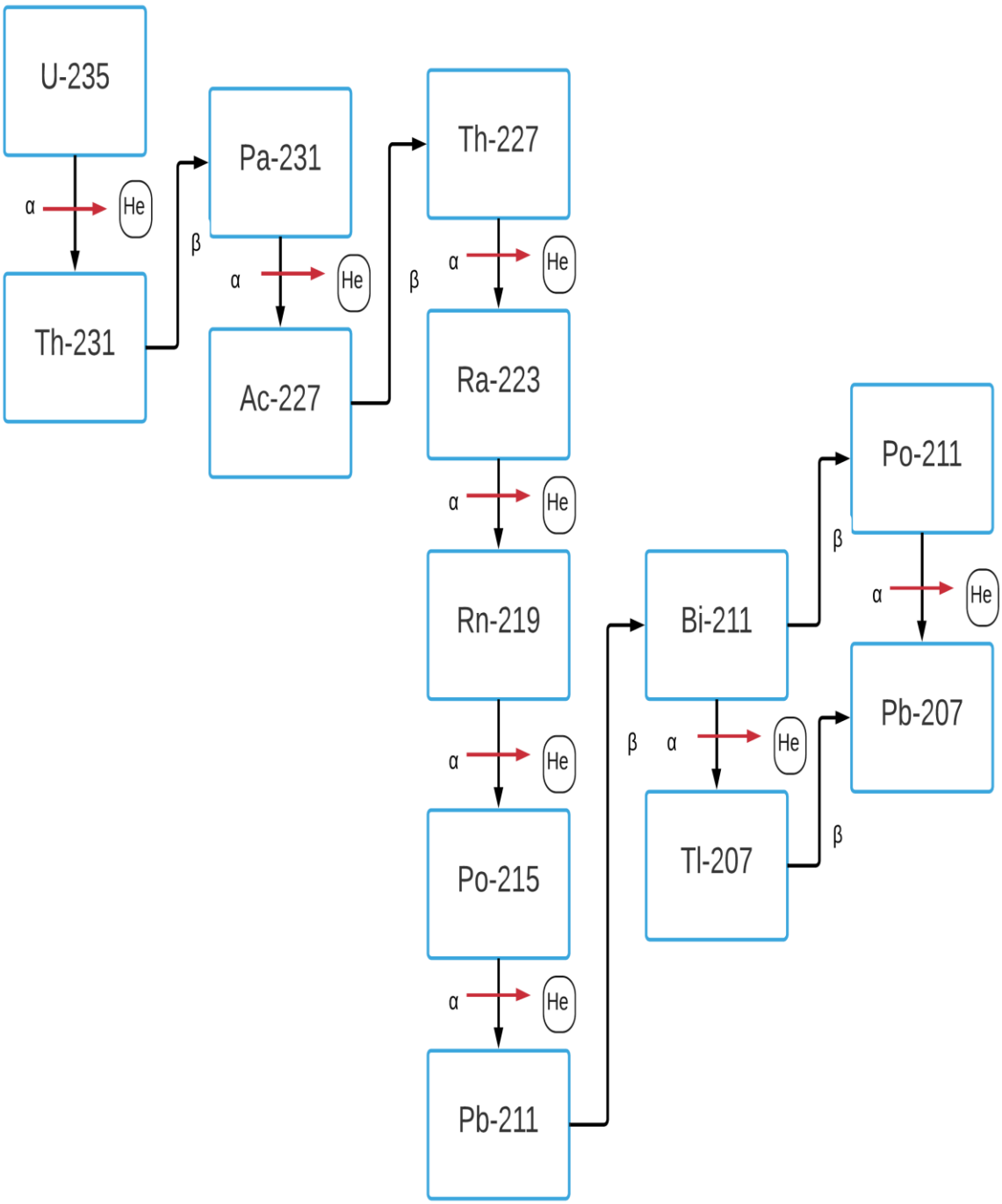

Figure 1.5: Uranium-235 decay series

Figures 1.3, 1.4 and 1.5 showing decay series of uranium-238, thorium-232 decay, and uranium-235, respectively.

Radon progeny is a decay product of radon gas. The half-life of radon is 3.82 days, which means the alpha particles intensity of radon in samples will reduce to half in 3.82 days. The longer half-life indicates that radon can migrate to a large distance in the air without decay.

1.3.1 Characteristics of Radon

Radon is an inert gas and has a high boiling point and melting point. It has high critical pressure and temperature. Due to its inert nature, it is not detectable by the chemical process. Radon has certain solubility in cold water but with increasing temperature the solubility of radon in the water decreases [15]. Chemical properties and characteristics of radon are listed in table 1.2.

Table 1.2: Characteristics of radon [16]

S. no.	Characteristics	Remarks
1.	Block	P
2.	Atomic Number	86
3.	Atomic Radius	1.34 Å
4.	Relative atomic mass	[222]
5.	State at 20° C	Gas
6.	Electrons Configuration	[Xe] $4f^{14}\,5d^{10}\,6s^{2}\,6p^{6}$
7.	Key Isotopes	^{211}Rn, ^{220}Rn, ^{222}Rn
8.	Mean Excitation Energy	794.03 eV
9.	Atomic Volume	50.52 cm^3/ mol
10.	Critical point	377K at 6.28MPa
11.	Boiling point	211.50K
12.	Melting point	202K

13.	Critical pressure	62atm.
14.	Critical temperature	104.2^0 C
15.	Heat of fusion	2.890 – 3.247kJ/mol
16.	Enthalpy of fusion	2.70 kJ/mol
17.	Heat of vaporization	16.40 – 18.10kJ/mol
18.	Enthalpy of vaporization	18.10kJ/mol
19.	Specific heat capacity	94J/kg.K
20.	Thermal entropy	176.10 J/ mol.K (298.15K)
21.	Heat capacity	20.786 J / mol.K (25^0 C)
22.	Thermal conductivity	3.610 mW/ m.K (at 300K)
23.	Electrical conductivity	0.1 mΩ-cm
24.	Crystal Structure	Cubic
25.	Ionization energy	10.744eV
26.	Oceanic abundance	6×10^{-16}mg / l
27.	Crystal abundance	4×10^{-13}mg/kg
28.	Polarizability	$5.3\times10^{-24}cm^3$
29.	Density	9.73g/ L(0° C)
30.	Production rate	0.001 cm^3/day per g of Ra^{226}
31.	Water solubility	230cm^3/kg (20°C)
32.	Vapour pressure at:	
	110K	1Pa
	121K	10Pa
	134K	100Pa
	152K	1kPa
	176K	10kPa
	211K	100kPa

33.	Coefficients of solubility	
	atmospheric pressure in water at:	
	0^{o} C	0.507
	10^{o} C	0.340
	20^{o} C	0.250
	30^{o} C	0.195
	37^{o} C	0.167
	50^{o} C	0.138
	75^{o} C	0.114
	100^{o} C	0.106

The atomic number of radon is 86 and its mass number is 222. It is the heaviest gaseous element among the rare gas group. Radon is weakly soluble in water but more soluble than any other inert gas. The solubility of radon in organic solvents is moderate but decreases with a rise in temperature. When it is cooled below freezing point, radon turns from yellow to orange-red [17].

The presence of radon at any place is dependent on many natural and artificial conditions. It is also dependent on height above the ground, time, place, and atmospheric geological conditions [18]. Soil porosity and type of cracks, holes and other openings in foundation material provide an easy path for radon to enter in a closed structure. In general radon gas entry into a closed structure is more at boundary area as compared to central areas.

The influx of radon in a closed structure is a function of

i) The permeability of the underlying soil
ii) The permeability of the medium above the soil
iii) Geological, meteorological and structural factors
iv) The rate at which radon is generated in sources materials
v) Ventilation rate
vi) Atmospheric pressure

vii) Varying environment conditions

viii) Construction type

Radon gas and its progeny concentrations in a given indoor air can vary over a substantial range. It depends on the various factor like radon gas released by building material and ventilation in the building.

1.3.2 Radon Release Mechanism

Radon is produced mainly due to the decay of radium but since this decay happens inside the mineral, the radon gets trapped inside and a small quantity of radon gets released during its production by the decay of the radioactive element.

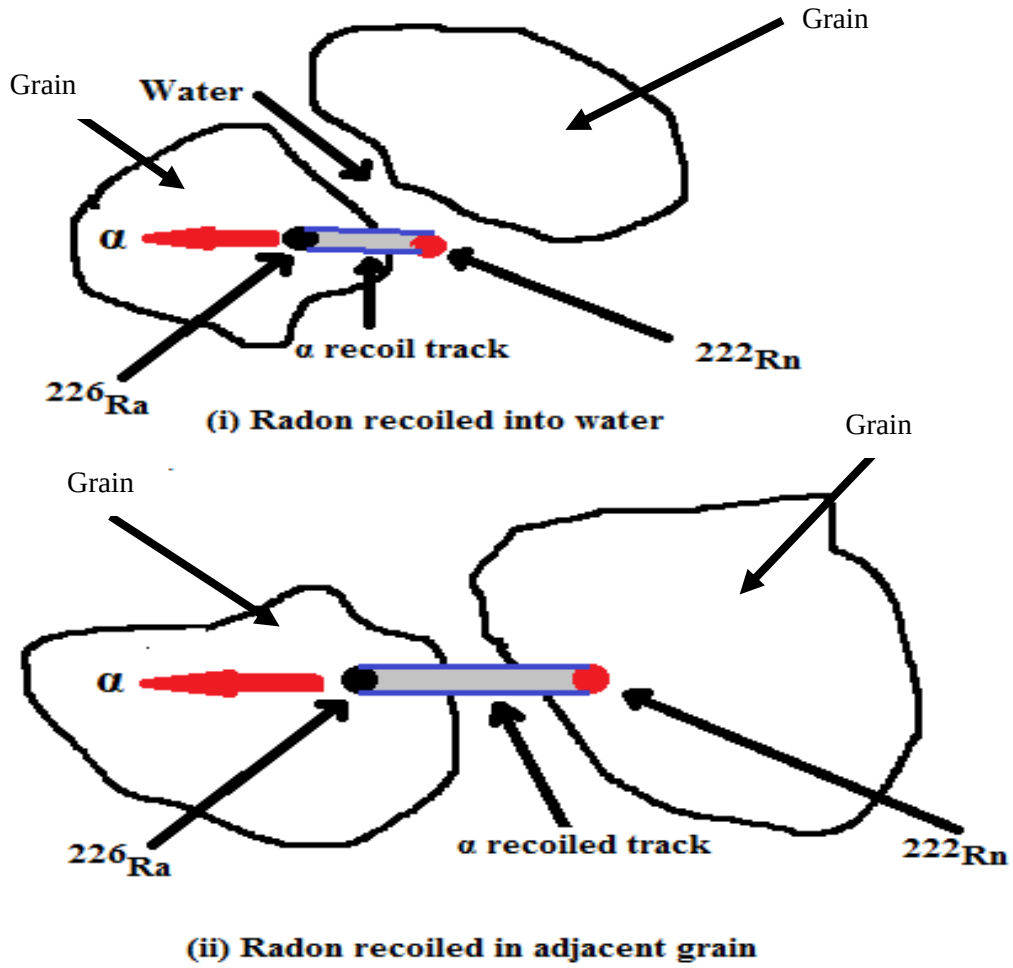

Figure 1.6: A Radon release mechanism [19]

Many studies [20-21] show the possible release mechanism of radon from the interior of minerals due to the recoil action of an alpha particle. The emission of alpha particles is carried out with 4.7MeV energy; the recoil energy computed for radon nucleus is of the order of 86keV. The study of cracks on mineral grain also reveals that these recoiled radon nuclei may trap enter into the adjacent grain and inner granular liquid, if any [22].

Due to the emission of high energy alpha particles the recoiled radon nucleus may come to rest in pore space where water is present. Radon can diffuse through water and may get transported to the environment. In the other mechanism, this radon may enter into interstitial sites of adjacent grain and cause damage to the adjacent grain [19]. This trapped radon can be released from the interstitial site through the water.

1.4 HEALTH HAZARDS FROM RADON EXPOSURE

When radiation interacts with living matter, it affects the smallest unit cell of a living organism. The cell which has nucleus contains chromosomes filled with a sack of fluid (cytoplasm) enclosed by a membrane. The chromosomes are responsible for holding the genes which are a part of DNA that carry life-sustaining information. The biological hazards produced by radiation occurs first at the cellular or subcellular level. It is due to ionization of the macromolecules within the cell of living matter or in the suspension [23]. In indirect action, radiation interacts with water molecules inside the cell which constitutes 70% to 85% of the cell content. In this process, water is ionized, a free radical is formed and the end by-product of that ionization can include hydrogen peroxide [24].

The mechanism is explained below:

For every molecule of DNA in the cell, there are 1.2×10^{7} molecules of water present. The ionization takes place when radiation interacts with matter; resulting in the generation of ion pair of HOH^{+} and an electron.

$$\textbf{Radiation} \longrightarrow \textbf{HOH} \longrightarrow \textbf{HOH}^{+} + \textbf{e}^{-1}$$

The electron produced in the above reaction can again reconnect with the HOH^{+} ion or it may attach to water molecule present in the proximity to create another free

radical. The free radical HOH tends to form OH and hydrogen. The free H radicals may form hydroperoxyl radicals by combining with oxygen. The oxygen and hydrogen peroxide are produced by hydroperoxyl radicals, which can produce harmful damage.

$$H + O_2 \longrightarrow HO_2$$

$$2\,HO_2 \longrightarrow H_2O_2 + O_2$$

$$HOH^+ + HOH \longrightarrow OH^- + H_30$$

$$HOH^+ \longrightarrow H^+ + OH^-$$

The HOH^+ can react with other HOH, and create OH and H_3O, which can further create a hydroxyl radical and hydrogen ion, H^+.

The chemically reactive hydrogen peroxide (H_2O_2) and free radicals so generated attack the cell membrane and damage it. Such cell damage is augmented in many ways and can result in the death of the cell, inability to reproduce and unlikely transformation of cells to another form all these results in the initiation of cancer.

1.4.1 Effect on Chromosomes and Genes

The human cell contains 46 chromosomes. Chromosomes are made up of DNA molecules that contain genes in a coded form. Due to the passage of ionizing radiation damage of chromosomes is possible through several mechanisms, for example, breakage, cross-linkage, and alteration in the sites of a particular gene [25]. In most cases re-substitution takes place and breaks are healed. But if breakage in chromosome takes place and mitosis is disrupted, the cell may die. The majority of this disruption may cause chromosome defects which can lead to miscarriage, congenital malformations, mental and physical defects etc. It is also possible that instead of fatal changes, a few mutations in some of the genes may occur which can appear in the offspring in subsequent generations [26-27].

The probability of gene mutations is frequent than chromosomes defects. The gene mutation may pass through one generation to another generation whereas chromosome mutations affect only individual [28].

The gene component, DNA is a double-stranded helical macromolecule. The stranded of DNA is supported by phosphate residue and sugar. The two strands are joined by a pair of 'nucleotide' bases. The radiation can create the radical which can alter the sequence of bases and hence can change genetic code [29-30]. This process is called a DNA mutation. Mutation is passed to progeny during reproduction and leads to genetic defects.

1.4.2 Radiation Terms and Units

Following are the terms and units generally used in the measurement of radiation

a) Activity

The rate of decay of nuclei of a radioactive sample is measured in terms of activity. A single disintegration of radionuclide per second is called 1 unit Bq. In past activity was also measured by curie (Ci). One unit Ci is approximately equal to 1 gram activity of radon or it is equal to 3.7×10^{10} disintegrations of radionuclide per second. Therefore

$1\,Ci = 3.7 \times 10^{10}$ Bq

b) Activity Concentration

The activity of radionuclide present per unit volume of the sample is known as activity concentration. The S.I. unit of activity concentration is $Bq.m^{-3}$.

c) Radon Emanation Factor

It is defined as the number of radon atoms released to free pore space to the number of radon atoms created.

d) Radiation Exposure

The amount of radiation present in the air at a point due to a radiation source is called radiation exposure. It is used to represent the radiation ionization ability of the radiation in the air. In past, radiation exposure is measured in Röentgen (R).

S.I. unit is C/kg (Coulomb per kilogram)

1 Röentgen is the amount of radiation exposure, which will produce 1 e.s.u. of charge (of either sign) in 1 c.c. of air at NTP. 1 C/kg is the amount of radiation exposure, which will produce a charge of 1 Coulomb (of either sign) in 1 kg of air at NTP.

$1\ R = 2.58 x l0^{-4}\ C/kg$

e) Exposure Rate:

The radiation exposure per unit time is defined as the exposure rate. It is measured in R/hr or C/kg/hr**.**

f) Absorbed Dose:

The absorbed dose is defined as the dose absorbed by a matter of material. In past, it is represented by rad.

S.I. unit is Gray (Gy) or J/kg.

1 rad is equal to 100 ergs of radiation energy which get deposited in one gram of irradiated matter. 1 Gray is equivalent to 1 Joule of radiation energy deposited in 1 kg of irradiated matter.

1 Gy=100 rad

g) Equivalent Dose:

The equivalent dose (H_T) is equal to the product of radiation weighting factor (Wr) to absorbed dose. In past, it is measured in rem. The S.I. unit of equivalent dose is Sievert (Sv).

$H_T = D \times W_r$

1 rem $= 10^{-2}$ Sv

h) Radiation Weighing Factor:

It represents the weight of radiation absorbed dose as the biological effect of radiation on a human is dependent upon type and energy of radiation and also on the amount of absorbed dose.

i) Tissue Weighting Factor:

Some organs are more sensitive to radiation than others; the probability of stochastic effect in tissue depends on its sensitivity to radiation. For example, bone marrow is much more sensitive than skin.

j) Effective Dose:

Effective dose is the product of tissue-weighting factor (W_T) to the equivalent dose.

$E = W_T H_T$

If multiple tissues are exposed to radiation then the effective dose is measured by taking all the weight doses of all the affected tissues.

$E = \sum W_T H_T$

1.5 SOURCES OF RADON

The major sources of radon are as follows

1.5.1 Soil and Rocks

A Naturally occurring radioactive materials uranium and thorium are found everywhere in the soil. These are the major source of radon. The concentration of radon in soil is approximately 10^3 times greater than that found in the environment [31]. Radon moves slower in water-saturated rocks or soil in comparison to dry rocks or soil. That is why the buildings constructed on porous soil are at more risk of radon

exposure in comparison to clayey soil [32]. The distribution of radon concentration depends upon many issues like initial uranium concentration in soil, the porosity of the soil, the ambient temperature of the soil, wind speed and direction, humidity, pressure and seasonal variation etc.[33-34].

1.5.2 Building Materials

Many building materials are a common source of indoor radon. Building materials originate from the rocks and soil which are rich in different types of radioactive elements. In building construction waste from different industries are also used. These waste products contain an enhanced concentration of radon. In cement, fly-ash is used commonly which is a waste product of coal based thermal power plants. Gypsum, by-product of the phosphate industry, is utilized in plasterboards and in concrete. Building materials contribute about 52% of the natural background radiations to which the public is exposed [35].

1.5.3 Water

Radon concentration in water has been found to vary in a wide range. The amount of radon in water mainly depends on the local geological character and type of water supply. Drilled wells, especially in granite areas, have been found to have the highest concentration of radon in water. The lowest concentration is found in water from dug wells and surface water [36-37]. Radon can be transferred to the indoor air through the use of water during household activities like showering, laundering, dishwashing etc.

1.5.4 Natural Gas

Natural gas is released from underground sources. Due to the excessive concentration of radon in oil-fields and the superior solubility in organic solutions, the presence of radon in natural gas is very high. Several studies indicate that the concentration of radon in natural gas is in the range of 150 to 53700 Bqm^3 [38]. The fresh LPG contains radon concentration in the range of 70 to 48000 Bqm^3 [38].

1.6 APPLICATIONS OF RADON STUDY

The radon gas can help in the study of physical geology as it can migrate inside the earth crust for very long distance [39] and also it can be measured with very precise accuracy. Many geophysical phenomena of earth can be better understood by using radon as study material.

A few applications areas where radon study can help are as follows

a) Study of atmospheric movement

Water or moisture in the soil helps in the transportation of the radon through soil. Hence by studying the transport mechanism of radon through soil the moisture content can be deduced. Pressure gradient, temperature gradient and wind pressure [40] are other factors on which the flow of radon depends. So the study of the flow of radon can assist in determining these factors.

b) Uranium exploration

The radon can transport within the atmosphere, water and earth and this characteristic of radon make it helpful to be used as tracer medium for tracing various geochemical and geophysical materials [41]. It can be used for exploration of hydrocarbons and uranium deep inside the earth as these materials contain a high amount of radon concentration. Detection of abnormally high radon from a specific place can ensure the presence of these materials in the earth.

b) Radon as a precursor for earthquake prediction

Radium and radon gas is found in all kind of stones or rocks all over the world. It has been observed that the concentration of radon increases markedly in soil gas or groundwater before or during the earthquake [42]. This abnormal behaviour can assist in the prediction of an earthquake.

c) Medical applications

It has been reported in many studies that radon is hazardous to human. Radon is a major contributor of radiation doses received by humans. Many surveys have reported that lung cancer, kidney diseases and skin cancer [43] were associated with exposure to radon. The radon study in the suspected area will provide ample scope for the detection of these diseases in advance.

1.7 MOTIVATION

In 1597, a high lung cancer rate was observed in miner by German physician Georgius Agricola [7]. Ludewig and Lorenzer in 1924 suggested that the high lung cancer rate in miners is due to the enhanced concentration of radon in the environment of mines [16]. Bale and Shapiro in 1956 suggested that radon is not the true cause of high lung rate but its daughter particle alpha is the main culprit [24]. Radon and its progeny can enter the human body is through inhalation of air and ingestion of food and water.

The major portion of natural radiation exposure received by humans is due to the inhalation of radon and its progeny, which occurs in the free state in the environment. During inhalation radioactive particles are accumulated in the lungs.

The other mechanism by which a human can be directly exposed to radon is through food. Natural radioactivity in soils comes from ^{238}U and ^{232}Th series, and natural ^{40}K. ^{238}U and ^{232}Th give rise to the radium and thorium atoms found in all humans, acquired from the food we eat and that food, of course, obtained from these materials from the soil in which it grew [44]. Radon generally enters into the human body through the food chain and also through the inhalation of the suspended dust in the air. When inhaled or ingested these elements accumulate in critical organs and deliver radiation doses. Hence the research in this domain is also necessary to establish the impact of radon on human health through food.

The Chernobyl nuclear disaster occurred on 26 April 1986 at the nuclear reactor in the Chernobyl Nuclear Power Plant, near the city of Pripyat in the north of the USSR [45]. It is one of only two nuclear energy accident rated as the maximum severity on

the International Nuclear Event Scale; the other being the Fukushima Daiichi nuclear disaster. The Great East Japan Earthquake and subsequent Fukushima nuclear disaster occurred on 11 March 2011, which caused the leakage of radioactive materials into the environment [46].

These disasters triggered the release of substantial amounts of radioactive contamination into the atmosphere in the form of both particulate and gaseous radioisotopes. Various studies on the populations that were exposed to radiation after these accidents have provided important data linking exposure to radiation and the future development of cancer [47-49].

Epidemiology is the study of health and disease in populations. The descriptive element of epidemiology comprises tracking of health and disease indicators and population risk factors. The epidemiologic modeling of radiation-induced health effects for the purposes of risk estimation in many cases relies on biological concepts developed from experimental studies with cultured cells and biological samples [49].

After seeing the importance of radon for human health many research groups became active in this area throughout the world. However, to best of our knowledge southern Haryana, India, remains an unexplored region. Faridabad, being a heavily populated industrial town, can be at high-risk zone due to Aravali mountain range passing through it. The present study has been aimed at investigating the effect of radon on the population of Faridabad and its neighbouring towns.

1.8 OBJECTIVES

The research work envisages the study of the environmental radioactivity in southern Haryana with the specific attention to the measurement of radon concentration, radon activity and their impact on human health. The major objectives of the present research work are:

1. To study the natural radioactivity in building materials like stone dust, natural dust, granite, cement, and fly-ash etc.
2. To study the natural radioactivity in rock samples collected from the Aravali Range.

3. To study the radon activity in vegetation (including cereals, fruits) samples in some districts of Haryana mainly Faridabad, Palwal, Gurugram, and Mewat.
4. To study the correlation between the concentration of ^{226}Ra and ^{222}Rn, in different radioactive materials.

For this purpose, the samples were collected from a different place of southern Haryana mainly Faridabad, Palwal, Gurgaon, and Mewat. The CAN Technique and Gamma Spectroscopy were used to study these samples and various parameters related to radon were measured. The estimation of radionuclides concentration is expected to be used as reference data to study the effects of radiation on the human population living in the sample collection area.

1.9 ORGANISATION OF THESIS

This thesis is organised in six chapters

Chapter-I: Introduction

This chapter starts with the introduction of radiation and the origin of radiation. Radon, the main contributor of radiation, its characteristics, and sources are discussed in details. The applications of radon are also discussed in detail. The effect of radiation on human health is also introduced.

Chapter-II: Literature Review

This chapter gives a brief history of radon and its measurements by the different research groups.

Chapter-III: Experimental Techniques

The “CAN technique” and “Gamma Spectroscopy” techniques are discussed in the chapter which is used for the measurement of the various parameter of radioactivity from the samples collected from different sites. The CAN technique is a passive method of measurement and the Gamma spectroscopy which is an active method of measurement and process is carried out at IUAC Delhi. This chapter also discusses the components and instruments used in these techniques for the estimation of radium concentration.

Chapter-IV: Measurement of Radon Exhalation Rate using CAN Technique

CAN Technique is the most efficient and cost-effective method for the measurement of the exhalation rate of radon. The exhalation rate of various samples which includes the soil samples collected from various districts of Haryana, the rock samples collected from Aravali range in Haryana, the cement and fly-ash mixtures sample, the natural dust and stone dust samples and the vegetation etc. samples collected from the local market in Faridabad. This chapter also discusses in detail the results obtained from all the experiments for the different samples under study.

Chapter-V: Measurement of Radioactivity Parameters using Gamma Spectroscopy

Gamma Spectroscopy technique has been discussed in this chapter. The samples which are collected from different places include

1. Vegetation samples which include wheat, rice, coarse grains samples and fruits, these samples were collected from the fields and the local market of the four districts viz. Faridabad, Palwal, Gurugram, and Mewat of Haryana State of India. The samples had been collected randomly from these different locations.
2. Different types of cereal which are commercially available and from the fields.
3. Rock samples from multiple places and altitudes of Aravali Range from Delhi, Haryana besides from Rajasthan, and Gujarat, for comparison purpose.
4. Fly-ash samples which are used in building materials are collected from the National Council for Cement and Building Materials (NCB), Faridabad.

The results obtained from the analysis of the above samples were also discussed to estimate the radiation risk to human.

Chapter-VI: Conclusion and Scope for future work

This chapter concludes the results obtained from the investigation in chapter 4 and chapter 5 carried out to measure various parameters of radioactivity. It also discusses the scope for future work in this area.

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CHAPTER-II

LITERATURE REVIEW

This chapter describes the history of radon and discusses the different studies and research work carried out previously in this field by different researchers. The various aspects of radionuclides like estimation of activity concentration of radionuclides, radon exhalation rate measurement and various related parameters of radon in different geophysical materials present in the earth crust are discussed.

2.1 INTRODUCTION

A German scientist Frederick Dorn in 1900 discovered that radium ^{226}Ra emits a gas, which is named as "radium emanation", later it was modified to Radon [1-2]. Ramsey and Whytlow-Gray in 1908 studied the physical characteristics of radium emanation and renamed it as Niton [3]. The Niton comes from Latin word which means the shining one. The unit of radon activity concentration was first introduced by Mache-Enheit and named as Mache. In 1910 the unit of radon activity concentration was replaced by Curie [4].

The chronicle order which shows the events related to radon and its progeny are as under

Year	Related Radon Research in this year
1597	High incidence of lung disease in metal miners was first mentioned by Agricola [5].
1879	The autopsy findings relating radon to lung cancer on underground miners was first documented by Harting and Hess [6].
1898	Radioactivity of radium, thorium, and polonium was discovered by Curie and Schmidt [2].
1898	The discovery of α and β particles by Rutherford [4].
1899	Ionization produced by radioactivity was demonstrated by Thomson and Rutherford [4].
1900	F. Dorn discovered Radon [2].
1901	Radon is a radioactive gas, demonstrated by Rutherford and Brooks [7].

1902	Transmutation process in which outer particle interacts with the nucleus was discovered by Rutherford and Soddy [4].
1902	Radon and its progeny measurements on screen made up of charged wire were done by Elster and Geitel [8].
1902	Radon was detected in tap water by Thomson [2].
1903	Condensation characteristics of radon at low temperatures were observed by Rutherford and Soddy [7].
1906	Rutherford demonstrated that charcoal adsorbs the radioactive emanations from radionuclide [9].
1907	Eve used radon trapped charcoal in an electroscope to measure radon [7].
1908	Whytlaw-Gray and William Ramsey studied the properties of radon by isolating it from radium emanation [10].
1909	Charcoal was used by Satterly to trap radium emanation from the environment and carried out a measurement of radium emanation concentration [10].
1908	Ashman determined atmosphere radium emanation by using charcoal [2].
1909	Regener set up a scintillation counter system for the determination of decay ^{210}Po [11].
1913	Squamous cells which are responsible for lung cancer identified by Arnstein from the miner's autopsy [6].
1913	Rutherford elaborated various physical and chemical properties of radiation emanations and showed that α particles are emitted during emanation, these α particles get condensed at very low temperature –150^{o}C and can be absorbed by coconut charcoal [12].
1914	The first time, Radium emanation was used for medical purpose [5].
1925	Radon name was used for the first time and symbol Rn was assigned to all the isotopes with atomic number 86 [4].
1932	Evans used two ionization chambers to evaluate very small quantities of radon [9].
1943	First reported work on radon in the environment and river water was carried out by Hess [6].
1940	The informal relationship was established between lung cancer and radon [13-14].

2.2 RADON MEASUREMENTS IN EARLY DAYS

Scientists in the early 1890s used only two devices to identify the ions produced by radioactive materials [2]. The electrometer was used as the initial measuring instrument for the measurement of radioactivity of radioactive materials whereas other instrument included electroscope. These instruments were basically ion chambers in which the sample was filled for measurement. The chambers were updated to include various components like dc amplifier, electrometer tubes, and recorder. Elster and Geitel in 1902 first measured the decay products of radon by accumulating these decay products on a single charged wire [8]. The electroscope became the prime choice for measurement since 1914. In 1908 Rutherford and Geiger demonstrated that α particle was a He nucleus [7]. Ionization chamber was later developed by Geiger to detect ions, so these chambers were named as Geiger counter [15].

In 1909, Regener used detector made up of ZnS materials and used it to count the alpha particles emitted by polonium [16]. Two chamber systems built by Robley Evans in which one chamber was filled with air for the subtraction of background radiation [9].

Curtis and Davis in 1943 measured the total ion current using a single ion chamber [17]. In 1947, a subsidiary of U.S. Atomic Energy Commission, HASL (Health and Safety Laboratory) and Davis opts for pulse ionization chambers as these were capable of measuring even small concentration of radon in different conditions [18].

Many methods and techniques were developed to measure radon in the mid of 1950s to fulfill the requirement of research scientists and who carried out measurements in outdoor and indoor environments, occupational sites, underground area [19-20]. At present, there are many types of techniques as well as instruments for radon estimation using the continuous methods of sampling, integrating and grab sampling. Research has always continued in this field so as to build precise and accurate radiation measurement instruments.

2.3 RADON AND ITS PROGENY MEASUREMENTS

Radon measurements studies were conducted on different samples like soil, groundwater, building materials, rock, and vegetation etc. by various researchers all

over the world.

Duggan et al., 1970 [21] investigated the exposure of radon on miners in the United Kingdom. The research conducted on Schneeberg and Jachymov mining communities during the 1920s and 1930s for epidemiological studies, recognized lung cancer may be caused by radon. The measurement was conducted for a total of 22 mines. The reported work shows that the highest concentration of radon was found in hematite mines.

Pritchard et al., 1977 [22] adopted the most accepted method which is a commercial liquid scintillation counter for radon measurement in water. The report suggested that the charged particles (alpha) from decay series of Po-218 and Po-214 interact with scintillation material and produce photons which can be detected by a photomultiplier tube.

Hayakawa et al., 1985 [23] reported the presence of short half-life radon progeny concentration in cloud and rain. The data for the period of 7 years was obtained from NaI (Tl) scintillation counter continuous monitoring system from Fukui in Japan. The experiment was conducted at different locations and sites of Japan to study the seasonal variations effects on the specific activity of radon daughters in rainwater. In winter, the value of the specific activity of radon in rain water was found higher than the value of specific activity of radon in summer. This study demonstrates that the radon progeny specific activity due to seasonal variations rain water depends on rainfall rate and radon concentration in the cloud.

Jarad et al., 1988 [24] demonstrated the radon measurement using SSNTDs. The study summarized the previous work done in the field of radon measurement. In the study SSNTDs were used in different techniques for radon measurement like CAN Technique which can be used for measurement of radon concentration and emanation from building materials. Working Level Monitor, which can be used for measurement of working levels in houses over a short period and passive radon dosimeters which can be used for radon emanation for longer period of time.

Ramachandran et al., 1989 [25] measured the radionuclides present in foodstuffs

consumed by the Indian population at large. The samples for the study included the main components of Indian thali like rice, wheat, pulses, potatoes, fish, mutton, and fenugreek. The samples were collected from the market and were ashed to obtain radioactive equilibrium. The NaI(TI) gamma-ray spectrometry was used for measurement. The study concluded that the daily intake value of radium and thorium in the sample region was higher than the western countries. The study on the basis of obtained results showed that the natural radioactivity content in the non-vegetarian diet was found to be lower than plant origin foodstuff.

Choubey et al., 1997 [26] measured radon level in Garhwal Himalaya located in Uttarakhand state of India. Different samples of soil and spring water were collected from hill region. A scintillation cell detector was used for measurement.

Lubin et al., 1997 [27] discussed the findings of studies which was carried out on underground miners. The lung cancer probability in miners in mines is high because they are open to highly radioactive materials radon and their progeny. The study regarding the carcinogens, which are capable of causing cancer and affect public health, were related to exposure from radon. It was estimated on the basis of the report of underground miners studies that around 6000–36000 deaths due to lung cancer every year in the U.S were because of indoor radon. Some studies show positive or weakly positive results, while others report related to prolonged exposure of radon. The meta-interpretive analysis was applied to case studies to gather information. This study further was applied to individual findings of 200 cases in which a long-term method for indoor radon measurements was used.

Jarad et al., 1997 [28] used CR-39 nuclear track detectors as dosimeter to detect solar radiation. These detectors were bombarded with Cf-252 before exposing them to sunlight. The two methods used in the study include radiation grabbing from the sun by using solar tracking, while in another method the sample gets fixed radiation from the sum. The experiment was conducted for 1 week to 8-week duration in Saudi Arabia. The result indicated the linear correlation between track diameter and fission fragments. The result also indicated that SSNTDs could be utilized for environment detection.

Tufail et al., 1999 [29] used CAN technique for the determination of exhalation rate of radon for coal and shale materials collected from various mines in Chakwal and Makarwal region of Pakistan. It can be observed from the result that the samples collected from targeted mines have low uranium content and hence pose no risk to mines workers. The radon exhalation results obtained from this experiment are within the safe limit specified by OECD. The obtained results were also compared with other researchers results for the same target site.

Kant et al., 2001 [30] presented the radiological impact due to radon from cement, coal and fly-ash samples. The fly ash and coal samples were reported to be collected from the power generation unit of Northern India and cement samples were taken from NCB, Faridabad. These samples were analyzed for radon concentration using SSNTDs. The value of lifetime fatality risk and annual effective dose was also computed during reported research. The various observed data were found below the global safe limit as specified by OECD.

Kant et al., 2003 [31] presented a study on the beneficial effects of low-level ionizing radiation (LLIR), the process is also called radiation hormesis. The results obtained from the experimentation on animals suggested the role of LLIR to protect against radiation-induced cancer. The various studies establishing the benefit of exposure to LLIR were also presented in the analyses. The paper also presents the chronicle order of the events carried out in the field of LLIR and their application in the various field. The LLIR significantly reduces the cancer mortality is established in the paper through various studied carried out in the field of LLIR.

Nikezica et al., 2004 [32] reviewed a few applications and characteristics of SSNTDs for the radon and its decay product measurement. Many models for the alpha track creation and calculation of track etch rate and bulk etch rate were illustrated and compared to each other for the obtained results. The multiple models were verified for the major and minor axis calculation of tracks created during exposure from radon samples. The review also focused on the bulk etching rate and its dependence on etching conditions, preparation methodology and irradiation before the etching process.

Scheibel et al., 2005 [33] measured radioactivity concentrations of ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs in commercial samples of South-Brazilian cereal flours (soy, wheat, corn, manioc, rye and oat flour) to verify the radiological safety of these foodstuffs. The cereal flours are the major component of the Brazilian diet and are also important exportation products. The measurements were carried out by gamma-ray spectrometry using a 66% relative efficiency HPGe detector. The highest concentrations levels of ^{232}Th and ^{226}Ra were 0.69±0.04 Bq. kg^{-1} and 0.44±0.03 Bq. kg^{-1}, respectively, in soy flour.

Shukla et al., 2005 [34] analyzed the increased radioactivity in building materials by utilizing gamma-ray spectroscopy. The reported work found an enhanced level of radioactivity in the construction utilizing fly-ash and phosphor-gypsum in building materials. The fertilizer industry generally produces phosphor-gypsum during the manufacturing of phosphoric acid in and fly-ash is obtained from burning of coal in a thermal power plant. The results demonstrated that the bricks made of phosphor-gypsum which had about 90% of original ^{226}Ra content and flyash which is a by-product of coal, contains non combustible mineral matter, which have most of radionuclides, and hence bricks made up of these two materials had enhanced the level of the radioactivity.

Tuccimeia et al., 2005 [35] investigated the influence of humidity content, surface exhalation and precursors concentration of radon and thoron exhalation rates from samples of construction materials using SSNTD and accumulation chambers. The technique used a continuous monitor having an SSNTD alpha detector, available in a small chamber. The back diffusion on ^{222}Rn effects and chamber leakage effects were computed in this study. The study emphasized the mechanism for marking the building materials with a radiation performance index on the basis of radiation dose contribution from them. It can be concluded from this study that the conditions for experimental should be fixed as the exhalation rates values are dependent and highly variable according to indicators as air temperature, humidity, grain size.

Amaral et al., 2006 [36] determined the activity concentration of uranium and ^{226}Ra in foodstuffs cultivated in this area, where the phosphate mineral has been extracted. The activity concentrations found for uranium and ^{226}Ra in the foodstuffs analyzed

varied from 13 to 186 $mBq.kg^{-1}$ (wet weight), with a mean value of 46 $mBq.kg^{-1}$ and from 43 to 2209 $mBq.kg^{-1}$ (wet weight), with a mean value of 358 $mBq.kg^{-1}$, respectively. The annual intake of these radionuclides, for rural residents, was 7.45 Bq for uranium and 69.3 Bq.for ^{226}Ra.

Kant et al., 2006 [37] measured the radon concentration, annual effective dose, PACE, lifetime fatality risk and annual exposure in the Indian dwelling made up of fire mud brick, fly ash brick, mud and concrete blocks using SSNTDs. The effects of plaster, paint, and whitewash were also analyzed in the research. The range of calculated data for the above said parameters were found under the safe limit as recommended by various global environmental agencies (OECD, UNSCR, ICRP).

Mehra et al., 2007 [38] analyzed the activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K in soil samples. Malwa region of Punjab was the sample collection site. The measurement was carried out using HPGe (High Purity Germanium detector) high resolution gamma-ray spectrometry. The measurement of absorbed dose rate was carried out by using the value of activity concentration of ^{222}Ra, ^{232}Th and ^{40}K. The obtained value of H_{ex} is less than 1 which is considered as safe limit.

Iimoto et al., 2007 [39] developed a radon exhalation measurement technique from an activated charcoal collector. In this technique cuvette of 2.1 litres was used for sampling by covering the soil on the target site for 1 day. A liquid scintillation counting system (PICO RAD) and activated charcoal were used for radon concentration measurement. The modified technique demonstrates high reliability. It can be utilized for radioactivity concentration measurement from natural environments.

Sahoo et al., 2007 [40] calculated emanation factors for different construction materials. The analysis of the building materials samples was carried out for ^{226}Ra, ^{232}Th, and ^{40}K contents. AlphaGUARD, a monitoring system was used for instantaneous or continuous measurement for radon mass exhalation rates. From the obtained result of ^{226}Ra content, ^{222}Rn emanation factor and ^{222}Rn mass exhalation rates were calculated for these samples. Gamma dose rate is more thanthe normal values in these samples. The silica-fumes has high amount of radioactivity.

Kant et al., 2008 [41] investigated the effect of the occurrence of radon, thoron and their decay product in the environment to the workers of LPG (Liquified Petroleum Gas) bottling plant. Annual exposure, annual effective dose and lifetime fatality risk for the worker of LPG plant were measured and calculated for the four seasons of the year. The twin cup dosimeter technique employing SSNTDs were used in this research. It is reported that radon activity maximum value was measured in the winter season and the minimum value was measured during the summer season in the range of 7.78 $Bq.m^{-3}$ to 59.01 $Bq.m^{-3}$. The similar trends were reported in the value of thoron activity and PACE due to thoron and radon. The data also indicated the lifetime fatality risk varied in the range $0.31x10^{-4}$ to $0.65x10^{-4}$. The reported work suggests the probable health risk due to external irradiation and inhalation of the progeny of radon.

Gray et al., 2009 [42] reported about the deaths due to lung cancer caused by indoor radon and explored the cost-effective methods to decrease the level of indoor radon. The estimation for the number of deaths from lung cancer before and after the suggested remedial corrections to limit the indoor radon in homes was done. The computation of the lifetime risk or death and the potential of these methods to reduce the mortality rate due to lung cancer were carried out. It is known fact basic defense mechanism to reduce entry in the dwelling is very costly hence cheap methods of radon reduction were experimented.

Naureen et al., 2010 [43] discussed that the exposure of radon for longer duration enhances the lung cancer development risk in non-smokers. The study was carried out to determine radon exhalation level in different 53 samples used as interior decoration in houses. Samples of materials like tiles, drywall, and granite available in the Canadian market were collected. The study concludes that decorative building materials used in homes show a notable change in radon concentration with appropriate air ventilation.

Abo-Elmagd et al., 2010 [44] calibrated the CR-39 for radon-related values for the measurement of radon mass and area exhalation rates of soil and rock samples and effective radium content of the samples was carried out. The CR-39 in a sealed cup (passive method) was used for the measurement. The correlation in the calibration of

Lucas cell, which is an active method was done. The study established a reliable calibration factor for each sample. It can be concluded that calibration factor optimization is very difficult for a sample in experimental conditions as it is considered that each sample has its own calibration factor. The study also demonstrated the advantage of a back diffusion correction method. It was also demonstrated that if the back diffusion corrections were carried out, the constant calibration factor can be obtained for the 1/3 of the volume of the sealed cup.

Rahman et al., 2010 [45] carried out the radon measurement in workplaces of Rawalpindi region and Islamabad region of Pakistan. The indoor radon measurement was done at 105 workplaces of the sample area. The study was conducted to assess the indoor radon health hazard. Obtained result values were lower than the recommended by ICRP.

Hassan et al., 2011 [46] investigated the concentration of radionuclides, ^{226}Ra, ^{232}Th and ^{40}K using γ-ray spectroscopy in a different variety of granites used as building materials in Japan. The different health hazard indices were calculated from the obtained results. The accumulation chamber having solid-state alpha particle detector was employed for the calculation of exhalation rates of radon.

Scott et al., 2011 [47] studied the relation of indoor radon level with lung cancer. The LNT was applied for the risk-assessment paradigm and the LNT theory was not verified in many cases. The hormetic relative risk (HRR) model was also applied for the radon exhalation measurement in dwellings.

Boukhenfouf et al., 2011 [48] studied the natural radioactivity concentration of radium, thorium, and potassium in the fertilizers and soil. The active method was used for measurement and the results were compared with other published results for different radioactivity at different values of radium equivalent concentration.

Al-Alawy et al., 2011 [49] proposed two experimental set ups for surface measurements of radon concentrations in underground well water and oil-produced water separated from oil in Karbala Governorate in Iraq. In the first measurement active technique using RAD7, which is a fast electronic technique, was used and in

second measurement passive technique using SSNTD was carried out. The experiment demonstrated a good correlation among the results from both active and passive measurement techniques.

Xiofeng et al., 2011 [50] measured the radon exhalation rate using professional radon monitor RAD7. Single story dwelling was selected as a research site in Shenzhen city. The radon exhalation rate in the soil surface was higher than radon exhalation rate of the room's floor made up of cement. It was observed that the surface exhalation rate of radon of the floor made of ceramic tile was lower than the cement floor. The research concluded that indoor radon level can be minimized by proper natural ventilation and selecting the right building materials to avoid high indoor radon concentration.

Bavarnegin et al., 2012 [51] used a radon gas analyzer having a container for the measurement of exhalation rates of radon. The samples used in the study were construction materials collected from Ramsar in Iran. The measurement of contents of ^{40}K, ^{232}Th and ^{226}Ra was carried out by employing the HPGe gamma spectrometer system. It can be observed from the result that an excellent correlation coefficient in radium concentration and the exhalation rate of radon was obtained. The results of the study indicate that radium content and exhalation rate of radon in some stones samples which were used in basements were higher than the world average. These samples can be considered as the major sources of indoor radon emanation. It can be concluded from the results of this study that, use of the bricks, cement etc. are better than the use of hot spring's stones.

Ahmad et al., 2012 [52] determined the value of concentration of radon, radon exhalation rate, annual effective dose and radium activity in building materials used as construction materials in the dwelling of Dera Ismail Khan in Pakistan. The cement, unbaked and baked brick samples were taken for measurement. Active and passive method RAD7 and SSNTDs based radon dosimeters were employed. It was concluded that radium activity, radon concentration, exhalation rate was observed highest in unbaked brick in comparison to other samples in the study; but the overall values of different radioactivity falled for safe limit values below 370 Bq kg^{-1} for radium activity, 57.6 Bq $m^{-2}h^{-1}$ and 1100 µSvy^{-1} for an annual effective dose.

Hussein et al., 2013 [53] measured radon activities and exhalation rate of radon in different materials like cement, gravel, sand, ceramic tile, gypsum, gypsum board and block which are used in public hospitals in Iraqi Kurdistan region. The passive method using the CR-39 SSNTD technique was carried out. It was concluded that the radon exhalation rates in sand samples had higher values in comparison to that for gravel block, gypsum, ceramic tiles, gypsum board, and cement.

Mehdizadeh et al., 2013 [54] collected water samples from the Fars province which is located in Iran the south-west region. The nuclear power plant Bushehr is located in this area. A total of ninety-two samples of spring water of Fars province and the water supply of Shiraz city were investigated. Measurement of concentration of natural radioactive elements by ^{222}Rn emanation method was carried out. The uranium concentration was measured by fluorimetry analyzer. The evaporation method was used for gross α and β concentrations measurement. The results of the experiment demonstrate that the radon activity concentration was found in the safe limit as determined by the WHO and the US EPA for the water samples.

Kumar et al., 2014 [55] observed the effect of moisture and additive in concrete for radon exhalation rate. The building materials contribute radon significantly after soil in an indoor environment. The indoor radon level depends upon the amount of radon flux emitted by building materials, which were mainly used in wall, ceiling, and roofs of the dwellings. The radon flux of concrete depends upon many factors like porosity, radium content, moisture, the ratio of material for construction of concrete and age of concrete. The study was carried out using a continuous radon monitor active method. The results showed that variation in the porosity of concrete and moisture content affects the radon exhalation rate.

Abu-Jarad et al., 2014 [56] analyzed the risk of lung Cancer in smokers from the cigarette in a closed room in Iraq by taking uranium contents and carrier of indoor radon products. The measurement was done to calculate the effective dose of radon from these cigarettes. Sealed can technique using SSNTDs was utilized for the experiment. The observed results showed an excellent correlation between lung cancer and radon concentration.

Kumar et al., 2014 [57] assessed the activity concentration of radon and radon exhalation rate in drinking water and soils sample. The collection of the samples was carried out from the Pathankot district of Punjab. RAD 7 electronic detectors were used to measure radon concentrations and CAN technique with α sensitive nuclear track detector was used to measure the exhalation rate of radon. The obtained results were found under safe limits.

Amin et Al., 2015 [58] measured the exhalation rates of radon and radium content in the construction materials. The measurement was carried out by applying SSNTDs in CAN technique. It was observed that radium content value for all soil samples was found below the recommended safe limit of 370 $Bqkg^{-1}$.

Abd-Elmoniem et al., 2015 [59] investigated soil samples for the exhalation rate of radon. The samples collection was done from three towns El-Hosh, El-Managil, and Medani in the Gezira State of Central Sudan. The report demonstrates that the concentration of radon in soil depends upon the presence of soil permeability, the levels emanation from rocks and soil. The passive technique, using CR-39 was used to estimate the radium concentration and radon exhalation rate. The obtained values of the exhalation rate of radon and radium content was found very much below permissible safe limit by OECD.

Khan et al., 2015 [60] studied soil samples for the radium content and exhalation rate of radon. The samples collection was carried out from different sites of Shahjahanpur district of Uttar Pradesh state of India. Cylindrical can technique (CCT) based on SSNTDs was used for the experiment. The value of radium content were found below the safe limit as specified by OECD.

Chauhan et al., 2016 [61] analyzed the radioactivity in tobacco plants. Tobacco smoke is related to pulmonary cancer and is considered producing carcinogenic effect on human health. The analysis of radioactive elements present in samples was done using the ICAP-AES technique. It is shown that densities of alpha tracks vary in the bottom and the top side of the tobacco leaf. It is observed that the value of alpha track density is high in the lower side because of the high amount of trichomes present on the lower side which attract radon daughter and dust from the environment.

Sung-Soo et al., 2016 [62] reported that radon was considered as the second most reason for lung cancer after smoking. It was observed that lung cancer rate has been steadily increasing especially in non-smokers. To address this problem, the epidemiological studies were carried out to assess human health risks from radon exposure. The reported work includes cohort mortality studies, residential case-control studies, and ecological studies. It was concluded from the data that elevated levels of radon concentration increase lung cancer risk in non-smokers.

Yousef et al., 2016 [63] used LR-115 and CR-39 nuclear track detectors to calculate the value of surface exhalation rate and radon concentration in phosphate samples. These samples were collected from taken from Abu-Tartur and El-Sebaeya, Egypt. The results suggest the higher concentration of radon in the phosphate sample of Abu-Tartur in comparison to samples of El-Sebaeya.

Abd-Elmoniem et al., 2016 [64] studied soil samples for concentration of exhalation rate of radon. The collection of samples was carried out from Kassala town in Sudan. The SSNTDs in CAN technique was used for this purpose. The results indicate an excellent correlation between radium concentration and exhalation rates of radon for soil samples.

Khan et al., 2016 [65] performed the radon measurement studies by using the nuclear track detector in Pakistan occupied Kashmir. It was concluded after many epidemiological studies that the lung cancer rate was related to radon and radon decay products. The report gives a clearer view for understanding the roadmap for radon measurements in Pakistan Occupied Kashmir (POK).

Abd et al., 2017 [66] developed an equation that links the relationship between the theoretical and experimental value of activity concentration of radium. The reported work was to design a methodology for the characterization of radioactive materials. The samples of radioactive materials included radioactive waste used for transport and storage was taken to determine the radiation dose rate. The report measured the activity concentration of radiation describing them in the gamma analysis system.

Durusoy et al., 2017 [67] measured the activity concentrations of uranium, thorium, potassium, and ^{137}C which were induced in Chernobyl accident. The measurement was carried out in the soil samples collected from the Rize Province of Turkey. Rize city was significantly affected by the Chernobyl accident in 1986, and its effects are still visible. The main production plant of this region is Tea and that is used for measurement. It is considered that tea plant and the soil gets contaminated by radioactivity from artificial source. The collected samples were analyzed for activity measurements using gamma spectrometry having a Nal(TI) as a detector. The activity concentrations of radionuclides present in soil samples were compared to previous finding on this area and other surrounding areas.

Choudhary et al., 2017 [68] analyzed the soil samples for exhalation rates of radon and radon activities. The soil for experimentation was collected from sites of Banda district of Uttar Pradesh. The SSNTD LR-115 type-II detector was placed in sealed CAN. Based on the assessment of soil samples, the effective doses from indoor inhalation exposure works out to be less than 0.3mSv/year. It was concluded that variation in soil samples was due to the difference in porosity, uranium and radium concentration of soil samples.

Subber et al., 2017 [69] analyzed soil samples for radium concentration and exhalation rates of radon. The measurement was carried out in the area of Basarh Governorate in Iraq. The measurement was done using SSNTDs. The exhalation level of radon from soil samples collected in this experiment was significantly lower than the safe limit recommended by UNSCEAR 2000.

Avinash et al., 2017 [70] determined the concentration of ^{226}Ra, ^{232}Th and their daughter product in dwellings of Gogi region located in Yadgir District of Karnataka state, India. Dwellings were chosen in the 5 km range near the mining area. The measurement of radon and thoron activity concentrations was done using pinholes based dosimeter. The observed data shows that the average value of radon concentration is well within the range of Indian average and world average value. The result shows a higher concentration of radon in comparison to thoron. The average annual inhalation dose was found within the UNSCEAR limits.

Abbasi et al., 2017 [71] measured radioactivity of granite which is the commonly used building materials all over the world. The two types of methods used for measurement were passive and active methods. In passive method concentration of radium was measured by high purity germanium detector HPGe gamma-ray spectroscopy, after sample preparation. In the active method, an alphaGUARD model PQ 2000 was used. The calculated values of radon concentration and exhalation rate of radon from both the method were compared and the results were found to be the same.

Ruano-Ravina et al., 2017 [72] considered Galician municipalities as a study unit and designed an ecological study. Municipalities had done three major radon measurements. The relation between radon concentrations and mortality rate in municipal during the period 1999–2008 was shown. The study observed a correlation between indoor radon with mortality due to brain cancer for females and males. The intensity of female mortality was found higher.

Abujassim, 2017 [73] emphasized that hair dyes are frequently used materials nowadays by the general public all over the world. The samples of hair dye were investigated for the radium content and exhalation rate of radon. The measurement was carried out using a sealed can technique. The study finds a very good correlation between effective radium content and the exhalation rate of radon.

Nwankwo et al., 2018 [74] deduced the activity concentration of radon with the depth of the mines as miners are directly exposed to radiation from earth crust. The measurement for activity concentration was carried out for the different samples collected from different depth 0-222ft in Komu and 0-30 ft in Olode mining sites, Oyo State. The gamma-ray spectroscopy was used to determine radionuclides. ^{238}U and ^{232}Th concentrations at Komu, increased with increased depth but the value of ^{40}K did not show any defined pattern. ^{238}U and ^{232}Th concentrations at the Olode site was found decreasing with depth and value of ^{40}K was almost constant. The value of internal hazard indices at Komu sites was observed higher than the acceptable limits for workers.

Han et al., 2018 [75] analyzed the concentrations of uranium, thorium, and potassium in the aerosols. The atmospheric samples of PM10 were taken from Gosan city of Jeju Island. The analysis of activity concentration was done using ICP-DRC-MS to find the annual effective dose. The study was carried out to find the presence of radionuclides in the flowing air to the island. The island has a slight presence of fine dust arriving from China otherwise the Jeju Island has low pollution and low population density. The research proposed the results are primary data to study the impact of fine dust Koren peninsula.

Mugahed et al., 2018 [76] determined the annual effective doses and exhalation rates of radon of ^{222}Rn exposure in the construction material procured from Yemen. 33 samples of construction materials were processed. The SSNTD was used in the experiment to measure the effective radium concentration and exhalation rate of radon and of the samples. The result of effective radium concentration for all samples was found in the safe limit as suggested by OECD.

Mehra et al., 2018 [77] analyzed the soil samples for the exhalation rate of radon by SSNTD. For this study, the samples were collected from Western Haryana. It can be observed from the obtained values that radiological risk due to natural radiations can be reduced by choosing the right building material for the dwellings.

Abo-Elmagd et al., 2018 [78] evaluated the various parameters related to radon from the samples of environmental collected from Saudi Arabia. The soil, building materials used for construction as well as decoration materials are considered as the main radon sources in indoor environment. To determine radium content and radon exhalation rate sealed cup technique was used. The back diffusion mechanism was utilized for a quality result. The relationship between back diffusion and radon exhalation rate was demonstrated in the research.

Lekshmi et al., 2018 [79] determined the emanation factor and exhalation rates of radon of soil samples. These soil samples were collected from the seashore of Kerala state of India. It is considered that due to the erosion of rocks and weathering, the mineral gets deposited on the beaches. The sand samples were collected from the coastline of Kerala as the high radiation materials are also gets deposited on the sand

of the beaches causing high environmental radiation. The results show that many soil samples demonstrate high specific activity of ^{226}Ra and the correlation exist to ^{222}Rn mass exhalation rate.

Gillmore et al., 2018 [80] presented a study of final outputs of the five-year UNESCO / IUGS / IGCP Project and presented a new data on radon in the built and natural environments, radon as a diagnostic tool of geophysical phenomena, reflections and a critique of radon's asserted use as a therapy. The study proposes that radon science has the potential to be a useful tool in understanding our environment as well as its impacts on human health. Concerning natural hazards, large-scale data analysis and detailed modeling, based on knowledge of the variability and complexity of the natural and human processes, will ensure radon physics as one of the most reliable tools for the surveillance and the protection of the environment.

Razumov et al., 2015 [81] presented some applications of radon for medical rehabilitation of patients. One of its application is Radon therapy, which is the mechanism of action which consist of the influence of the small radiation doses of radon and its daughter products on the nervous, vascular, and immune apparatuses of the skin and mucosal membranes that eventually enhances the protective and adaptive potential of the body and thereby its ability to resist pathological impacts. Presently, the high effectiveness of radon therapy is universally recognized and used for the treatment of various diseases in different fields of medicine. These include locomotor disorders, neurological disorders, cardiological disorders, gastrointestinal disorders, gynecological problems in the patients. On the basis of available data, the research presents excellent modern applications of radon in the field of medical treatments.

Park et al., 2016 [82] investigated the relationship between indoor radon concentrations and lung cancer. Initially, the indoor radon concentration was assessed and the factors affecting indoor radon concentrations were observed. The related mathematical models based on the mass balance equation and the differential equations were also presented in the reported work. Most of the models reviewed in this article utilized solutions at a steady-state and considered parameters to be constant in order to estimate indoor radon concentrations. Such approaches are often inadequate, as a steady-state is rarely attained in actual buildings because of time-

dependent factors. For this reason, time-dependent solutions may be more accurate than steady-state solutions in the assessment of actual building conditions.

Yang et al., 2019 [83] experimentally recorded the radon exhalation rates from the soil as well as corresponding meteorological and soil parameters for two subsequent years. Based on long-term field data, a statistical regression model for the radon exhalation and the most important influencing parameters soil water content, temperature of soil and air, air pressure and autocorrelation of the exhalation rate was prepared. The result showed that the multivariate model developed by the authors can explain up to 61% of the variation of the exhalation rate. The results also indicate that the air temperature had a positive effect while the soil temperature had a strong negative effect on the exhalation rate, indicating their different influencing-mechanisms on the exhalation. The air pressure was negligible. The variable shows strong autocorrelation hence the lagged values of radon exhalation were included in the model.

2.4 EFFECTS OF RADON ON HEALTH OF UNDERGROUND WORKERS

Since the discovery of radon around 300 years ago, a German physician Georgius Agricola had figured out a high rate of mortal lung diseases to local miners [1]. In 1879, Harting and Hess, two physicians, observed that the mortality rate was around 75% in miners of Czechoslovak and Germany [6]. Lung cancer was found mostly in those miners who had worked for more than 10 years in mines. Margaret Uhlig in 1921 reported that radium might be the reason behind this disease [10]. Ludewig and Lorenser in 1924 suggested that lung cancer in these miners may be due to the radon gas [13-14]. Radon was attributed to be a major reason for lung cancer to the miners of Schneeberg in Germany and Joachimstal in Czechoslovakia in the reported research carried out from 1924 to 1932 [5-6]. Sikl and Pirchan in 1932 suggested that lung tumors among Jachymov miners caused by radium emanation [10]. Around 50% of people who died due to lung cancer had worked in these mines [13].

Lorenz in 1944 reported that radon is not the main culprit but its daughter product α particle plays the main role [17]. In 1951 Bale and in 1952 Harley, suggested that α particles in the decay series of radium cause the lung cancer [12]. Bale and Shapiro in 1956, calculated the expected dose to the lung by deposition of radon and its decay

products via inhalation [12]. The idea that α particle can cause lung cancer was not welcomed by many scientists, until 1950 to 1960 when the study was carried out to epidemiology on underground miners [13].

The BEIR VI report based on the underground mines data reported that from 1941-1990, approximately 60,000 miners who had worked in eight different countries, 2,600 were detected with lung cancer in comparison to expected 750 [14]. Eleven studies of epidemiology out of 20 studies carried out on underground miners established the link between lung cancer risks to radon emanation.

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CHAPTER-III

MATERIALS AND METHODS

3.1 INTRODUCTION

In the early 1900s, the researchers used two types of instruments for the detection of radioactive materials. These instruments used to detect the ions produced by radioactive materials. These instruments were ion chambers along with the sample kept inside the closed vessel. The modified ion chamber included electrometer tubes, besides recorder and amplifier were also there. Elster and Geitel in 1902 were successful in collecting radon decay products on the wire [1]. In 1909, Regener built a counting system, which uses a ZnS medium as a detector that counts the alpha particles produced by Po-210 [2]. A two chambers system was developed by Robley Evans, which had one chamber sealed with air that responded to gamma radiation and also allowed subtraction of background radiation developed in another chamber [2].

In 1940s scintillation electroscopes and ionization chambers were accessible for detection of alpha and beta particles. In the era of the 1950s to meet the needs, scientist developed many other instruments for the detection of radiation. Today, many instruments and techniques are available for radon measurement in the air and in the environment.

The major factors on which the selection of radon measurement technique depends are:

I. Sample to be measured for radon or its progeny

A radon measurement technique depends on the type of sample to be detected. e.g. in case of foodstuff radon produced by radium will be lost, therefore gamma ray spectroscopy is the best suitable technique for this purpose. In another case, for example in earthquake prediction, continuous monitoring is necessary [3]. In the measurement of hydrocarbon exploration, the time-integrated radon measurement technique is always preferred. The ZnS scintillation cell method is suitable to count low concentration radon in water samples.

II. Types of emission type of radioactive material (alpha/beta/gamma radiations

1. Detection of alpha particles only.

2. Detection of all alpha, beta and gamma liberated by a radioactive element.
3. Detection of the only gamma rays.
4. Detection of beta particles only.

III. Resolution time

The resolution time is defined as the time taken by an experiment to resolve a problem.

Measurement techniques can be divided into three categories on the basis of resolution time.

(a) Grab sample technique

The sample of air or water, in which radon or its progeny measurement is to be carried out, is collected during the short interval of time in comparison to mean life of radon [4].

(b) Continuous Monitoring Technique

In this technique, the measurement of radon concentration is carried out in time series. This technique involves the collection of the sample from the source i.e. air, soil gas, water etc. and measured simultaneously. A few instruments like Scintillation Radon Monitor and Smart Radon Duo are usually employed for detection and measurement of radon parameters [5].

(c) Time-Integrated Radon Measurement Technique

This technique includes the measurement of radon concentration and its integration over a specific period of time. In this technique time interval may be as large as few months to a year, hence this method includes detection of radon concentration for a large time so as to find average radon concentration at a specific location [6]. This process is usually carried out using passive detectors.

3.2 MEASUREMENT TECHNIQUES

Radon measurement parameters mainly depend upon the detection of α-particles released during the decay of radon. Measurement technique can be classified as active and passive as shown in figure 3.1 [6].

3.2.1 Active Techniques

In these techniques, radon measurement and its level of working are continuously monitored. This involves continuous measurement and recording of the presence of

radon and its progeny in the samples. Unusual swings in radon concentration can be measured using these techniques [7]. Different types of technique includes an electronic integrating device (EID), continuous radon monitor (CRM), RAD7 etc

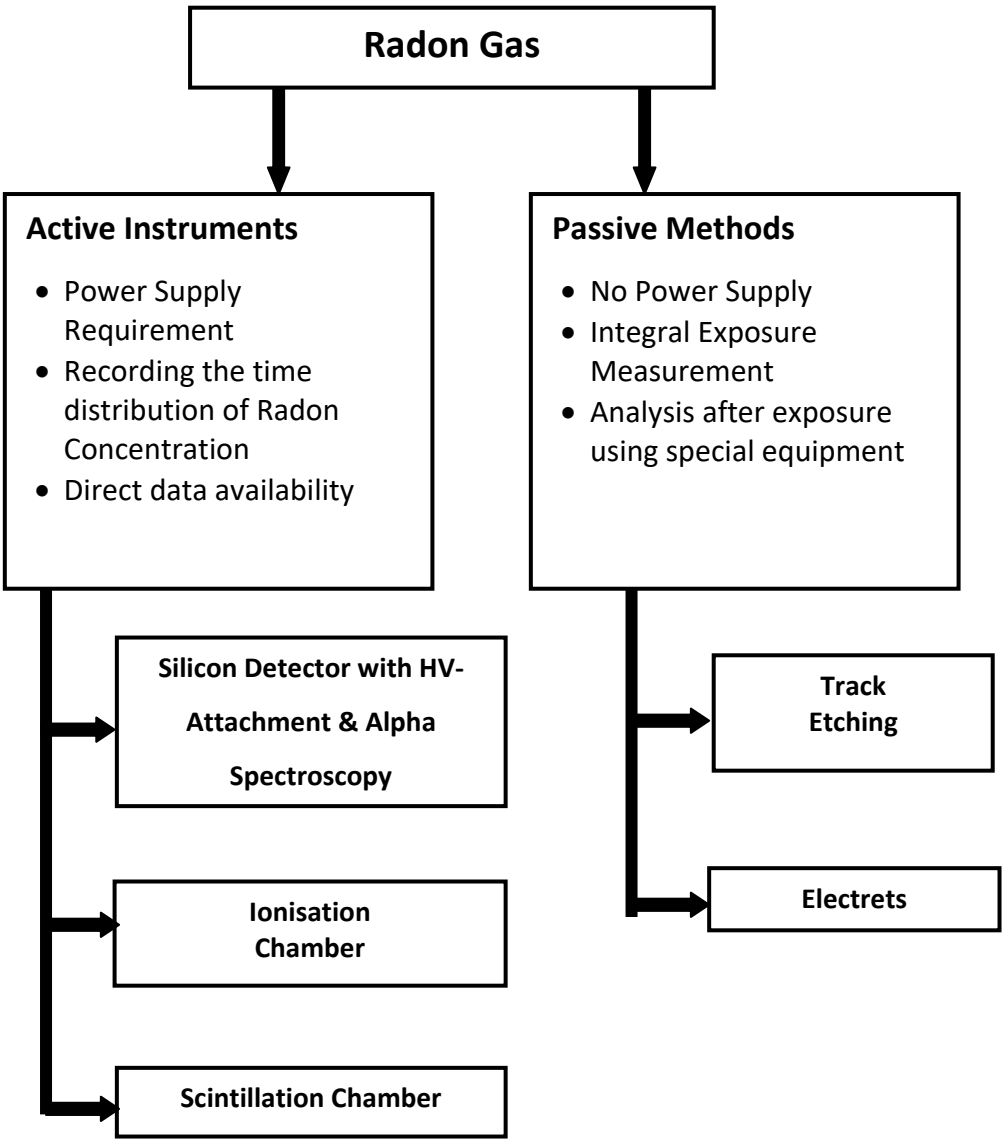

Figure 3.1: Active and passive techniques of radon measurement

3.2.2 Passive Techniques

In passive techniques, the sample is exposed to air for a very short interval of time [8]. These radon testing methods include AlphaGuard, alpha-track detectors, electret ion chamber detectors, charcoal canisters, thermo-luminescent detectors etc.

3.3 TECHNIQUES USED IN PRESENT INVESTIGATIONS

The present research work utilizes CAN Technique (passive technique) and Gamma spectroscopy (active technique) for the measurement of different parameters. The different components used in CAN technique are as follows:

3.3.1 Solid State Nuclear Track Detectors (SSNTDs)

Time-integrated measurements technique using SSNTDs for a slightly longer time period (around 2-3 months) has been extensively used for radiation detection and measurement. The short term measurement may not result in an accurate value, but the long-term radon measurement with the help of an active technique is expensive than the passive technique. Mainly two types of SSNTDs are used in the measurement of radon parameter.

1. CR-39 detector

The CR-39 detector is one of the most versatile, sensitive, and widely used SSNTD nowadays. It has been recognized by Cartwright and his colleagues as a track detector since 1978 [9]. CR-39 is an abbreviation of Columbia Resin-1939 [10]. It is an amorphous polymer consisting of short poly-allyl chains joined by links containing carbonates and diethylene glycol groups [11] into a dense three-dimensional network with an initiating monomer unit [12]

2. LR-115 type-II plastic track detector

The LR-115 type-II detector is used for the investigation in the present work.

3.3.1.1 LR-115 Detector

LR-115 type-II plastic detectors are manufactured by Kodak. It is a two-layered detector which consists of a red-coloured cellulose nitrate CN $(C_6H_{10}O_5)_n$ layer and an about 100μm thick polyester support as shown in figure 3.2. These detectors have been used to detect alpha particles all over the world for integrated measurements as it is alpha sensitive.

The thickness of the sensitive layer of the detector, LR-115 films (Type II) is 12 μm. The tracks of α-particles, in the energy range of 1.9–4.2 MeV, can be revealed through etched holes [13]. The recommended track etchant is usually 6N NaOH, at

60°C, to reduce the thickness from about 12 µm to 7–8 µm. If the track is etched completely through the sensitive CN layer, the track appears as bright holes Thus the etched through tracks (holes) can also be evaluated with jumping spark counter [14]. Etching makes track counting easier. Kodak LR-115, type II films are of two types, pelleculable and non-pelleculable. For pelleculable films, spark counter can be used to count the tracks, which is less time-consuming. For non-pelleculable films, the counting of tracks has to be carried out using an optical microscope. These detectors are a very useful and economical for radon measurement.

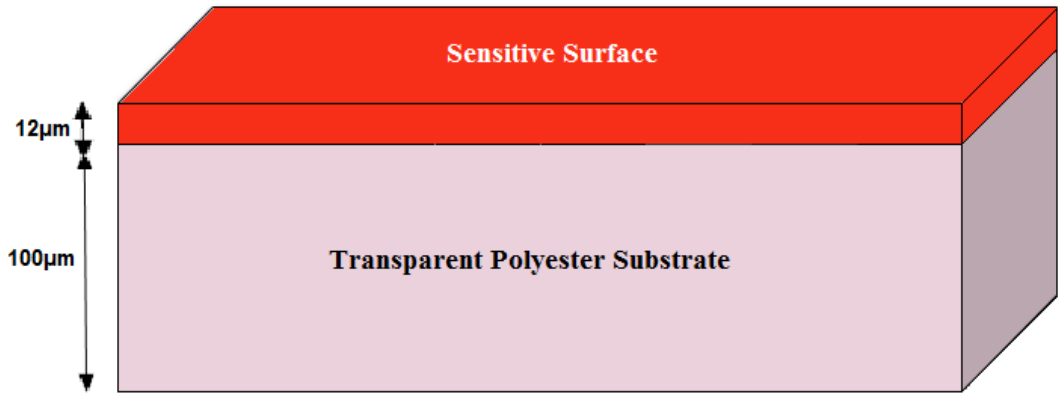

Figure 3.2: LR-115 type-II SSNTD

3.3.1.2 Advantages of SSNTDs

SSNTDs are preferred over other detectors because:

(i) These are cost effective and are available in varying sizes.
(ii) They are quite suitable for personal dosimetry in the environment and in congested surroundings.
(iii) Their fabrication is very simple compared to other track detectors like nuclear emulsions, bubble, spark and cloud chambers [15].
(iv) The tracks formed are permanent and are stable under severe environmental conditions like mechanical vibrations, humidity and pressure but can be annealed thermally [16].

(v) These detectors are highly robust and do not affect health when a human comes in contact with them.

(vi) The angular measurements can be carried out as these detectors are very flexible.

(vii) The major advantage is that these detectors do not require sophisticated electronics instruments.

3.3.1.3 Formation of Particle Tracks

Track formation in solids depends mainly upon the rate of energy loss (dE/dx) of the track forming particles. When a charged particle travels through solid, depending upon the velocity and charge of the particle, it loses its energy [17]. A low energy particle loses its energy by collision process and this lost energy is confined around the particle trajectory. In case of high energy particle, the particle loses its energy by the production of high energy electrons and these electrons increases the ionization and energy are deposited in the extended larger volume [18]. This ionizing process creates free radicals and other charges. These generated radicals and charges create damaged sections on radon sensitive material which are called latent tracks [19-21]. Each solid have a critical rate of energy loss, a particle produces no tracks if the lost energy per unit length is below this critical rate of energy loss. The value of critical energy loss is different for different solids; but in a given material, its value is uniform for all particles.

The particles can lose energy in different ways when they pass through a solid. There are several models which explain the track formation mechanism in SSNTDs, predicting different modes of energy loss [22-24].

Different models to predict track formation are:

(i) Direct atomic displacement model

(ii) Thermal spike model

(iii) Total energy loss model

(iv) Confined / Restricted energy loss model

(v) Primary energy loss model

(vi) Secondary energy loss model

(vii) Primary ionisation model or ion explosion model.

The primary ionisation model or ion explosion spike model are considered a satisfactory model that describes the various conditions to be fulfilled for track

formation by ions. This model is based on the postulate that the particle first loses energy to the electrons of the atoms in its path in the solid. The energized electrons leave the region as positive ions and form a latent track.

The formation of track formation is carried out in the following two steps:

(a) The defects creation

(b) The defect relaxation

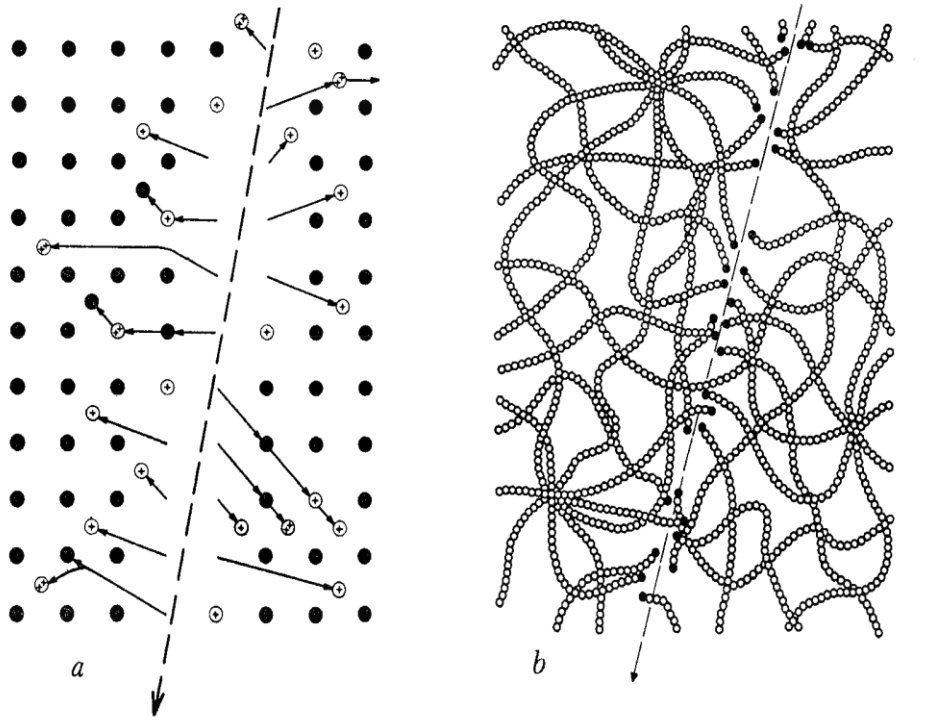

Figure 3.3: Tracks characteristics of in (a) Crystalline solid (b) Polymeric Solid [20]

3.3.1.4 Restriction on Track Formation

The formation of track recording can be restricted under the following two conditions: The first condition is related to the availability of electrons in the proximity of the ionized track [25]. The track will not appear if already available electrons are present to exchange the electrons which are released by highly energetic charged particles. In-advert track formation can be reduced by the expulsion of electrons available in the cylindrical region around the ionized track. This process should be completed in less than 10 seconds, as the electrons needed 10 seconds to displace the ions from their site [26]. In case of metal, electron takes even less than 10 seconds to replace the ejected electron, so no track would result in metal, where as in case of insulators, the electrons cannot replace the ejected electrons in time less than 10 seconds, hence tracks would result.

The second condition depends on the mobility of holes. High holes concentration is found along an ionized region in the tracks, which may result in away movement of holes and hence may hinder the permanent formation of the track. It is reported that when the hole mobility is more than 102 cm^2/V-sec, the track will not appear in materials [27]; hence metals and many semiconductors are not considered as track recording materials. The insulators are considered as good track recording materials because of less hole mobility (102 cm^2/V-sec).

3.3.2 General Aspect of Track Etching

The radiation damages are carried out by highly charged ionized particles. These damages are not only etched in mica but also in many other insulating solids such as glasses, crystals, inorganic minerals as well as in plastics or organic polymers [28] [1]. When chemical etching solutions such as 2.5 N NaOH/KOH are used, the damaged region can also be etched. A bigger track is formed when the etching solution reacts with the damaged region. This track can be seen using an optical microscope [29].

3.3.2.1 Chemical Etching

This process requires a water bath which maintains a temperature in the range of 60°C. The 2.5 N aqueous NaOH/ KOH solutions are used as the chemical etchant which means an approximately 250g solution of NaOH is added to distilled water. The etching is carried out for about 90 minutes. The size of the damaged track varies when the etching time, etching temperature or molarity of the etchant changes [30].

During the etching process, the detectors are suspended using wires at a fixed distance. The detectors are dipped into the etchant inside the beakers which are placed into constant temperature bath. The mouth of the beaker is kept covered to stop any foreign impurity to enter into the solution besides to avoid evaporation of the water used in etchants. After the completion of etching time, the detectors are taken out from the chemical etchant and washed in fresh water, afterward these detectors are kept for drying. The tracks in the detectors which are enlarged after the chemical etching process can be easily counted by using an optical microscope.

3.3.2.2 Etching Apparatus

The most general method for observation of the particle tracks in solids is subjecting the film to chemical etching. In chemical etching, the damaged tracks due to radiation are enlarged with suitable chemical reagent [31-33].

The apparatus used for etching is constant temperature water bath as shown in figure 3.4. A 2.5 N NaOH solution for etching is prepared by using distilled water. Then this solution is placed in the water bath. The water bath has a digital temperature controller. When the desired temperature is achieved, then samples are properly dipped in the solution. The samples are marked properly so that they are not mixed during etching. The beaker with etching solution is kept covered with lids during the time of etching so that there is no change in concentration of solution due to evaporation. After etching time (90 minutes), the samples are taken out from the water bath and washed first with distilled water and then in running water for 20-30 minutes. Then after washing, the films are peeled off from the base and stored for counting in paper envelopes with care so that these are not mixed.

Figure 3.4: Constant etching bath tub

3.3.2.3 Etching Conditions

The conditions that should be followed for the better etching results are:

(a) Bulk etching is necessary to enlarge the tracks to optically visible size. The etchant should be so chosen that it slowly dissolve away the material at a constant rate [34].

(b) The etching rate, which is controlled by the temperature and concentration of the etchant, can be varied by changing the concentration or by adding other chemicals to the etchants [35].

(c) It is important that the uniformity of the temperature of the etchant is maintained. Stirring the etchant during etching ensures uniformity of temperature [36].

(d) The surface to be detected should be optically smooth so that there is no confusion between sub-microscopic cracks or scratches and tracks [36].

3.3.3 Tracks Counting Techniques

The information is stored in detectors in the form of damaged tracks and it is important required that this information should be retained accurately without much complexity [37-39]. The track counting process may be carried out using a high power microscope for accuracy or using a spark counter for the prompt operation of the counting process while using appropriate LR-115 film.

There are many reported techniques in the literature for tracks counting in SSNTDs. The optical microscope and spark counter technique is used in the current work.

3.3.3.1 Track Counting using Optical Microscope

After etching, for counting the tracks samples are usually scanned with an optical microscope as shown in figure 3.5 at ordinary magnification. For normal or near normal incidence the microscope can be focussed on the surface of the detector, where the intersection of the track with the surface is seen as a dark circular spot [40-41]. The depth of the track can be analysed by changing the focus. In the case of oblique incidence, the tracks can be seen by changing the focus of the microscope. Area of etched detector surface is scanned to find the track density or a total number of tracks by using an eyepiece, equipped with a grid marked graticule [42]. The view

of the field is calibrated with the help of a stage micrometer glass slide to find out the scanned area. Figure 3.6 shows the tracks created by α-particles on the LR-115 detector when seen from the optical microscope.

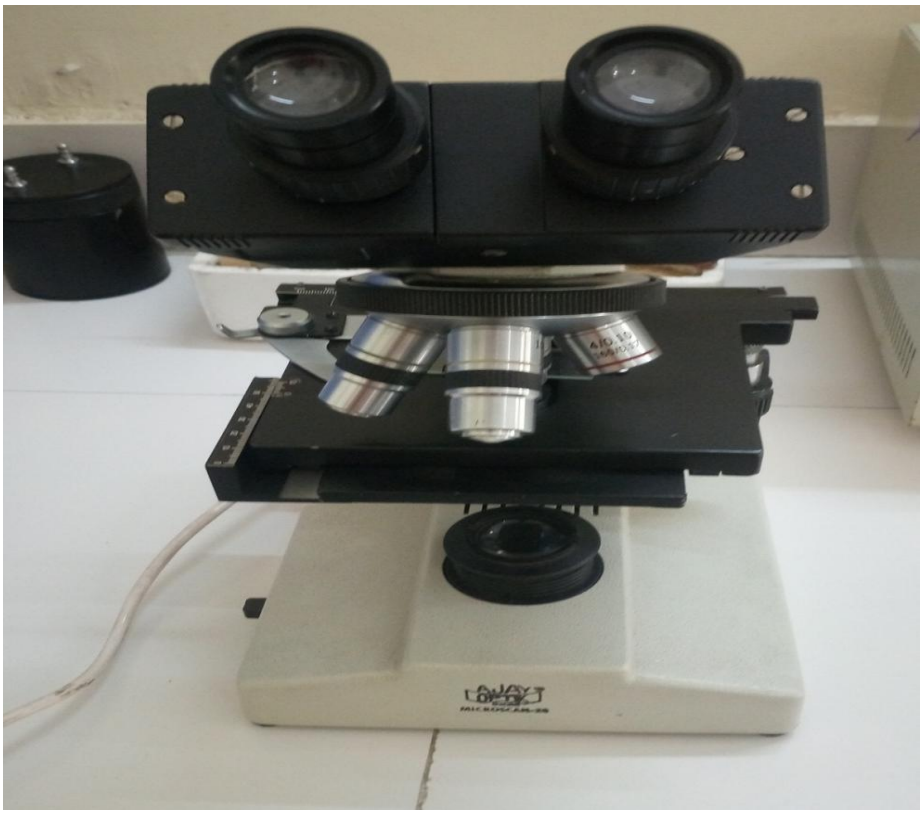

Figure 3.5: Optical Microscope

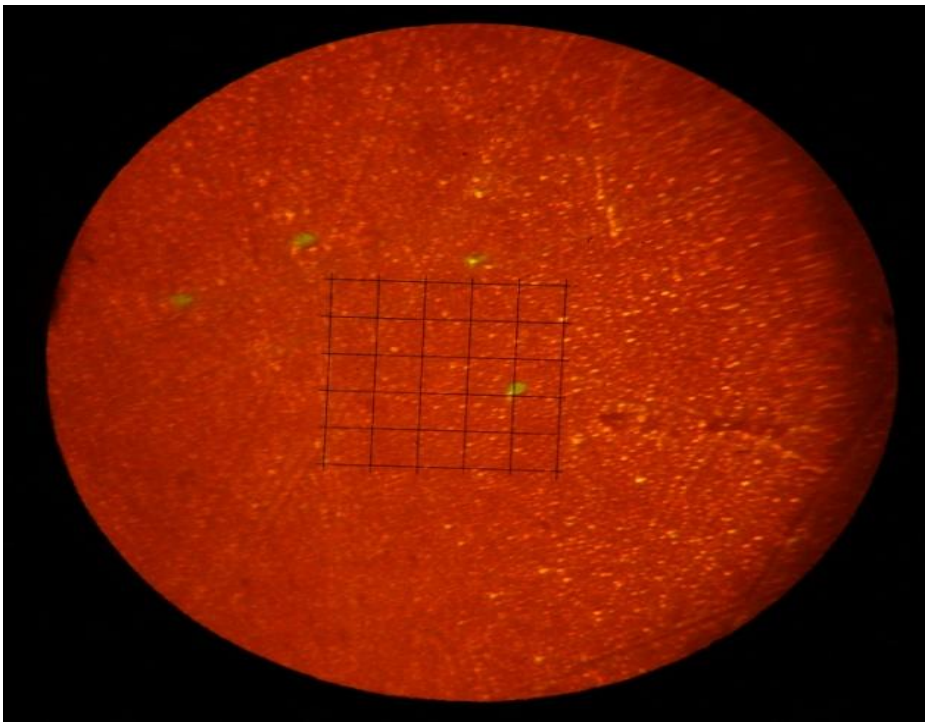

Figure 3.6: Track produced by alpha particles as seen from Microscope

3.3.3.2 Track Counting using Spark Counter

The spark counting technique is the most widely used technique for counting the large amount of tracks formed by radon on plastic track detectors. A spark counter shown in fig 3.7 counts the tracks on these detectors. The spark counter has a counts display, spark head, wave shaping circuits, and adjustable HV circuit. The sensitive red top layer of LR-115 detectors is used counting the alpha particles tracks [43-45]. The process of track counting begins with the placement of a thinly peeled detector between the two electrodes of the spark counter [46]. The two electrodes are covered with a thin aluminium sheet on the mylar layer.

The plastic foil is kept in contact with the very thin detector from the aluminium side. Mostly brass made thick electrodes are used in spark counter. The plastic foil is covered with weight so that there is firm contact between the electrode and thin detector [47]. The block diagram for spark counter set up is shown in figure 3.8. When a high voltage is applied across capacitor C, the spark is generated through a track hole. The voltage used for this purpose is also recorded in the system.

Figure 3.7: Spark Counter

The holes were created when the chemical etching of the track detector was carried out. During the process of counting, the spark which passes through the created hole has sufficient energy to evaporate thin aluminium coating and generate a bigger hole in aluminium electrode [48]. Due to the creation of holes, the electrode comes in

contact with each other and hence cannot produce the spark again. The process of spark generation is hence stopped and the capacitor is again fully charged. This process is repeated again for another hole and the process is continued until all the holes are counted. The number of sparks is equal to evaporated spots of size 100 μm or higher on aluminium, which was equal to holes in plastic track detectors [49]. The holes on aluminium can easily be counted by microfiche reader or optical microscope.

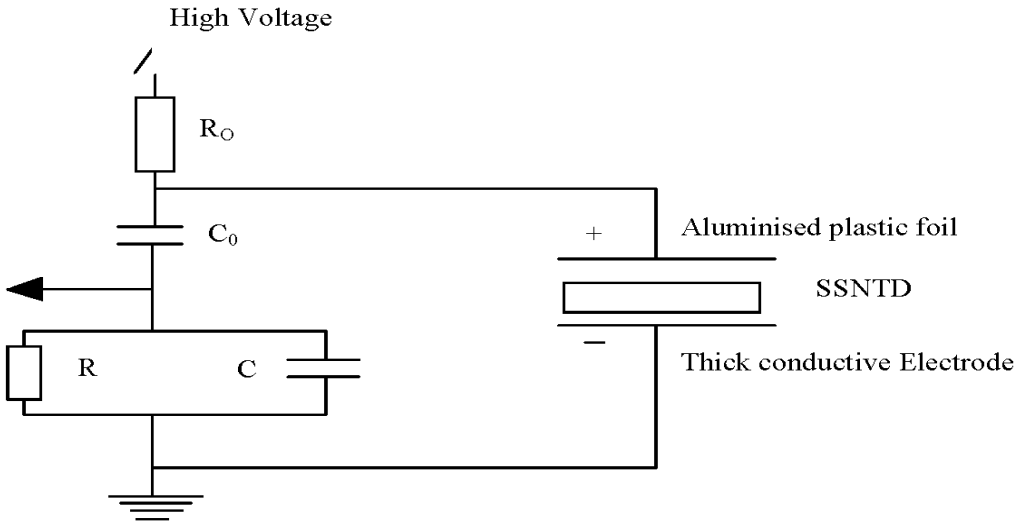

Figure 3.8: A schematic diagram of the Spark Counter

3.3.3.3 Spark Counter Characteristics

Operating voltage of a spark counter is that specific voltage at which the counting of tracks should be done. When the graph is plotted between applied voltages and counts, a plateau region is produced. The corresponding middle voltage of this plateau is taken as the operating voltage for that spark counter. This plateau region shows that even with a small change in the applied voltage, the number of tracks count by the counter remains constant [50]. Hence, with the fluctuation of the applied voltage, the counts will remain the same and equal to the number of holes in track detectors.

3.3.3.4 Plateau Characteristics of Spark Counter

The voltage at the centre of the plateau region, i.e. 480 V, was selected as the operating voltage. The detector film is kept on spark counter head for the counting of tracks. The mylar film is cut in size such that it extends to the circular electrode and

reaches up to another electrode. Its aluminized side faces the film [51-54]. A cylindrical weight is kept above the electrodes to press the contact which is spring loaded, then about 900V is applied between electrodes. The pre-sparking process is initiated by pressing the on switch available on the spark counter; with this, the counting of tracks is started. The pre-sparking step is necessary to clear improperly developed holes during etching. When the counting stops, as indicated by an LED on the front panel, the mylar film is changed with a new mylar film without any disturbance to the detector film. Now the voltage across the electrodes is set to operate voltage i.e. 480 V and again the counting is started [55]. At the end of counting, the counts obtained are noted down and the process is carried out again with new mylar each time. The average of three readings is taken of the final count. Usually, the central area of the film is used for counting [56-57].

The spark counter works on the basis that slight energy is dissipated when the spark dashes. It fuses the walls of the track in the detector, the small portion of mylar films also get heated up which result in local sublimation of the aluminium. This is done to reduce the inadvertently recounting the same track but it may also result in electro-isolation of adjacent track [58]. The mylar films record the sublimated aluminium spots.

The effect of spark energy is evident on the sublimated area and its area varies according to applied electric field and capability of the spark counter and film.

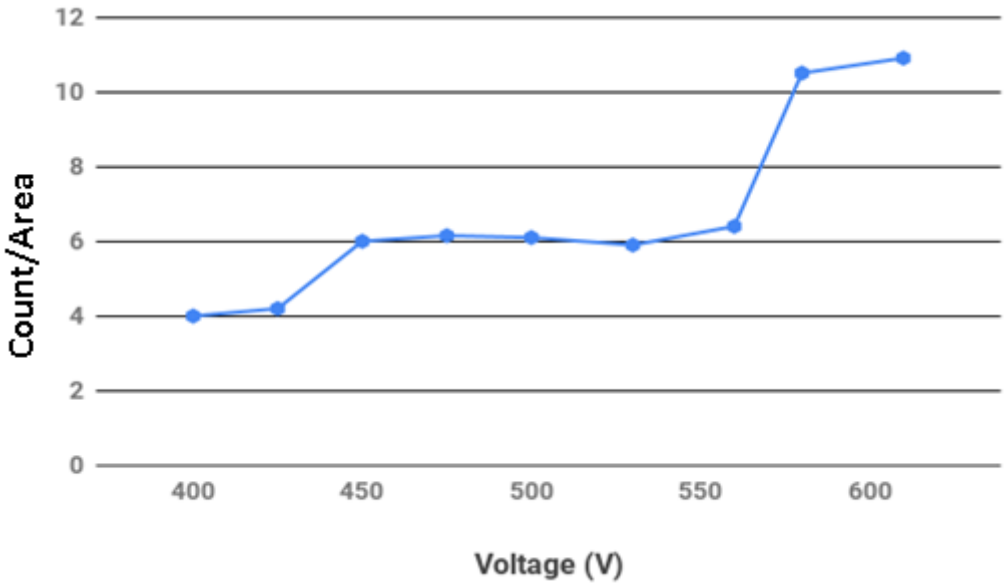

Figure 3.9: The graph between applied voltage and counts.

3.4 CAN TECHNIQUE METHODOLOGY

In this technique, samples are first dried in an oven to remove moisture content, and then, these samples are sieved through a sieve to get uniform grain size. The measurement starts with taking equal amounts of respective samples and placing them in different cans.

The CAN used in the experiment has a diameter of 8.1cm and a height of 11.3 cm. An LR-115 type-II detector is affixed on the inner side of the CAN using transparent cello tape [59-62]. The size of the detector is kept 2cm x 2cm. The track detector is affixed in such a manner that its sensitive side faces the sample as shown in figure 3.10 so that alpha particles which result by the decay of radioactive material are recorded in it. The radon and its progeny take about 7 days to reach the equilibrium state. The activity of exhalation of radon can be calculated from the dimensions of the CAN and the exposure time.

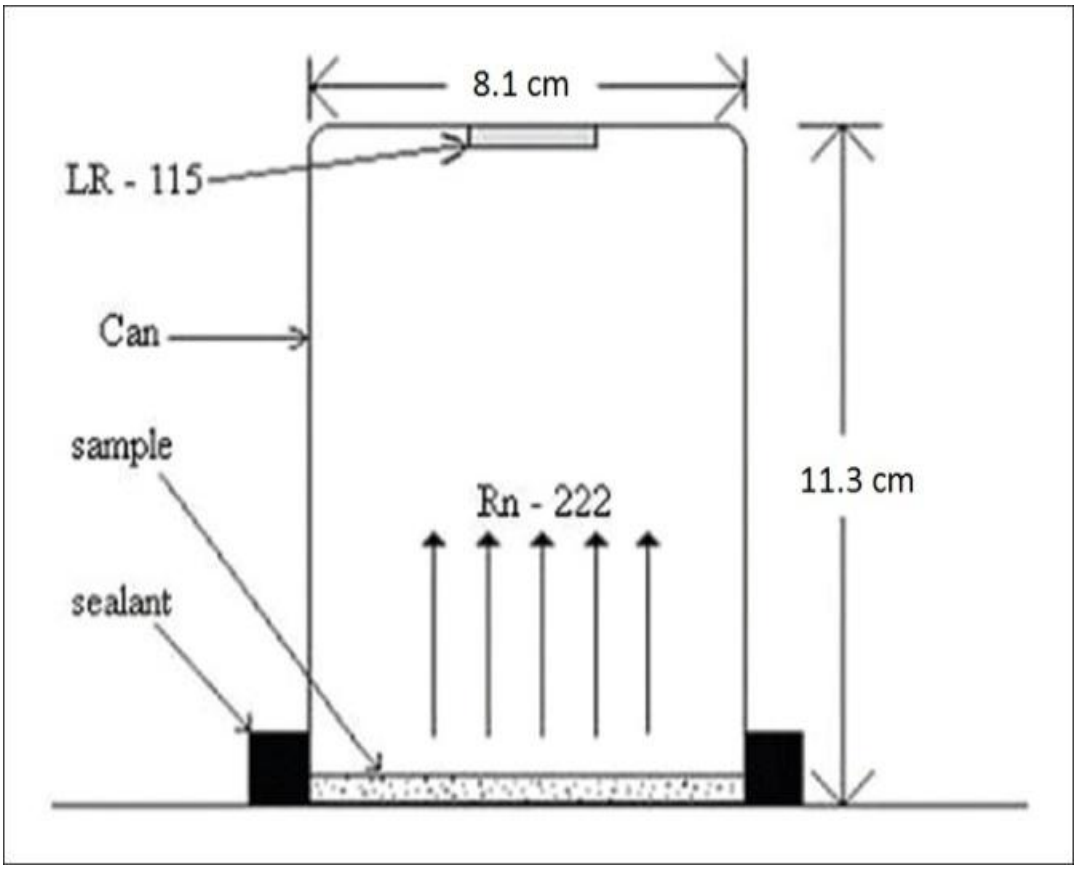

Figure 3.10: Arrangement for measurement of radon exhalation rate using CAN technique

The sample is kept for exposure for 100 days and after the exposure time is over, the detectors from different samples are detached from the CAN lid. Afterward, the etching process of the detector is carried out this for 90 minutes. A 2.5N NaOH solution is used for etching at 60 ºC, the process is carried out in constant temperature bath which is maintained at 60°C ± 0.5°C [63]. After this process of etching, the enlarged alpha tracks can be seen and counted using an optical microscope having a magnification of 400x or higher. The counting of alpha tracks can also be done using spark counter depending upon the type of LR-115 films used.

3.5 GAMMA SPECTROSCOPY

Gamma spectrometry is a very robust process for the detection of natural radionuclides present in diverse solid samples. In gamma spectroscopy, the gamma spectrum of the required material is seen on PC by using a Nal(Tl) or HPGe detector. The detector is bound to be a multichannel pulse height analyzer. Thus the technique offers a constructive multi-component analysis. This analysis is widely used for environmental samples. In the present work, the gamma activity of the sample was measured by using the high-resolution gamma spectrometry system using an HPGe detector situated at Inter University Accelerate Centre (IUAC), New Delhi. An n-type coaxial Germanium of very high impurity is used as a detector. The detector has specific resolution of 2.0 keV and for 1.332MeV gamma energy of Co-60, its efficiency is 20% [64-67]. The output is taken on the computer through a 4K MCA system. To reduce the background level, the detector is sealed using 4″ lead from all over [68].

In this process, the natural radioactive samples are first crushed to a fine powder using pestle and mortar for obtaining natural radioactivity. Then scientific grade sieve of 150-micron mesh size is used to obtain a fine powder. The samples are dried by keeping in the oven at 110º C (383K) for 24 hours. Afterward, each sample is left for 4 weeks in PVC container so that equilibrium is established between radon and its progeny. Approximately, 300 gm of the sample is required for the measurement process. An HPGe detector, a high-resolution gamma spectrometric system is used for finding the radioactivity of the given sample [69]. The employed detector is a highly pure n-type Germanium detector (made by EG & G, OR TEC, Oak Ridge, US). The

indigenous developed “CANDLE” (Collection and Analysis of Nuclear data using Linux network) software of IUAC (Inter University Accelerator Centre, New Delhi) was used for spectrum analysis. Figure 3.11 shows the physical setup of the gamma-ray spectrometer.

Figure 3.11: Gamma Ray Spectrometer (ORTEC)

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CHAPTER-IV

MEASUREMENT OF RADON EXHALATION RATE USING CAN TECHNIQUE

4.1 INTRODUCTION

Radon and its progeny are mainly liberated from Naturally Occurring Radioactive Materials (NORMs) like uranium, which is found in soil, building material and rocks. The average global content of uranium in soil and earth crust is approximately 4 ppm [1]. The uranium in its radiation series decays to radon, which further generates radon progenies, hence radon is also found wherever the uranium is present i.e. soil, earth's crust etc. The content of radon and its progeny vary from material to material and place to place. The human may receive radon from the air or through food chains. Since radon and other radioactive materials may emit alpha particles, which can damage various organs of the human body and immunity system [2]. These radiations may cause to reduce production essential red blood cells (RBCs), which can result in further serious impacts and life-threatening effects on the human body. The larger presence of radioactive materials results in a significant concentration of radon at that specified place. Hence the usages of buildings with a high concentration of radioactive materials will increase the presence of radon inside the building [3]. Thus, the estimation of radon concentration in different samples is an important parameter which needs a detailed study.

The estimation of radon concentration in various samples of soil, rocks and food items is carried out around the globe by various researchers [4-6] and many official reports by various environmental agencies [7-9] are available in this regard. However, only a few researchers [10-12] have reported work on subtropical regions of northern India.

The soil is considered as the major source of radon emanation, the exhalation of radon from the soil depends upon various factors such as presence of radioactive elements in soil, geographical locations, grain size of soil, moisture in soil, environment above soil, atmospheric pressure and vegetation over and around the soil samples [13-16]. The study of radon exhalation from soil samples is important so as

to ascertain public radiation dose and keeping reference data to study the variation in environmental radiation due to industrialization and other radiation activities due to human intervention.

As soil is omnipresent and humans come in contact with soil daily, hence the experiment was carried out to measure the radon mass exhalation and radon surface exhalation rate from the soil samples collected from four districts of Haryana viz. Palwal, Faridabad, Gurugram, and Mewat. These geographical locations are situated in southern Haryana in India. To our best of knowledge, there is no previous data available for radon exhalation rate for soil sample of these geographical locations.

4.2 MATERIAL AND METHODS

4.2.1 Sample Collection Area

The sample collection areas are situated in Haryana, state of India, which is situated in India's northwest between 27º39` to 30º35` northern latitude and 74º28` to 77º36` eastern latitude. The plane is fertile and the height above the sea is 700-900 feet [17], the slope is from north to south. The climate of the area is of pronounced character, it is very hot in summer and markedly cold in winter. The temperature varies in the range of 46ºC in summer and 2ºC in winter. The Haryana state has a total area of 44212 km^2 and ranked 21^{st} in terms of area in India [18]. It has a population of 253.5 lakh (Census 2011) with Faridabad as a most populous city [18]. The world's oldest and largest ancient civilization, Indus valley civilization existed here about 9000 years back [19].

The river Yamuna tributary of Ganga flows along the eastern boundary of the state. Ghaggar river and Sutlej river in the north and Sahibi river in the southeast are the famous rivers in this area. Since the state is situated towards the depression of river Yamuna and Indus, the soil is made up of alluvium entirely [20]. It has a vast ground of moist land. The alluvium contains clay, sand, slits and hard calcareous concentration. In southwestern part, due to windblown sand, sand piled up to form sand dunes of few meters in height and significant kilometers in area.

Faridabad, Gurugram, Palwal and Mewat districts of the state of Haryana in the southern part of the state are situated at 28º26` N and 77º19` E, 28º27`8` N and 77º1` E, 28º14`8` N and 77º 33`0` E, 28º1`6`` N and 77º5`4`` E longitude respectively. These districts face Aravali range from the southern part, the geographical location of sample collection sites is shown in figure 4.1.

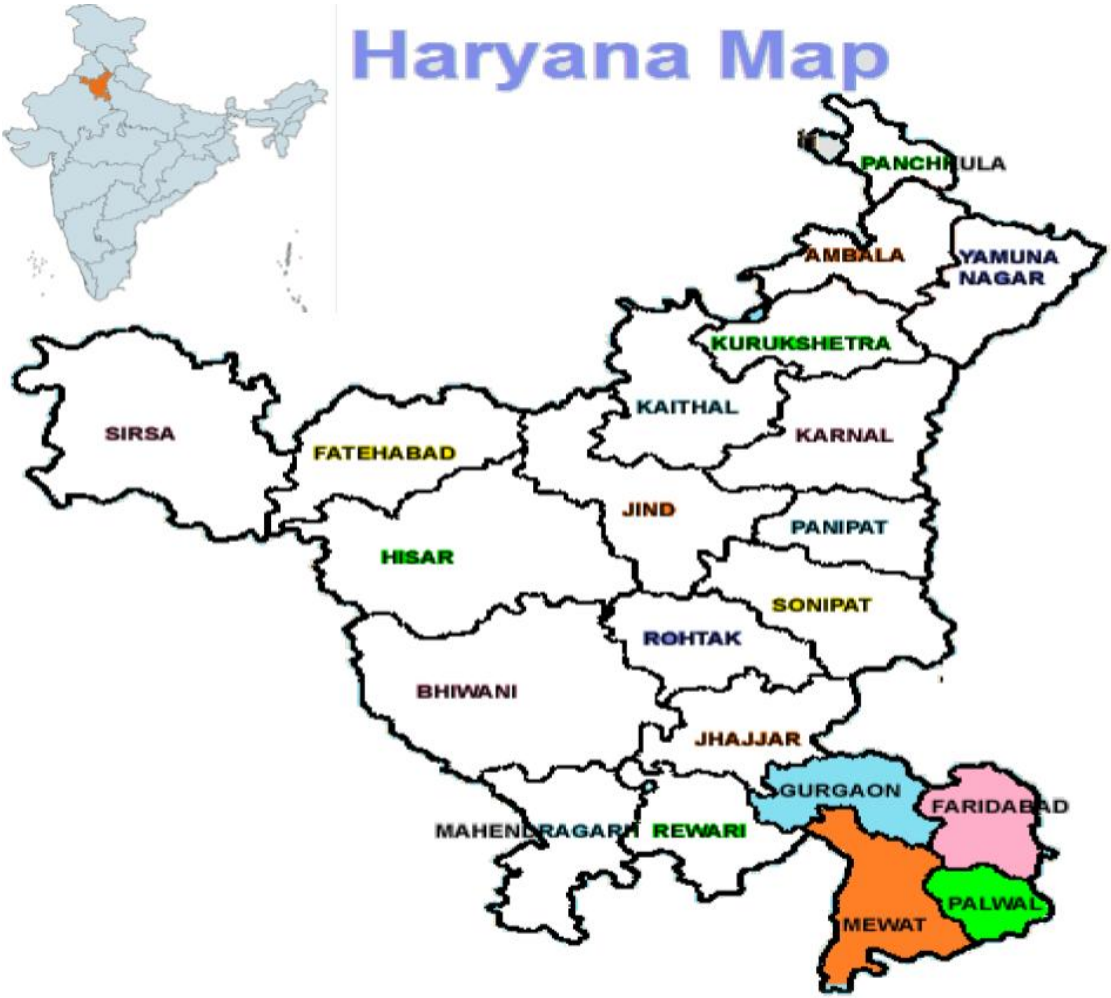

Figure 4.1: Geographical location of sample collection sites (shown in colour) [20]

4.2.2 Soil Samples Processing and Experimental Detail

Alter and Price in 1972, first proposed "CAN Technique" to study the radon exhalation rate and radium concentration. It was modified to sealed CAN technique by various researchers [21-25]. This method is also called an alpha track method [26]. Initially, CAN with the sample are sealed and left for one month so that it will

acquire an equilibrium concentration [27] of radon and its decay products. Once the equilibrium is established, the nuclear track detector (2x2 cm) [28] is fixed in the inner side of the CAN's lid and sealed again. At the end of exposure time, detectors were removed from the CAN. The detectors record the tracks produced by alpha particles. These alpha particles are generated due to the decay of radon gas and its progeny.

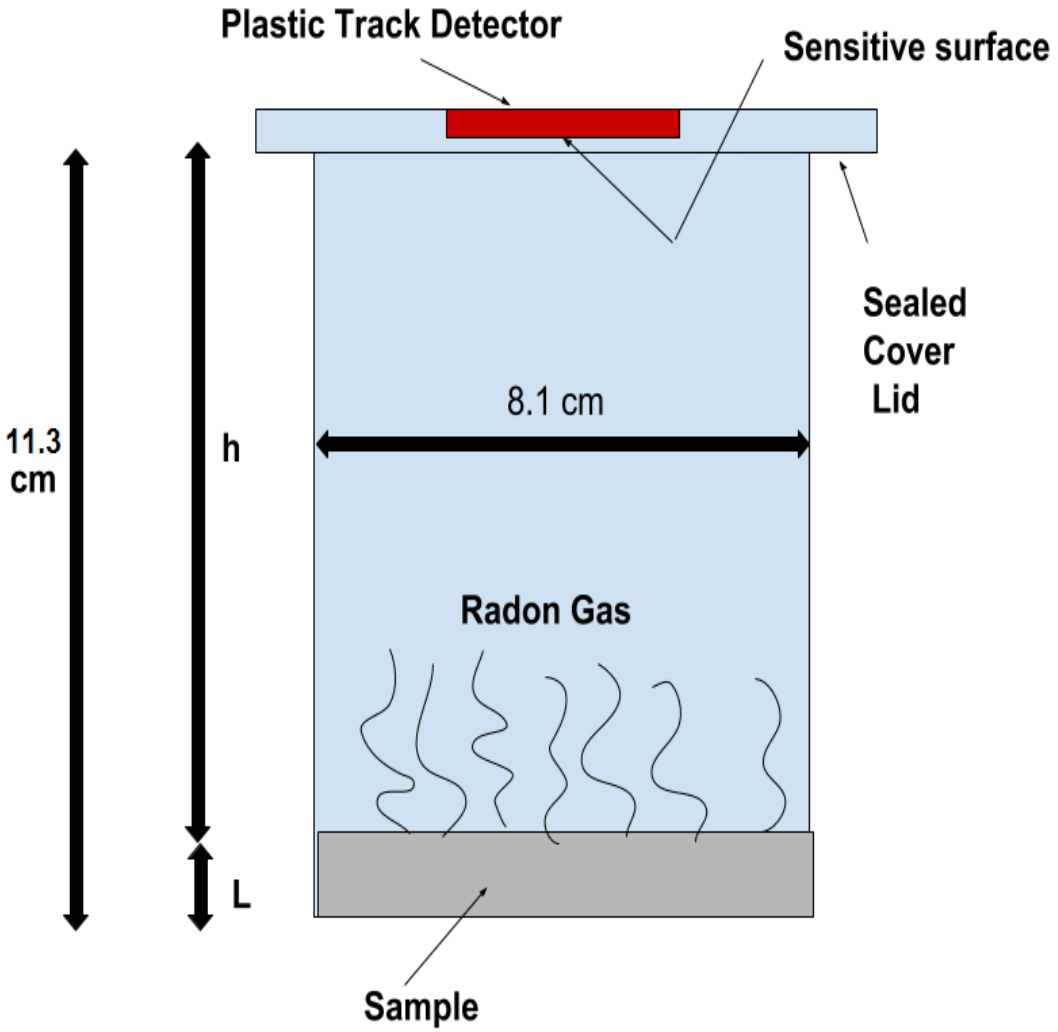

Figure 4.2: CAN as used in CAN Techniques

In this work, the etching process for these nuclear track detectors (LR-115 type-II) was carried out in the constant temperature bathtub maintained at the temperature of 60ºC, where 2.5 N NaOH solution was used for 90 minutes for etching. This etching process results in the production of holes in these nuclear track detectors at alpha

tracks. These holes were counted using a microscope of a magnification of 100X or by using spark counter [29].

4.3 THEORETICAL BACKGROUND OF PROCESSING

Radon (^{222}Rn) has the half-life of 3.82 days, whereas the parent entity Radium (^{226}Ra) has the half-life of 1620 years [4]. The sample is initially kept in a closed container to achieve equilibrium as it is considered that in a period of 30 days the effective equilibrium about 98% [21] for radon decay series will be achieved. The plastic track detectors LR-115 types II are used and they behave as bare detectors.

The relation between track density ρ (track cm^{-2}), exposure time T (seconds) and activity concentration of radon C_{Rn} ($Bq.cm^{-3}$) is given by the equation [30]:

$$\rho = \mathcal{K}. C_{Rn}. T \qquad (4.1)$$

Where $\mathcal{K}$ is the constant of proportionality and known as a sensitivity factor of LR-115 detector having ±15% tolerance [31]. This sensitivity factor [$\frac{1}{30}$ tracks cm^{-2} s^{-1}(Bq cm^{-3}]$^{-1}$ is given by:

$$\mathcal{K} = \frac{1}{4}(R_{max}-R_{min}) \cos^2 \theta_C \qquad (4.2)$$

Where $R_{max,}$ R_{min} are alpha particles range having energy 4.2 MeV and 1.9 MeV respectively as alpha particles require energy in the range of 1.9-4.2 MeV [32] to produce alpha tracks on LR-115. The air present on the surface acts as a source. The θ_C is the average value of critical angle for the plastic track detectors (40º±5º) [31].

Range-energy formula for a-particles is given by [33]

$$R_{air} \text{ (in cm)} = 0.322 (E_\alpha \text{ in MeV})^{1.5} \qquad (4.3)$$

Keeping average critical angle θ_C =40°, The value of sensitivity factor for the plastic track detector calculates to be 0.024 tracks $cm^{-2}d^{-1}$(Bq m^{-3})$^{-1}$.

From equation (4.1) unit of K should be in Track.cm^{-2} $s^{-1}(Bq\ cm^{-3})^{-1}$ and from equation (4.2) unit of K should be in length and can be easily converted into cm as shown below:

$$\text{Track } cm^{-2}\ s^{-1}(Bq\ cm^{-3})^{-1} = \frac{\text{Number}}{cm^2} \times \frac{1}{s} \times \left[\frac{\text{Number}}{s \times cm^{-3}}\right]^{-1}$$

$$= \frac{\text{Number}}{cm^2} \times \frac{1}{s} \times \left[\frac{s \times cm^{-3}}{\text{Number}}\right]^{-1}$$

$$= cm$$

By using equation (4.3), values of R_{max} and R_{min} are calculated to be 2.77cm and 0.84cm respectively.

Taking all these values R_{max}, R_{min} and θ_C the value of K can be calculated as

$$K = \frac{1}{4}(R_{max}-R_{min})\cos^2\theta_C$$

$$= \frac{1}{4}(2.77-0.84)\cos^2(40)$$

$$= 0.283 \text{ cm}$$

Track $(cm^{-2}\ s^{-1}\ (Bq\ cm^{-3})^{-1}) = 1$ cm

Converting the unit of Track $(cm^{-2}s^{-1}\ (Bq\ cm^{-3})^{-1})$ where time is in days.

$$1 \text{ track } (cm^{-2}s^{-1}\ (Bq\ cm^{-3})^{-1}) = 1 \text{ track } cm^{-2}\left(\frac{1d}{24 \times 3600}\right) - 1\ [\ Bq\ (10^{-2}\ m)^{-3}\ ]^{-1}$$

$$= \frac{1 \times 24 \times 3600}{10-6} \text{ track } [cm^{-2}d^{-1}\ (Bq.cm^{-3})^{-1}]$$

$$= 0.0864 \text{ track } [cm^{-2}d^{-1}\ (Bq\ cm^{-3})^{-1}]$$

Thus the value of K, which was 0.283 cm, will now have the value

$$K = 0.283 \times 0.0864 \text{ track } [cm^{-2}d^{-1}\ (Bq\ cm^{-3})^{-1}]$$

$K = 0.0245$ track $[cm^{-2}d^{-1}(Bq\ cm^{-3})^{-1}]$

The value of radon activity concentration C_{Rn} (Bqm^{-3}) can be obtained from equation

$$C = \frac{\rho}{KT} \quad (4.4)$$

$K = 0.0245$ track $[cm^{-2}d^{-1}(Bq\ cm^{-3})^{-1}]$

T = total time of exposure in days.

ρ = background corrected track density $(track.cm^{-2})$

After closing the CAN, radon activity concentration increases and given by relation [34]

$$C_{Rn} = C_{Ra}(1-e^{-T.\lambda_{Rn}}) \quad (4.5)$$

Here C_{Ra} is effective radium concentration in the sample. The number of alpha disintegrations in unit volume of can in time T and with sensitivity factor as K the track density is given by

$$\rho = KC_{Ra}T_e \quad (4.6)$$

$$\rho = KC_{Ra}[T-\lambda(1-e^{-\lambda T})] \quad (4.7)$$

T_e is the effective exposure time and given by

$$T_e = [T-\lambda_{Rn}^{-1}(1-e^{-\lambda_{Rn}T})] \quad (4.7)$$

The sample is kept for exposure to detector for 100 days hence T_e = 100 days

The effective radium content can be calculated as [35]

$$C_{Ra}\ (Bq.kg^{-1}) = \left(\frac{\rho}{KT_e}\right)\left(\frac{hA}{M}\right) \quad (4.8)$$

h is the spacing between the top surface of the sample and detector affixed on the lid of the CAN, A is the effective cross-sectional area of the CAN in m^2 and M is the mass of solid sample in Kg.

The mass exhalation rate of the sample for radon release can be obtained as

$$C_{Rn}\,(Bq\ kg^{-1}d^{-1}) = C_{Ra} \times \frac{\lambda_{Ra}}{\lambda_{Rn}} \times \frac{1}{T_e} \qquad (4.9)$$

Similarly surface exhalation rate for radon release is given by

$$C_{Rn}\,(Bqm^{-2}d^{-1}) = C_{Ra} \times \frac{\lambda_{Ra}}{\lambda_{Rn}} \times \frac{M}{A.T_e} \qquad (4.10)$$

4.4 RADON EXHALATION STUDIES IN SOIL SAMPLES

As stated earlier, radon gas, which is naturally occurring gas generated due to the decay of radium is omnipresent, it is found in rocks, building materials, soil etc. It has a half-life of 3.82 days and hence can reach to the human environment through migration from the soil, which is a major source of radium content. The effects of radon on human health necessitate the study of radon exhalation from the soil. The present study was conducted to measure the radon exhalation rate using CAN technique and tracks were counted using spark counter. The soil samples were collected from the four different districts namely Faridabad, Gurugram, Palwal and Mewat of the southern part of Haryana state.

4.4.1 Result and discussion

In the experiment to measure the exhalation rate using CAN, samples were collected from different locations and 150gm content of each sample was placed in different cans. Five samples of soil were collected from a location and four such locations were selected in Faridabad district, four locations in Palwal District and Gurugram District each and three locations were selected in Mewat District.

Table 4.1: Radon exhalation rate and radium concentration in soil samples

S. No.	Location	Sample code	Radon exhalation rates		Radium Concentration Ra ($Bq.kg^{-1}$)
			EA (Bq. $m^{-2} h^{-1}$) x 10^{-3}	EM (Bq. $kg^{-1}h^{-1}$) x 10^{-3}	
1	Faridabad	Soil-1	643.43±11.8	24.3±0.3	14.9±0.1
2		Soil-2	468.69±18.8	16.9±0.2	10.4±0.2
3		Soil-3	375.21±15.1	13.9±0.4	8.9±0.2
4		Soil-4	401.63±14.8	15.8±0.2	9.8±0.1
5	Palwal	Soil-5	546.32±9.8	22.4±0.1	13.4±0.1
6		Soil-6	643.42±7.1	23.7±02	14.6±0.3
7		Soil-7	509.54±7.1	19.9±0.3	12.3±0.4
8		Soil-8	469.76±8.9	19.3±0.3	11.8±0.2
9	Mewat	Soil-9	499.83±5.6	19.8±0.4	11.9±0.1
10		Soil-10	434.63±8.9	16.2±0.3	10.2±0.2
11		Soil-11	384.25±4.7	15.1±0.4	9.6±0.2
12	Gurugram	Soil-12	363.3±15.8	13.4±0.2	8.8±0.3
13		Soil-13	516.15±21.7	20.3±0.2	12.7±0.2
14		Soil-14	678.5±19.4	28.6±0.1	15.7±0.3
15		Soil-15	568.49±13.4	22.7±0.5	13.6±0.5
Minimum			363.3	13.4	8.8
Maximum			678.5	28.6	15.7
Average			500.21	19.4	11.9

Table 4.1 shows the values of radon mass exhalation rate, surface exhalation rate and radium concentration in soil samples collected from different sites of a district of Haryana state. The study was carried out to measure the radon exhalation rate and

radium concentration from the specific soil samples. The value shown against each sample is the average of values of 5 samples collected from a single location i.e. 5 samples of soil were taken from a location in the urban area of Faridabad and coded as Soil-1. The samples soil-1 and soil-2 were collected from an urban area of Faridabad and soil-3 and soil-4 were collected from a rural area of Faridabad. The samples soil-5 and soil-6 were collected from an urban area of Palwal and soil-7 and soil-8 were collected from a rural area of Palwal. The samples soil-9 and soil-10 were collected from an urban area of Gurugram and soil-11 and soil-12 were collected from a rural area of Gurugram. The sample soil-13 and soil-14 were collected from an urban area of Mewat and soil-15 was collected from a rural area of Mewat. The observed radium concentration of these samples ranges from 8.8 to 15.7 $Bqkg^{-1}$.The radon mass exhalation rate varies in the range of 13.4 to 28.6 $mBqkg^{-1}$ h^{-1}. The surface exhalation rate of radon was found to be varied in the range of 363.3 to 678.5 $mBqm^{-2}$ h^{-1}. The mean value of the mass exhalation rate and surface exhalation rate was found to be 19.4 $mBqkg^{-1}$ h^{-1} and 500.21 $mBqm^{-2}$ h^{-1}.

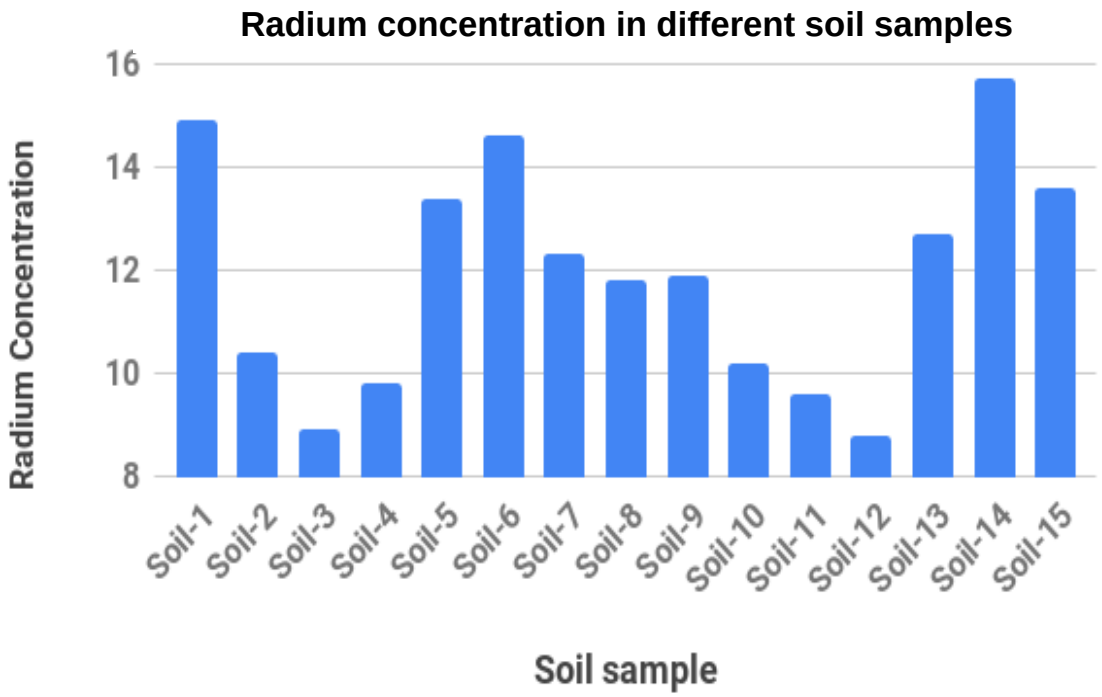

Figure 4.3: Radium concentration in different soil samples

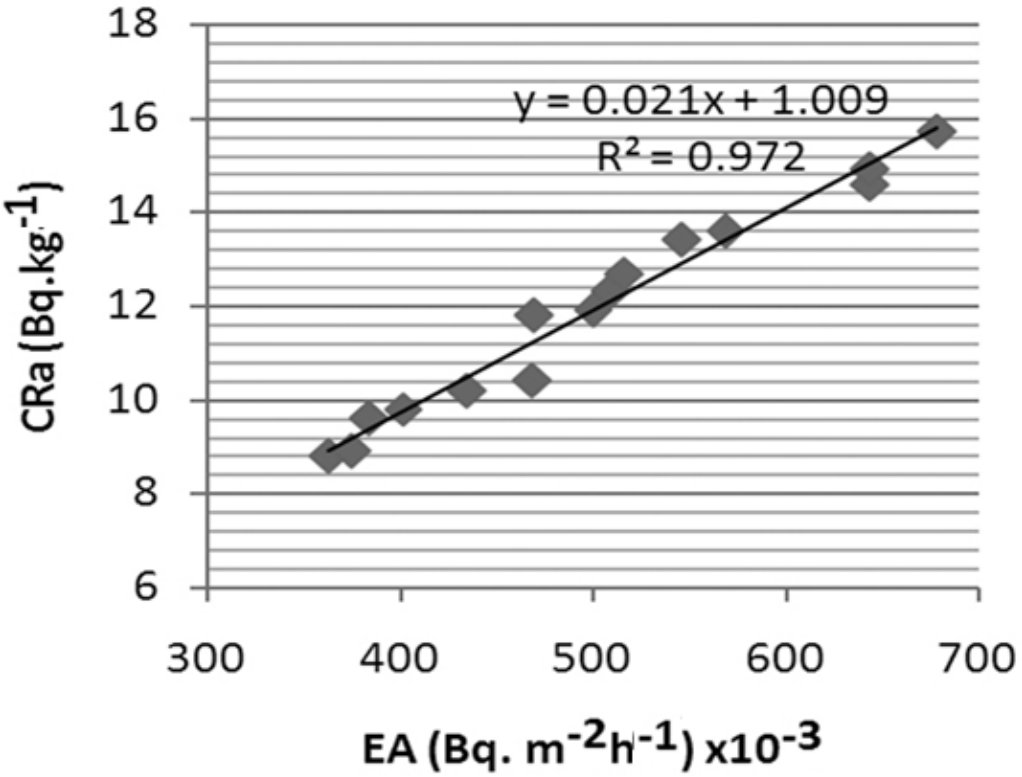

Figure 4.4: Correlation coefficient between radium concentration and radon exhalation rate

Figure 4.3 graphically represents table 4.1. As shown in figure 4.4 an excellent correlation coefficient has been observed between radium concentration and radon exhalation rate. The observed values of an experiment for radium concentration in all soil samples were found to be lower than safe limit as recommended by Organization for Economic Cooperation and Development (OECD) [36] and comparable to the global average of radium concentration value of soil samples. The result shows that the sample collection area is safe as far as the health hazard effect of radium is concerned.

4.5 RADON EXHALATION STUDIES OF BUILDING MATERIALS

The assessment of radium concentration and radon exhalation rate analysis for building materials is of immense importance as most of the human population spend 80% of their time indoors. The main contributor of indoor radium and radon concentration are soil gases, which are liberated from the ground of the indoor dwellings [37] and also the building materials used in the construction of the dwellings [38]. The probability of radon exposure increases exponentially if uranium-rich materials are used as building materials. The ambient temperature, pressure, and texture of the grain also affect the production of radon gas [39-40]. The emanation of soil gases from ground depends on various parameters [41-42] like soil permeability, soil moisture, entry route, environmental condition and most importantly the concentration of radon in soil. The parameters like rainfall [43], wind direction [44], dwelling occupancy pattern [45], environmental seasonal conditions [46], geological pattern [47], and proximity to ocean, river, and mountain [48] are also known to effects the presence of indoor radon concentration. Most of the studies carried out to measure the indoor radon concentration enlist two major contributors of radon gas as radon entering into dwelling through cracks etc. in the form of soil gas and radon generated by building materials. The building materials contribute a significant quantity of radon inside the dwellings. The analysis of different building materials for radon exhalation provides an estimation of indoor radon concentration levels. The building materials are the sources of indoor radiation but also protect us from outdoor radiation. The common building materials like fire and unfired bricks, concrete, marvel, sandstone, granite etc. are used as major construction materials and as decorative items in a dwelling, thus a study of radon exhalation and radium concentration determination was carried out on above-mentioned building materials. The purpose of the study is to mark the probably dangerous building materials emanating radon on the above safe limit.

The inhalation of radium (^{222}Rn) and its progeny like ^{214}Po and ^{218}Po is expected to pose a health hazard to humans [49-50]. The majority of radiation dose through the respiratory system gets deposited within the lungs [51]. The alpha radiations due to these radioactive elements may damage sensitive inner cells of lungs, hence causing lung cancer [52]. Different studies have predicted that almost 9 % of all deaths were

due to lung cancer and 2% of all deaths due to cancer were caused by radon exposure in dwellings [53]. Further many studies have established the link between high radon concentration in dwellings and the possibility of developing lung cancer in dwellings [54-55]. The building material samples for this experiment were collected from the local market of Gurugram district. A total of 10 samples of each building materials were taken for better measurement of the result. The experiment was conducted using a sealed CAN technique and LR-115 type II detectors. The samples were processed as per the requirements of the experiment like all the samples were first powdered using mortar and pestle and then dried at 100º C for two hours. These samples were further sieved through the 150-micron scientific sieve and kept in CAN for experimentation as discussed earlier. The exposure period like the previous experiment was 100 days. After the completion of exposure time, the detector was removed etched. The track created by alpha particles was calculated using the spark counter.

4.5.1 Results and discussion

The mass exhalation rate of radon has been observed to vary from 0.87±0.15 $mBqkg^{-1}h^{-1}$ to 8.03±0.46 $mBqkg^{-1}h^{-1}$, while the surface exhalation rate of radon varies from 77.44±14.17 $mBqm^{-2}h^{-1}$ to 793.98±46.51 $mBqm^{-2}h^{-1}$. The calculated values of radon concentration and radon exhalation rate were plotted figure 4.5. The granite sample contains the highest amount of radon concentration while ceramic contains the lowest amount of radon concentration. Figure 4.6 shows a correlation between the surface exhalation rate and the concentration of radon.

Table 4.2: Radium concentration and radon exhalation rate for building materials

Building Material	**Radon concentration (C_{Rn} Bq·m^{-3})**	**Surface exhalation rate (mBq·m^{-2}·h^{-1})**	**Mass exhalation rate (mBq.kg^{-1}h^{-1})**
Gypsum	78.35 ± 12.88	100.94 ± 16.49	1.89 ± 0.29
Cement (Black)	194.72 ± 20.23	251.01 ± 25.76	3.14 ± 0.34
Stone dust	146.04 ± 17.59	188.25 ± 22.54	1.77 ± 0.20

Cement (White)	70.38 ± 12.04	90.67 ± 15.46	1.14 ± 0.19
Ceramic	61.41 ± 11.40	79.12 ± 14.68	0.87 ± 0.15
Marble	192.66 ± 20.20	248.28 ± 26.02	2.86 ± 0.29
Bricks	163.06 ± 18.58	210.08 ± 23.83	2.68 ± 0.29
Granite	616.12 ± 36.12	793.98 ± 46.51	8.03 ± 0.46
Tiles	148.09 ± 17.59	190.99 ± 21.90	2.81 ± 0.31
POP	62.11 ± 11.14	79.44 ± 14.17	1.16 ± 0.21

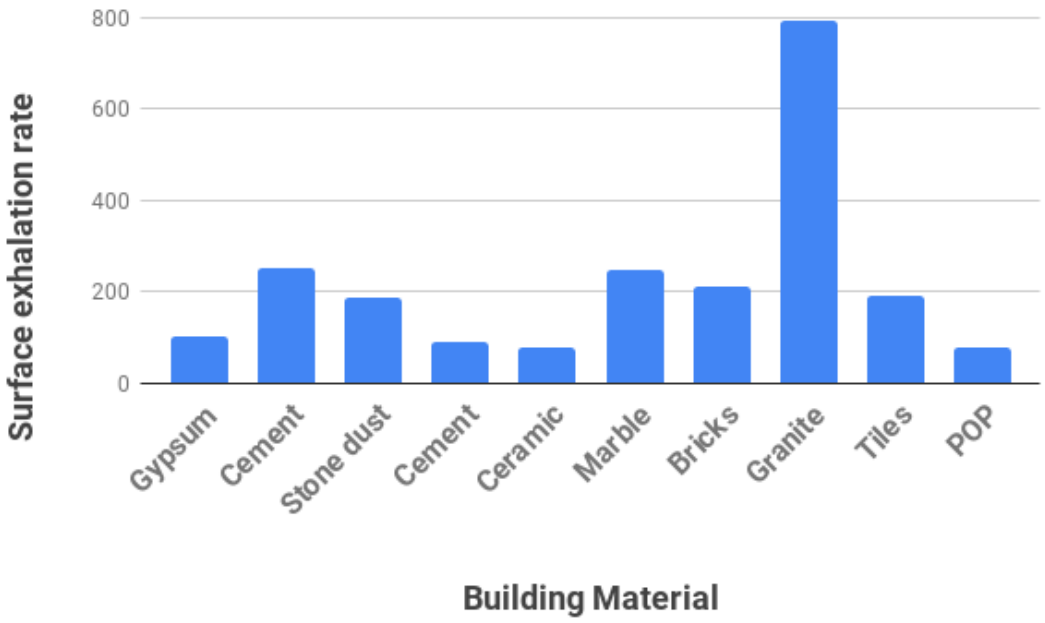

Figure 4.5: Radon surface exhalation rates in different building materials

It can be observed from the figure 4.5 that granite emanates the highest amount of radon to indoor air, the reason for this can be attributed to the high amount of uranium present in mines of granite [56].

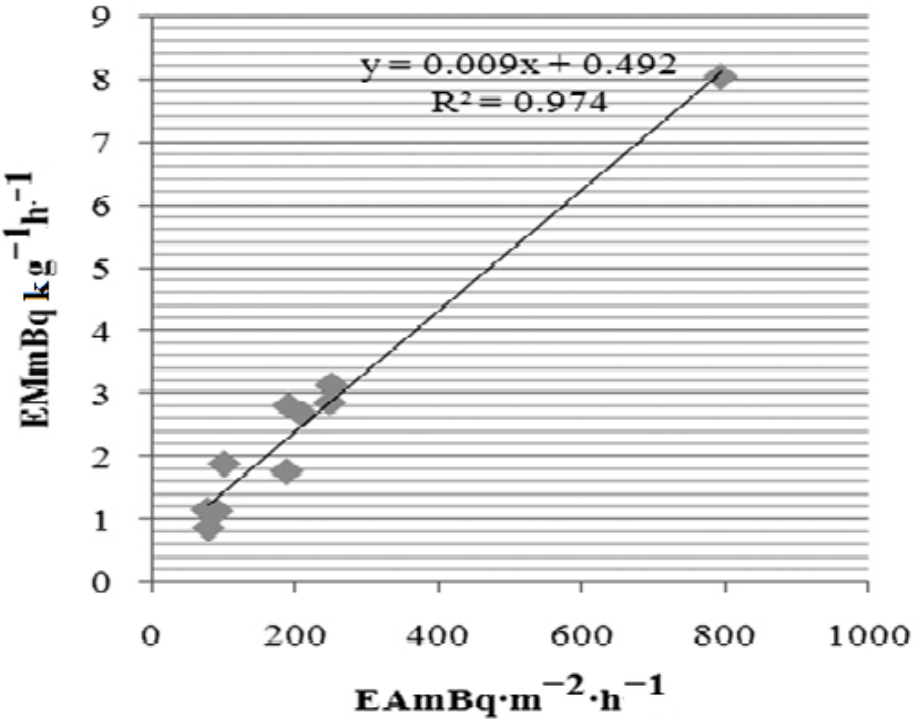

Figure 4.6: Correlation between radon mass and surface exhalation rates of building materials

Figure 4.6 depicts the excellent correlation between surface exhalation rate and radon concentration for building materials. The maximum value of effective radium content is well below the safe limit suggested by OECD [36].

4.6 THE EFFECT OF GRAIN SIZE ON RADON EXHALATION RATE

The value of radon concentration depends upon many environmental parameters like soil, stone and dust particles present on a location. The research work of radon exhalation rate measurement can be extended to analyse the exhalation rate measurement pattern in different samples of dust i.e. natural dust and stone dust. Since the radioactivity also depends upon the particle size of grain [57] hence by studying the radon concentration pattern from natural dust and stone dust for different grain sizes, a choice of dust as building materials having minimum radiation hazard to human can be made.

It is a well-known fact that dust particles contaminated with ^{226}Ra and radon pose the long term hazard to human health. So this fact necessitates the study of radon presence on dust particles. Studies show that radium and radon deposited in dust particles when inhaled get deposited inside the body. The inhalation risk is primarily due to alpha particles which damages the cell lining inside the lungs and potentially causes lung cancer in many cases [52]. The natural and stone dust which is used as building materials can contribute to the radiation dose received by dwellers [57]. Many studies [58-59] have suggested remedial actions to reduce radon exposure to human due to building materials; it mainly includes very well ventilated dwellings and uses of materials with low radon exhalation.

The present study was conducted to determine the exhalation rate of radon and its dependence on the grain size of samples. The samples selected for this purpose were natural dust and stone dust, which are extensively used as building material. The samples of natural dust and stone dust were collected from National Council for Building and Cement Board (NCB) Faridabad (Haryana) India and some other stone mining area near to Faridabad. CAN technique was used for radon exhalation rate determination. The exhalation rate depends upon the sample material, quantity of sample under test and dimensions of the experimental CAN [22]. The equilibrium concentration of radon and its progeny was achieved in a week. The LR-115 type II detector affixed on the CAN lid is exposed for 100 days, afterward, the detectors were removed carefully, etched in 2.5 N NaOH at 60º C for 90 minutes. The resultant

tracks on the detector's exposed face were counted using spark counter which is pre-sparked at 900 volts and operated at 450 volts.

4.6.1 Results and Discussion

The exhalation rates of radon for natural dust and stone dust are determined in this study. Each sample of natural dust and stone dust were divided into four parts on the basis of grain size i.e. grain size of 75 to 150 µm, 150 to 300 µm, 300 to 600 µm and further above 600 µm in dimension. The samples were also divided in terms of weight of the samples kept inside the CAN i.e. 100 grams, 150 grams, and 200 grams.

It can be seen from table 4.3, that mass exhalation rate of radon varies in the range of 1.05 $mBq.kg^{-1}hr^{-1}$ to 19.64 $mBq.kg^{-1}hr^{-1}$ with an average of 9.02 ± 5.37 $mBq.kg^{-1}hr^{-1}$ for stone dust and 3.66 $mBq.kg^{-1}hr^{-1}$ to 10.39 $mBq.kg^{-1}hr^{-1}$ with an average of 5.95 ± 2.7 $mBq.kg^{-1}hr^{-1}$ for natural dust. The value of surface exhalation rate of radon varies in the range of 119 $mBq.m^{-2}\ hr^{-1}$ to 522 $mBq.m^{-2}\ hr^{-1}$ with an average of 360 ± 67 $mBq.m^{-2}\ hr^{-1}$ for stone dust and 152 $mBq.m^{-2}\ hr^{-1}$ to 381 $mBq.m^{-2}\ hr^{-1}$ with an average of 286 ± 36 $mBq.m^{-2}\ hr^{-1}$ for natural dust for the used samples.

It can also be observed from table 4.3 that with the increase in grain size the exhalation rate of the sample increases. This phenomenon can be attributed to the fact that as the grain size increases the porosity of the sample increases [60] and hence radon exhalation rate of the samples also increases. The radon concentration in all the samples was found to be within the limit as prescribed by different environmental agencies hence these dust can be used as building material safely.

Table 4.3: Radon exhalation rate of natural dust and stone dust samples

Sample Size (µm)	Mass (g)	Natural Dust		Stone Dust	
		Em ($mBqkg^{-1}hr^{-1}$)	Es ($mBqm^{-2}hr^{-1}$)	Em ($mBqkg^{-1}hr^{-1}$)	Es ($mBqm^{-2}hr^{-1}$)
≤150 and ≥75	100	3.66	152	1.05	119
	150	4.87	198	4.05	176
	200	7.89	367	8.60	289
≤300 and ≥150	100	3.89	159	3.26	167
	150	5.32	245	4.69	189
	200	9.12	378	9.91	343
≤600 and ≥300	100	4.10	162	7.45	256
	150	6.56	321	7.29	245
	200	9.35	378	15.08	489
≥600	100	4.23	177	11.58	289
	150	6.18	315	13.31	467
	200	10.39	381	19.64	522

4.7 MEASUREMENT OF RADON EXHALATION RATE IN STONE MINING AREA

Radioactive materials like radium and radon exist in decay series of uranium and thorium. These radionuclides are used in tracing studies and environmental dating processes. Radium is a naturally occurring radioactive material which exists in earth's crust in very low amount and in uranium mines in very high amount. The mining of stones are carried out for a long time and many workers are always involved in this work. Stones are important building materials and are used in the floor and walls inside the dwellings. The stones are excavated from deep inside of the earth surface where the presence of radionuclide is considered very high. For the mine workers and residents in general, the radiation exposure beyond permissible safety limit can have dangerous effects on human health. These facts necessitate the extensive studies of natural radioactivity levels in the stone mining area of Aravali range of Faridabad, India. The proximity of Aravali mine to fast developing urban area is the reason for carrying out the study to measure radon concentration in these mining areas of Aravali. The geographical coordinates of samples collection sites are 28º 252′593″ N latitude and 77º 192′ 003″ E longitude.

Measurements were carried out using the CAN technique. Different types of stone and soil samples were collected from the above-mentioned site. The samples were first crushed, sieved and then oven dried to remove moisture. Ten samples of 150 grams each were placed in plastic CANs. As already discussed LR-115 type II plastic track detectors were used for this experiment. The detectors were exposed for 100 days and after the end of exposure time, the detectors were removed. The detectors were subjected to a chemical etching process with a 2.5 N NaOH solution at 60º C for 90 minutes. The detectors were then washed and dried. The tracks produced on detectors by alpha particles were counted using spark counter.

4.7.1 Results and Discussion

All samples were collected from the Aravali mining area viz. SA-1 to SA-3 from residential sites near Aravali region, SA-4 to SA-6 from the fresh mine area where the mining is under process, SA-7 to SA-10 transportation sites of the mines. Equation

4.9 and 4.10, the radon mass exhalation rate and radon surface exhalation rates were computed and listed in table 4.4.

Table 4.4: Radon exhalation rates in some stone samples from the Aravali range

Sample Code	**Mass (gm)**	**Radon Mass Exhal. Rate (mBq $kg^{-1}h^{-1}$)**	**Radon Surface Exhal. Rate (mBq/$m^{-2}h^{-1}$)**
SA-1	150	3.41	75.90
SA-2	150	4.03	89.70
SA-3	150	3.98	88.58
SA-4	150	9.11	202.77
SA-5	150	6.82	151.8
SA-6	150	8.98	199.87
SA-7	150	5.98	133.10
SA-8	150	5.43	120.86
SA-9	150	4.78	106.39
SA-10	150	5.78	128.65

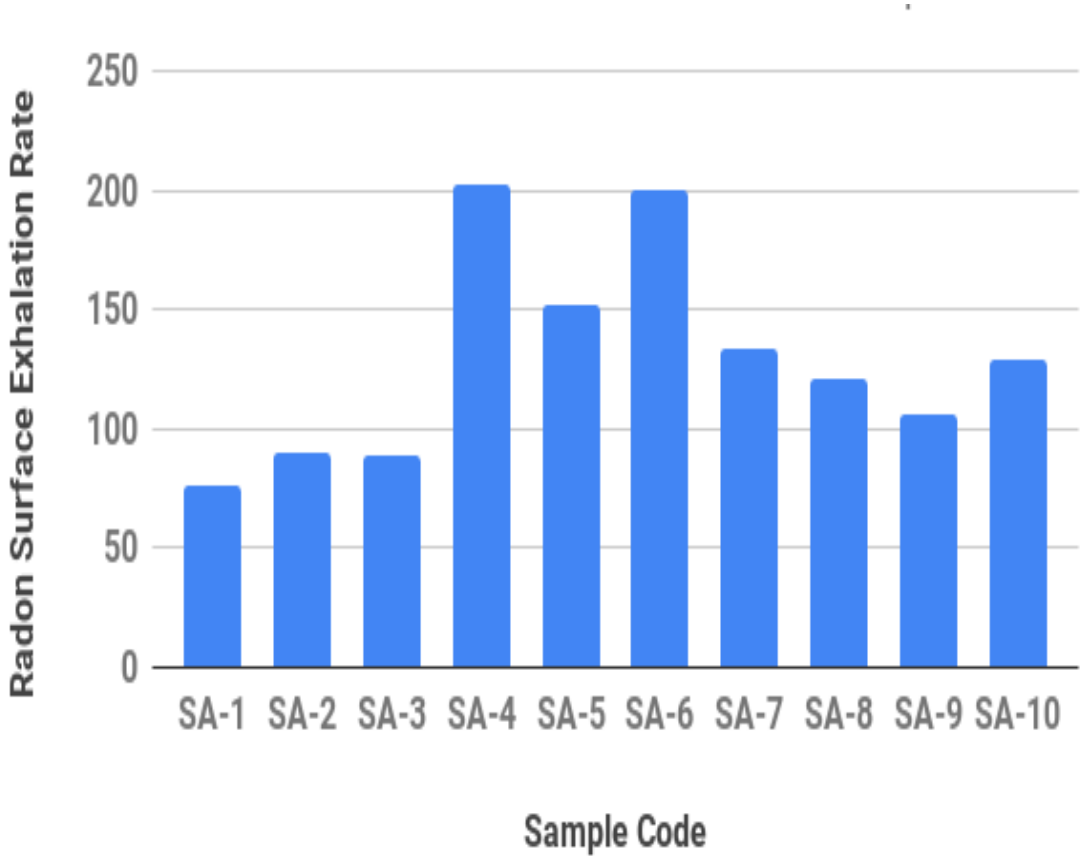

Figure 4.7: Radon surface exhalation rates for different stone samples

The mass and surface exhalation rates from the stone samples vary in the range of 3.41- 9.11 mBqkg-^{1}h^{-1} and 75.9- 202.7 mBq.m^{-2}h^{-1}, respectively. Figure 4.7 depicts of radon surface exhalation rate for the different stone samples. The study reveals the significant presence of radon concentration in the collected samples from the fresh mines area, however, the range is found within the safety limits as prescribed by environmental agencies for radon concentration [36].

4.8 THE EFFECT OF ADDITION OF FLY-ASH TO CEMENT ON RADON EXHALATION RATE

India majorly depends upon coal reserves for the generation of electricity. The coal-based power plants produce about 72% of total power in India [61]. Coal, which is found very deep inside the earth, is rich in radioactive materials like uranium, thorium, and potassium. When the coal is combusted in thermal power plants, the radionuclides are released in the surrounding ecosystem. The by-product fly-ash does contain a high amount of these radionuclides and their daughter products. The disposal of fly-ash poses serious environmental problems it requires a large area for large disposal, causes pollution to air, soil, and water in the surrounding area where ash is stored. Thus it is suggested that ash should not be stored and innovative methods should be explored to utilise this by-product. It provides an economic advantage as it can be used as a cheap resource as construction material. It can be used as a cavity filling material and also can be used for the production of cement [62] however using it in cements is also harmful. This utilisation and the presence of fly-ash in the environment lead to a study of radon exhalation from fly-ash to estimate the radiation risk. In the current study, the Ordinary Portland Cement (OPC) and fly-ash samples were collected from the National Council for Cement and Building Board (NCB) Faridabad in Haryana.

The study of radon exhalation rate for fly-ash and cement samples were carried out using the CAN technique. The known amount (100gm) of the sample is placed in the lower part of the sealed CAN. The samples were prepared and processed as required for the CAN technique. For this experiment, 11 different samples of cement and fly-ash mixture were prepared. The samples starting 100% cement and 0% fly-ash to 100% fly-ash and 0% cement were prepared by increasing the content of fly-ash by 10% in every subsequent sample. The LR-115 type II detectors were used in the process as discussed earlier. The samples were sieved through 300-micron mesh scientific sieve and kept in CAN for detector's exposure for 100 days. When the exposure time ends, the etching of the detectors was carried out as already discussed in CAN technique. The tracks produced by alpha particles were counted using spark counter.

4.8.1 Results and Discussion

Total 11 samples are processed for this experiment as shown in table 4.5. Equation 4.9 and 4.10 were used to calculate that radon mass and surface exhalation rate for these 11 samples.

Table 4.5: Content of cement sample and their respective exhalation rate (C here represents cement and FA represents fly-ash)

Sample no.	**Sample Content**	**Radon Mass Exhal. Rate ($mBq/kg^{-1}h^{-1}$)**	**Radon Surface Exhal. Rate ($mBq/m^{-2}h^{-1}$)**
1	100% C	2.51354	48.803
2	100% FA	4.44247	86.2552
3	10% FA	3.48298	67.6256
4	20% FA	3.61114	70.114
5	30% FA	3.93942	76.4879
6	40% FA	5.58085	108.358
7	50% FA	5.19774	100.919
8	60% FA	5.03531	97.7656
9	70% FA	4.82148	93.6139
10	80% FA	3.85718	74.8911
11	90% FA	2.57145	49.9274

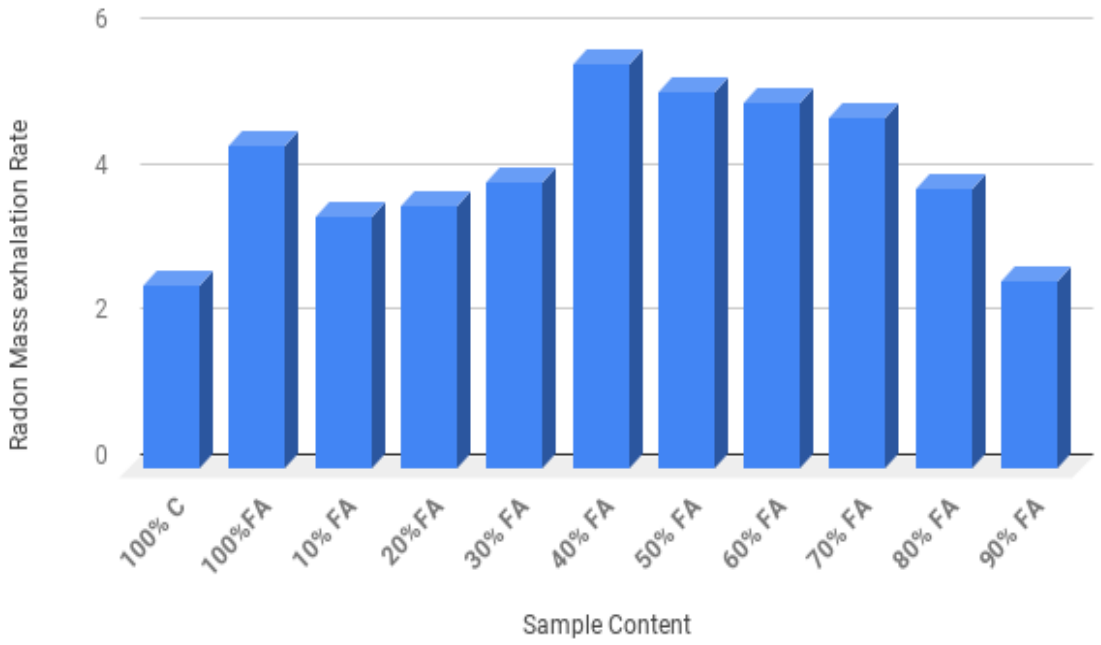

Figure 4.8: Radon Mass exhalation rates at different concentration of fly-ash in cement samples

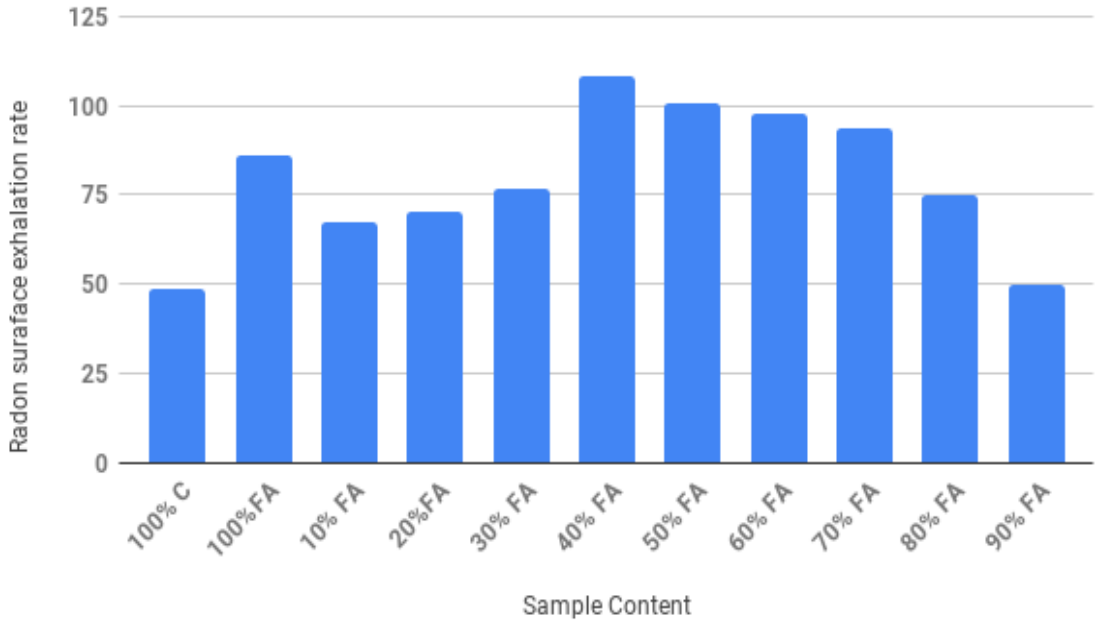

Figure 4.9: Surface exhalation rates of radon at different concentration of fly-ash in cement samples

It can be seen that from figure 4.8 and 4.9 that the radon exhalation rate increases to sample number 6 (40% fly-ash and 60% cement) and then start decreasing gradually. The reason for this can be understood as the fly-ash itself is a radioactive material, adding it in cement will result in an increase in radon exhalation rate. But after a certain limit, exhalation rate starts to decrease, since the grain size of fly-ash is small and mixing it in higher concentration will decrease the porosity, which eventually will result in a decrease of radon exhalation rate [60].

It was quite important to estimate the health risk due to radiation, by mixing fly-ash with cement in different proportions. As can be observed by the table 4.5 the maximum surface exhalation rate and mass exhalation rate of radon are 108.3 $mBqm^{-2}h^{-1}$ and 5.58 $mBqkg^{-1}h^{-1}$ respectively, which is under prescribed safety limit and hence fly-ash mixed with cement and used as a building material will not pose any health hazard.

4.9 MEASUREMENT OF RADON EXHALATION RATE AND RADIUM CONTENT IN CEREAL SAMPLES

The natural radioactivity is ubiquitous. The radionuclides such as radium, uranium, thorium, and potassium exist in rocks, soil, water, and air etc. The radionuclides decay to form radon gas which is absorbed by plants from the soil and then it may get transferred to the human body through the food chain [52]. These cereals products are either directly or indirectly consumed by humans. Hence it is very essential to study the concentration of radon exhalation rate and radium concentration in cereal samples. The purpose of this work is to analyse the cereal samples for radon concentration.

The cereal samples were collected from Faridabad in Haryana state, India. The experiment to measure radium content and radon exhalation rate for cereal samples were conducted using the CAN technique. In this analysis, 10 different samples were collected from the local market of Faridabad. All the samples were washed to remove dust and dried in an oven at 100ºC for 2 hours to remove moisture. For the processing of samples using the CAN technique, samples were crushed and sieved to obtain uniform grain size. The 50 grams of different samples were placed on the bottom of the CAN. The plastic track detectors LR-115 type II were exposed to sample for 100 days. After the end of exposure time, the detectors were removed and then etched by the process used in CAN technique. Tracks produced by alpha particles were counted using spark counter.

4.9.1 Results and Discussion

A total of 10 different samples were analyzed in this experiment. The effective radium content and radon exhalation rate were calculated using equations 4.8 and 4.9 respectively.

Table 4.6: Radium content C_{Ra}, the radon area exhalation rate and radon mass exhalation rate in cereal samples

Sample	Sample Code	ρ (Track/cm^2.d)	CRa (mBq/kg)	J_A (μBq/m^2.s)	J_M (μBq/kg.s)
Corn	SC	4.56 ± 0.12	167.13 ± 7.665	7.18± 1.45	0.26 ± 0.07
Wheat	SW	2.31 ± 0.45	66.09 ± 8.22	3.98 ± 1.68	0.14 ± 0.11
Brown Lentil	SBL	3.69 ± 0.89	112.46 ± 4.69	5.78 ± 1.79	0.27 ± 0.08
Rice	SR	6.34 ± 0.34	256.36 ± 7.19	9.62 ± 1.66	0.38 ± 0.15
Kidney Beans	SKB	4.45 ± 1.23	157.56 ± 8.56	10.34 ± 1.87	0.48 ± 0.14
Semolina	SS	6.02 ± 1.15	243.45 ± 8.11	9.87 ± 1.54	0.38 ± 0.19
Black eyed Beans	SB	5.84 ± 0.98	198.61 ± 5.43	8.76 ± 1.64	0.45 ± 0.16
Green Gram	SGG	3.45 ± 1.34	82.76 ± 5.44	6.43 ± 1.07	0.19 ± 0.07
Black Gram	SBG	5.19 ± 3.70	189.02 ± 8.98	9.23 ± 1.43	0.39 ± 0.16
Pigeon Pea	SPP	1.76±1.01	46.34 ± 6.45	2.64 ± 1.32	0.12 ± 0.07
Maximum		6.34 ± 0.34	256.36 ± 7.19	10.34 ± 1.87	0.48 ± 0.14
Minimum		1.76±1.011	46.34 ± 6.45	2.64 ± 1.32	0.12 ± 0.07
Mean		4.053 ± 0.67	151.35 ± 6.82	6.49 ± 1.59	0.30 ± 0.11

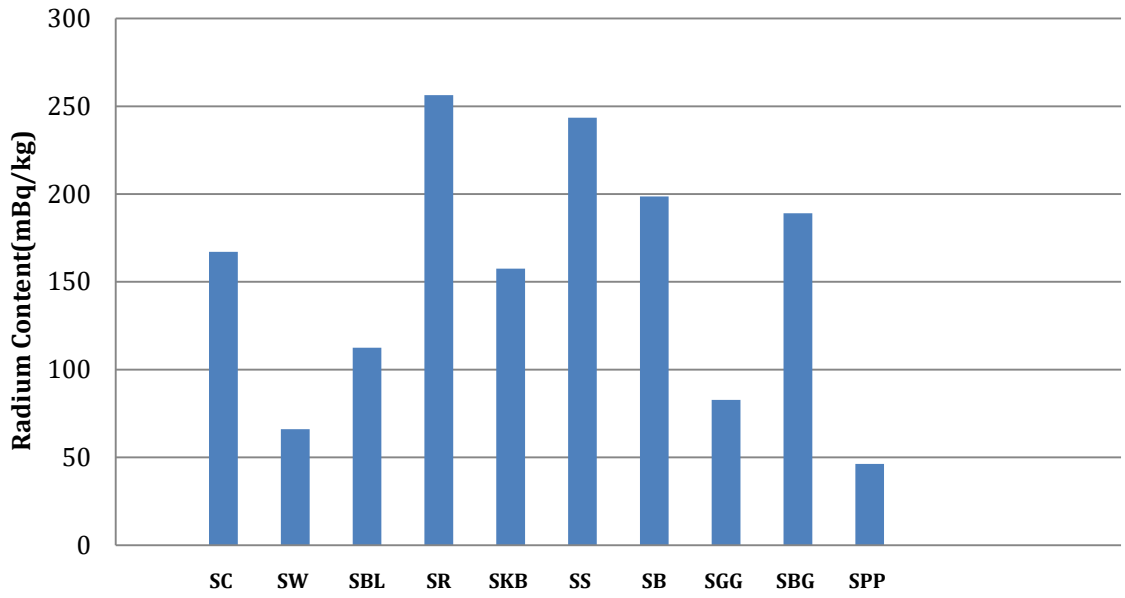

Figure 4.10: Radium content in cereals samples

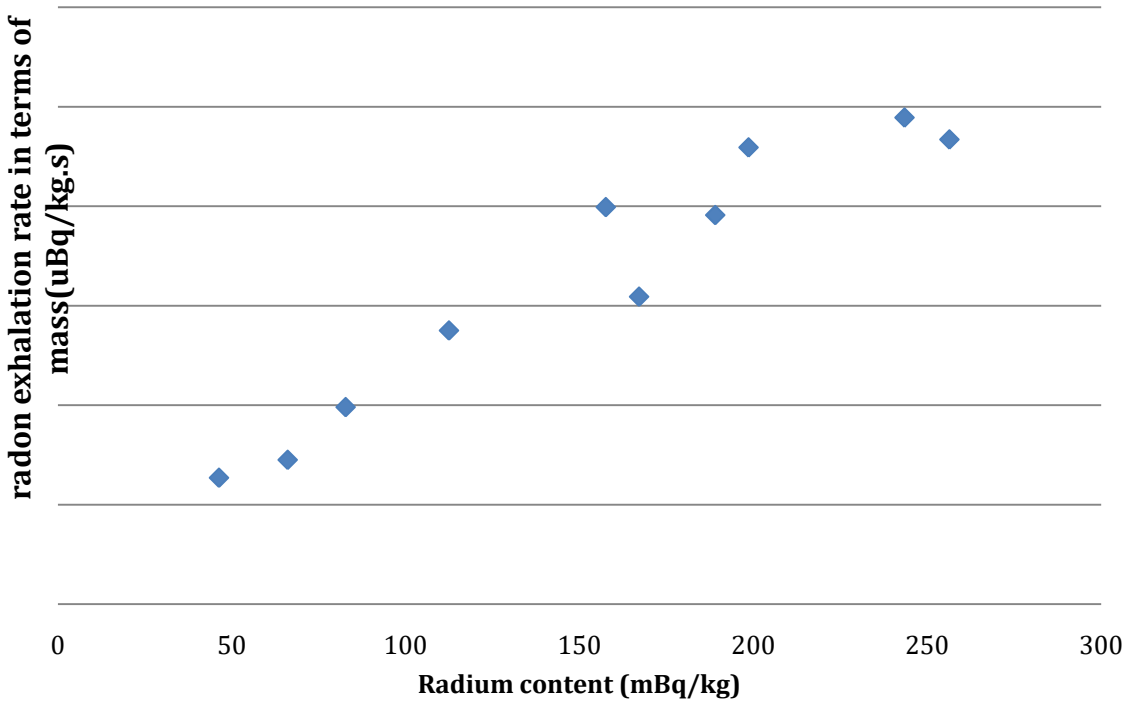

Figure 4.11: Correlation between radium content and radon exhalation rate

It is observed from table 4.6 that the highest value of radium content was found in rice samples which is equal to 25.36 ± 7.19 mBq kg^{-1}, whereas the lowest value has been

found in pigeon pea sample, which is equal to 46.34 ± 6.45 mBq.kg^{-1} with average of 15.35 ± 6.82 mBq.kg^{-1} for all the samples. The result of radon exhalation rate for the same samples ranges from 2.64 ± 1.32 to 10.34 ± 1.87 µBq m^{-2}s^{-1} with an average of 6.49 ± 1.59 µBqm^{-2}s^{-1} and from 0.12 ± 0.07 to 0.48 ± 0.1 4 µBqm^{-2}s^{-1} with an average of 0.308 ± 0.110 µBqm^{-2}s^{-1}, respectively for area and mass parameter. Figure 4.10 shows the radium concentration for the different cereals samples. Figure 4.11 shows the excellent correlation obtained between radium content and radon exhalation rate for cereals. The radon exhalation rate increases whenever the radium content increases and vice versa. The value of radium content calculated as in table 4.6 corresponds with the value reported by many researchers [63]. The values of radium content for all the samples under study was found in the normal range and are expected not to pose any radiation regarding risk for human beings.

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CHAPTER-V

MEASUREMENT OF RADIOACTIVITY USING GAMMA SPECTROSCOPY

5.1 INTRODUCTION

Another experimental technique used in this work is gamma spectrometry with the HPGe detector [1]. This technique is applied to the multiple samples of foodstuff, stone and building material (fly-ash) [2-3]. Radon transports to humans can be attributed to two factors as i) ingestion through foodstuff ii) inhalation air from the environment [4]. So it is very important to have the proper knowledge of the distribution of radionuclides and natural radioactivity in these samples for the estimation of radiation exposure to human and to establish the national standard in the context of international suggestions in this area [5].

Gamma spectrometry is a very powerful and robust technique for the detection of radionuclides in various types of solid samples [6]. In the present work, the HPGe detector based on the high-resolution gamma spectrometry system was deployed for the measurement of the gamma activity. The gamma-ray spectrometry facility at Inter University Accelerate Centre (IUAC), New Delhi was availed for experimentation.

5.2 RADIOACTIVITY MEASUREMENTS THROUGH GAMMA SPECTROSCOPY

In this technique, the gamma spectrum of the desired sample is obtained using an HPGe detector which is connected to a multi-channel pulse height analyzer [7]. The gamma spectrum thus obtained is often complex and the various components are separated by different analytical techniques. Thus the technique provides a robust multi-component analysis which is extensively used for environmental samples [8-9]. The gamma spectroscopy technique for the measurement of radioactivity is used as discussed in chapter 3.

5.3 SAMPLE PREPARATION

Initially, the samples were crushed into fine powder by using mortar and pestle. The fine quality of the sample was obtained using the scientific sieve of the 250-micron mesh size. The samples were dried in an oven at a temperature of 383 K for 24 hours. The samples were then packed and sealed in an airtight PVC container and kept for about 4 week period to allow radioactive equilibrium among the daughter products of radon [10]. On an average 300 gm of the sample is required for measurement.

5.4 THEORETICAL BACKGROUND OF PROCESSING

5.4.1 Measuring Activity Concentration of Uranium, Thorium, and Potassium

In the spectrometry process the concentrations of ^{238}U, ^{232}Th, and ^{40}K were determined by using the following equation [11]:

$$\text{Activity} = \frac{\text{CPS} \times 100 \times 100}{\text{B.I} \times \text{E}_{\text{ff}}} + \frac{\text{CPS}_{\text{error}} \times 100 \times 100}{\text{B.I} \times \text{E}_{\text{ff}}} \quad (5.1)$$

Where CPS - Net count rate per second, B.I. - Branching Intensity and E_{ff} - Detector Efficiency.

5.4.2 Radium Equivalent Activity

It is the weighted sum of activities of the ^{238}U, ^{232}Th and ^{40}K radionuclides on the basis of the estimation that 10 Bq/kg of ^{238}U, 7 Bq/kg of ^{232}Th and 130 Bq/kg of ^{40}K produce the same gamma-ray dose radiation [11-12]. It is represented by Ra_{eq} and is given by the equation.

$$\text{Ra}_{\text{eq}} = \text{C}_{\text{U}} + 1.43\text{C}_{\text{Th}} + 0.077\text{C}_{\text{k}} \quad (5.2)$$

Where C_U, C_{Th} and C_k are the specific activities of ^{238}U, ^{232}Th, and ^{40}K in Bq/kg respectively.

5.4.3 Assessment of Radiological Hazards

Radiological hazards in terms of various hazard index like external hazard index (H_{ex}),

Internal hazard index (H_{in}), Gamma index (I_γ) and Alpha index (I_α) have been determined using the following equations [13-14].

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (5.3)$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (5.4)$$

$$I_\gamma = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \quad (5.5)$$

$$I_\alpha = \frac{A_{Ra}}{300} \quad (5.6)$$

The samples are considered safe from the health point of view when the value of H_{ex} and H_{in} are less than unity. Value of $I_\gamma \leq 2$ corresponds to a dose rate of 0.3 mSv/y, whereas $2 \leq I_\gamma \leq 6$ corresponds to 1 mSv/y [14].

5.4.4 Estimation of Annual Effective Dose and Absorbed Dose

The measured activity concentration of ^{238}U, ^{232}Th and ^{40}K were converted into doses ($nGyh^{-1}$ per Bq kg^{-1}) and the total absorbed gamma dose rate in air at one meter above the ground level was calculated using the equation [15].

$$D(nGy/h) = 0.462C_U + 0.604C_{Th} + 0.0417C_k \quad (5.7)$$

Where C_U, C_{Th} and C_K are the activity concentrations (Bq/kg) of uranium, thorium, and potassium; 0.462, 0.604 and 0.0417 are the conversion factors for uranium, thorium, and potassium, respectively in the samples.

The factors of indoor occupancy pattern and conversion coefficient (absorbed dose in the air to effective dose) must be taken care of while estimating the annual effective dose [16].

A conversion factor of 0.7 Sv/Gy is considered to calculate the annual average effective dose received by a human while assuming 80% indoor occupancy and 20% outdoor occupancy. [17-19].

The indoor and outdoor annual effective doses are calculated using the equations (5.8) and (5.9) respectively [19].

$$\text{Indoor(mSv)} = \text{(Absorbed Dose)nGyh}^{-1} + 8760\text{h} \times 0.8 \times 0.7\ \text{SvGy}^{-1} \times 10^{-6} \quad (5.8)$$

$$\text{Outdoor(mSv)} = \text{(Absorbed Dose)nGyh}^{-1} + 8760\text{h} \times 0.2 \times 0.7\ \text{SvGy}^{-1} \times 10^{-6} \quad (5.9)$$

5.4.5 Annual Effective Dose (D_{eff})

Effective dose due to ingestion is a useful concept that enables the radiation doses from different radionuclides to be added. Estimation of the radiation-induced health effects associated with the intake of radionuclides in the body is proportional to the total dose delivered by the radionuclides while present in various organs [20]. The annual effective ingestion dose to the population from the intake of radionuclides in foodstuffs (cereals) is determined using the following relation [21,22].

$$D_{eff}\ (\frac{Sv}{y}) = A \times C \times R \quad (5.10)$$

Where D_{eff} is the effective dose by ingestion of the radionuclide present in the food (Sv/y), A is the activity concentration of radionuclides in the sample (Bq kg^{-1}), C is the internal dose conversion factor by ingestion of the radionuclides (Sv Bq^{-1}) as 2.8×10^{-7}, 4.5×10^{-8}, 2.3×10^{-7} and 6.2×10^{-9} for ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K, respectively.

5.4.6 Excess Lifetime Cancer Risk (R_C)

The risk associated with an intake of radionuclides in the body is proportional to the total annual internal dose delivered by the radionuclides as per the LNT hypothesis [23]. The risk factor states that the probability of a person dying of cancer increases by 5% for a total dose of 1 Sv received during his lifetime. Therefore, the probability of death from cancer due to 'natural incidence' increases from about 25% to 30% following a total lifetime exposure of 1 Sv [24]. The cancer risk for an adult person can be estimated by using the following relationship [25].

$$R_C = C_d \times R_F(Sv^{-1}) \quad (5.11)$$

Where R_F is a risk factor (Sv^{-1}), fatal cancer risk per sievert. For stochastic effects,

ICRP 60 uses values of 0.05 for the public [26]. The C_d is the lifetime effective dose that is a measure of the total effective dose received over an average lifetime of 50 years following ingestion of a radionuclide was calculated as [27].

$$C_d = 50 \times D \quad (5.12)$$

Where D is the total effective dose to an individual.

5.5 MEASUREMENT OF NATURAL RADIOACTIVITY IN DIFFERENT VEGETATION SAMPLES

Vegetation (foodstuffs) which include vegetables and fruits, are grown in the ground having soil full of radioactive elements, these radioactive elements may get deposited to plants either due to by absorption from the soil or due to radioactive fallout [28]. This vegetation when consumed can pose serious health hazards to the human [29], so experiments were conducted for natural radioactivity measurement of vegetable samples to analyze the radionuclides emanation from these samples.

5.5.1 Samples collection Sites

The geographical location of the samples collection sites belongs to the Haryana state of India, which is located between 27º37´ and 30º35´ Northern Latitude and 74º28´ to 77º36´ East Longitude in the northern part of India. The four districts of Haryana namely, Faridabad, Palwal, Gurgaon, and Mewat were selected for samples collection. The sample collection sites are shown in figure 5.1

5.5.2 Sample Collection and Preparation

In this investigation, various samples of commercially available vegetables (Cabbage, sweet potato, onion, radish, jackfruit, tomato, beetroot, potato, carrot, spinach and cucumber) and various sample of fruits (papaya, mango, apple, grapes, pears, guava, banana)were taken from the market and fields. The measurement activity concentrations of the natural radionuclides were carried out in these samples.

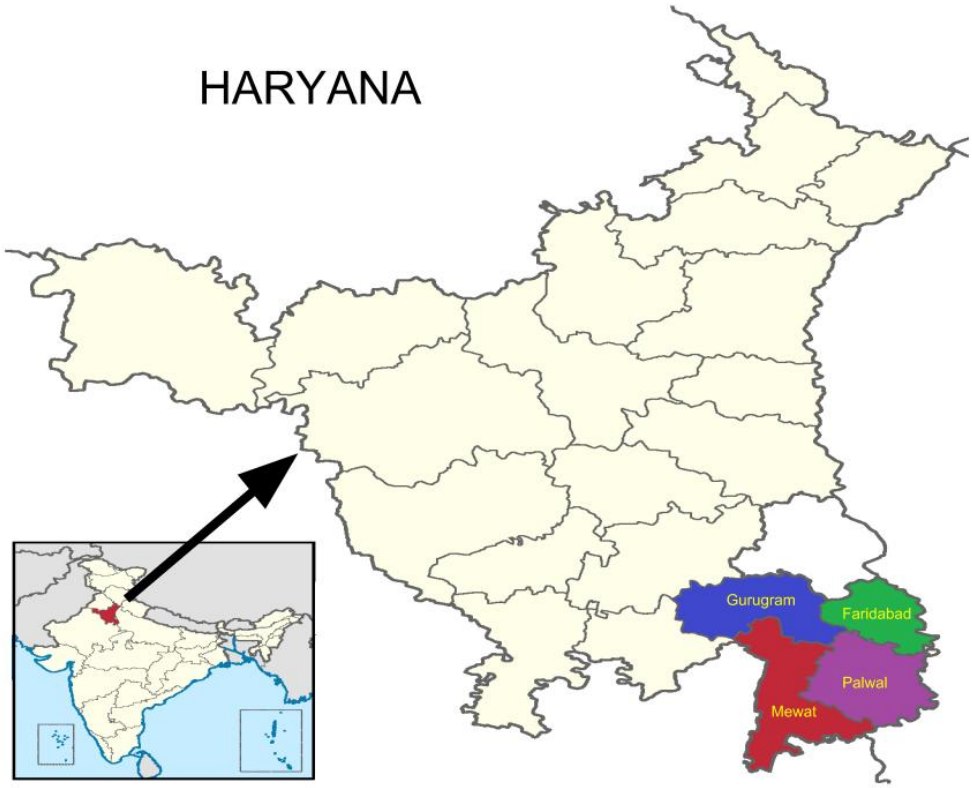

Figure 5.1: Geographical location of sample collection sites in Haryana, India

The collected samples were washed and dried to remove moisture and any soil left out further to obtain a constant dry weight, these samples were kept in the electric oven at 110°C for 10 to15 hours [30]. The muffle furnace was used to obtain white ash from samples which were charred at 450°C under low flame. The samples were sieved in 250-micron mesh size sieve to obtain the particles of size less than 250 microns. Sieved samples were sealed in 300 ml airtight PVC container. These sealed PVC containers then left for four weeks to settle radioactive equilibrium between the radon (^{222}Rn), thoron (^{220}Rn) and their short-lived progenies [31]. Each sample contains approximately 300 grams of ash. A secondary standard was obtained for the low background counting system calibration; it is made compatible with the primary standard which is designed as per the recommendation of the International Atomic Energy Agency [32]. The concentration of ^{226}Ra was calculated at the photon peak of 609 keV (46.1%) from ^{214}Bi on the analyzer. The 185.7 keV peak of ^{235}U interfere with 186 keV peak of ^{226}Ra hence the 186keV peak is ignored. The gamma transitions

of 583 keV (86%) from ^{208}Tl are used to determine the ^{232}Th concentration [32]. The gamma transitions of 1461 keV (10.7%) are used to determine the ^{40}K concentration.

5.5.3 Results

The radionuclides concentration and radium equivalent activity calculated using equations (5.1) & (5.2) for the vegetable samples and for fruit samples are presented in table 5.1 and table 5.2, respectively.

Table 5.1: Activity concentration of ^{238}U, ^{232}Th, and ^{40}K and Radium equivalent Concentration in vegetable samples.

S. No.	Sample code	Activity concentration (Bq / kg)			Radium equivalent Concentration Ra (Bq.kg^{-1})
		^{238}U	^{232}Th	^{40}K	
1	VC-1	28.08 ± 1.28	42.31 ± 2.17	1158.4 ± 26.05	177.78
2	VC -2	15.55 ± 1.77	57.52 ± 2.34	1579.2 ± 17.58	219.40
3	VC -3	22.72 ± 1.63	47.32 ± 1.35	1268.5 ± 15.05	188.06
4	VC -4	29.13 ± 0.69	22.20 ± 2.46	1885.1 ± 28.03	206.03
5	VC -5	28.22 ± 1.62	26.48 ± 1.41	1590.9 ± 19.21	188.59
6	VC -6	10.25 ± 0.94	36.37 ± 1.20	1665.4 ± 17.63	190.49
7	VC -7	19.94 ± 0.40	49.17 ± 1.31	1820.6 ±27.24	230.44
8	VC -8	17.74 ± 1.59	58.21 ± 1.15	1962.2 ± 18.17	252.07
9	VC -9	18.38 ± 1.15	50.82 ± 1.17	1698.5 ± 28.04	221.84
10	VC -10	11.40 ± 0.90	39.85 ± 2.32	1775.0 ± 26.48	205.06
11	VC -11	19.21 ± 1.61	26.53 ± 1.89	1625.2 ± 18.28	182.29

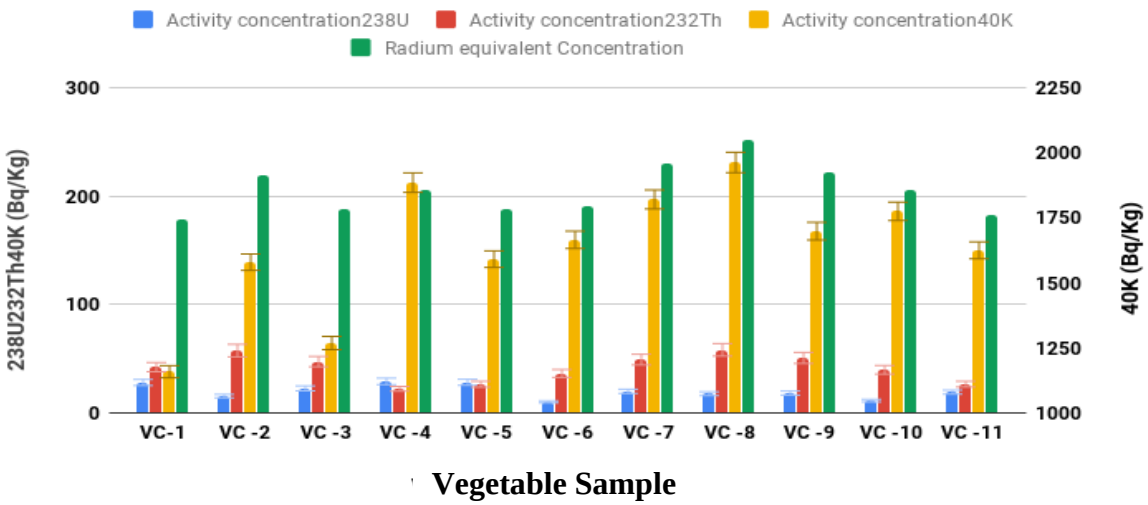

Figure 5.2: Radium equivalent concentration and Activity concentration of ^{238}U, ^{232}Th and ^{40}K in vegetable samples.

Table 5.1 shows the activity concentration for ^{238}U, ^{232}Th, and ^{40}K for various vegetable samples under investigation in the present work found to vary in the range of 10.25±0.94 Bq/kg to 29.13±0.69 Bq/kg, 22.20±2.46 Bq/kg to 58.21±1.15 Bq/kg, and 1158.4±26.05 Bq/kg to 1962.2±18.17 Bq/kg respectively. The radium equivalent activity was found to be varied from 177.78 Bq/kg to 252.07 Bq/kg.

Table 5.2: Activity concentration of ^{238}U, ^{232}Th, and ^{40}K, and Radium equivalent Concentration in fruit samples.

S. No.	Sample code	Activity concentration (Bq/kg)			Radium equivalent Concentration Ra (Bq.kg^{-1})
		^{238}U	^{232}Th	^{40}K	
1	FR-1	8.1 ± 0.15	12.3 ± 1.49	687.13 ± 12.23	78.60
2	FR-2	2.5 ± 0.16	15.1 ± 1.67	554.74 ± 14.09	66.81
3	FR-3	3.9 ± 0.25	13.0 ± 1.32	487.13 ± 11 .23	60.00
4	FR-4	4.6 ± 0.23	09.6 ± 1.22	384.13 ± 9.23	47.91
5	FR-5	9.8 ± 0.15	11.4 ± 1.29	287.13 ± 11.23	48.21

6	FR-6	5.6 ± 0.15	13.4 ± 1.76	437.56 ± 12.39	58.45
7	FR-7	4.6 ± 0.29	08.3 ± 1.53	737.39 ± 9.37	73.25
8	FR-8	3.5 ± 0.27	07.4 ± 1.24	815.72 ± 12.50	76.89
9	FR-9	8.5 ± 0.12	18.4 ± 1.39	587.13 ± 16.23	80.02
10	FR-10	7.4 ± 0.18	08.1 ± 1.34	511.25 ± 8.17	58.35

MADL (Minimum Activity Detection Limit) is considered as 2 Bq/kg, 2Bq/kg and 4Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K respectively.

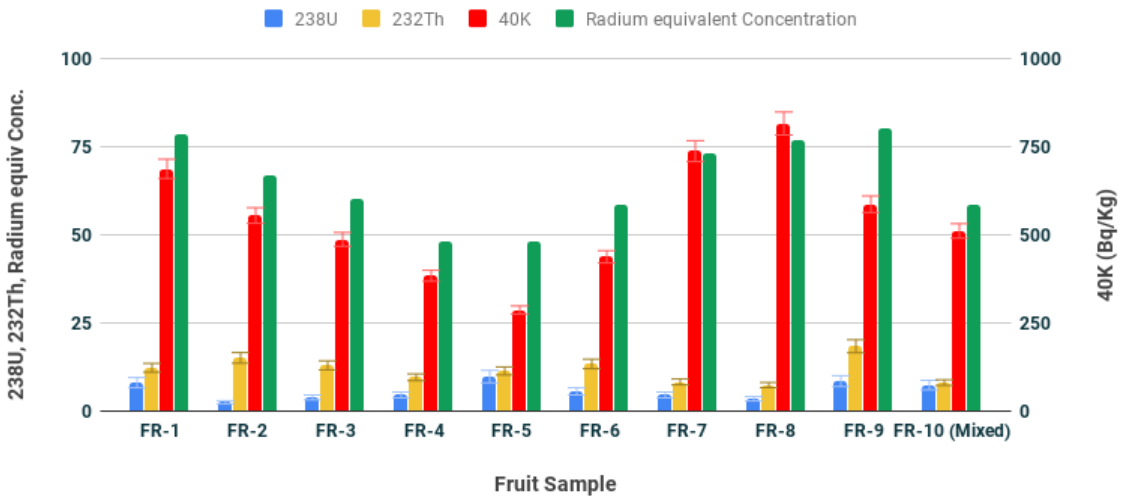

Figure 5.3: Radium equivalent concentration and Activity concentration of ^{238}U, ^{232}Th and ^{40}K in fruit samples.

It can be seen from table 5.2 that the activity concentration for uranium, thorium, and potassium for various fruit samples varies in the range of 2.5±0.16 Bq/kg to 9.8±0.15 Bq/kg, 7.4±1.24 Bq/kg to 18.4±1.39 Bq/kg, and 287.13±11.23 Bq/kg to 815.72±12.50 Bq/kg respectively. The radium equivalent activity varies in the range of 47.91 Bq/kg to 80.02 Bq/kg.

The radiological hazards in terms of external hazard index (H_{ex}), Internal hazard index (H_{in}), Gamma index (I_γ) and Alpha index (I_α) for the samples calculated using equations (5.3), (5.4), (5.5) & (5.6) are given for vegetable samples and for fruit samples in table 5.3 and 5.4 respectively.

Table 5.3: External hazard index (H_{ex}), internal hazard index (H_{ex}), gamma index **(**I_{γ}) and alpha index (I_{α}) in vegetable samples.

S. No.	Sample Code	H_{ex}	H_{in}	I_{γ}	I_{α}
1	VC-1	0.48	0.57	0.69	0.14
2	VC -2	0.59	0.64	0.87	0.08
3	VC -3	0.51	0.59	0.74	0.11
4	VC -4	0.56	0.54	0.84	0.15
5	VC -5	0.51	0.68	0.76	0.14
6	VC -6	0.51	0.73	0.77	0.05
7	VC -7	0.62	0.65	0.92	0.10
8	VC -8	0.68	0.58	1.00	0.09
9	VC -9	0.60	0.54	0.88	0.09
10	VC -10	0.55	0.57	0.83	0.06
11	VC -11	0.49	0.64	0.74	0.10

Table 5.4: External hazard index (H_{ex}), internal hazard index (H_{ex}), gamma index **(**I_{γ}) and alpha index (I_{α}) in fruit samples

S. No.	Sample Code	H_{ex}	H_{in}	I_{γ}	I_{α}
1	FR-1	0.21	0.23	0.32	0.04
2	FR-2	0.18	0.19	0.27	0.01
3	FR-3	0.16	0.17	0.24	0.02
4	FR-4	0.13	0.14	0.19	0.02
5	FR-5	0.13	0.16	0.19	0.05

6	FR-6	0.16	0.17	0.23	0.03
7	FR-7	0.20	0.21	0.30	0.02
8	FR-8	0.21	0.22	0.32	0.02
9	FR-9	0.22	0.24	0.32	0.04
10	FR-10	0.16	0.18	0.24	0.04

Table 5.3 and 5.4 shows that external hazard indices varies from 0.48 to 0.68 for samples of vegetable and it varies in the range of 0.13 to 0.22 for various fruit samples; the internal hazard index varies from 0.54 to 0.73 for various vegetable samples and varies from 0.14 to 0.24 for various fruit samples, the gamma index varies from 0.69 to 1.00 for various vegetable samples and varies from 0.19 to 0.32 for various fruit samples, the alpha index varies from 0.05 to 0.15 for various vegetable samples and varies from 0.01 to 0.05 for various fruit samples studied in the present work.

Table 5.5: Annual effective dose and radiation absorbed dose rate of vegetable samples

S. No.	**Sample code**	**Absorbed dose rate ($nGyh^{-1}$)**	**Annual effective dose (mSv)**	
			Indoor	**Outdoor**
1	VC-1	86.83	0.425	0.106
2	VC -2	107.78	0.528	0.132
3	VC -3	91.97	0.451	0.112
4	VC -4	105.48	0.517	0.129
5	VC -5	95.37	0.467	0.117
6	VC -6	96.15	0.471	0.118
7	VC -7	114.83	0.563	0.141
8	VC -8	125.18	0.614	0.153
9	VC -9	110.01	0.539	0.135
10	VC -10	103.35	0.507	0.126

11	VC -11	92.67	0.454	0.113

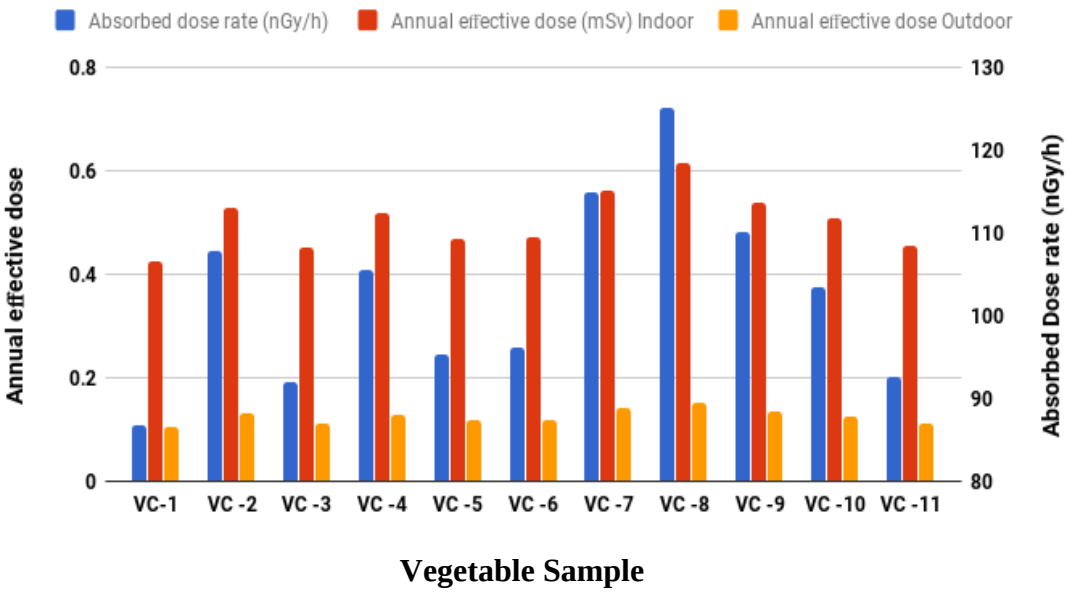

Figure 5.4: Annual effective dose and radiation absorbed dose rate in vegetable samples

Table 5.6: Annual effective dose and radiation absorbed dose rate of fruit samples

S. No.	Sample code	Absorbed dose rate ($nGyh^{-1}$)	Annual effective dose (mSv)	
			Indoor	Outdoor
1	FR-1	39.82	0.195	0.048
2	FR-2	33.41	0.163	0.041
3	FR-3	29.97	0.147	0.036
4	FR-4	23.94	0.117	0.029
5	FR-5	23.39	0.114	0.028
6	FR-6	28.93	0.141	0.035
7	FR-7	37.89	0.185	0.046
8	FR-8	40.10	0.196	0.049
9	FR-9	39.52	0.193	0.048

10	FR-10	29.63	0.145	0.036

Annual effective dose and radiation absorbed dose rate from vegetables samples and fruit samples were calculated using equations (5.7), (5.8) & (5.9) and are given in table 5.5 and table 5.6 respectively. Table 5.5 shows that the values of absorbed dose varied from 86.83 $nGyh^{-1}$ to 125.18 $nGyh^{-1}$, indoor annual effective dose varied from 0.425 mSv to 0.614 mSv and outdoor annual effective dose varied from 0.106 mSv to 0.153 mSv for various vegetable samples under investigation. Table 5.6 shows that the calculated values of absorbed dose varied from 23.39 $nGyh^{-1}$ to 40.10 $nGyh^{-1}$ and annual effective dose (indoors and outdoors) varied from 0.114 mSv/y to 0.196 mSv/y and 0.028 mSv/y to 0.049 mSv/y, respectively for fruit samples.

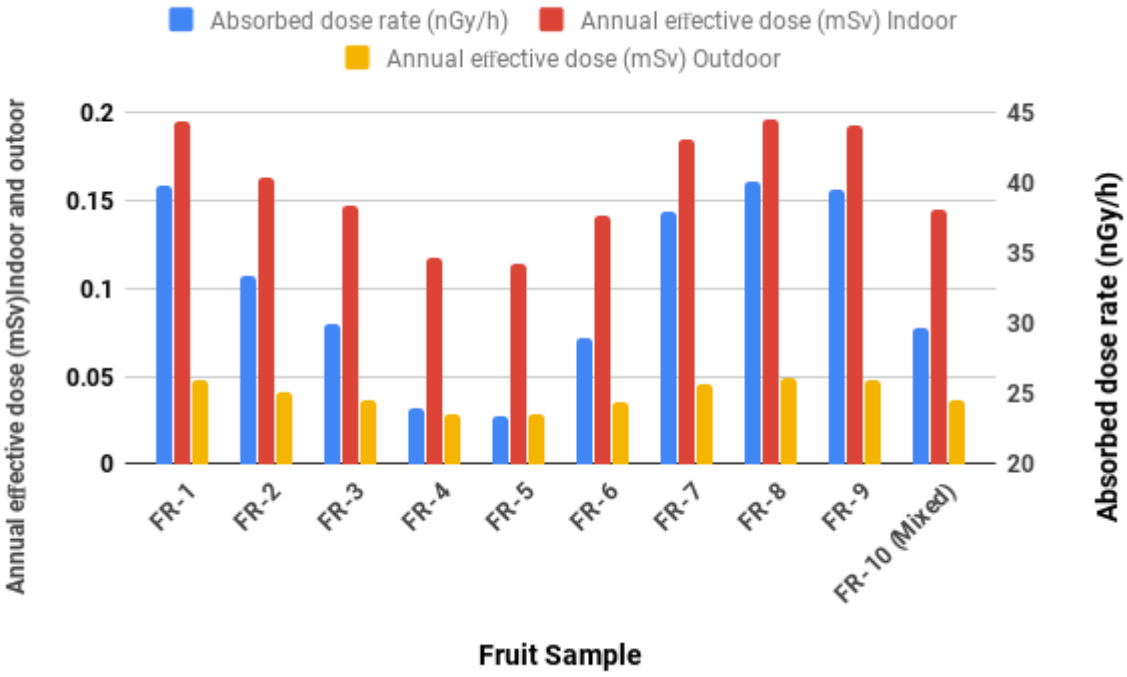

Figure 5.5: Annual effective dose and radiation absorbed dose rate of fruit samples

5.5.4 Discussion

Results from the experiments indicate that the concentration of radium, thorium and potassium and the radium equivalent activity varied significantly for various samples,

one of the reason for this variation can be the quantity and type of fertilizer used in the fields [33]. The radium equivalent activity and activity concentration in the fruit samples in which or less fertilizer was used, is found to be significantly lower than that in the vegetable samples collected from market and crop fields where fertilizers were used to enhance the crop yield.

The findings of the experiments are in agreement with the findings of other researchers reported in the literature [33-37]. The concentration of the radionuclide ^{40}K is higher in all the vegetable samples in comparison to fruit samples. It can also be seen that the value of radium equivalent activity is under the safe limit of 370 Bq/kg [38].

The calculated values of external hazard index and internal hazard index for all samples are less than unity and therefore these vegetations are safe for consumption [38]. The value of gamma index I_γ less than 2 corresponds to a dose rate criterion of 0.3 mSv/y, whereas when the value of gamma index I_γ is between 2 to 6 it is equivalent to 1 mSv/y. Thus the vegetation with gamma index I_γ greater than 6 should be avoided for consumption as this value of dose rate lies in the range is higher than the recommended safe limit values [39]. It can be seen that for all the samples the 'I_γ' values follow the criterion ($I_\gamma \leq 2$) therefore it can be concluded that the samples are safe from health and hygiene point of view and will not pose any significant health hazards to the consumers. When the value of I_α is 1, it indicates that the concentration of ^{226}Ra is 200 Bq/kg, which is prescribed safe limit [40]. The obtained value of I_α for all the samples is under unity hence it can be concluded that the samples are safe for consumption and will not pose any environmental radiation hazards.

The indoor annual effective dose for all the samples was found to be less than the safety limit of 1 mSv/y as recommended for the general public [41]. The absorbed dose and uranium activity shows a positive correlation between them. It was observed that in vegetable where fertilisers were used have a higher absorbed dose and annual effective dose in comparisons to fruit samples.

5.6 ACTIVITY CONCENTRATION AND ANNUAL EFFECTIVE INGESTION DOSE ASSESSMENT DUE TO NATURAL RADIONUCLIDES PRESENT IN CEREAL SAMPLES

This study includes the measurement of activity concentration and annual dose assessment due to radionuclides in cereal samples. Radioactivity present in the foodstuffs gets enhanced due to the use of phosphate fertilisers to enhance the crop yield. In NPK fertilisers (nitrogen, phosphorus, and potassium), potassium component augments the natural radioactivity [42]. It is the predominant radioactive component in human tissues and in most of the food items [43]. So the activity concentration of radionuclides in foodstuffs should also be measured to predict their effects on human health.

In this investigation, cereal samples (wheat, rice and coarse grains) commercially available in the market and from the fields were collected randomly from different locations of the four districts Faridabad, Palwal, Gurgaon, and Mewat of Haryana State of India, shown in figure 5.1. These samples were analyzed for the activity concentration of the natural radionuclides namely ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K using gamma spectrometry as carried out by other researchers [44-47]. All the samples were processed following the standard procedures as per the International Atomic Energy Agency (IAEA) guidelines [48]. The samples were processed using gamma-ray spectroscopy.

5.6.1 Results and Discussion

The activity concentration of uranium, thorium, and potassium and radium equivalent activity calculated using equations (5.1) & (5.2) in the cereal samples. The values of the results are shown in table 5.7 and represented graphically in figure 5.6. The uncertainty in all measured values has been also calculated and reported.

Table 5.7: The activity concentration of uranium, thorium, and potassium in cereal samples

S. No.	Sample code	Activity concentration (Bq/kg)			
		^{226}Ra	^{238}U	^{232}Th	^{40}K
1	VC-1	28.08 ± 1.28	19.02 ± 0.48	42.31 ± 2.17	1158.4 ± 26.05
2	VC-2	15.55 ± 1.77	11.12 ± 0.77	57.52 ± 2.34	1579.2 ± 17.58
3	VC-3	22.72 ± 1.63	16.75 ± 1.48	47.32 ± 1.35	1268.5 ± 15.05
4	VC-4	29.13 ± 0.69	20.08 ± 1.32	22.20 ± 2.46	1885.1 ± 28.03
5	VC-5	28.22 ± 1.62	16.74 ± 1.08	26.48 ± 1.41	1590.9 ± 19.21
6	VC-6	10.25 ± 0.94	7.04 ± 0.13	36.37 ± 1.20	1665.4 ± 17.63
7	VC-7	19.94 ± 0.40	11.32 ± 0.47	49.17 ± 1.31	1820.6 ±27.24
8	VC-8	17.74 ± 1.59	10.64 ± 0.89	58.21 ± 1.15	1962.2 ± 18.17
9	VC-9	18.38 ± 1.15	12.15 ± 0.92	50.82 ± 1.17	1698.5 ± 28.04
10	VC-10	11.40 ± 0.90	8.23 ± 0.43	39.85 ± 2.32	1775.0 ± 26.48
11	VC-11	19.21 ± 1.61	10.83 ± 0.88	26.53 ± 1.89	1625.2 ± 18.28
12	Average ± SE*	20.06 ± 4.82	13.08 ± 1.31	41.53 ± 3.77	1639 ± 75.55

Notes: SE* is the standard error which is calculated as $\sigma/\sqrt{N}$, where σ is the standard deviation and N is the total no. of observations.

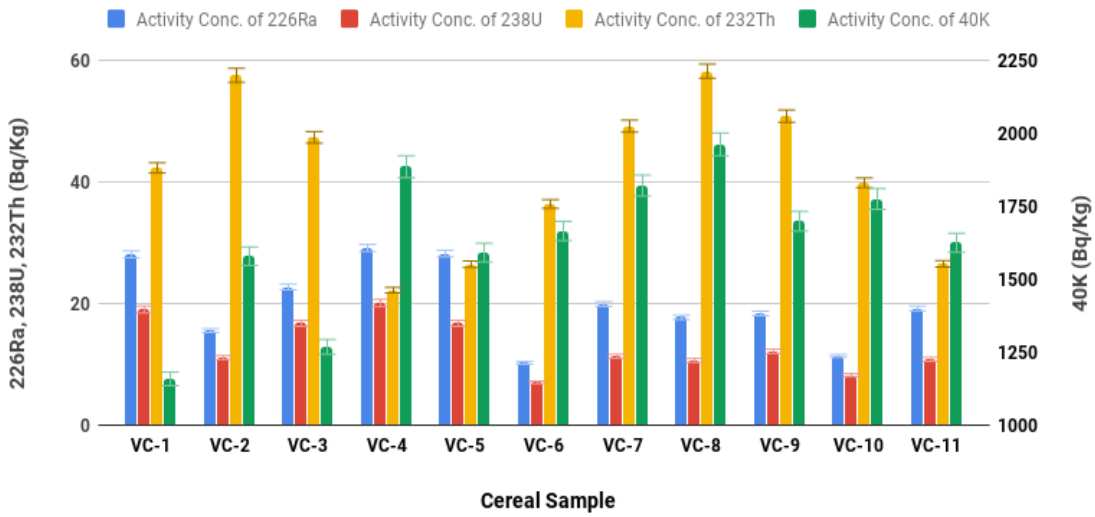

Figure 5.6: Bar diagram showing the activity concentration of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K in cereal samples

MADL (Minimum Activity Detection Limit) = 2 Bq/kg, 2Bq/kg and 4Bq/kg for ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K respectively.

From table 5.7, it is clear that the activity concentration for ^{226}Ra,^{238}U, ^{232}Th, and ^{40}K varied from 10.25 ± 0.94 Bq/kg to 29.13 ± 0.69 Bq/kg with an average value of 20.06 ± 4.82 Bq/kg, 7.04 ± 0.13 Bq/kg to 20.08 ± 1.32 Bq/kg with an average value of 13.08 ± 1.31 Bq/kg, 22.20 ± 2.46 Bq/kg to 58.21 ± 1.15 Bq/kg with an average value of 41.53 ± 3.77 Bq/kg, and 1158.4 ± 26.05 Bq/kg to 1962. ± 18.17 Bq/kg with an average value of 1639 ± 75.55 Bq/kg respectively in various cereal samples studied in the present work. From figure 5.7 it is clear that the activity concentration of ^{40}K was significant and the highest in all samples and much higher than that of uranium, radium, and thorium. This was probably due to the higher concentrations of ^{40}K, a micronutrient in the soil whose mobilization and subsequent migration might be facilitated to the plant through its roots [49-50] and also due to the heavy use of fertilisers by the farmers to enhance the crop yield. However, ^{40}K is a key element in regulating many functions and it is homeostatically controlled in living organisms. For that reason, its potassium content is held constant by metabolic processes [51].

Different values of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K in cereal samples may be due to the variation in the levels of natural radioactivity in soil and the different geology of the

area as the samples were collected from different locations in the study area. The second reason for the different activities in the sample may be due to the value of transfer factor because it is affected by the meta-selective function of the plant during the uptake of elements so as to maintain the mechanism of homeostasis in a normal environment [52]. The radionuclides concentrations in root vegetables, leafy vegetables, and grain products are less than 1Bq/kg [53] around the globe, except for a few countries [54]. This clearly suggests that all the samples under investigation have higher radionuclides concentration, but the annual effective ingestion dose is considered as a crucial parameter to study the effect of radionuclides on the human being.

Using the above equations (5.10), (5.11) and (5.12), the annual effective doses from ingested food (cereals) and cancer risks were calculated as shown in table 5.8 and figure 5.7.

Table 5.8: Annual effective ingestion dose, total ingestion dose and cancer risk due to the intake of ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K radionuclides in the cereal samples.

S. No.	Sample code	Annual effective ingestion dose				Total ingestion dose	Cancer risk x 10^{-4}
		^{226}Ra	^{238}U	^{232}Th	^{40}K		
1	VC-1	0.99	0.10	1.23	0.90	3.22	80.5
2	VC-2	0.55	0.06	1.67	1.23	3.51	87.8
3	VC-3	0.80	0.10	1.37	1.00	3.27	81.8
4	VC-4	0.10	0.11	0.64	1.47	2.32	58.0
5	VC-5	1.00	0.09	0.77	1.24	3.10	77.5
6	VC-6	0.36	0.03	1.05	1.30	2.74	95.9
7	VC-7	0.70	0.06	1.42	1.42	3.60	90.0
8	VC-8	0.63	0.06	1.69	1.53	3.91	97.8
9	VC-9	0.65	0.07	1.47	1.33	3.52	88.0

10	VC-10	0.40	0.05	1.15	1.39	2.99	74.7
11	VC-11	0.68	0.06	0.77	1.27	2.78	69.5
12	Average ± SE*	0.62 ±0.08	0.07 ±0.006	1.20 ±0.11	1.28 ±0.06	3.18 ±0.14	79.5 ±3.55

Note: SE* is the standard error which is calculated as $\sigma/\sqrt{N}$, where σ is the standard deviation and N is the total number of observations.

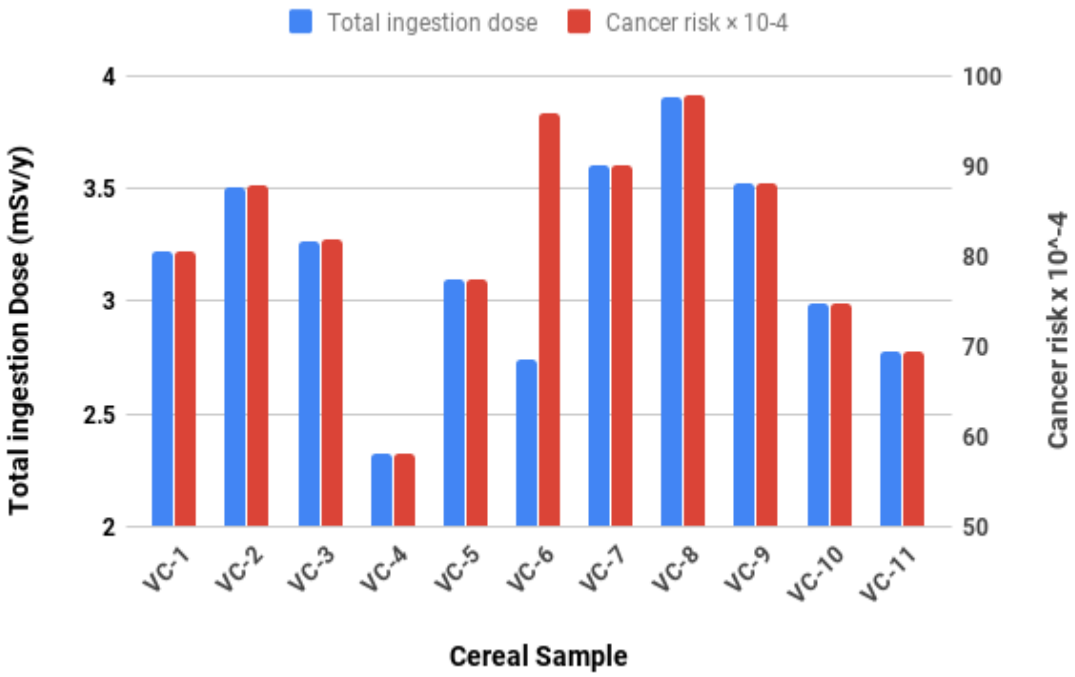

Figure 5.7: Total ingestion dose and cancer risk

Table 5.8 shows that the minimum and maximum values of annual effective ingestion dose from ingested food varies from 0.36 mSv/y to 0.99 mSv/y with an average value of 0.62 ± 0.08 mSv/y, 0.03 mSv/y to 0.11 mSv/y with an average value of 0.07 ± 0.006 mSv/y, 0.64 mSv/y to 1.69 mSv/y with an average value of 1.20 ± 0.11 mSv/y and 0.90 mSv/y to 1.53 mSv/y with an average value of 1.28 ± 0.06 mSv/y from ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K, respectively in various cereal samples under investigation. The value of total annual ingestion dose varied from 2.32 mSv/y to 3.91 mSv/y with an average value of 3.18 ± 0.14 mSv/y, and cancer risk varied from 58.0×10^{-4} to 97.8×10^{-4} with an average value of $(79.5 \pm 3.55) \times 10^{-4}$. The correlation between total

ingestion dose and cancer risk is shown in figure 5.8. This shows a strong positive correlation with $R^2 = 0.559$ between the total ingestion dose and cancer risk.

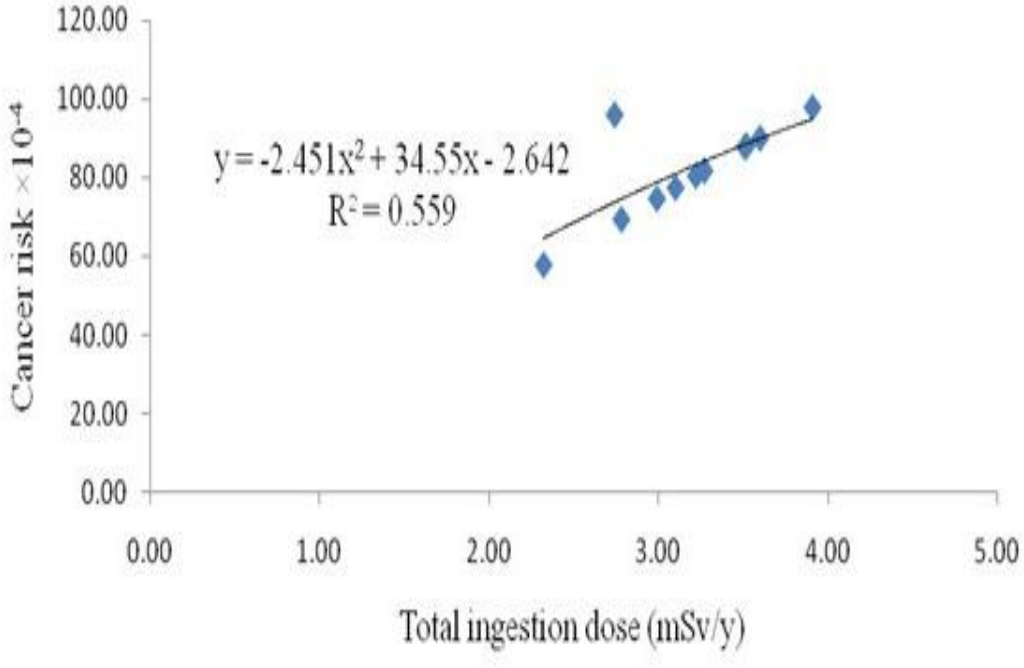

Figure 5.8: The correlation between the total ingestion dose and cancer risk

The higher values of the total annual effective ingestion doses due to the intake of cereals can be attributed to the scale of the annual intake (126 kg/year) [55] when compared with a few kilograms per year for the total food intake in terms of fruits, spices etc. When compared with the global dose due to ingestion of naturally occurring ^{226}Ra, ^{238}U, ^{232}Th, ^{40}K radionuclides excluding radon and thoron as reported by UNSCEAR (2000) [56], most samples show significant increases in their ingestion doses over the reported values. The average value of cancer risk is significantly higher than that of health risks which gives a risk factor of 0.48×10^{-4} due to spices [57] and higher than the world average (2.9×10^{-4}) based on annual dose limit of 1 mSv for the general public [58]. The findings of the present study are in good agreement with the results of other studies carried out in India and abroad [59-64].

5.7 NATURAL RADIOACTIVITY IN ROCK SAMPLES OF ARAVALI HILLS IN INDIA

The natural radionuclides exist in different geological formations like air, water, plants, soils, and rocks. These radionuclides exist since the inception of the earth. The study of natural radioactivity level is carried out to have information about the current levels of radiological pollution which is discharged to the environment or to living beings. It is very necessary to determine the effects of radiation exposure on humans. To estimate the possible hazard to human health, many studies for the measurement of natural radioactivity from various natural resources had been carried out [65-70]. The present study to estimate the radiation exposure from the rock samples of Aravali hills in India is also a part of those studies. In this investigation, 20 rock samples were collected from various altitudes and places of Aravali Range in Haryana, Delhi, Rajasthan, and Gujarat and analyzed using gamma spectroscopy.

The samples were prepared for the process of gamma spectroscopy by initially crushing the samples into a fine powder using mortar and pestle. A 250 micron-mesh sieve is used to obtain the fine quality of particle size less than 250 microns. The samples were kept in the oven at 110^{o}C for 24 hours to make sample free from moisture. The airtight container is used to pack and seal the samples for about 4 weeks to establish the radioactive equilibrium among the radon, thoron and their progeny. About 300 grams of the sample is taken in powder form.

5.7.1 Sample Collection Sites

The Geographical locations of the sample collection sites are shown in figure 5.9. The collection of rock samples were carried out from the Aravali Range, this Aravali range exists about 695 km in a northeastern direction across India, through the states of Gujarat, Rajasthan, Haryana, and Delhi [71]. The Guru Shikhar in Mount Abu (Rajasthan) is its highest peak at a height of 1722 meters [72]. The Aravali ranges are sources of copper and other metals and it is estimated that mining in these ranges dates back to 5th century BC [73]. The mostly found mines in Haryana are of slate stone, limestone, china clay, building stone, dolomite, and marble.

Figure 5.9: Rock sample collection sites in India

5.7.2 Result and Discussion

The radium equivalent activity and activity concentration of uranium (^{238}U), thorium (^{232}Th), and potassium (^{40}K) were calculated using equations (5.1) & (5.2) in the rock samples. The values of the results are shown in table 5.9 and represented graphically in figure 5.10.

Table 5.9: The activity concentration uranium (^{238}U), thorium (^{232}Th), and potassium (^{40}K) and radium equivalent activity in rock samples

S. No	Sample code	Sample collection site	Activity concentration (Bq/kg)			Radium equivalent Concentration Ra (Bq.kg^{-1})
			^{238}U	^{232}Th	^{40}K	

1	ARS-1	Aravali Biodiversity Park, Gurugram (Haryana)	3.45	17.29	58.01	32.64
2	ARS -2		5.78	45.32	123.33	80.08
3	ARS -3		7.58	67.56	89.87	111.11
4	ARS -4		4.24	34.41	243.47	72.19
5	ARS -5		6.89	56.86	555.08	130.94
6	ARS -6	Silsher Lake, Alwar (Rajasthan)	5.56	43.23	857.58	133.41
7	ARS -7		16.12	12.34	933.20	105.62
8	ARS -8		13.78	18.18	1547.49	158.93
9	ARS -9		19.21	56.25	1822.78	240.00
10	ARS -10		22.56	72.89	687.56	179.73
11	ARS -11	Bhatti Mines, Tughlaqabad, Delhi	11.76	32.56	876.32	125.80
12	ARS -12		8.24	28.32	432.43	82.03
13	ARS -13		9.67	10.17	257.78	44.06
14	ARS -14		6.53	16.12	98.17	37.14
15	ARS -15		7.29	24.27	376.09	70.96
16	ARS -16	Taranga hills, Vadnagar (Gujrat)	23.31	53.47	521.23	139.91
17	ARS -17		32.18	42.21	289.18	114.81
18	ARS -18		17.33	37.34	1088.28	154.52
19	ARS -19		11.59	102.18	1470.56	270.94
20	ARS -20		9.83	132.45	456.37	234.37
Range			3.45-32.18	10.17-132.45	58.01-1822.78	32.64-270.94
Mean ± SE			12.15 ±1.68	45.17 ±6.92	639.24 ±115.86	125.96 ±14.94

Considering N is the total no. of observations, σ is the standard deviation the SE is the Standard Error which is given as σ/√N.

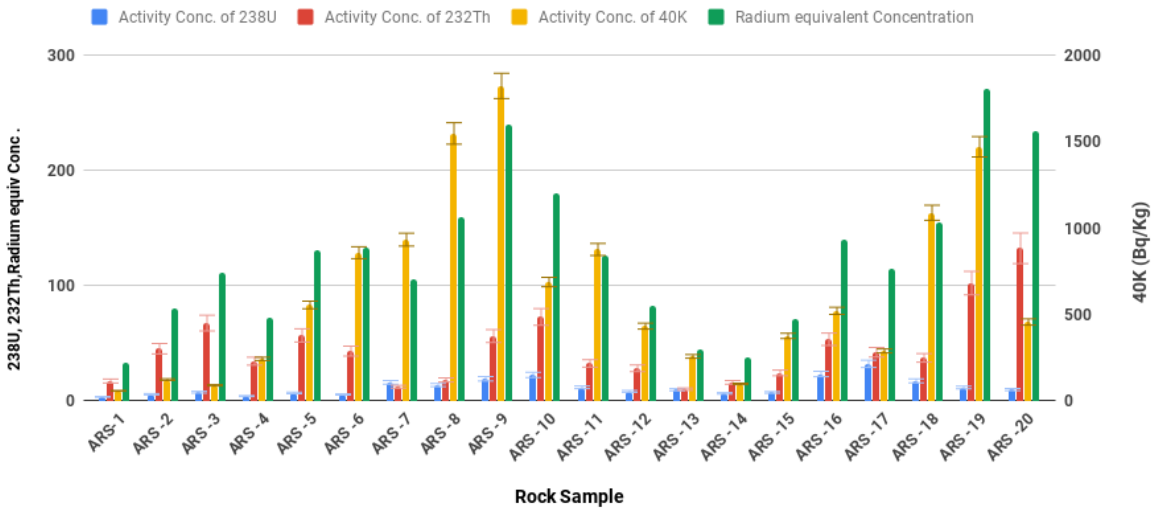

Figure 5.10: The activity concentration of uranium, thorium and potassium and radium equivalent activity in rock samples

It can be seen that from table 5.9 that the activity concentration for uranium (^{238}U), thorium (^{232}Th), and potassium (^{40}K) varies from 3.45 Bq/kg to 32.18 Bq/kg with a mean value of 12.15 ± 1.68 Bq/kg, 10.17 Bq/kg to 132.45 Bq/kg with a mean value of 45.17 ± 6.92 Bq/kg and 58.01 Bq/ kg to 1822.78 Bq/kg with a mean value of 639.24 ± 115.86 Bq/kg respectively. The mean values of concentration of the radionuclides are found within the safe limit as compared with the world average of 33, 45 and 420 Bq/kg, respectively and Indian average is of 28.67, 63.83 and 400 Bq/kg respectively [74-75], however the activity concentration of ^{40}K was found slightly higher. This was probably due to the higher concentrations of ^{40}K, a micronutrient in the soil whose mobilization and subsequent migration might be the reason for these higher values.

The value of radium equivalent activity ranges from 32.64 Bq/kg to 270.94 Bq/kg having a mean value of 125.96 ± 14.94 Bq/kg which is under the safe limit of 370 Bq/kg [38]. It can be seen that the radium equivalent activity and concentration of uranium (^{238}U), thorium (^{232}Th), and potassium (^{40}K) radium varied significantly in different samples. The values of various hazard indexes are shown in table 5.10.

Table 5.10: External hazard index (H_{ex}), internal hazard index (H_{in}), gamma index **(**I_{γ}) and alpha index (I_{α}) in rock samples

S. No.	Sample code	H_{ex}	H_{in}	I_{γ}	I_{α}
1	ARS-1	0.09	0.10	0.12	0.02
2	ARS -2	0.22	0.23	0.29	0.03
3	ARS -3	0.30	0.32	0.39	0.04
4	ARS -4	0.19	0.21	0.27	0.02
5	ARS -5	0.35	0.37	0.49	0.03
6	ARS -6	0.36	0.38	0.52	0.03
7	ARS -7	0.29	0.33	0.43	0.08
8	ARS -8	0.43	0.47	0.65	0.07
9	ARS -9	0.65	0.70	0.95	0.10
10	ARS -10	0.49	0.55	0.67	0.11
11	ARS -11	0.34	0.37	0.49	0.06
12	ARS -12	0.22	0.24	0.31	0.04
13	ARS -13	0.12	0.15	0.17	0.05
14	ARS -14	0.10	0.12	0.14	0.03
15	ARS -15	0.19	0.21	0.27	0.04
16	ARS -16	0.38	0.44	0.52	0.12
17	ARS -17	0.31	0.40	0.41	0.16
18	ARS -18	0.42	0.46	0.61	0.09
19	ARS -19	0.73	0.76	1.04	0.06
20	ARS -20	0.63	0.66	0.85	0.05
Range		3.45 -32.18	10.17 -132.45	58.01 -1822.78	32.64 -270.94
Mean ± SE		12.15 ±1.68	45.17 ±6.92	639.24 ±115.86	125.96 ±14.94

Considering N is the total no. of observations, σ is the standard deviation the SE is the Standard Error which is given as σ/√N.

The recommended limit for the concentration of ^{226}Ra is 200 Bq/kg, for this value gamma index is 1 [76]. Most of the observed values are less than unity except for one sample. Hence it can be said that most of the samples do not pose any environmental radiation hazards and are safe from the health point of view. The radiation absorbed dose rate and annual absorbed dose from rock samples is calculated using equations (5.10), (5.11) and (5.12) and are given in table 5.11.

Table 5.11: Annual effective dose and radiation absorbed dose and from rock samples

S. No.	Sample code	Absorbed dose rate ($nGyh^{-1}$)	Annual effective dose (mSv)	
			Indoor	Outdoor
1	ARS-1	14.46	0.07	0.02
2	ARS -2	35.19	0.17	0.04
3	ARS -3	48.06	0.24	0.06
4	ARS -4	32.90	0.16	0.04
5	ARS -5	60.67	0.30	0.07
6	ARS -6	64.44	0.32	0.08
7	ARS -7	53.82	0.26	0.07
8	ARS -8	81.88	0.40	0.10
9	ARS -9	118.86	0.58	0.15
10	ARS -10	83.12	0.41	0.10
11	ARS -11	61.64	0.30	0.08
12	ARS -12	38.94	0.19	0.05
13	ARS -13	21.36	0.10	0.03
14	ARS -14	16.85	0.08	0.02
15	ARS -15	33.71	0.17	0.04
16	ARS -16	64.80	0.32	0.08

17	ARS -17	52.42	0.26	0.06
18	ARS -18	75.94	0.37	0.09
19	ARS -19	128.39	0.63	0.16
20	ARS -20	103.57	0.51	0.13
Range		14.46-128.39	0.07-0.63	0.02-0.16
Mean ± SE		59.55±7.16	0.04±0.29	0.07±0.00

Considering N is the total no. of observations, σ is the standard deviation the SE is the Standard Error which is given as σ/√N.

It can be seen that from table 5.11 that absorbed dose rate varies in range of 14.46 nGyh^{-1} to 128.39 nGy h^{-1} with a mean value of 59.55 ± 7.16 nGy h^{-1} which is close to the World (57 nGyh^{-1}) and Indian (56 nGyh^{-1}) average values [77].

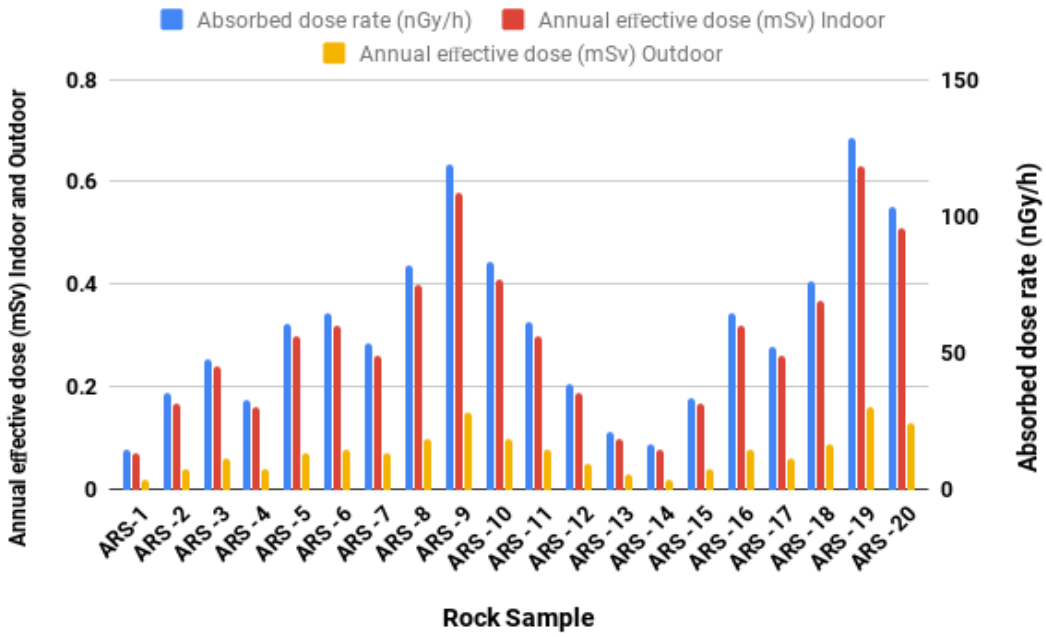

Figure 5.11: Radiation absorbed dose and annual effective dose from rock samples

The annual effective dose calculated using equations (5.11) and (5.12) varies in range of 0.07 mSv to 0.63 mSv with a mean value of 0.29 ± 0.04 mSv and 0.02 mSv to 0.16 mSv with a mean value of 0.07± 0.00 mSv respectively. The indoor annual effective dose was found less than the recommended safety limit of 1 mSy/y for all the samples [78]. A positive correlation with $R^2 = 0.99$ can be seen between the radium equivalent activity and the absorbed dose as shown in figure 5.12.

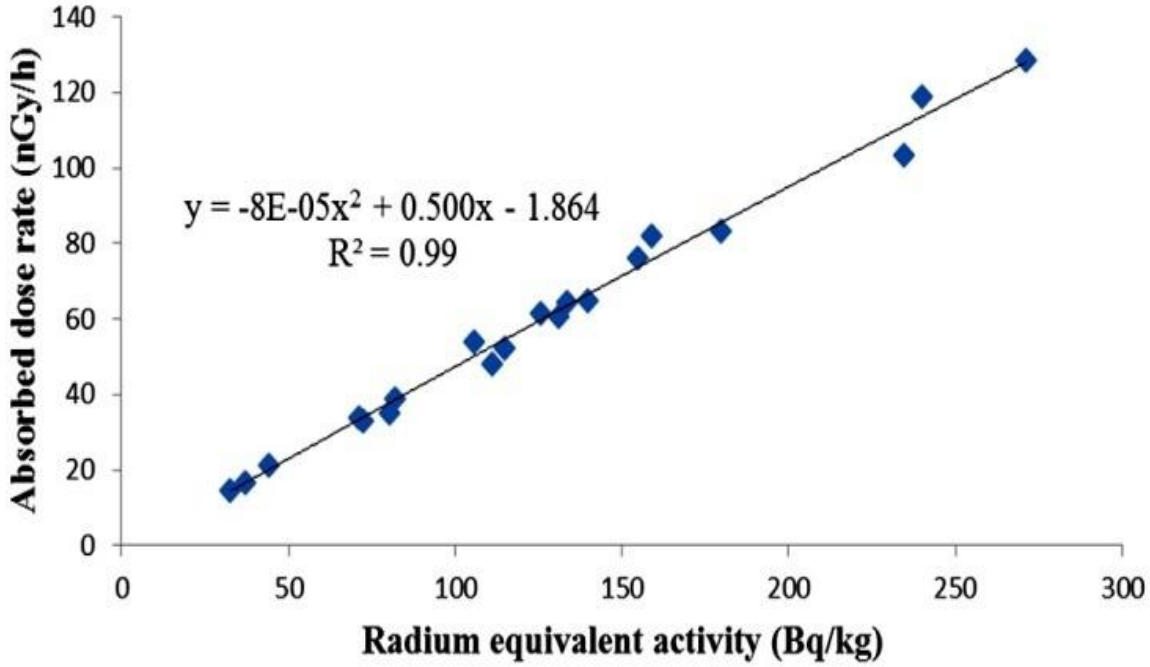

Figure 5.12: The correlation between absorbed dose and radium equivalent activity

5.8 MEASUREMENT OF NATURAL RADIOACTIVITY IN FLY-ASH SAMPLES

The building materials used in construction contain radioactive materials. Now-a-day industrial by-products containing Technologically Enhanced Naturally Occurring Radioactive Materials (TENORMs) are widely used in the construction materials [79]. Coal ash or fly-ash, a kind of TENORM, is a byproduct produced during the combustion of coal. The fly-ash is used as an additive to concrete, in bricks and mostly in cement [80]. It has been reported that the combustion of coal in various thermal power plants results in the release of only some natural radioactivity to the environment and the largest part of the coal radioactivity remains with the ashes [81-82]. When used in building construction materials it becomes potentially hazardous to humans. Measurement of radioactivity in fly-ash samples in terms of activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, radon concentration and radon exhalation rate in fly-ash is due to its health hazards posed to human and environmental pollution.

Emanation rate of radon from construction materials has also been investigated in many studies [83-86] as the construction materials are a considerable source of radon in the dwelling, hence from a radiation protection point of view, the measurement of radioactivity in these samples is very important.

In this investigation, fly-ash samples were collected from various thermal power plant in Haryana and Delhi viz. samples FA-1 to FA-4 were collected from Badarpur Thermal Power Plant Delhi, samples FA-5 to FA-8 were collected from Rajiv Gandhi Thermal Power Project, Hisar (Haryana), samples FA-9 to FA-12 were collected from Deen Bandhu Chhotu Ram Thermal Power Station, Yaumna Nagar (Haryana), samples FA-13 to FA-16 were collected from Panipat Thermal Power Station I, Panipat (Haryana) and samples FA-17 to FA-20 were collected from Panipat Thermal Power Station II, Panipat (Haryana). These samples were powdered and sieved to obtain particles of size less than 250 microns. The constant dry weight of the sample is obtained by keeping the samples at 110°C for 10 – 15 hours in an electric oven. Sieved samples were packed and sealed in 300 ml airtight PVC container and left for four weeks to establish radioactive equilibrium between the radon (^{222}Rn), thoron (^{220}Rn), and their progenies [53-54]. During the experiment, about 300 grams of ash was taken for each sample.

5.8.1 Results and Discussion

The concentration of the radionuclides, ^{238}U, ^{232}Th and ^{40}K, and radium equivalent activity has been calculated using equations (5.1) and (5.2) for the fly-ash samples under investigation are shown in table 5.12 and represented graphically in figure 5.13.

Table 5.12: Activity concentration of uranium (^{238}U), thorium (^{232}Th), and potassium (^{40}K) in fly-ash samples.

S. No.	Sample code	Activity concentration (Bq/kg)			Radium equivalent Concentration Ra (Bq/kg)
		^{238}U	^{232}Th	^{40}K	
1	FA-1	89±3	103±6	344±1	263
2	FA-2	76±3	123±2	154±2	264
3	FA-3	23±6	197±3	342±5	331
4	FA-4	89±4	212±1	786±6	453
5	FA-5	221±2	154±4	698±4	495
6	FA-6	89±1	123±5	BDL	453
7	FA-7	230±2	67±3	455±4	361
8	FA-8	98±5	103±2	654±4	296
9	FA-9	56±3	99±5	412±5	229
10	FA-10	112±4	45±1	BDL	243
11	FA-11	56±3	109±2	543±6	254
12	FA-12	69±1	125±2	876±5	315
13	FA-13	114±5	167±1	345±3	379
14	FA-14	109±6	67±4	453±2	240
15	FA-15	234±6	54±6	BDL	365
16	FA-16	169±3	BDL	BDL	432
17	FA-17	154±2	76±5	BDL	342
18	FA-18	64±4	123±3	659±5	291

19	FA-19	89±4	107±4	352±3	269
20	FA-20	106±5	BDL	230±2	321

BDL is below detection limit; MADL (Minimum Activity Detection Limit) is taken as 2Bq/kg, 2Bq/kg and 4Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K respectively.

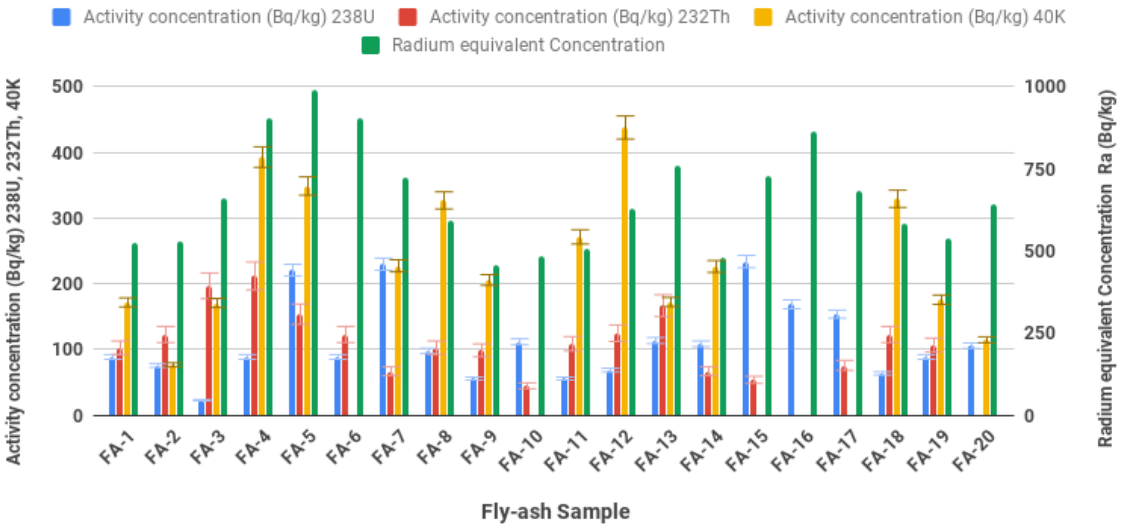

Figure 5.13: Activity concentration of uranium (^{238}U), thorium (^{232}Th), and potassium (^{40}K) in different fly-ash samples

The radiological hazards in terms of external hazard index (H_{ex}), internal hazard index (H_{in}), Gamma index (I_γ) and Alpha index (I_α) for the samples calculated using equations (5.3), (5.4), (5.5) and (5.6) are given in table 5.13.

Table 5.13: External hazard index (H_{ex}), internal hazard index (H_{ex}), gamma index **(**I_γ) and alpha index (I_α) in fly-ash samples

S. No.	Sample code	H_{ex}	H_{in}	I_γ	I_α
1	FA-1	0.71	0.95	0.93	0.45
2	FA-2	0.71	0.92	0.92	0.38
3	FA-3	0.89	0.96	1.18	0.12
4	FA-4	1.22	1.46	1.62	0.45
5	FA-5	1.34	1.93	1.74	1.11

6	FA-6	0.72	0.96	0.91	0.45
7	FA-7	0.97	1.60	1.25	1.15
8	FA-8	0.80	1.06	1.06	0.49
9	FA-9	0.62	0.77	0.82	0.28
10	FA-10	0.48	0.78	0.60	0.56
11	FA-11	0.69	0.84	0.91	0.28
12	FA-12	0.85	1.04	1.15	0.35
13	FA-13	1.02	1.33	1.33	0.57
14	FA-14	0.65	0.94	0.85	0.55
15	FA-15	0.84	1.47	1.05	1.17
16	FA-16	0.46	0.91	0.56	0.85
17	FA-17	0.71	1.13	0.89	0.77
18	FA-18	0.78	0.96	1.05	0.32
19	FA-19	0.73	0.97	0.95	0.45
20	FA-20	0.33	0.62	0.43	0.53

The annual effective dose and radiation absorbed dose of fly-ash samples were calculated using equations (5.7), (5.8) and (5.9) are given in table 5.14 and represented graphically in figure 5.14. The findings of the study are in good agreement with the findings of other researchers [87-89]. The concentration of the radionuclide ^{40}K is higher in some of the samples but it can be seen that the value of radium equivalent activity is well within the safe limits of 370 Bq/kg [38].

Table 5.14: Annual effective dose and radiation absorbed dose from fly-ash samples

S. No.	Sample code	Absorbed dose rate (nGy h^{-1})	Annual effective dose (mSv)	
			Indoor	Outdoor
1	FA-1	118	0.58	0.14
2	FA-2	116	0.57	0.14
3	FA-3	144	0.71	0.18
4	FA-4	202	0.99	0.25
5	FA-5	224	1.10	0.27
6	FA-6	115	0.57	0.14
7	FA-7	166	0.81	0.20
8	FA-8	135	0.66	0.16
9	FA-9	103	0.50	0.13
10	FA-10	79	0.39	0.09
11	FA-11	114	0.56	0.14
12	FA-12	144	0.71	0.18
13	FA-13	168	0.82	0.21
14	FA-14	110	0.54	0.13
15	FA-15	141	0.69	0.17
16	FA-16	78	0.38	0.09
17	FA-17	117	0.57	0.14
18	FA-18	131	0.64	0.16
19	FA-19	120	0.59	0.15
20	FA-20	59	0.29	0.07

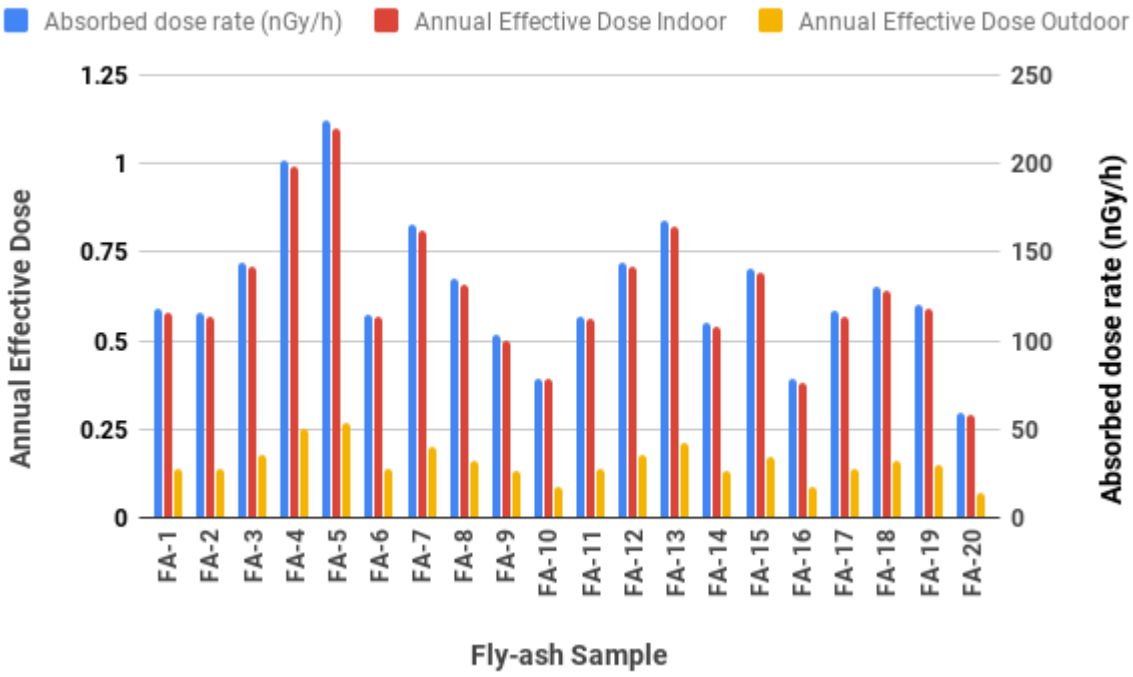

Figure 5.14: Annual effective dose and radiation absorbed dose from fly-ash samples

The H_{ex} and H_{in} for the studied samples are less than unity and therefore these samples are safe from a health point of view [90]. All the current 'I_γ' values follow the criterion ($I_\gamma \leq 2$) and $I_\alpha < 1$for concentration of ^{226}Ra to 200 Bq/kg, can be seen in the table 5.14 hence it may be concluded that the most of the samples are safe from health and hygiene point of view and don't pose any significant health hazards to the human. In all the samples, the indoor annual effective dose was found less than the recommended limit of 1 mSy/y for the general public [91].

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CHAPTER-VI

CONCLUSION AND SCOPE FOR FUTURE WORK

This chapter provides the conclusions of the thesis. In the present work, an extensive study was carried out to assess radioactivity related parameters like surface and mass exhalation rates and gamma activity of radon, annual effective dose, radionuclide concentration and health hazard index due to radioactivity.

Radon contributes maximum radiation dose to humans compared to all natural and manmade sources taken together, hence it is necessary to study the radon exhalation rates and gamma activity from different materials to minimize the radiological impact of radon on human health. Keeping these issues under consideration, the study was oriented into two different aspects.

1. The radon and its progeny can enter the human respiratory tract by inhalation; hence study was focussed on the materials which emanate radon and lies in the proximity of humans like soil, building material, and rocks. The materials which emanate radon and its progeny beyond the critical value, will pose a serious risk to human health. Hence the study was carried out to measure the radon emanation and radium content in these materials.
2. The other mechanism by which radon and its progeny can enter the human respiratory tract is via ingestion. The presence of radionuclides in soil escalate the radon and its progeny in plants and when these plants based foods like cereals, fruits, and vegetables are consumed directly or indirectly (when these affected plants were consumed by animals and human consume these animals either as non-vegetarian diet or consume milk/egg from these animals), the radionuclides can reach human respiratory tract. The fertilisers which are used to enhance the yield of crops also contain enhanced radioactive elements. Hence it becomes essential to assess the foodstuff for the presence of radioactive elements.

The present study covers the aspect of the radiological impact of radon on human health due to inhalation of radon and its progeny through air emitted from the soil,

rocks and building materials and ingestion of cereals, vegetables, and fruits which contains radionuclides.

6.1 CONCLUSION

The various conclusion which can be drawn from the obtained results are listed as

1. Mass and Surface Exhalation Rate

1.1. In the experiment involving soil samples, the radon area exhalation rate ranges from 363.3- 678.5 $mBq.m^{-2}h^{-1}$ and radon mass exhalation rate range from 13.4-28.6 $mBqkg^{-1}h^{-1}$. The values of radium concentration in all the soil samples were found to be lower than the safe limit and are comparable to the global average value of radium in the soil. Therefore the use of soil of this area in brick manufacturing for buildings construction is expected to be safe.

1.2. In the experiment involving various building materials, the value of surface exhalation rate in building materials varies in the range of 77.44 -793.98 $mBq.m^{-2}h^{-1}$ and value of mass exhalation rate varies in the range of 0.87-8.03 $mBqkg^{-1}h^{-1}$. The minimum value of surface and mass exhalation rate is found for the ceramic samples. The maximum value of surface and mass exhalation rates is found for granite samples, hence the minimal use of granite in the indoor environment is suggested.

1.3. In the experiment involving stone dust and natural dust, the value of exhalation rate is found to be increasing with a grain size of the samples because the porosity of the material increases with grain size, which results in the increase in emanation rate. The increased emanation rate causes an increased exhalation rate of the sample.

1.4. In the experiment involving rock samples collected from Aravali range in Faridabad, the measured value of mass exhalation rate varies in the range of 3.4-9.1$mBqkg^{-1}h^{-1}$. The value of the surface exhalation rate varies in the range of 75.9-202.7$mBqm^{-2}h^{-1}$. The highest value of exhalation rate was found from the samples collected from the fresh mine however the value of radon exhalation rate was found within the safe limit as recommended by OECD.

1.5. In the experiment involving the addition of fly-ash in different proportions to cement, the measured values of mass and surface exhalation rates of fly-ash and

cement are found to be in the range of 2.5-5.5 $mBqkg^{-1}h^{-1}$and 48.8-108.3 $mBqm^{-2}h^{-1}$ respectively. The minimum value of exhalation rate was found when no fly-ash was added and maximum exhalation rate was found in a sample having 60% cement and 40% fly-ash. As the ash is itself radioactive material adding it to cement will increase the radon exhalation rate; however the grain size of the fly-ash is small and mixing it in higher concentration will reduce the porosity of the mixture, hence the radon exhalation rate starts to decrease.

2. Activity Concentration of Uranium, Thorium, and Potassium

2.1. In the experiments to measure activity concentration in different vegetation samples using gamma-ray spectroscopy, the results of experiments reveal that the activity concentration and the radium equivalent activity in the fruit samples in which no fertilizer or less fertilizer was used, is significantly lower than that in the vegetable samples, where fertilizers were used to enhance the crop yield.

2.2. It was also observed that the higher value of activity concentration of ^{40}K is obtained as compared to uranium, radium, and thorium. This may be due to the higher concentrations of ^{40}K in the soil, which is a micronutrient whose mobilisation and subsequent migration is very less and it is facilitated to the plant through its roots and also due to the heavy use of fertilisers by the farmers to enhance the crop yield.

2.3. Different values of activity concentration of ^{238}U, ^{232}Th and ^{40}K in cereal samples were obtained which may be due to the variation in the levels of natural radioactivity in the soil and different geologies of the area as the samples were collected from a different location in the study area. The transfer factor can be a second reason for different activities of radionuclides in the samples, due to the effects of the plant's meta-selective function during element uptake to maintain the homeostasis mechanism of a normal environment.

2.4. In the experiment to study activity concentration of natural radionuclides in rock samples, the calculated value of activity concentration for ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K varies from 3.45-32.18 Bq/kg with a mean value of 12.15±1.68 Bq/kg, 10.17 -132.45 Bq/kg with a mean value of 45.17±6.92 Bq/kg and 58.01 Bq/kg-1822.78 Bq/kg with a mean value of 639.24±115.86 Bq/kg, respectively in various rock samples under investigation. This higher value of activity

concentration for ^{40}K may be due to less mobilization and subsequent migration of ^{40}K in soil or rock.

2.5. The result of present investigations to measure the activity concentrations of fly ash shows inborn radioactivity in coal samples was modified during technological enhancement (converting coal into ash) but all the fly-ash samples were found to have activity concentration in the safe limit.

3. Radium Equivalent Activity

The calculated value of radium equivalent activity in various samples is well within the safety limit of 370 Bq/kg [1-2].

3.1. The radium equivalent activity varied from 32.64 Bq/kg to 270.94 Bq/kg with a mean value of 125.96 to 14.94 Bq/kg in the rock samples.

3.2. The radium equivalent activity varied from 177.78 Bq/kg to 252.07 Bq/kg in the vegetable samples.

3.3. The radium equivalent activity varied from 47.91 Bq/kg to 80.02 Bq/kg in fruit samples.

4. Absorbed dose, Indoor, and Outdoor Annual Effective Dose

4.1. The value of total ingestion dose in cereal samples varies in the range of 2.32-3.91 mSv/y with an average value of 3.18 ± 0.14 mSv/y, and cancer risk varies from 58.0×10^{-4} - 97.8×10^{-4} with an average value of $79.5 \pm 3.55 \times 10^{-4}$.

4.2. The average annual effective ingestion dose from cereals was 3.18 mSv/y, which is 10.96 times higher than world safe value of total exposure per person resulting from the ingestion of terrestrial radioisotopes. The results clearly indicate that use of fertilizers to enhance crop yield results in an increase in the radioactivity and the annual effective ingestion dose.

4.3. The absorbed dose and annual effective dose for the vegetable and cereal samples in which fertilizers were used to enhance the crop yield were found to be higher than that in fruit samples where less fertilizer was used.

4.4. The annual effective dose in most of the rocks and fly-ash samples was found to lies within the safe limit of 1 mSv/y [3-4].

5. Hazard Index

It is found that all the index i.e. external hazard index, internal hazard index, gamma index and alpha index for most of the samples have a value less than 1, hence it is estimated that these samples are safe for use [5-6].

6.2 FUTURE SCOPE

In the present work, an attempt has been made to study in detail about the radioactivity in Naturally Occurring Radioactive Materials (NORMs). The investigation has brought some interesting points worthy of opening new areas of research

1. The Aravali range lies in the seismic zone and the underground radon activity can be a major precursor of an earthquake; further investigation can be carried out to find the relationship between the earthquakes and anomalous radon concentration.
2. A detailed study about the radioactivity from the technically enhanced radioactive materials may also be carried out to estimate the radioactivity from these materials and estimating the health hazards from those materials
3. Vegetation samples like cereals, vegetables, and fruits were covered in present work. However, research can be extended to dairy products and meat etc.
4. The study area of research can be broadened. So that location wise data of radon activity can be analysed.

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Raj Kumari is presently working as Assistant Professor, Physics Department in DAV Centenary College, Faridabad. She has obtained bachelor's degree in Physics Honours from Delhi University in 2007 and Master's degree in Physics from Jamia Milia Islamia University, Delhi in 2009. She is pursuing Ph.D. under the guidance of Dr. Maneesha Garg, Assistant Professor, HAS Department in J.C. Bose University of Science and Technology YMCA, Faridabad and Dr. Krishan Kant Gupta, Principal, Aggarwal college Ballabgrah (Faridabad). She has 9 years of teaching experience.

List of Publications

S.No.	Title of published paper	Journal	Whether peer reviewed
1.	Natural radioactivity in rock samples of Aravali in India, 2017, 15(4) (Impact factor: 1.69)	International Journal of Radiation Research	Yes
2.	Natural Radioactivity in Indian Vegetation Samples, April 2015, Volume 13, No 2, pp. 143-150 (Impact factor: 1.69)	International Journal of Radiation Research	Yes
3.	Activity concentration and annual effective ingestion dose assessment due to natural radio-nuclides present in cereal samples consumed by inhabitants of India, 2015 Vol.10, No.2, pp.155 – 168 (Impact factor: 0.21)	International Journal of Low radiation	Yes
4.	The effect of grain size on radon exhalation rate in natural- dust and stone- dust samples", 80, 2015, pp.128 –130. (Impact factor: 0.70)	Physics Procedia	Yes
5.	Measurement of radium concentration and radon exhalation rates of soil samples collected from some areas of district Faridabad, Haryana 7(1), 2016, pp. 6-8.	ISST Journal of Applied Physics	Yes
6.	Study of radon and thoron concentration in the dwellings of Faridabad, Southern Haryana, India 7(1), 2016, pp.13-15.	ISST Journal of Applied Physics	Yes
7.	Measurement of radon exhalation rate in some building materials using plastic track detectors 7(1), 2016, pp.16-18.	ISST Journal of Applied Physics	Yes
8.	Study of Effect of Addition of Fly Ash on Radon Exhalation Rate in Cement Samples. Volume 5, No. 2, 2014, pp. 64-66.	ISST Journal of Applied Physics	Yes
9.	Study of Radon Exhalation and Emanation Rates from Fly Ash Samples, Volume 5(2), 2014, pp.96-100.	ISST Journal of Applied Physics	Yes