ENVIRONMENTAL RADIOACTIVITY STUDIES IN WAYANAD, KERALA, INDIA

A THESIS

Submitted to the University of Calicut in partial fulfillment

of the requirements for the award of the degree of

DOCTOR OF PHILOSOPHY IN PHYSICS

by

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under the guidance of

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October 2018

DECLARATION

I hereby declare that the work presented in this thesis entitled **"*Environmental Radioactivity Studies In Wayanad, Kerala, India*"** is based on the original work done by me under the guidance of Dr. C D Ravikumar, Associate Professor, Department of Physics, University of Calicut, Calicut University P. O., Kerala, India. The subject matter presented in this thesis has not previously been formed the basis for the award of any degree, diploma, associateship, fellowship or any other similar title except where the due acknowledgement has been made in the text.

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Instances where selected sources appear:

4

to

my Parents

for giving me the torch

and my Teachers

for showing me the light

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It was my interest to travel (read travel, purely for academic purpose!!) that prompted me to take a subject like this. The day I presented the synopsis, for approval, one big revelation came my way, 'this is not going to be an easy route to travel'. Every step of this work was completed only because many were willing to lend their hand, unconditionally. Since what are visible in the thesis are the results of the study, I mention and acknowledge the kind hearted support received from various quarters, at different stages, here.

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SYNOPSIS

Nature and its inhabitants are exposed to different forms of radiation. It is found that all elements with atomic number greater than 80 have radioactive isotopes, and all isotopes of elements with atomic number greater than 83 are radioactive. Major exposure to radiation comes from the naturally occurring radioactive materials. Main sources of these are (a) cosmic radiation (b) terrestrial radiation causing external exposure (c) terrestrial sources causing internal exposure by ingestion and inhalation.

The unstable nuclei present on the earth are called "primordial" radionuclides. They existed ever since the earth was created. Those having a long half-life period of more than 10^8 years are still present in the environment. Most commonly occurring radionuclides ^{40}K, ^{238}U, ^{235}U, and ^{232}Th together with their unstable decay products contribute to about one half of the natural radiation exposure to mankind. Along with those already present in nature, the activities of man have contributed to this radiation exposure by the introduction of the Technologically Enhanced Naturally Occurring Radioactive Materials. Radon and thoron which are the daughter products in the Uranium and Thorium series, respectively, form more than 50% of exposure to natural sources of radiation.

Study of the natural sources of radiation throws light on the distribution of radioactivity in a region which in turn is used for the epidemiological studies to understand the effect of radiation. It was such studies which led to the understanding that the underground mine workers were affected by radon gas in the mines which led to lung cancer. Such studies and several others in the indoor and outdoor environment have thrown light on the effect of various types of exposure on human beings. Not only is the data of natural radioactivity of a region significant from the point of view of its effect on the inhabitants, it also is significant to understand any increase in the levels of background

radiation due to man-made causes. Any nuclear reactor accidents or nuclear weapon trials not only affects the site concerned but also several 100 kilometers in the vicinity. Hence, generation of the radioactivity map of a region helps in understanding the future changes and aftereffects there off.

Unlike the well-studied coastal regions of Kerala, Wayanad and Idukki, which are the hill stations of Kerala, do not have any data of background radioactivity. Hence this study aims to generate baseline data for the terrestrial radioactivity of Wayanad district and also to get a brief idea about the indoor radon thoron and their progeny concentrations.

The first chapter of this thesis gives a brief introduction to radioactivity in general and the health effects of the same. Radiation protection needs and the various regulatory requirements are also discussed.

The second chapter gives a review of the available literature to get a better understanding of the way the work on natural radioactivity analysis progressed over the years in several parts of the world.

The third chapter deals with the methodology of the work. It explains the sampling sites and the details of the sampling procedure. Different types of detectors that were used and the various mathematical formulae which were used to arrive at the results are also mentioned.

Fourth and fifth chapters of this thesis discuss the results of the work. Distribution of terrestrial radioactivity in the soil and rocks of Wayanad district has been elaborated. Based on the data generated, terrestrial radioactivity of Wayanad district has been mapped. An effort has been made to gain an understanding of the hazards if any, present due to the presence of these radionuclides in the fourth chapter. In the fifth

chapter, a more intensive discussion on the results of the radon, thoron and progeny measurement is given.

The sixth chapter concludes the thesis with a brief summary of the overall results and further studies which the authors intend to take up in the future in this region.

PUBLICATIONS

1) **Reshma Bhaskaran,** C D Ravikumar, Visnuprasad A K, Jojo P J, Danalakshmi B, B. Danalakshmi, Chitra N, S Balasundar, M T Jose, Rosaline Mishra. *Inhalation Dose and Source Term Studies in a Tribal Area of Wayanad, Kerala, India,* Journal of Environmental and Public Health, Volume 2017, Article ID 1930787. https://doi.org/10.1155/2017/1930787***.(Published)***

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2) *Soil Radioactivity Measurement and Calculation of Hazard Indices in Wayanad District, Kerala, S.India.* Reshma Bhaskaran, C D Ravikumar, Proceedings of 37th National Conference of AMPI, Nov. 2016, pp 219.
3) *Diurnal Variation of Indoor Radon Concentration in Wayanad District, Kerala*. Reshma Bhaskaran, C D Ravikumar, Proceedings of 17th Asia Oceania Congress of Medical Physics and 38th Annual Conference of Medical Physicists of India, 2017.
4) *Computational Fluid Dynamics Analysis for Radon Dispersion Study and Mitigation*, A. K. Visnuprasad, P.J. Jojo Reshma Bhaskaran, 19th International Conference of Radioactivity and Radiation Protection, Paris, May 18-19, 2017, International Journal of Physical and Mathematical Sciences, Vol:11, No.5, 2017.

CONTENTS

Chapter 1	RADIATION AND THE ENVIRONMENT	1-33
	1.1 Primordial Radionuclides	3
	1.1.1 Series primordial radionuclides	3
	1.1.2 Non Series Primordial Radionuclides	6
	1.2 Cosmic Rays	8
	1.2.1 Primary cosmic rays	8
	1.2.2 Secondary cosmic rays	8
	1.3 Man- Made Sources	9
	1.4 Exposure to Natural Sources of Radiation	10
	1.4.1 External Exposure	10
	1.4.2 Internal Exposure	14
	1.5 Emission Mechanism of Radon/Thoron	16
	1.6 Normal Background Radiation Areas and High Background Radiation Areas	19
	1.7 High Background Radiation Areas In India	22
	1.8 Radiobiology And Radiation Protection	23
	1.9 Radiation Protection	27
	1.10 Purpose Of Study	30
Chapter 2	LITERATURE REVIEW	34-43
	2.1 Main High Background Radiation Areas of The World	37
	2.1.1 Brazil	37
	2.1.2 China	38
	2.1.3 Iran	39
	2.1.4 India	39

Chapter 3	LOCALE AND METHODOLOGY	45-74
	3.1 Geology	46
	3.2 Sample Collection	47
	3.3 Soil Gammaray Spectrometry	50
	3.3.1 Calibration of Gamma Ray Spectrometer	52
	3.3.2 Activity	54
	3.3.3 Annual effective dose	57
	3.3.4 Hazard Indices	58
	3.4 Radon Thoron measurements	
	3.4.1 Passive indoor measurement	60
	3.4.2 Radon and thoron progeny concentration	64
	3.4.3 Equilibrium factor for radon and thoron	66
	3.5 Annual Effective Dose	67
	3.6 Source Term Studies	68
	3.6.1 Portable Radon Monitor – SRM for indoor active measurement of Indoor Radon and thoron	71
Chapter 4	GAMMARAY SPECTROMETRY	75-94
	4.1 Concentration Of Terrestrial Radionuclides In Soil	76
	4.1.1 Potassium	79
	4.1.2 Radium	80
	4.1.3 Thorium	82
	4.2 Concentration Of Terrestrial Radionuclides In Rock	84
	4.3 Dosimetric Parameters	87
	4.3.1 Dose rate	87
	4.3.2 Annual Effective Dose	90
	4.3.3 Hazard Indices	90

	4.4 Discussion	91
Chapter 5	RADON THORON MEASUREMENTS	95-118
	5.1 Phase I	97
	5.2 Phase 2	101
	5.2.1 Active measurements	103
	5.2.2 Exhalation measurements	106
	5.3 Discussion	113
	5.3.1 Active measurements and exhalation studies	116
Chapter 6	CONCLUSION	119-123
	6.1 Important Results	120
	6.2 Scope for Future Studies	122
	Appendices	
	A. List of sampling sites for soil Gammaray spectrometry.	124
	B. Result of soil radioactivity analysis	126
	C. Result of rock radioactivity analysis	128
	D. Dosimetric parameters of Soil.	129
	E. Dosimetric parameters of rocks.	132
	F. Concentration of Radon in phase 1 study.	133
	G. Concentration of thoron in phase 1 study.	134
	H. Average Annual Effective Dose due to Radon and Thoron.	135
	I. The result of the soil Gammaray spectrometry and indoor Radon, Thoron concentration of phase 1 study.	137
	J. Radon Concentration for the phase 2 study.	138
	K. Thoron Concentration for the phase 2 study	139

L.	EERC, EETC, F_R and F_T for phase 2 study.	140
M.	Average annual effective dose for Amblavayal region of Bathery, Wayanad	141
	Bibliography	143-157

LIST OF TABLES

Table No.	Title	Page No.
1.1	Concentration of primordial radionuclides in different environmental matrices	7
1.2	Principal cosmogenic radionuclides and their half lives	11
1.3	Intake of Cosmogenic Radionuclides Effective Dose to adults annually	12
1.4	Radioactivity in building materials in few countries	13
1.5	World average annual effective dose due to various sources of natural radioactivity	18
1.6	High background radiation areas of the world	21
1.7	Average annual effective dose due to natural radioactivity in India and the world	23
1.8	Threshold dose for radiation effects	26
1.9	Radiation weighting factors for different types of radiations	28
1.10	Indoor radon concentration and action levels in different countries	30
3.1	Data for energy calibration of NaI (Tl) detector	52
3.2	Heads of decay chain segments in ^{238}U decay chain and the respective grow up times	55
3.3	Heads of decay chain segments in thorium decay chain and the respective grow-up times	56

4.1	Statistics of soil and rock samples from Wayanad, Kerala	85
4.2	Comaprison of the concentration of terrestrial radionuclides in Wayanad with other places in India, available in literature.	86
4.3	Comparison of the concentration of rock samples in Wayanad, Kerala in Bq/Kg with available literature	87
4.4	Dose rate, AED and Hazard Indices for soil and Rock samples at Wayanad, Kerala	90
5.1	Average concentration of radon and thoron in phase 1 study	101
5.2	Average concentration of radon, thoron and progeny in Wayanad, Kerala, India. Result of the phase 2 study	102
5.3	Comparison of indoor radon concentration measured using active and passive methods	103
5.4	Comparison of soil exhalation studies, passive measurement of indoor radon thoron concentration and terrestrial radioactivity content in soil.	107
5.5	Comparison of active and passive indoor radon concentration with soil gas radon concentration.	109
5.6	Concentration of indoor Radon (in Bq/m^3) with reference to the various wall materials	111

LIST OF FIGURES

Fig. No	Title	Page No.
1.1	Major pathways of exposure to background radiation	10
1.2	Methods of radon emission from soil matrix to atmosphere	16
3.1	Map of the study area	47
3.2.	Soil sampling locations in Wayanad District	49
3.3	Schematic diagram of a NaI(Tl) scintillation detector	51
3.4	Energy calibration of NaI (Tl) detector.	53
3.5	Schematics of the pin hole dosimeter	61
3.6	Sample etching bath	62
3.7	Spark Counter	63
3.8	Direct Radon and Thoron progeny sensors	65
3.9	Crossection of DRPS and DTPS	65
3.10	RAD 7 detector	69
3.11	Smart RnDuo	72
3.12	Schematic diagram of Smart RnDuo	73
3.13	Picture of soil gas radon measurement	74
4.1	Spectrum of soil sample (1)	77
4.2	Spectrum of soil sample (2)	78
4.3	Concentration of ^{40}Kin the soil of Wayanad, Kerala.	79
4.4	Histogram for activity concentration of ^{40}K	80
4.5	Concentration of ^{226}Ra in the soil of Wayanad, Kerala	81
4.6	Histogram for activity concentration of ^{226}Ra.	81
4.7	Concentration of ^{232}Th in the soil of Wayanad, Kerala	82

4.8	Histogram for activity concentration of ^{232}Th.	83
4.9	Correlation between AED from soil and survey meter	88
4.10	Map of Wayanad district depicting the distribution of Gammaray dose rate.	89
4.11	Dose rate due to soil and rock samples from Wayanad, Kerala.	92
4.12	Correlation of ^{40}K concentration in rock and soil	93
4.13	Correlation of ^{232}Th concentration between rock and soil	93
4.14	Correlation of ^{226}Ra in soil and rock	94
4. 15	Correlation of ^{226}Ra and ^{232}Th in rock	94
5.1	Correlation between indoor Radon and Thoron in 25 samples of Bathery, Wayanad	97
5.2	Distribution of Radon concentration in phase 1 study	98
5.3	Distribution of Thoron concentration in phase 1 study	99
5.4	Q-Q plot for radon concentration	99
5. 5	Q-Q plot for thoron concentration	100
5.6	Correlation between Active and Passive measurements	104
5.7	Diurnal variation of indoor Radon concentration	104
5.8	Diurnal variation of indoor Radon concentration	105
5.9	Diurnal variation of Radon in the house showing maximum Radon concentration.	105
5.10	^{232}Th versus thoron exhalation rate in soil	108
5.11.	Correlation between soil gas radon concentration and indoor radon concentration using active measurements	110
5.12	Correlation between soil gas radon concentration and indoor radon concentration using passive measurements	110

5.13	Comparison of Radon and Thoron concentration with type of construction material	112
5.14	Distribution of exposure to total radon concentration	116

RESHMA BHASKARAN “ ENVIRONMENTAL RADIOACTIVITY STUDIES IN WAYANAD, KERALA, INDIA”. THESIS. DEPARTMENT OF PHYSICS, UNIVERSITY OF CALICUT, 2018.

Chapter 1
RADIATION AND THE ENVIRONMENT

"Life on earth has developed with an ever present background of radiation. It is not something new, invented by the wit of man: radiation has always been there"

Eric J Hall (Radiation and Life)

Radiation is one of the modes of transfer of energy in space. The most common form of radiation, which common man is aware of, is visible light or the radiation from sun. Radiation from the electromagnetic spectrum, visible light is a part of which, can be divided into ionising and non-ionising radiation. Amongst the ionising radiation are x and gamma rays. Besides this particulate radiation in the form of alpha, beta, positrons, neutrons and mesons are also common knowledge.

The discovery of high energy electromagnetic radiation dates back to 1895 by Wilhelm Conrad Roentgen. These radiations, named x-rays could affect photographic plates and could easily penetrate dense matter. The discovery of x-rays was followed by the discovery of natural radioactivity by Henri Becquerel in the year 1896. Marie and Pierre Curie identified and isolated Polonium and Radium in the succeeding year. In 1899 particle radiations were introduced by Ernest Rutherford with the discovery of the so called "Alpha" and "Beta" radiations. In 1900, Paul Villard and Henri Becquerel noted that the radioactive materials blackened the photographic films. This property was identified to be because of the emission of gamma rays by Rutherford in 1903. Thus all the three major radiations, namely Alpha, Beta and Gamma were identified.

The phenomenon of radioactivity results from the instability in the nucleus. A nucleus in the excited state de-excites by the emission of gamma rays or alternately by internal conversion. The hole left in the electron cloud by the ejected electron is later filled by an orbital electron in the outer orbit. The transition of this electron is followed by the emission of x-rays or by the ejection of another electron similar to the internal

conversion. The emitted electrons are called Auger electrons. Radioactive decay is the result of the nuclear stability curve, the number of neutrons versus number of protons curve. Any nucleus which lies on the curve has equal number of neutrons and protons. Those above the curve are neutron rich and hence decay by proton and electron emission. Those below the stability curve are proton rich and hence decay by the emission of neutron and positrons.

The sources of radiation are many, ranging from natural to artificial. Both natural and artificial sources cause exposure to ionising radiation. However exposure to natural sources of radiation exceeds that from all man made sources of radiation. There are more than 60 radionuclides found in the environment. These can be categorised into Primordial (formed before the creation of earth), cosmogenic (formed due to cosmic ray interaction with the earth's atmosphere) and man-made (produced as a result of different human actions).

1.1 PRIMORDIAL RADIONUCLIDES

The primordial radionuclides are believed to have persisted since the origin of the solar system. These radionuclides have very long half-lives. These can be further sub divided into series and non-series radionuclides.

1.1.1 Series primordial radionuclides

The series primordial radionuclides are the four independent sets of unstable heavy atomic nuclei that decay through a sequence of alpha and beta decays until a stable nucleus is achieved. The four chains are the Uranium series, Thorium series, Actinium series and the Neptunium series. The radionuclides in the Neptunium series are headed by ^{237}Np. This has a half-life which is less than the age of the earth. Hence the members of this series are no longer available naturally and are hence produced artificially by

nuclear reactions. The other three members of the series primordial radionuclides are still present in the earth's crust.

A. Uranium series

The parent isotope of this series is the ^{238}U isotope. It has a half-life of 4.5×10^{10} years. Since the half-life of this nuclide is very long, this series is still present in the earth's crust. The series attains stability by decaying to ^{206}Pb by the emission of 8 alpha particles and 6 beta particles accompanied by gamma radiation. The series is in secular equilibrium because all the daughter products following ^{238}U have shorter half-life than the parent nuclide. ^{226}Ra and ^{222}Rn are both daughter products on this series. ^{238}U is the most abundant form of natural uranium with a relative abundance of 99.274%. It is found in all soil and rocks with varied concentration. Since it is present in soil it enters the food chain as well thus causing internal exposure. One of the main sources of chain disequilibrium is the occurrence and behaviour of uranium in aqueous environment. Uranium exists in both hexavalent and tetravalent state. Hexavalent state is very soluble (carbonate, phosphate and sulphate complexes) whereas the tetravalent state is insoluble. The daughter product of ^{238}U, ^{226}Ra exhibits only +2 oxidation states in solution and forms water soluble chloride, bromide and nitrate salts. ^{226}Ra, ^{222}Rn, ^{210}Pb (which head the three sub series of ^{238}U series) and ^{210}Po, which belongs to ^{210}Pb sub series, are the other important radionuclides in ^{238}U series. ^{226}Ra has a half-life of 1620 years and becomes ^{222}Rn through alpha decay. Chemical properties of radium are similar to that of calcium and therefore enter the human body through food chain easily and gets concentrated in bones. More than 70% of the radium in the body is contained in bones, the remaining fraction being distributed rather uniformly in soft tissues. The average annual dietary intake of ^{226}Ra in areas of normal background radiation is 15 Bq

B. Thorium series

The parent radionuclide of this series is ^{232}Th. Both the thorium and Uranium series present strong similarities. Both have isotopes of the same elements and contain a large proportion of alpha emitters. The series decays to a stable ^{208}Pb by alpha and beta emissions. ^{232}Th is not as widely distributed as ^{238}U in the rocks and soil. However, in certain rocks like the igneous rocks, ^{232}Th is found to be four times that of ^{238}U. ^{228}Ra and ^{220}Rn are part of this series. Thorium occurs in tetravalent oxidation state and is insoluble except near neutral pH or in the presence of organic compounds like humic acid. When the pH is near neutral Thorium is removed from solution by precipitation as highly insoluble hydrated oxide phase and the co-precipitation with hydrated ferric oxides with sorption reactions. These two phenomena along with the low solution rate of Thorium- bearing minerals form the major reasons for the generally low concentration of Thorium in natural waters. Radium (^{228}Ra) has the same geochemical properties as radium (^{226}Ra) in the uranium chain. Thoron having shorter half-life as compared to radon finds it difficult to escape in to the atmosphere and hence is expected to be in lesser concentration as compared to radon in the indoor atmosphere.

C. Actinium series

The parent radionuclide of this series is ^{235}U. Hence it is also known as the ^{235}U series. ^{235}U decays by a series of alpha and beta emissions to ^{207}Pb. Since the relative abundance of ^{235}U is only 0.72%, its contribution to the dose received by human beings due to the exposure to terrestrial radioactivity is negligible. Hence ^{235}U is not taken into account while doing measurements of radiation exposure.

1.1.2 Non Series Primordial Radionuclides

The major non-series radionuclides are ^{40}K and ^{87}Rb. Potassium is widely distributed in nature and hence is the seventh most abundant element in the crust of the earth. It is also the sixth most abundant element in solution in oceans. It is found in all plant and animal tissues. Two stable isotopes of potassium exist namely, ^{39}K and ^{41}K. ^{40}K is one of the radioactive isotopes of potassium and forms only a small fraction (0.018%) of the natural potassium. Besides this there are several radioactive isotopes of potassium. However, all of them have half-lives less than one day and hence are not of significance from the exposure point of view. ^{40}K has a half-life of 1.3 billion years. ^{40}K is the isotope which undergoes both types of beta decay. It decays to ^{40}Ca with the emission of beta particle. Another type of decay is one in which the isotope undergoes electron capture and decays to ^{40}Ar. Very rarely ^{40}K decays to ^{40}Ar by emitting a positron and a neutrino. Potassium is soluble in water and hence gets lost through solution during weathering.

Rubidium is chemically similar to Potassium. The concentration of natural rubidium is only 1% of that of potassium. Radioactive rubidium ,^{87}Rb is only 60% of that of ^{40}K [2] and contributes to the internal radiation exposure by the emission of beta rays. Table 1.1 lists the concentration of primordial radionuclides in different environmental matrices.

Table 1.1
Concentration of primordial radionuclides in different environmental matrices (Reproduced from [3]**)**

Environmental Matrix	^{238}U	^{226}Ra	^{40}K	^{87}Rb
Igneous rock (Bq/kg)	0.04	0.048	1.2	-
Phosphate rock (Bq/kg)	1.60	1.50	0.4	-
Lime stone (mBq/kg)	16.0	5.0-20.0	30.0-150.0	-
Soil (mBq/kg)	37.0	16.0	100.0	-
Air (µBq/m3)	1.2	1.5	22.0	-
Surface water (mBq/l)	0.18-62.9	0.4-111.0	3.7×10^2- 2.4×10^5	0.9
Ocean surface water (mBq/l)	44.4	1.3-3.1	1.1×10^4	100.0
Ocean Bottom water (mBq/l)	40.0	3.0-5.6	1.1 x 104	-
Human (Bq)	1.3-1.6	1.0-1.5	6300.0	455.0
Daily intake by human (mBq)	13.0	190.0-270.0	1×10^5 – 1.4×10^5	7000.0
Annual effective dose (µSv)	1.2	7.0	180.0	6.0

1.2 COSMIC RAYS

Only a small portion of the dose from natural background radiation comes from cosmogenic radionuclides. Cosmic rays can be classified as the primary cosmic rays and the secondary cosmic rays. The high energy radiation entering the earth's atmosphere from outer space is referred to as primary cosmic rays. These radiations after entering the earth's atmosphere interact with the atomic nuclei there, thus forming secondary particles and electromagnetic radiations. These secondary particles and electromagnetic radiation are called secondary cosmic rays [4]

1.2.1 Primary cosmic rays

The origin of the primary cosmic rays is mostly from outside the solar system. They are of two types: the primary galactic cosmic rays and the primary solar cosmic rays. The primary galactic cosmic rays consist of high energy protons which enter the solar system from the interstellar space, together with the ^{4}He ions, heavier particles and electrons, photons and neutrons. The primary flux density of these particles is affected by the earth's magnetic field. The other type is the primary solar cosmic rays. These are charged particles, mainly protons and alpha particles, which are released during the solar flares.

1.2.2 Secondary cosmic rays

Secondary cosmic rays are produced when the primary cosmic rays enter the earth's atmosphere and undergo spallation reactions with the nuclei of the atoms present in the air. The by-products of these reactions are neutrons, protons, pions and kaons along with cosmogenic nuclides like ^{3}H, ^{7}Be, ^{10}Be, ^{22}Na and ^{14}Na. These high energy particles further interact with the nuclei in the air to form more secondary particles.

1.3 MAN- MADE SOURCES

Over the years the activities of man also contributed to radioactivity in the atmosphere. Several industries, weapon testing, nuclear power plants, etc., have played a significant role in the exposure of radiation to common man [1]. Following are the few man made sources of radiation.

- Medical Sources (by far, the most significant man-made source)
  - Diagnostic x-rays
  - Nuclear medicine procedures (iodine-131, cesium-137, and others)
- Consumer Products
  - Building and road construction materials
  - Combustible fuels, including gas and coal
  - X-ray security systems
  - Televisions
  - Fluorescent lamp starters
  - Smoke detectors (americium)
  - Luminous watches (tritium)
  - Lantern mantles (thorium)
  - Tobacco (polonium-210)
  - Ophthalmic glass used in eyeglasses
  - Some ceramics

Besides the above mentioned sources, common man is also exposed to radiation from the nuclear fuel cycle, from uranium mining and milling to disposal of spent fuel. Minimal exposure is also caused due to the transfer of radioactive material and due to fallout radiation from nuclear weapon testing.

1.4 EXPOSURE TO NATURAL SOURCES OF RADIATION

Exposure caused by naturally occurring radioisotopes is both internal as well as external. Radioisotopes of terrestrial origin are the source of external gamma ray exposure. At the same time, radionuclides, when ingested in the form of food cause internal exposure due to both alpha and beta irradiations. The daughter product of Uranium and Thorium series ^{222}Rn and ^{220}Rn cause exposure to the bronchus during inhalation. The main exposure pathways are as shown in Fig.1.1.

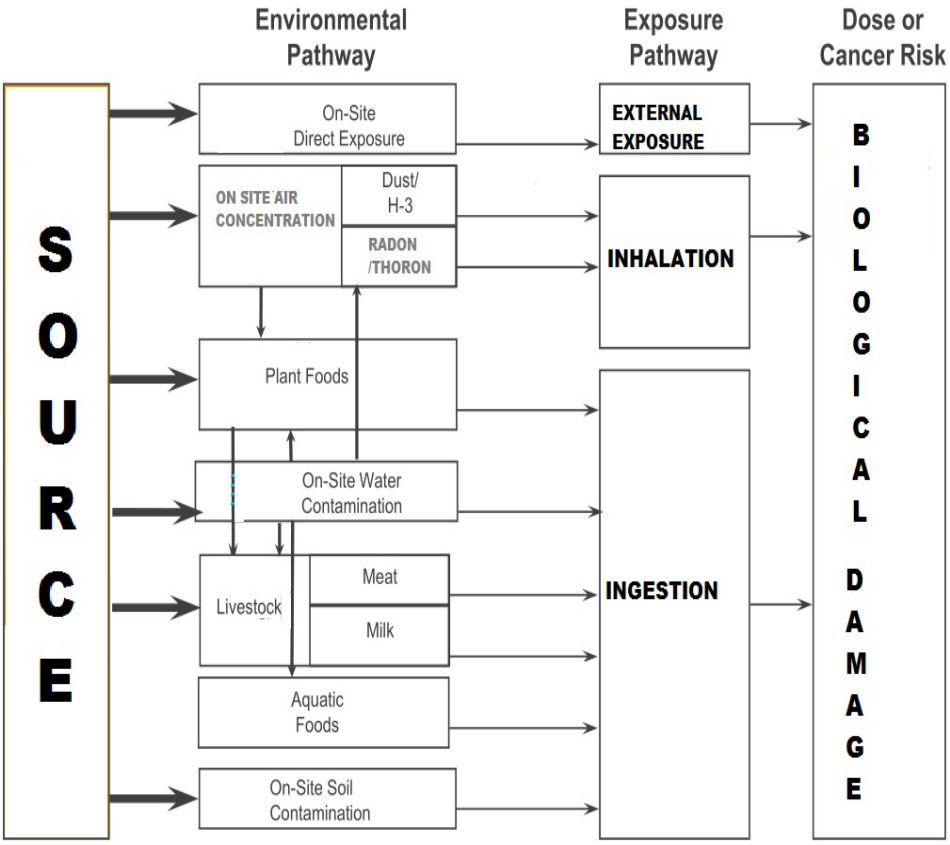

Fig 1.1 Major pathways of exposure to background radiation

1.4.1 External Exposure

External exposure to natural sources can be caused due to both cosmic as well as terrestrial radiation sources. The atmosphere which is approximately 10,000 kg/m^2 shields a major part of the earth from cosmic radiation. Hence at sea level the cosmic rays contribute only 10% of the total dose rate from naturally occurring radiation.

However, as the height from the sea level increases, exposure due to cosmic sources of radiation also increases. Exposure due to cosmic sources of radiation is affected by both latitude and altitude. Due to the shape of the earth's magnetic field only particles from the cosmic radiation which have higher energies can penetrate at lower geomagnetic latitudes. Thus the intensities of the radiation as well as the dose rates are minimal at the equator and maximal near the geomagnetic poles. UNSCEAR has adopted an average dose rate of 31 nGy/h at sea level due to cosmic radiation [1]. Muons contribute 80% of this dose rate and the remaining comes from electrons produced by the muons. Using a shielding factor of 0.8 and an indoor occupancy fraction of 0.8, the worldwide average annual effective dose due to ionising radiation from the cosmic rays is 0.28 mSv. Table 1.2 lists the principal cosmogenic radionuclides in the atmosphere and their half-lives. The intake of few cosmogenic radionuclides by human adult and the dose received due to their intake is listed in Table 1.3.

Table 1.2
Principal cosmogenic radionuclides and their half lives

Radionuclide	Half-life (Year/days)	Radionuclide	Half-life (Year/days)
^{3}H	12.3y	^{129}I	1.6×10^{7} y
^{7}Be	53d	^{40}K	1.3×10^{9} y
^{10}Be	1.6×10^{6}	^{41}Ca	1×10^{5} y
^{14}C	5730 y	^{46}Sc	84 d
^{22}Na	2.6 y	^{48}V	16 d
^{26}Al	7.1×10^{5} y	^{53}Mn	3.7×10^{6} y
^{32}S	87 d	^{54}Mn	312 d
^{36}Cl	3×10^{5} y	^{55}Fe	2.7 y
^{37}Ar	35 d	^{56}Co	79 d
^{39}Ar	269 d	^{59}Ni	7.6×10^{4} y
^{61}Kr	2.1×10^{5} y	^{60}Fe	1.5×10^{6} y

Table 1.3

Intake of Cosmogenic Radionuclides Effective Dose to adults annually

(Reproduced from [5]**)**

Radionuclides	Intake (Bq/y)	Annual Effective Dose (micro Sv)
^{3}H	500	0.01
^{7}Be	1000	0.03
^{14}C	2000	12
^{23}Na	50	0.15

Terrestrial sources of radiation are mainly the radionuclides in the Uranium and Thorium series in addition to Potassium. Worldwide average of ^{238}U is 33 Bq/kg, ^{232}Th is 45 Bq/kg and ^{40}K is 412 Bq/kg. The concentration of the terrestrial radionuclides shows large variation from place to place. Worldwide average outdoor dose rate due to terrestrial radionuclides is taken as 58 nGy/h.

Indoor exposure due to terrestrial radionuclides is caused mainly by the outdoor concentration of radionuclides as well as those present in building materials. The contribution to indoor exposure varies due to the type of the building material used as well as the ventilation available in the buildings. Out of all the available building materials granite and marble show [1] the highest average ^{226}Ra content (77 Bq/kg). Worldwide average indoor absorbed dose rate in air is taken as 84 nGy/h. Ratio of indoor to outdoor exposure rate has an average value of 1.4. UNSCEAR has adopted a conversion rate from absorbed dose rate in air to effective dose received by adults of 0.7 Sv/Gy. Taking 0.8 as the occupancy fraction indoors, the average annual effective dose due to terrestrial sources of radiation amounts to 0.48 mSv, out of which 0.41 mSv is due to the indoor occupancy and 0.07 mSv is due to outdoor occupancy. Radioactivity in building materials in few countries [2] is reproduced in Table 1.4.

Table 1.4

Radioactivity in building materials in few countries

Building Material	Radioactivity concentration (Bq/kg)			Country
	^{40}K	^{226}Ra	^{232}Th	
Cement	290.8	39.6	28.9	Iran
	422	41	27	Brazil
	241	20	13	Greece
	432	37	24	India
Gypsum	116	8	2	Iran
	40	9	4	Turkey
	173	6	13	Pakistan
	39	13	5	Bulgaria
Brick	851	37	12	Iran
	675	65	51	Algeria
	710	35	45	Greece
	585	35	72	Sri Lanka
	714	59	50	China

Terrestrial radionuclides are absorbed by plants. Thus they enter the food chain and cause exposure due to ingestion. ^{238}U is mainly retained in the skeleton after ingestion and inhalation. Its concentration differs in different kinds of bones. Similarly, ^{232}Th gets deposited on bone surfaces. Potassium is nearly uniformly distributed in the body. Concentration of Potassium is said be under homeostatic control. For adults the body content of potassium is 0.18% and for children 0.2%. The total annual effective

dose due to inhalation and ingestion of radionuclides of terrestrial origin is taken to be 0.29 mSv. Out of this ^{40}K contributes more as compared to the radionuclides in the Uranium and Thorium series combined together.

1.4.2 Internal Exposure

A. Ingestion

Radionuclides enter the human body both through ingestion and inhalation. Natural radioactivity concentration in food is often in the range of 40 – 600 Bq/kg of food. The concentration of radioactive potassium in different food varies from 45.9 to 649 Bq/kg. ^{226}Ra varies from 0.01 to 1.16 Bq/kg and ^{228}Th from 0.02 to 1.26 Bq/kg [6]

Even though the concentration of radioactivity shows a particular range the amount of radioactivity retained in the various organs depend on both the physical and biological half-life of the radioisotopes. Hence the exposure due to ingestion varies from person to person for the same concentration of the activity being ingested as the factors like sweating, secretion through saliva and other discharges from the body etc.

B. Inhalation

Radon and Thoron are the major sources of the inhalation dose. Radon was the fifth radioactive element to be discovered. In 1900, Friedrich Ernst Dorn discovered some radium compounds emanate a radioactive gas. He named it Radium Emanation (Ra Em). Later on it was known as radon. Radon is colourless and odourless noble gas. It is about 7.5 times heavier than air and about 100 times heavier than hydrogen. The atomic number of radon is 86 and it has three isotopes namely, ^{222}Rn (radon), ^{220}Rn (thoron) and ^{219}Rn (action). ^{222}Rn occurs as immediate decay product of ^{226}Ra (radium) in the decay series of ^{238}U (uranium). It has a half-life of 3.824 days. ^{220}Rn occurs as immediate decay product of ^{224}Ra in the decay series of ^{232}Th (thorium) while ^{219}Rn occurs as

immediate decay product of ^{223}Th in the decay series of ^{235}U (uranium). Earlier radon was identified as a risk to the underground miners. However, recently, several studies have shown that radon might carry a risk due to domestic exposure as well [7].

The daughter product in the ^{232}Th series, Thoron was discovered by R.B Owens at McGill University in collaboration with Ernest Rutherford [8]. Thoron is also an inert gas. ^{220}Rn has a half-life of 55s. Until recently the exposure due to thoron was underestimated. However several studies have pointed towards the significance of the study of thoron along with radon in assessing the total inhalation exposure [9–11]. Hence several detectors and various measurement techniques were devised to differentiate thoron from radon and find the dose due to each separately. Together radon and thoron contribute more than 50% of the radiation dose due to natural sources.

Radon was recognised as a health hazard around 100 years back among the underground miners by Harting and Hesse [12]. Higher cases of lung cancer were diagnosed among the miners as compared to the normal population. After 45 years of this discovery, Ludewig and Lorenzer [13] suggested that the higher incidence of lung cancer among the miners may be due to the high concentration (10^3 to 5×10^4 Bq/m^3) of Radon in these mines. Later, after 30 years it was found that it was not radon but the progeny of radon, which when inhaled, causes lung cancer [14].

As already mentioned, Radon and Thoron occur in the environment due to releases from natural sources and also due to sources from anthropogenic activities. The natural sources include the emission from soil, rocks and ore bodies in the earth crust. Besides these, the emission of radon and thoron also happens from the building materials like bricks, cement, etc. Besides these, the activities of man like uranium mine and milling facilities and some other industries form the anthropogenic sources of radon and

thoron. These sources are called technologically enhanced natural occurring radioactive materials (TENORMs)

1.5 EMISSION MECHANISM OF RADON/THORON

Radon is formed in the rocks and soils as a daughter product of ^{226}Ra. Radon thus formed is released into the surrounding water or air only partially. A fraction of the radon thus formed escapes into the pore spaces and is transported to nearby surroundings. Several factors like the porosity of soil, moisture content etc effect the diffusion of radon from pores. Besides these, mechanical factors like tides and earthquakes also effect the transportation of radon within soils. The processes involved in the emanation of radon (Fig.1.2) are as follows:

1. Emanation – Radon atoms formed from the decay of radium escape from the grains by recoil. These then reach the interstitial space between the grains by the process called emanation.
2. Transport- The radon is transported through the pore spaces of the soil matrix to the ground surface by diffusion and advective flow.
3. Exhalation- It is the process of escape of the radon from the ground surface to the atmosphere.

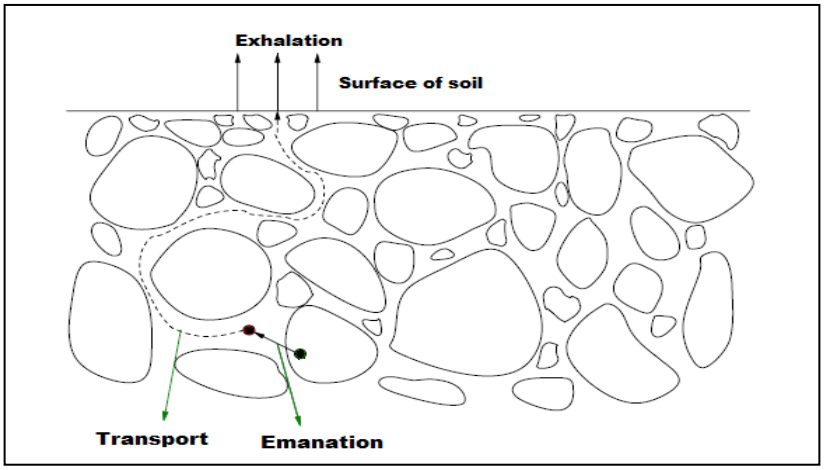

Fig1.2: Methods of radon emission from soil matrix to atmosphere

Movement of radon in air depends on the vertical temperature gradient, the strength and direction of wind and air turbulence. Since the major part of the dust air mass, etc., are found in the troposphere, radon and decay products are also found there. The variation of radon depends on the temperature and wind condition during a day. The maximum radon levels are found during early mornings and minimum during noon [15–17]. Similarly, the maximum radon concentrations are found during winter and minimum during summer.

Major portion of the inhalation dose due to radon comes from the indoor environment. Source of radon in indoor air may be soil and rocks in the surroundings of the building or the presence of ^{226}Ra in the construction material. Along with these, natural gas and well water, if they contain high concentration of radon, may lead to emanation of radon into the indoor atmosphere. Some of the common entry points of radon in the indoor atmosphere are joints and cracks in the floor and walls, openings in sills above hollow block walls, sump holes, drains and piping in electrical penetrations in walls and floor.

The aerosol in the atmosphere absorbs the radon progeny present in the atmosphere. During inhalation these aerosols get stuck to the bronchus thus irradiating the lung. Later on it was found that not only the miners but also the people living indoors, near the granite quarries, etc., face hazard due to the inhalation of radon and thoron. The construction materials also contain ^{238}U and ^{232}Th radionuclides and their daughter products. Radon escapes from the surface of building materials and can stay inside rooms if ventilation is not proper. This radon and its decay products thus enter the lungs due to inhalation. The concentration of radon at a particular place is influenced by several factors like concentration of the parent radionuclide in the building material and

the surrounding atmosphere, temperature and humidity, ventilation of the rooms, aerosol concentration of the rooms, presence of radon in the tap water, etc.

Out of all the radioisotopes of Radon, ^{222}Rn has the longest half-life of 3.8 days. ^{219}Rn is the shortest and is produced in much smaller amounts. Hence its contribution to the exposure to natural sources of radiation is generally ignored. ^{220}Rn too has a half-life of 56 seconds only. The health hazard due to radon and thoron is primarily due to the progeny which sticks to the walls of the bronchus along with the aerosols in the atmosphere.

All together the annual effective dose due to naturally occurring radionuclides is as given in Table 1.5.

Table 1.5

World average annual effective dose due to various sources of natural radioactivity (Reproduced from [18]**)**

Source of exposure		**Annual effective dose (mSv)**	
		Average	Typical range
Cosmic radiation	Directly ionizing and photon - component Neutron component Cosmogenic radionuclides	0.28 0.10 0.01	
	Total cosmic and cosmogenic	0.39	0.3-1.0[a]
External terrestrial radiation	Outdoors Indoors	0.07 0.41	
	Total external terrestrial radiation	0.48	0.3-1.0[b]
Inhalation	Uranium and thorium series Radon (^{222}Rn) Thoron (^{220}Rn)	0.006 1.15 0.10	
	Total inhalation exposure	1.26	0.2-10[c]

Ingestion	^{40}K Uranium and thorium series	0.17 0.12	
	Total ingestion exposure	0.29	0.2-1.0[d]
Total		2.42	1.0-13

a. range from sea level to high ground elevation

b. depending on radionuclide composition of soil and building material

c. depending on indoor accumulation of Radon gas.

d. depending on radionuclide composition of foods and drinking water.

1.6 NORMAL BACKGROUND RADIATION AREAS AND HIGH BACKGROUND RADIATION AREAS

Depending on the concentration of terrestrial radionuclides as well as the contribution from cosmic rays, the environmental radiation level varies from place to place. Some regions of the world show radiation levels far higher than the other regions. Depending of the exposure rate at a particular place, the place may be classified as Normal Background Radiation Area (NBRA) and High Background Natural Radiation Area (HBNRA). According to UNSCEAR[18], 95% of world population resides in the NBRA. These areas may be defined as areas receiving outdoor absorbed dose rate of 30 to 70 nGy/h [19]. Some regions, on the other hand, have dose rates significantly higher than the values mentioned for the normal background radiation. Such regions with elevated background radiation level are called High Background Radiation Areas. Ramsar (Iran), Yangjiang (China), Guarapari (Brazil), the northern Flinders rangers (Australia) and the south west coastal belt of Kerala and Tamilnadu (India) are the HBNRAs of the world. These regions show high background radiation due to the presence of high terrestrial radioactivity.

In Kerala, major reason of HBNRA is the monazite sand present in the west coast belt. Monazite is the ore of ^{232}Th and its concentration is high in the beach sand. However, these high concentrations are seen only in patches. Thus marking a particular area as high or low background becomes difficult as there is a large variation amongst the minimum values found in the area and the patches of high values found in the same region. To resolve this issue, some researchers have come forward with criteria to define regions as high and normal background radiation areas.

Cullen et al proposed in 1977 [20] that a region may be identified as High Level Natural Radiation Area if the region satisfies any or more of the following:

1) The exposure rate from external terrestrial sources, over extended areas, is greater than 2 mGy/y;
2) The long-lived alpha activity ingested through the local diet and water is greater than1.85 Bq/d;
3) The radon concentration of potable water is greater than 185 kBq/m^3;
4) The ^{220}Rn and ^{222}Rn concentrations in the atmosphere are greater than 37 Bq/m^3 .

U. C. Mishra [21] also proposed some criteria which are as follows:

1) The exposure rate from external terrestrial sources, over extended areas, should be greater than 4 mSv/y
2) The long-lived alpha activity ingested as a result of a local diet and water should be greater than 2 Bq/d
3) The ^{222}Rn concentration of the potable water should be greater than 200 kBq/m^3
4) The ^{220}Rn and ^{222}Rn concentrations of the atmosphere should be greater than 40 Bq/m^3

5) The area should have at least a population of 1000 to be considered of significance for epidemiological investigations

A large amount of work has been going on since the past 100 years to study the background radioactivity and its effect on humankind. Based on these studies few High background radiation areas and the radioactivity levels in those places are as given in Table1.6**.**

Table 1.6

High background radiation areas of the world

Site	Main source of exposure	Average absorbed dose rate in air (μGy/h)	Maximum annual dose (mSv)
Ramsar (Iran)	^{226}Ra deposits in hot spring waters	0.070 - 30	438
State of Espirito Santo-Guarapari, Meaipe and Cumuruxatiba, Brazil	Monazite sands	0.1 - 10,000	88 788 (beach)
Morro do Ferro- Minas Gerais	Thorium rich deposit hill	0.3 - 22	245
Coast near Mombasa	Thorium bearing Carbonalyte deposits	12	105
Kerala (India)	Monazite sands	0.2 - 4	76
Tamilnadu (India)	Monazite sands	0.19 - 4	53
Permian basin of Lodeve	Uranium minerals	0.01 - 10	88

1.7 HIGH BACKGROUND RADIATION AREAS IN INDIA

In India, the west coastal belt, Ullal In Karnataka and some parts of Uttar Pradesh, Bihar and Orissa have been found to show elevated levels of natural radioactivity[22–24]. Out of these the west coastal belt is counted amongst the highest background radiation areas of the world. Due to this a large amount of research has been conducted in this region of Kerala. A 57 km coastline of Kerala is one of the highest background radiation areas of the world. The main region of concern is the Chavara-Neendakara belt in Southern Kerala. Monazite ore has been stated as the prime reason for this enhanced radioactivity in this region. The monazite deposits present here is an orthophosphate of thorium and rare earths and contains thorium oxide and uranium oxide along with other rare earths and silicon dioxide (Chougaonkar et al 2004)**.** In Kollam district of Kerala, several studies have been going on since the past several years[26, 27]. One of the studies shows that the concentration of ^{232}Th there is as high as 1342 Bq/kg [28]. In Tamilnadu Agatheeswaram taluk and its adjoining taluk, Thovalai of Kanyakumari [29] district are amongst the high background radiation areas.

In the Northern part of India, the Gangetic alluvial regions of Uttar Pradesh, Bihar and West Bengal show relatively higher natural radioactivity. Studies are going on in the Kumaon, Garwal and other parts of Uttarakhand, Himachal Pradesh etc. to study the background radioactivity and also the effect temperature has on the radon thoron concentrations[30, 31].

Overall the annual effective dose due to different sources of natural radioactivity in the India and its comparison with the world average are as given in Table 1.7.

Table 1.7

Average annual effective dose due to natural radioactivity in India and the world

Radiation sources	INDIA		WORLD	
	Annual Effective Dose (mSv/y)	Percentage Contribution	Annual Effective Dose (mSv/y)	Percentage Contribution
External sources				
Cosmic Radiation	0.355	15.44	0.380	16.14
Terrestrial Sources	0.379	16.48	0.480	19.55
Internal Inhalation other than Radon and Thoron	0.015	0.65	0.010	0.41
Inhalation due to Radon and Thoron	1.235	53.72	1.275	51.94
Terrestrial	0.315	13.70	0.310	12.63
Total	20.3	100	2.5	100

1.8 RADIOBIOLOGY AND RADIATION PROTECTION

The adverse effects of radiation are common knowledge now. The physical interaction of ionising radiation produces free radicals. These free radicals interact with the DNA, either directly or indirectly. In the direct mechanism, the free radicals cause excitation and ionisation in the biological molecule thus creating single or double strand DNA breaks. It has been found that one third of the biological damage due to

gamma. radiation is caused by direct effects. The direct effect becomes more prominent if the Linear Energy Transfer of the radiation is high as in the case of neutrons and alpha particles.

Second method of biological damage is through indirect effect of radiation. Water forms 80% of the mass of a living cell. When radiation is incident on the body, a major portion of the energy will be absorbed by water. This causes water radiolysis and thus produces free radicals. These free radicals then interact with the DNA causing DNA strand damage.

Damage in the DNA can be repaired by natural mechanisms. However, if the damage remains unrepaired then it can cause chromosome damage. This chromosomal damage leads to damage of organs.

The damage to the organ or the organism may be of two types, namely stochastic effect and deterministic effect. In the case of stochastic effect, the probability of effect and not its severity depends on the dose. Examples of stochastic effects are malignancies including leukaemia and hereditary effects. There is no threshold for stochastic effect and the dose response is linear [32]. Stochastic effect is understood to arise either due to the effect of radiation in a single cell or a very small number of cells in which a limited number of genes are implicated as part of series of events in the malignant transformation of normal cell.

Human population has not yet shown signs of hereditary effects. The survivors of Japanese atomic bomb exposed to large doses of radiation and the common public who were not exposed to any high doses even from natural radioactivity had almost the same genetic effect. However, the occurrence of genetic effects has been demonstrated in animal experiments. Hence studies on a larger scale are required to assess the hereditary

effects, if any, in human beings. If the embryo or foetus is exposed to ionising radiation, it increases the risk of leukaemia in infants. If it is during early pregnancy, with high amount of radiation, it may lead to mental retardation [26].

Cancer is another stochastic effect, the probability of occurrence of which increases with increase in the dose to which the individual is exposed. There is no difference between radiation induced cancer and the cancers normally found in unexposed individuals. However, one particular type may occur more frequently as compared to others after the exposure. Hence epidemiological studies are the base for the analysis of this kind of outcome. In the case of the atomic bomb survivors of Hiroshima and Nagasaki, an increase in the rate of leukaemia was observed in persons exposed to doses higher than 0.1Gy. An increase in solid tumours was observed above doses of 0.4Gy only. Since the results of cancer induction in persons exposed to low levels of radiation are in negative, the cancer risk coefficients are usually estimated by the extrapolation of data from observations on populations that received high doses of radiation [33].

In the case of deterministic effect, the severity of effect increases with the dose to which the individual is exposed. Deterministic effect has a threshold beyond which the effect is sure to happen. Unlike the stochastic effect deterministic effect is the result of damage to a large number of cells in an organ or tissue. As the dose to the organ increase the damage also increases thus leading to higher loss of function. Besides this radiation exposure may cause genetic effects as well like mutagenesis or teratogenesis. The damage to DNA because of radiation is more severe than that due to chemical or thermal processes.

Examples of the threshold dose for few deterministic effects of radiation are as shown in Table 1.8.

Table: 1.8

Threshold dose for radiation effects

WHOLE BODY EXPOSURE OF				
Dose (cGy)	Response	Symptoms	Expected Outcome	Risk
<100	Prodromal Syndrome	Nausea Vomitting Diarrhea	Complete recovery	Late effects
100- 1000	Hemetopoetic syndrome	Decrease in blood count	Below LD50/60 some recoveries. At 1000 cGy no survivors	Increase in dose increases risk of death and decreases mean survival time. Late effects are possible for all survivors
1000 – 5000	Gastrointestinal syndrome	Severe bloody diarrhea, sluffing of the mucosa layer	**Death** Mean survival time 4 to 10 days	**Death**
>5000	CNS syndrome	Loss of vision, loss of consciousness, seizures, coma	**Death** Mean survival time few hours to a day or two	**Death**

1.9 RADIATION PROTECTION

Since the harmful effect of radiation is well known as far as the deterministic effects are concerned, several international bodies have proposed threshold doses and protection procedures for the staff as well as member of the public. One of the most important organisations, namely International Commission for Radiation Protection (ICRP), gave its first general recommendations in 1928, concerning the protection of the medical profession through the restriction of working hours with medical sources [34]. In 1954, the commission recognised the possibility of stochastic effects for which threshold doses may or may not exist and hence came up with the recommendations that every effort should be made to reduce the exposures to all types of ionising radiation to the lowest possible level.

In 1956, commission set the annual dose limit of 50mSv for the workers, which was reduced to 20 mSv in 1990. This reduction was based on the revision of the risk for stochastic effects estimated from the life span study of the Hiroshima Nagasaki atomic bomb survivors. The annual dose limit of 1 mSv in a year was proposed for the members of the public.

Even though Gray (Joules/kg) is considered as the SI unit of dose, in the concept of radiation protection this unit is insufficient. The reason for it is that 1 Gray of photons and neutrons have different radiobiological effects. Similarly, the same dose of radiation of the same type has different effects on different organs. Hence for the sake of use in terms of radiation protection, few new quantities were introduced. **Equivalent dose:** Equivalent dose is the product of the absorbed dose and the radiation weighting factor. It takes into account the relative biological effect of the radiation in question. Radiation weighting factor is a unit less quantity. Following Table 1.9 gives the weighting factor of different radiation.

Table 1.9

Radiation weighting factors for different types of radiations

Radiation	Weighting factor
Photons, electrons and muons of all energies	1
Protons > 2MeV (except recoil protons)	2
Alpha particles and heavy ions	20

For neutrons the weighting factor is a continuum with the maximum value of 20 for 10 MeV.

Effective dose: Effective dose is the product of the equivalent dose and the tissue weighting factor. Tissue weighting factor is unit less quantity. Both equivalent dose and effective dose have the unit Sievert (Sv).

As per ICRP [35] recommendations, the exposure due to radiation has been separated into three categories, namely, occupational exposure, medical exposure of patients and public exposure. In ICRP 103, three different types of exposure situations have been defined namely, planned exposure situation, emergency exposure situations and existing exposure situations.

Out of these three, natural radioactivity is classified as existing exposure situation, situations which already exist when a decision on control has to be taken. This kind of exposure situation is the largest contributor to public exposure.

To restrict the individual doses due to various exposure situations three basic principles have been recommended namely, justification of the practice (the exposure situation should do more good than harm), optimisation of protection (the likelihood of incurring exposures, the number of people exposed, and the magnitude of their individual doses should be kept as low as reasonably achievable, taking into account

economic and societal factors) and application of dose limits (total dose from planned exposure should not exceed the dose limits proposed by the commission). In the existing exposure situation justification is used in making decisions as to whether to take action to avert further exposure. When an exposure situation has been justified, optimisation of protection is intended for that application. According to this principle the exposure to radiation should be as less as practically achievable keeping the societal and economic factors into consideration. The process of optimisation follows the following steps:

- Evaluation of the exposure situation, including any potential exposures
- Selection of appropriate value for the constraint and reference level
- Identification of possible protection options
- Selection of the best option under the prevailing circumstances
- Implementation of the selected option

In the case of exposure of the public to natural radioactivity, there is a necessity to have an awareness of the baseline data of exposure in the place of concern, so that any increase to the baseline may thus be identified in future studies. This forms the evaluation of the situation.

The idea of dose constraint and reference level is used along with optimisation of protection to restrict individual doses. The term dose constraint is used in the case of planned exposures. For emergency and existing exposure situation reference level is used. There is no particular dose constraint or reference level for the terrestrial radioactivity. The estimation of terrestrial radioactivity acts as a reference for further contamination if any. Besides this, the study of terrestrial radioactivity is mandatory to understand the intake of radioactivity in human beings through the food chain. However, in the case of exposure due to radon, certain measures may be taken to control and reduce the indoor exposures. Since lung cancer due to exposure to radon has been proved

in the miners in the past, several international bodies have suggested reference levels for radon. This reference level is used to identify situations where action is required to control the exposure to radon. The reference level as suggested by ICRP is 10 mSv. Following Table 1.10 gives a list of reference levels suggested by various countries

Table 1.10

Indoor radon concentration and action levels in different countries

Country	Average radon concentration in homes (Bq m^{-3})	Action Level (Bq m^{-3})
Czech Republic	140	200
Finland	123	400
Germany	50	250
Ireland	60	200
Norway	51-60	200
Poland	25	400
USA	46	150
Canada	20-50	800

1.10 PURPOSE OF STUDY

With regard to the radiation safety of the public, all exposure situations should be well studied and documented. This leads to an idea of the present hazards if any and also gives measures to be taken for any increase in exposures in the future. The study of terrestrial radioactivity is important not only to assess the hazards to the residents of that particular region but also to the people of other regions where the raw material from the region of study are used to construct buildings. One of the main advantages of setting up

a baseline data for the natural radioactivity is that exposure from natural sources of radiation remain relatively stable over time in contrast to artificial sources [36] . The chances of variation in the exposure from natural sources occur if the natural sources are affected by the artificial sources.

Immense research work is going on in India and other countries for the identification of the Naturally Occurring Radioactive Material present in the soil, rock, water and air. This has assumed significance from the point of view of mining of these elements as well as from the point of view of radiation dose being received by the population living in these areas [19]. For a vast country like India with different meteorological and geological conditions, the data of background radioactivity available remains scanty. Comprehensive region wise data has to be generated to get substantial idea about terrestrial and cosmic radioactivity as well as the radon/ thoron dose level and the inhalation dose rates.

Several reports have established that the southwest coast of India, more precisely the southern part of Kerala namely the Chavara-Neendakara belt are the very high background radiation areas [26, 37]. Monazite sand deposits have been described as the source of these radiations. On the basis of these reports several studies are being conducted in the southern part of Kerala. Detailed radioactivity analysis as well as epidemiological studies to analyse the health effects of these exposures are going on in the south of Kerala. However, knowledge of the background radiation levels with respect to the northern part of Kerala seems scarce.

In many cases coal and lignite present high concentration of naturally occurring radionuclide such as ^{238}U, ^{226}Ra ,^{210}Pb, ^{232}Th and ^{40}K [38, 39]. Several studies are also undergoing to study the Radon emanation from different types of Granites[40–42]. The Dept. of Mining and Geology, Govt of Kerala (http://dmg.kerala.gov.in/) has reported the

presence of heavy minerals, namely, Ilmenite, Rutile, Zircon, Monazite and Sillimanite in Kannur and Malapuram districts. Recently deposits of lignite have been identified in Kannur and Kasargode. Several types of granite are present in the Kannur – Kozhikode belt and in Wayanad. The granite from Wayanad is used for construction of buildings in the neighbouring districts as well.

Weathering of rocks in the Nilgiri hills and Western Ghats is the reason for monazite sand deposits in the coastal areas of Kerala [43, 44]. Wayanad is a part of the Western Ghats and have altitude ranging from 700 to 2100 m. It has also been reported that Kabani River which originates from Wayanad and is a main tributary of the river Kaveri has ^{210}Po concentration varying from 0.77 to 1.27 mBql^{-1} with a mean of 1.10 mBql^{-1} [15]. Hence a study of the radioactivity concentration of Wayanad is imperative to understand whether the weathering of the soil and rocks from this region are contributing to the radioactivity content in the west coast of Kerala.

The study done by Mishra [45] shows that the content of ^{232}Th is high in the soil of Udagamandalam (114.6 Bq kg^{-1}). Several other papers have proved the presence of significant amount of ^{238}U, ^{232}Th and ^{40}K in the Nilgiris [46–48]. Udagamandalam and the Nilgiris form the bordering districts of Wayanad. However, data regarding Wayanad is not available. These points emphasize the need for a detailed study of the background radiation levels in the Northern part of Kerala.

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THE AIMS OF THE STUDY ARE AS FOLLOWS:

1) In air radiation dose in the Wayanad district by survey.
2) Distribution of naturally occurring radioisotopes namely, Radium, Thorium and Potassium in the soil and rocks of Wayanad district.
3) Annual Effective Dose and hazard indices due to terrestrial radioactivity in Wayanad district.
4) Indoor concentration of radon, thoron and their progeny concentrations in areas of higher terrestrial radioactivity and their variation with seasons and time of the day.
5) Inhalation dose due to the indoor concentration of Radon and Thoron and compare with the reference levels prescribed by regulatory bodies.
6) Soil gas radon concentration in the regions of higher indoor radon concentration.

RESHMA BHASKARAN “ ENVIRONMENTAL RADIOACTIVITY STUDIES IN WAYANAD, KERALA, INDIA”. THESIS. DEPARTMENT OF PHYSICS, UNIVERSITY OF CALICUT, 2018.

Chapter 2

LITERATURE REVIEW

The story began in the 15th century when silver was mined at Joachimsthal, a town in the then Czechoslovakia. Silver was threaded through hard black greasy pitchblende. It was from the Joachimsthal pitchblende that the German chemist Klaproth isolated the compound of Uranium in 1789 [49]. Since 15th century it was common knowledge that there was a disease amongst the mining community called the Schoenberg Mountain sickness. In 1879 it was found that the disease was a kind of cancer. Later on in the twentieth century the cancer was identified as lung cancer which was the cause of 50% of the deaths among the miners. In the 1920s radon, the daughter product of Uranium was suspected as the reason for the lung cancer. In future, over the years, the mechanism by which radon caused cancer was identified as well. Thus the society started exploring the presence of the naturally occurring radioactive materials to find the exposure due to these radioisotopes and the health hazards thereafter.

The discovery of X-rays by Roentgen in 1895 followed by the discovery of radioactivity in Uranium by Henry Becquerel in 1896 exhibited radiation hazard of a different kind. By the time the first measures for precaution were adopted, nearly 170 cases of radiation injury were recorded [50]. By 1922 nearly 100 radiologists had died due to overexposure of the medical uses of X-rays. Delayed effects of radiation also got attention during this period. In 1927, Muller studied radiation induced mutations in germ cells of Drosophila Melanogaster [51] which won him the Nobel prize in 1946. Genetic studies on human were initiated soon after the end of World War II in the 1940s. However, none of these studies could prove the genetic effect on offspring due to parental irradiation.

In 1921, the first national organisations for radiological protection came into existence. These bodies gave their first recommendations for radiation protection. In Stockholm, during 1928, international congress of radiology was conducted which saw

the establishment of International Commission on Radiological Protection [50]. The discovery of nuclear fission in 1939 and its applications led to the atomic bomb explosions in Hiroshima and Nagasaki causing many human deaths. The contamination of the environment with the nuclear explosions, the discharge of nuclear waste arising from the nuclear reactors and the use of X rays and other radioisotopes for medical and industrial uses led to extensive radiation exposure of the public. The people exposed in Hiroshima and Nagasaki has been widely studied for not only cancer induction but also for genetic effects to the offspring. The analyses of mental disorders in the children who were exposed in utero showed that severe mental retardation may be induced. During 8 to 15 weeks of gestation the threshold dose for this is 0.1 to 0.2 Gy. However, studies supporting the dose effect relationship without threshold are also available [52]. It was then in 1955, that the General assembly of the United Nations decided to include "Effects of Atomic Radiation" as an agenda in the tenth session [50]. Since then several UNSCEAR reports have come up discussing the various aspects of Environmental radioactivity.

The study of data on human exposures serves two purposes. One is that it presents individual exposure data which indicates the possible level of risk to which an individual is subjected. The second is that it could be used to study the total consequences in radiation harm from the various sources of radiation [53]. For the second, source related studies are required which will give the collective dose commitments. The collective dose commitments help in taking precautionary measures to reduce the radiation detriment. The source related studies are the studies that have been done worldwide to analyse the various contributors to the background radioactivity. Extensive studies were done to assess the natural radioactivity content which lead to the division of the region of interest as High Background Radiation Areas and Normal

Background Radiation Areas. The classification of a region of interest as mentioned in the introduction is based on the concentration of different environmental parameters. However Sohrabi [54] suggested another criteria which depends on the unified system of limitation of annual effective dose. According to him,

1) Low Level Natural Radiation area (LLNRA) or Normal Level Natural Radiation Area (NLNRA) is one where the several environmental factors lead to annual effective dose which is equal to two times the average global annual effective dose from natural sources as given in UNSCEAR [5].
2) Medium level Natural Radiation Area (MLNRA) is one where the exposure to to the natural sources of radiation is higher than LLNRA but is less than or equal to the pre-established level of 20mSv/y.
3) High Level Natural Radiation Area (HLNRA) is one where the exposure due to natural sources of radiation is higher than 20 mSv y^{-1} but less than 50 mSv y^{-1}.
4) Very High Level Natural Radiation Area (VHLNRA) is one where the exposure due to natural sources of radiation is higher than 50 mSv y^{-1}. In this case he recommends evacuation as the first step in remedial action.

2.1 MAIN HIGH BACKGROUND RADIATION AREAS OF THE WORLD

2.1.1 Brazil

Pocos de Caldas, Arax'a, Tapira which are the volcanic alkaline intrusive zone and Guarapari are the high background radiation areas of brazil [55]. The reason for the elevated levels of radiation is the monazite sand deposits on the black sand beaches. The dose rate in these beaches is as high as 50 μGy/h. In Rio de Janeiro, near the monazite separation plant in Buena Lagoon, abnormal radium concentrations were found [56]

The radiation dose from the samples collected from the aforementioned sites range between 3.5 and 10 mSv/y [57] Among the Brazilian HBNRA, Pocos de Caldas has the highest average annual effective dose of 13 mSv. Brazil has witnessed epidemiological studies in these HBNRAs since the past 10 years. The studies related to mortality due to cancer and other causes were done. The result of the studies indicated that the expected cancer mortality was higher in the HBNRAs as compared to control areas. [58]. In Pocos de Caldas radon levels in air have been found to be high. The geometric mean of the radon levels has been reported as 200 Bq m^{-3} and 61 Bq m^{-3} for rural and urban areas respectively. It has also been found that 16% all deaths caused by lung cancer in POCOS de Caldas may be due to radon exposure. [59]

2.1.2 China

Yangjiang in China is identified as HBNRA. The average annual dose of external radiation has been found to be 3.5 mSv/y [60]. The primary source of this elevated radiation is Monazite. The internal dose annually to the residents has been found to be 4.27 mSv. Studies on thyroid nodularity were done in this area by Wang et al [61] The results show that the dose to the thyroid is twofold higher than that of the residents of NBRAs. However, no increase in the nodular diseases of thyroid was found as compared to the control. However higher exposure is expected to cause chromosome damage. Increase in frequency of dicentrics and ring chromosome aberrations have been found from the peripheral lymphocytes of the residents of HBNRA. Zou et al conducted epidemiological studies of cancer incidence among the residents of Yangjiang [60]. No significant increase in incidence of cancer mortality rates was found as compared to the controls. However, all these data require futher studies as the results of the earlier studies are based on scant data.

2.1.3 Iran

Ramsar in Iran is found to be another high background radiation area. The elevated natural radioactivity in this area is found to be due to ^{226}Ra and its decay products which have been brought to surface by the various hot springs. Extensive studies were done in this area by Sohrabi and Esmaili [62]. They reported that the annual effective dose to the public in Ramsar ranged from 0.6 to 131 mSv with a mean value of 6 mSv. Along with the hot water springs high indoor radon concentrations also contribute to the elevated radiation dose due to natural radioactivity to the residents. The residents of the hot spring areas use Ra-enriched rock from the hot springs as building materials [63]. The indoor radon concentration in Ramsar is 31 kBq m^{-3}. Hence the total internal and external exposure leads to an annual effective dose in the range of 3.0 to 202 mSv. Biological studies conducted in Ramsar have showed an increased frequency of chromosome aberration [64, 65]. Another study which investigated the alterations in eight tmor biomarkers in blood samples from residents of Ramsar found statistically significant correlation between chronic exposure and the concentrations of three of the eight investigated tumour markers [66]. However some other studies which oppose the findings suggest that the HBNRAs residents have developed a resistance to high radiation doses have also been reported [67].

2.1.4 India

Parts of Kerala, Tamilnadu and Orissa coasts in India are high background radiation areas. The west coast of Kerala, namely, the Chavara – Neendakara belt and the Manavalakurichi region of Kanyakumari are the well-studied high background areas of south India [23, 24]. In Kerala the effective doses ranged from 0.5 to 15 mSv y^{-1}. In the east coast of India, the absorbed mean gamma dose rates in air was found to be 1925 ±

718 nGy h^{-1}. The annual effective dose for the region was found to range from 0.78 to 3.086 mSv y^{-1}.

The residents of the HBNRA in India have been subjected to large scale epidemiological studies since the 1960s. High incidence of Downs syndrome was reported by Kochupillai et al., [68] in the Chavara- Neendakara region. From the studies in comparison to the control population they found that radiation induced anomalies occurred with above – average frequency in the HBNRA residents. However some other studies like those done by Das and Karuppasamy [69] concluded that radiation has no significant effect on the induction of micronuclei frequency among infants. Nair et al.,[26] also concluded that the cancer incidence in the HBNRA revealed no high-background-radiation-related excess of malignant tumours.

In India, studies have been conducted at several other places to find the terrestrial radioactivity as well as the dose due to internal and external sources of radiation. Mishra and Sadasivan [70] conducted a nationwide study of terrestrial radioactivity in 1971 itself. Their result projected a nation average dose rate of 710 μGy h^{-1} which included cosmic ray component of 290 μGy h^{-1}. Of the terrestrial component 49% was from ^{40}K, 34% from ^{232}Th series and 18% from ^{238}U series radionuclides.

The Himalayan terrains have been subject to extensive studies as it was found that the concentration of indoor radon and thoron was more in the colder regions of the world as compared to the well ventilated hotter regions. Manjulata Yadav et al., [31] studied the terrestrial radioactivity of Garwal. Their result suggests that the dose to residents of the area varied from 22 to 93 nGy h^{-1} with an average of 55 nGy h^{-1}. Gussain et al., [71] studied the Kumaon region and stated that the dose in these region ranged from 80 to 179.6 nGy h^{-1}.

Studies have been done in Rajasthan by Asha Rani et al., [72], Vikas Duggal et al., [73] etc. Their results show that the concentration of ^{40}K is high in Northern Rajasthan with a mean concentration of 1627 Bq kg^{-1}. The average annual effective dose was found to be 0.63 mSv in Northern Rajasthan whereas in western Rajasthan this value was 0.41 mSv.

Parts of Punjab and Himachal Pradesh were studied by Rohit Mehra and Manmohan Singh [74]. Their results showed that the average annual effective dose in both these states was less than the world average.

East coast regions of India, namely the Odisha coast have been studied as it was found that the background radioactivity was relatively higher in these regions. Gusain et.al.,[75] found that the dose rate due to terrestrial radioactivity varied from 77 to 1651 nGy h^{-1} with an average of 230 nGy h^{-1}. Sivakumar et al.,[76] studied the east coast of Tamilnadu and concluded that the annual effective dose rate ranged between 0.01 and 0.10 mSv with a mean value of 0.04 mSv y^{-1}. This value was found to be less than the world average. Studies by Senthilkumar et al., [77] in Thiruchirapalli, Tamilnadu showed that the outdoor annual effective dose ranged from 48.3 to 153.1 μSv y^{-1} with an average of 97.9 μSv y^{-1}.

Radon, thoron and their progeny form more than 50% of the radiation dose due to natural radioactivity. In India Subba Ramu et al,[78] did an initial survey over a period of three years of the houses in 25 different locations to estimate the concentration of indoor radon. Their results showed that the indoor radon concentration in Indian houses varied from 2.2 to 56 Bq m^{-3} with a geometric mean of 15 Bq m^{-3}. Thereafter widespread studies are on-going in India to estimate the contribution of radon and thoron to the dose due to natural radioactivity.

The HBNRAs have remained the main focus for the radon thoron measurements in India. Several studies have been conducted as mentioned earlier to study the effect of radiation on the health of the residents. The inhalation dose due to natural background radiation is mainly due to the radon and thoron progeny. Mayya et al.,[79] conducted studies in both the east and west coast HBNRAs of India to analyse the radon and thoron progeny concentration using the newly devised direct progeny sensors. Their results show that the maximum thoron progeny concentration was measured in Panmana as 3.32 Bq m^{-3} and the maximum radon progeny concentration was found in Neendakara. The ratio of thoron progeny dose to the radon progeny dose was found to be 0.6. Thus they concluded that the dose contributed due to thoron progeny forms a significant part of the inhalation dose. Similarly, in the HBNRA of east coast of India the ratio of the thoron progeny dose to the radon progeny dose was found to be 0.64. Thus they concluded that along with the study of concentration of radon and its progeny, the study of thoron and its progeny is also important to arrive at a correct estimation of the inhalation dose due to natural background radiation. Omori et al., [80] conducted long term measurement of residential radon, thoron and progeny concentration in Chhatrapur placer deposits in Odisha. They found that the inhalation dose in these regions ranged from 0.8 $mSvy^{-1}$ to 4.6 $mSvy^{-1}$ with an arithmetic mean of 1.8 mSv y^{-1} and a geometric mean of 1.7 $mSvy^{-1}$. As the exhalation of radon from the soil gas is considered as one of the main reasons of indoor radon concentration. Kanse et al.,[22] conducted study in the HBNRAs of Kerala and Odisha, to assess the thoron exhalation from the monazite rich sands of these regions. They found that the average in situ thoron exhalation rate in Kerala was about two times greater than the average thoron exhalation rates in Odisha. Their results also concluded that the thoron exhalation rate mainly depend on the ^{232}Th series radionuclide concentration in the sand.

Only few results are available from the north-eastern states of India. Mishra et al., [81] studied radon thoron concentration in Shillong, Meghalaya for a period of three years. The average radon concentration for the whole period was found to be 68 ± 22 Bq m^{-3} and the average thoron concentration was found to be 33 ± 14 Bq m^{-3}. The maximum concentration of radon was 166 Bq m^{-3} and that of thoron was 95 Bq m^{-3}. Zoliana et al., [82] conducted a study of radon concentration in the dwellings in Aizwal which is considered to be the region with highest cases of lung cancer amongst both male and females in India. However, their study could not find any correlation between the incidence of cancer and the radon exposure.

Due to results of high indoor radon concentration from Sweden and other countries with cold climate, the Himalayan regions of India have been widely studied. Ramola et al., [83] studied the diurnal variation of indoor radon concentration in the Budhakedar and Tehri regions of Garhwal. Singh et al.,[84] studied the correlation of terrestrial radionuclide contents with radon thoron concentration in Himachal Pradesh by studying the radon and thoron exhalation from soil in this region. They found positive correlation between ^{238}U and radon mass exhalation rate and ^{232}Th and thoron mass exhalation rates. Singh et al.,[85] studied the soil gas radon concentration in the upper Sivaliks using AlphaGUARD. They found that the radon concentration in the soil gas in Upper Sivaliks varied from 11.5 ± 0.9 kBq m^{-3} to 78.47 ± 3.1 kBq m^{-3}. They concluded that high concentration of radon in soil gas in this region was due to the fact that the region lies near the thrust fault of Nadah and Mandana. They also found positive but week correlation between the soil gas radon concentration and indoor radon concentration.

RESHMA BHASKARAN “ ENVIRONMENTAL RADIOACTIVITY STUDIES IN WAYANAD, KERALA, INDIA”. THESIS. DEPARTMENT OF PHYSICS, UNIVERSITY OF CALICUT, 2018.

Chapter 3

LOCALE AND METHODOLOGY

Since Kerala is amongst the HBNRAs of the world, the coastal region of Kerala has been very well studied. However, the annual effective dose to the public in this region cannot be used as a representation of the dose to the whole of Kerala. This is because the topography of Kerala is highly diverse which has the Western Ghats which forms the high altitude regions of the state as well as the southern coast. Not only the geology but also the temperature,humidity,seasons all are very diverse in the various parts of the state. Hence it is only reasonable to assume that there may be large variation in the NORM present in these places. Idukki and Wayanad form the main hill stations of Kerala. However, there is no literature available for these places with regard to NORM. Hence this work is one of its kind in Wayanad district to estimate the contribution of NORM to the radiation dose to the common man living in these areas. This will not only help in setting up a baseline data for Wayanad district but also help in getting a clear picture of the variation other parts of Kerala has with respect to the HBNRA.

As mentioned earlier the dose due to NORM include three main forms namely, external dose due to the terrestrial radionuclides, ingestion dose due to the terrestrial radionuclides and inhalation dose due to radon, thoron and their progeny. Hence the present work was divided into three main parts. First a general radiation survey of the region was conducted. This helped in getting a picture of the overall variation of background radiation in the district. This was followed by sample collection of soil and rocks in the region and their study to analyse the concentration of the terrestrial radionuclides. Once the concentration of the major terrestrial radionuclides was assessed radon, thoron and their progeny were studied in the region of relatively higher terrestrial radioactivity.

3.1 GEOLOGY

Wayanad district came into existence as the 12th district of Kerala. The plateau is situated between 700 metres and 2100 metres above mean sea level. The district forms a part of the Western Ghats and shares its borders with Kozhikode, Kannur and Malapuram districts of Kerala. It shares its borders with two other states of India namely Tamilnadu and Karnataka. Out of the total area of 2131 km^2 approximately 787.87 sq. km. area of the district is under protected forest.

The name Wayanad originated from the word "Vayal Nadu" which means land of Paddy field, now it is popular for coffee and spices. Kabani is the main river in Wayanad which is one of the three east flowing rivers in Kerala. The Panamaram, Mananthawady and Thirunelli rivers are its main tributaries. Besides Kabani, the Chaliyar and Valapattanam rivers also form the drainages of Wayanad.

Wayanad experiences an average rainfall of 2786 mm. Southern, Southwest and northeast regions receive more than 3000 mm of annual normal rainfall [86]. Climate of Wayanad is generally hot and humid. March and April are the hottest months. January and February are the coldest. The month of June has the maximum rainfall. The main soil types of Wayanad are laterite soil, brown hydromorphic soil, forest loam and riverine alluvium. The laterite soil has very less organic matter and the pH ranges between 5.5 and 6.5. Forest loam is found in Manathawady, Kalpetta and Sulthan Bathery blocks. They are rich in organic matter and are slightly acidic in nature.

Brown hydromorphic soil is formed by the transportation and sedimentation of materials from hill slopes. The pH ranges from 5.2 to 6.3 and is slightly acidic in nature. Alluvial soils are found along the banks of Chaliyar, Kabani and its tributaries. The riverine alluvium contains moderate organic matter, nitrogen, phosphorus and potash.

Gold, steatite and molybdenite are some of the economic minerals available in the district.

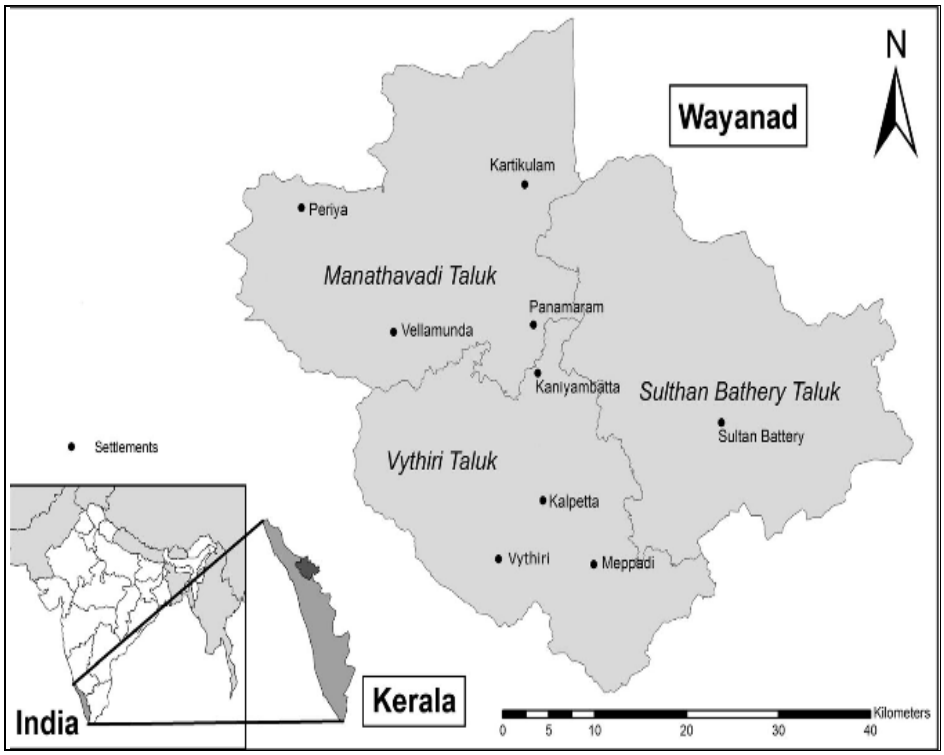

Fig 3.1 Map of the study area

The district is blessed with a large variety of granites. These can be broadly classified into two main types namely the Ambalavayal granites and the Kalpetta granites. The Ambalavayal granite is of four types, viz., foliated granite, pink granite, grey granite and aplitic granite [87]**.** These granites are composed of quartz, pink feldspar, hornblende and biotite. Rocks in the Kalpetta granites are grey in colour, medium grained and homogenous biotite granite. These granites have sharp contact with the country rock.

3.2 SAMPLE COLLECTION

In air radiation survey was the first phase of the work. A general survey of the district was done using High Pressure Ionisation chamber based survey meters. The

survey meter used was Fluke 451B. The survey meter detects alpha above 7.5 MeV, beta above 100 keV and gamma above 7keV. The operating ranges are 0 to 5mR/h, 0 to 50 mR/h, 0 to 500 mR/h, 0 to 5 R/h and 0 to 50R/h. The detection volume is 349 cc air ionisation chamber. The survey meter was calibrated using ^{137}Cs sources before every field visit.

After the initial radiation survey the soil and rock samples were collected from different parts of Wayanad district. A total of 58 soil samples and 13 rock samples were collected. The specifications of the sampling sites are as given in the Appendix A. Each sampling site was divided into a grid of 2 x 2 m^2. Surface soil along with vegetation covering the surface was removed. The soil samples were collected from surface (10-15 cm depth) at four corners and centre of the grid at every place. These were then mixed to form one sample representing the area. The sample was thoroughly dried to attain uniform dry weight. The vegetation and roots were separated. The sample was sieved with a 1mm mesh to attain uniform grain size. It was then filled in a container of 7 cm height and 5 cm base diameter. The containers were filled to the brim for uniformity. These bottled samples were sealed thoroughly, weighed and kept for a period of 30 days to attain secular equilibrium. The code, weight and date of packing were marked on each sample.

The granites in Wayanad district can be divided into two main types, namely the Ambalavayal granites and the Kalpetta granites. The rock samples were collected from all accessible granite quarries and include both Ambalavayal and Kalpetta granites. Powdered rock samples were dried, sieved and stored in a hermetically tight container of the same dimension as mentioned earlier to attain secular equilibrium. To find any correlation between the soil and the rock samples of the area, soil samples were collected from a distance of 2 km away from the quarries from where the rocks were collected.

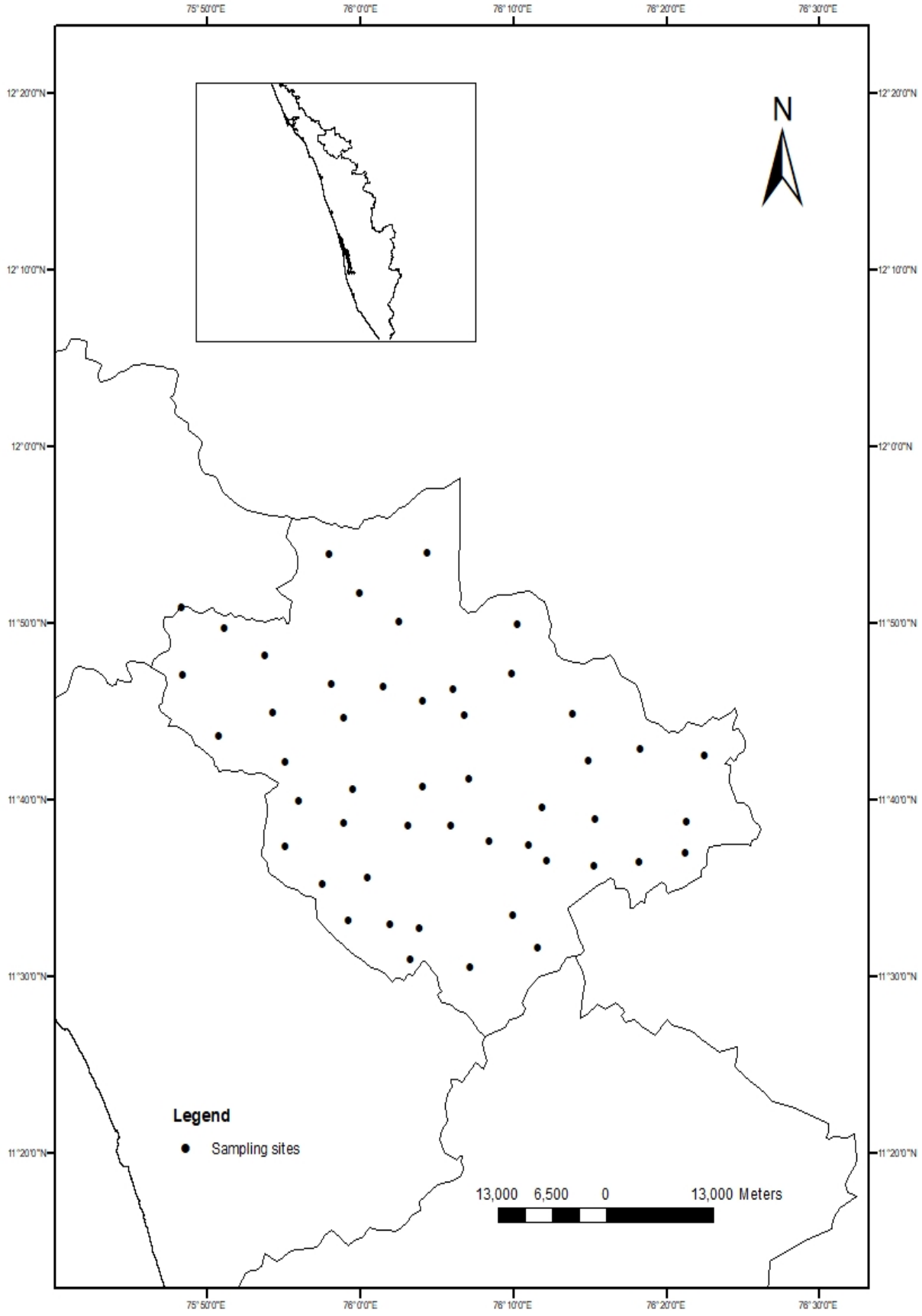

Fig: 3.2. Soil sampling locations in Wayanad District

3.3 SOIL GAMMARAY SPECTROMETRY

The fundamental principle of gamma ray spectrometry is that gamma ray photon has a discreet energy. This energy is characteristic of the source isotope from which it is emitted. Hence by measuring the energy of the gamma ray photon we can identify the radiation source.

The NaI (Tl) detectors are one of the most common detectors used for gamma ray spectrometry. A gamma ray interacting with a scintillator produces a light pulse which is converted to an electric pulse by the photomultiplier tube. The high atomic number of Iodine gives good efficiency for gamma ray detection. Maximum emission in pure NaI corresponds to a wavelength of 303 nm. Thallium is used to activate the crystal. A small amount of thallium when added increases the light emission. Thallium activator shifts the wavelength of emission from 303 nm to 410 nm, which matches with the sensitivity of the photo multiplier tube. NaI is a dominant material for the detection of gamma rays because of its better resolution and low cost. The amount of light produced in the detector is proportional to the gamma energy deposited.

The detector used in the present work is also NaI (Tl) detector with 3” x 3” crystals available at Indira Gandhi Center for Atomic Research (IGCAR), Kalpakkam. The Scintillation detectors consist of three main parts namely, the scintillator, the photomultiplier tube and the signal processing electronics. Fig 3.3 shows the schematic diagram of a scintillation detector.

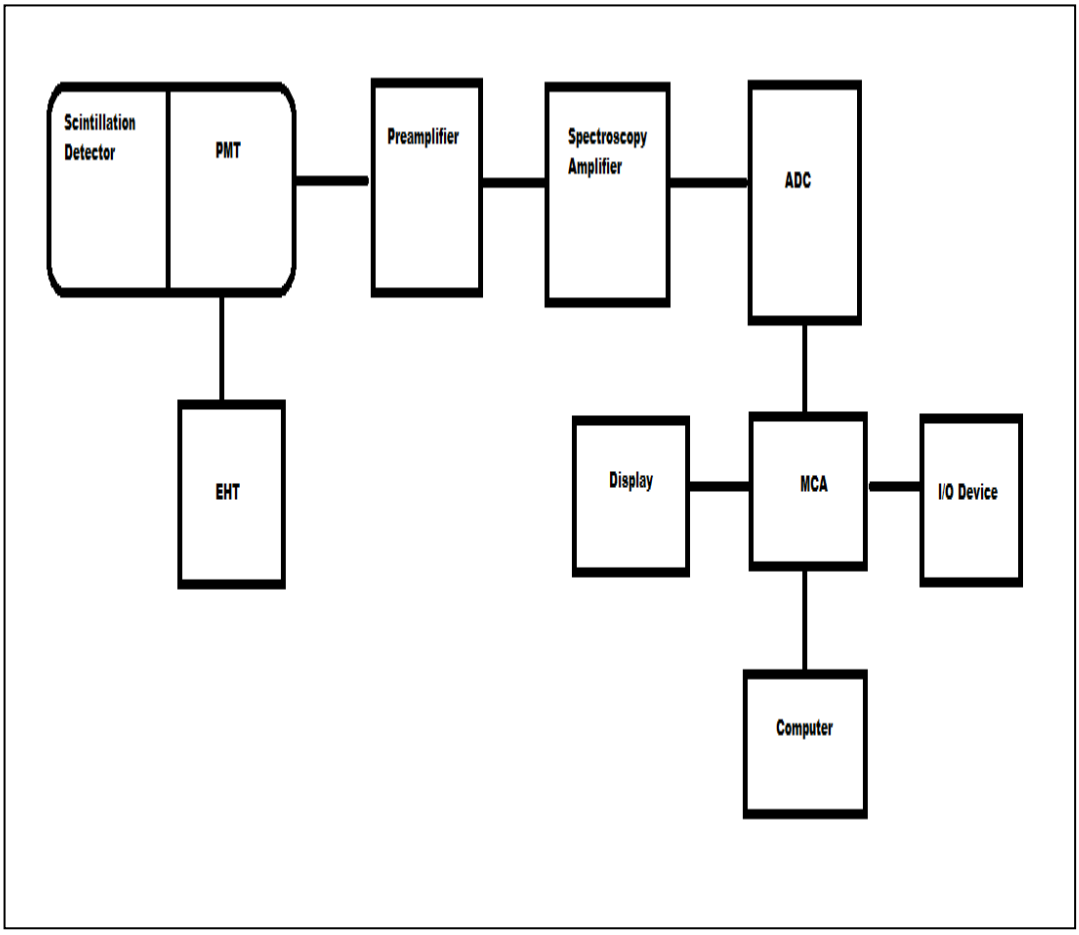

Fig: 3.3 Schematic diagram of a NaI(Tl) scintillation detector

The photocathode converts the scintillation photons to electrons. The emitted electrons are then multiplied using the photomultiplier tube with the help of dynodes in it. Even though many scintillation crystals are used as detectors, the advantage of NaI(Tl) crystals is that they have a linear response to electrons and gamma rays over a large energy range [88]. The resolution of the crystal is 7% for 662 keV ^{137}Cs gamma rays. The detector was sealed to the background radiation using a 15 cm thick lead shielding on the sides and 10 cm lead shielding on the top. Nucleus PCA – II 8 k multichannel analyser was used in 1K mode for spectrum analysis. Multichannel analyser collects the data and produces it in the form of a quantifiable output.

3.3.1 Calibration of Gamma Ray Spectrometer

The Gammaray spectrometer was calibrated for both energy and efficiency. Energy calibration is necessary to identify the gamma energy corresponding to the respective channel number. Efficiency calibration of the detector is essential to identify the activity of the radionuclide in the sample.

A. Energy calibration

Energy calibration is carried out to identify various radioisotopes present in the sample by their corresponding gamma ray energy. The linear dependence of Gammaray energy with the channel number is used for this purpose. Channel numbers obtained using multichannel analyser are converted to energy in keV by energy calibration.

The spectrum of ^{137}Cs, ^{60}Co, ^{54}Mn, ^{133}Ba, ^{214}Bi and ^{208}Tl sources, the gamma ray energies of which are already known, were used for energy calibration. The measured peaks in the spectrum of these sources were compared with the corresponding energies as given in Table 3.1.

Table 3.1
Data for energy calibration of NaI (Tl) detector

Radioisotope	Energy (keV)	Channel Number
^{133}Ba	356	105
^{137}Cs	661.51	221
^{54}Mn	834.83	284
^{60}Co	1173.24	409
^{60}Co	1332.50	466
^{214}Bi	1764.5	618
^{208}Tl	2614	922

Energy calibration then is the relationship between the channel number and the photon energies. Once the energy calibration has been established over the range of interest, the calibration graph is plotted by the photon energies with the channel number, as shown fig.3.4.

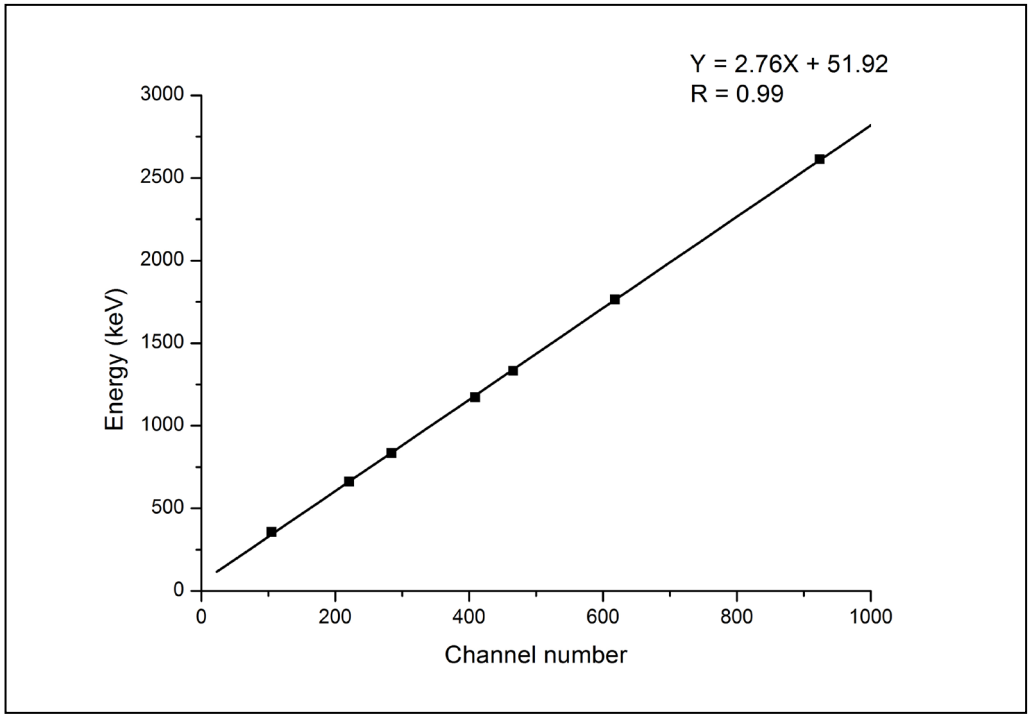

Fig: 3.4 Energy calibration of NaI (Tl) detector.

B. Efficiency calibration

Efficiency calibration was done using standard sources with known activity provided by IAEA namely, RGU-1 for ^{238}U, RGTh-1 for ^{232}Th and RGK-1 for ^{40}K. Once the activity of the radionuclide and the Gammaray abundance for the particular energy is known, efficiency for the specific energy can be calculated using the following formula (3.1)

$$\eta = \frac{CPS}{A \times \gamma} \qquad 3.1$$

Here η = detector photopeak efficiency for gamma energy

CPS = gamma peak count rate (cps)

A = activity of radionuclide (Bq)

γ = Gammaray abundance

The absolute efficiency of the detector for ^{238}U is 0.34%, for ^{232}Th is 0.31% and for ^{40}K is 0.15%. Efficiency calibration was done using the container geometry similar to the one used for the samples.

C. Minimum Detectable Activity

Minimum detectable activity is used to estimate the smallest amount of radioactivity that can be determined reliably. The minimum detectable activity of the Gammaray spectrometer was determined using an empty bottle of the same geometry to get the background Gammaray spectrum. From the spectrum the counts of the isotopes corresponding to the ^{238}U, ^{232}Th and ^{40}K were noted. These form the background counts for these sources. The MDA was calculated for the radionuclides using the equation

$$MDA = \frac{(4.65\,\sigma B + 2.706)}{(\varepsilon\,\gamma\,t)} \qquad (1)$$

Here σ_B is the standard deviation of background gamma counts for the energy of the radionuclide being studied, ε is the absolute detection efficiency, γ is the radiation yield per disintegration for the particular radionuclide and *t* is the counting time in seconds for the sample. The MDA of ^{40}K, ^{238}U and ^{232}Th was found to be 8 Bq/kg, 2 Bq/kg and 2 Bq/kg respectively.

3.3.2 Activity

The main radioisotopes in terrestrial radioactivity which are studied using Gammaray spectrometry are ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K. However, determination of activity concentration of ^{238}U, ^{226}Ra and ^{232}Th cannot be done directly due to their low

relative gamma ray intensity following decay. In the uranium decay chain some long lived radionuclides namely, ^{238}U, ^{226}Ra and ^{210}Pb can be the head of the decay chain segments. These can be measured through secular equilibrium grow up conditions. Table 3.2 gives the principle isotopes forming decay chain segments in the uranium decay chain and are used to study the secular equilibrium through gamma ray spectrometry measurements.

Table 3.2

Heads of decay chain segments in ^{238}U decay chain and the respective grow up times

	Chain segment Head	Grow up time interval (>99%)	Isotope	Energy (keV)	Intensity
1	^{238}U	168.7(d)	^{234Th}	63.9[e]	4.80
				92.4[e]	2.81
				92.8[e]	2.77
			$^{234}Pa^{m}$	1001.0[b]	0.84
2	^{234}U	Not detected	-	-	-
3	^{230Th}	Directly detected	^{230}Th	67.7[d]	0.38
4	^{226}Ra	Directly detected	^{226}Ra	186.2[c]	3.59
				351.9	37.6
			^{214}Pb	295.2	19.3
				242.0	7.43
				609.3	46.1
			^{214}Bi	1764.5	15.4
				1120.3	15.1
5	^{210}Pb	Directly detected	^{210}Pb	46.5[d]	4.25
6	^{210}Po	Not detected	-	-	-

a. Grow up time calculated as 7 times half-life of the longest lived isotope of the chain segment
b. Low yield gamma energy
c. Generally recorded as doublet with 185.7 keV (^{235}U)
d. Low energy (requires n type HPGe detector for best sensitivity and accuracy)
e. Recorded as doublet between 92.4 keV and 92.8 keV from ^{234Th}

The principle isotopes which may be used to study the secular equilibrium in the thorium chain are given in Table 3.3

Table: 3.3
Heads of decay chain segments in thorium decay chain and the respective grow-up times

	Chain segment Head	Grow up time interval (>99%)	Isotope	Energy (keV)	Intensity (%)
1	^{232}Th	Not detected	-	-	-
2	^{228}Ra	1.8 (d)	^{228}Ac	911.1 968.9 338.4	26.0 16.2 11.3
3	^{228}Th	25.7 (d)	^{212}Pb ^{212}Bi ^{208}Tl	115.2 238.6 300.1 727.2 1078.6 1620.56 583.1 860.5 2614.0	0.592 43.3 3.28 6.58 0.564 1.49 84.5 12.42 99

The activity concentration of the radionuclides was determined from the spectrum of each sample. 1.46 MeV photopeak was used to determine the counts corresponding to the ^{40}K concentration. Similarly, the 1.76 MeV peak corresponding to ^{214}Bi was used to identify the activity of ^{226}Ra and 2.614 MeV peak of ^{208}Tl was used for ^{232}Th.

The activity concentration, *A* of different radioisotopes in the soil was calculated using the equation [89]

$$A = \frac{N_E}{t\gamma\varepsilon M} \qquad (3.3)$$

Here N_E is the net area of the peak at energy E, t is the total counting time, γ is the gamma energy yield per disintegration of the radionuclide, and ε denotes detection efficiency of the detector for the energy E and M is the mass of the sample.

The activity of the radionuclides in soil causes outdoor radiation exposure to the natives of the place. Radiation exposure to an individual is described in terms of "effective dose". The annual effective dose due to any source of radiation to the residents of a region includes both external and internal doses and is usually reported in milliSieverts (mSv).

3.3.3. Annual effective dose

Both the internal and external effective doses depend on the data available from the environmental measurements. Absorbed dose rate in air, expressed in nanoGray per hour (nGy/h), from natural sources is a main quantity to characterize external exposure from natural sources. The absorbed dose rate in air at a distance of 1m from the soil surface was calculated using the equation [18].

$$\text{Dose Rate } \left(\frac{\text{nGy}}{h}\right) = 0.604\, C_{Th} + 0.462 C_{Ra} + .0417 C_K \qquad (3.4)$$

Here C_{Th}, C_{Ra} and C_K are the activity concentration in Bq/kg of ^{232}Th, ^{226}Ra and ^{40}K, respectively.

The dose rate obtained using the above equation can be converted to Annual Effective Dose in adults by using a transformation factor of 0.7 Sv/Gy [1]. This factor converts the absorbed dose in air to external effective dose to adults. Also it is generally assumed that an average adult spends nearly 80 % of his time indoors and only 20 % outdoors. Hence the Annual Effective Dose (AED) outdoor was calculated using an

occupancy factor (OF) of 0.2 (20% occupancy outdoors in a day) and the Gray to Sievert conversion factor of 0.7 $GySv^{-1}$. For the calculation of AED indoors the OF used was 0.8 (80% occupancy indoors in a day).

$$\mathrm{AED} = \mathrm{D}\left(\frac{\mathrm{nGy}}{\mathrm{hr}}\right) \times 8760 \times \mathrm{OF} \times 0.7 \times 10^{-6}\mathrm{mSv} \tag{3.5}$$

The nuclides of interest in terrestrial radioactivity contribute to the external exposure of persons roughly according to the ratios of their specific exposure rate constants namely ^{40}K: ^{226}Ra: ^{232}Th: 1: 10: 15. This was used to arrive at a generalised treatment of exposure to gamma rays. Thus gamma representative index [90] was defined as

$$I_\gamma = \frac{C_K}{1500} + \frac{C_{Ra}}{150} + \frac{C_{Th}}{100} \leq 1 \tag{3.6}$$

Here I_γ is the gamma representative index and C_K, C_{Ra} and C_{Th} are activity concentration of Pottassium, Radium and Thorium respectively. The value of the sum should be either equal to or less than unity to conclude that the hazard due to radiation is negligible.

3.3.4 Hazard Indices

Way back in the 1970s itself it was found that the construction materials made up of mud and other raw products which contained measurable quantities of Uranium and Thorium radioisotopes led to increased levels of indoor radiation exposure. To quantify the effect of the radiation exposure due to materials used for building construction certain hazard indices were defined. These were defined for both indoor and outdoor exposures as per the following equations[91]

$$\mathrm{H_{ex}} = \frac{\mathrm{C_{Ra}}}{370} + \frac{\mathrm{C_{Th}}}{259} + \frac{\mathrm{C_K}}{4810} \leq 1 \tag{3.7}$$

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (3.8)$$

Here H_{ex} is the external hazard index and H_{in} is the internal hazard index.

3.4 RADON THORON MEASUREMENTS

Radon gas emanates from the rocks and soils and concentrates in enclosed spaces like rooms in the houses. One of the important sources of residential radon is soil gas infiltration. It forms a major contributor to the ionising radiation dose received by the general population.

Indoor measurements in this study were divided into two phases. In the first phase (2014-2015), indoor radon and thoron concentrations were measured using Pinhole dosimeters[92]. This study included all the villages in Bathery Taluk. Along with the indoor measurements, soil samples were collected from the top surface (of 15 cm depth) from the vicinity of the houses. These samples were analysed using Gammaray Spectrometry for assessing terrestrial radioactivity. Radon and thoron exhalation studies were also done on the samples to obtain the mass exhalation rate for radon and the surface exhalation rate for thoron.

Second phase consisted of 19 houses in the Ambalavayal region which were studied during 2015-2016. Out of these 19 houses 10 were studied using both active and passive measurement techniques. The second phase study included the measurement of both radon and thoron concentrations and also their progeny. It is understood that the concentration of radon and thoron are effected by the ventilation, temperature and pressure of the rooms. This means that the indoor concentration of radon and thoron varies not only as per the seasons in a year, it also varies at different times in a day and

night. The diurnal variation of radon and wall and floor exhalation was also studied by using active measurement techniques.

3.4.1 Passive indoor measurement

Passive measurement of radon and thoron relies on the method of time integrated monitoring. Alpha particles when it passes through certain plastics leave a microscopic track of damage. Solid State Nuclear Track Detectors (SSNTD) are used as detectors in the alpha particle dosimetry. Exposure to the radiation causes damage in the film. The damages develop as a track by the chemical etching process. The number of tracks registered is proportional to the activity of the alpha emitting radionuclides.

To get a correct measurement of the radon concentration, the mode in which the SSNTD is used is significant. Initially SSNTD in bare mode was used for these measurements. However, in bare mode the response in the film included tracks from radon and thoron as well as their progeny. The contribution from the decay products to the net counts was filtered out by using a diffusion chamber with filter paper. The filter paper filtered the particulate decay products. However, in this detector it was not possible to differentiate between radon and thoron. Hence to differentiate between radon and thoron the twin cup dosimeters were developed by Mayya et al. [93]. The twin cup dosimeters even though efficient in separating radon and thoron showed erroneous results at times [92]. This was due to the variation in the entry rates from the two entrances of the dosimeter due to turbulence or air flow in one direction only. This ambiguity of different entry rates of radon was removed by the development of pin hole dosimeters [92]. LR115 films based pinhole dosimeter has a single face for gas entry and gives the time integrated measurement of radon and thoron [94]. The dosimeter consists of two cylindrical diffusion chambers, each of length of 4.1 cm and radius

3.1 cm separated by the pin hole based radon/thoron discriminating plates. The gas enters the first chamber after passing through a filter paper. This filter paper separates the particulate decay products of radon and thoron. The first chamber collects both radon and thoron. From this chamber, also called the 'radon + thoron' the gas diffuses to the second 'radon' chamber through four pin-holes (0.5mm radius). During diffusion from the first chamber to the second, the thoron is cut off due to its very short half-life (56s). The LR – 115 films kept in the first chamber registers tracks due to both radon and thoron and their decay products, whereas the second chamber registers the tracks due to radon and its decay products only. The pin holes have been designed in such a manner that the transmission of thoron is cut off to 99% while keeping the transmission of radon at about 98% into the 'radon' chamber. Construction of the pin hole dosimeter is as shown in figure.3.5.

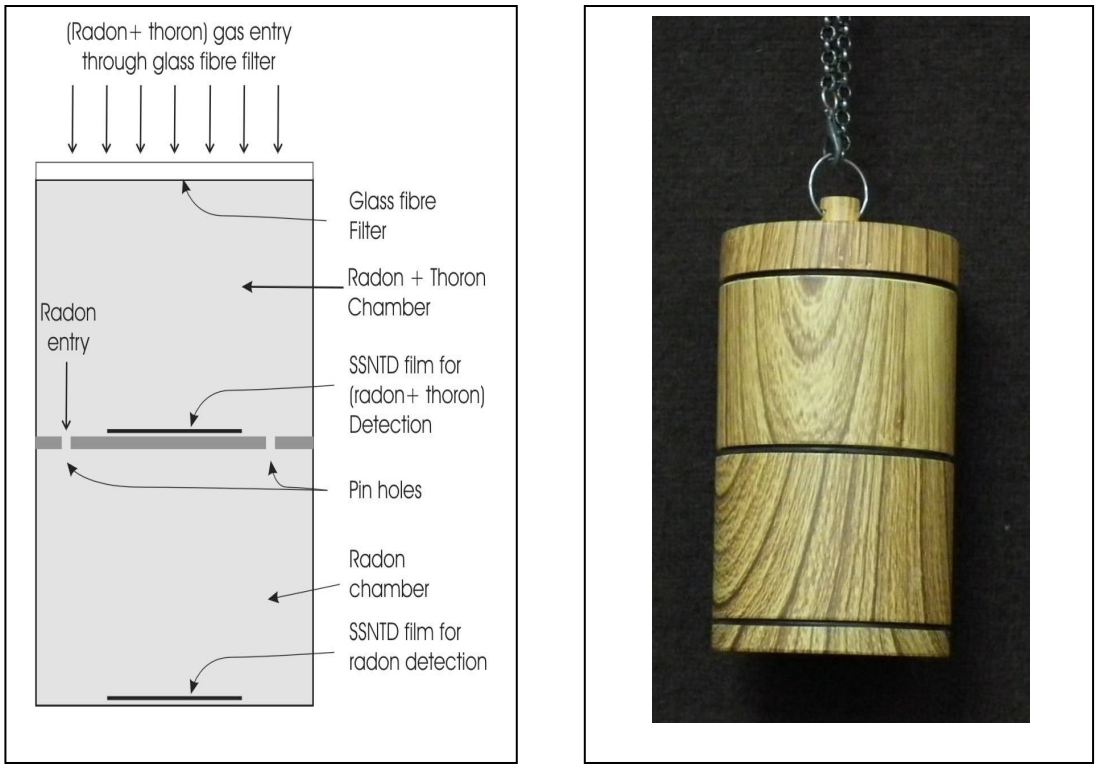

Fig: 3.5 Schematics of the pin hole dosimeter

In this work the Pin hole dosimeters equipped with LR-115 Type II Solid State Nuclear Track Detectors (SSNTD) with 2x2 cm^2 area were hung in the rooms of the houses (at least 30 cm away from nearby wall) for a period of three months. The dosimeters were hung at a height of around 2 m from the floor. The dosimeters were replaced with new ones at the end of every three months until a cycle of 1-year was completed. The retrieved dosimeters were processed in the laboratory and the developed tracks were scanned using a spark counter by the standardised protocol [95].

A. Chemical etching and counting

After exposing for a period of three months the detectors were retrieved and replaced with another set of detectors. The retrieved detectors were etched with 2.5 N NaOH solutions at a temperature of 60^0 C for 90 minutes without stirring. It is only after etching that the tracks created by the emitted radon, thoron and their progeny are visible for counting. Image of a sample etching bath is represented in Fig. 3.6

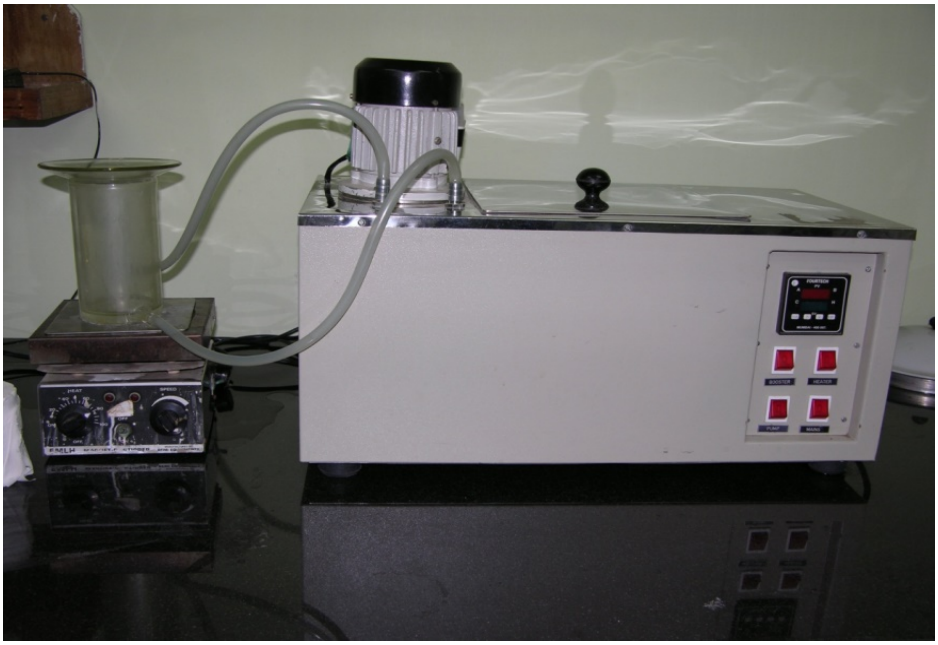

Fig: 3.6 Sample etching bath

After etching the detectors were peeled off from their cellulose acetate base. The counting of the tracks was carried out by spark counter. Spark counting technique was invented by Cross and Tommasino [96]. When the LR115 film is chemically etched the tracks enlarge and are converted to holes. This film is placed between the electrodes of the spark counter forming a capacitor. A thin aluminium coated mylar film is placed on top of the film to complete the circuit. With the application of a high voltage across this capacitor, an electrical spark takes place through the hole. The spark produced can be counted electronically [95]. The spark passing through the aluminised mylar evaporates it producing a spot of diameter about 100 μm. This prevents the production of second spark on the same track hole. Fig.3.7 shows the spark counter and the counting set up. Operating voltage of the spark counter is determined by plotting a graph between the applied voltages and counts. A plateau is formed after a particular voltage. The center of this plateau is taken as the operating voltage since any fluctuation from this voltage will not bring significant change in the number of counts.

Fig: 3.7 Spark Counter

The counts of the exposed film after the deduction of the background counts obtained from the control films were used to find the concentration of Radon and Thoron as per the following equations [94].

$$C_R = \frac{(T_1 - B)}{(d.k_R)} \quad (3.9)$$

$$C_T = \frac{(T_1 - d.\ C_R.k_R')}{(d.k_T)} \quad (3.10)$$

Here

T_1 is the track density observed in the radon compartment, k_R is the calibration factor of radon in radon compartment (0.017 tr. cm^{-2} d^{-1} / Bq m^{-3}) d is the number of days of exposure, T_2 is the track density observed in the radon + thoron compartment, k_R' is the calibration factor of radon in the radon + thoron compartment (0.0172 tr. $cm^{-2}d^{-1}$ /Bqm^{-3}). k_T is the calibration factor of thoron in radon + thoron compartment (0.010 tr.cm^{-2} d^{-1}/ Bqm^{-3}). B is the background tracks arising due to intrinsic properties and exposure during transit period. The Minimum Detectable Limit for radon in pin hole dosimeter is 2 Bq/m^3 and that for thoron is 6 Bq/m^3.

3.4.2 Radon and thoron progeny concentration

Inhalation dose due to radon and thoron are mainly contributed by their progeny. Hence knowledge of the cumulative decay product concentration is necessary to get the actual measure of exposure. The decay products are expressed in Equilibrium Equivalent Radon Concentration (EERC) and Equilibrium Equivalent Thoron Concentration (EETC). In this work, deposition based Direct Radon and Thoron progeny sensors (DRPS and DTPS) were used (Fig. 3.8). The sensors selectively detect α- particles from ^{214}Po and ^{212}Po.

Fig: 3.8 Direct Radon and Thoron progeny sensors

DRPS and DTPS have been developed by BARC Mumbai [97]. These are made up of LR-115 films (2.5 x2.5 cm^2) mounted with absorbers of sufficient thickness. The thickness of the absorber required is decided by alpha particle energy of the daughter product. In DTPS 50 μm aluminised mylar is used as the absorber. This permits only alpha particles of energy 8.78 MeV emitted from ^{212}Po. For ^{222}Rn progeny a combination of aluminised mylar (25 μm) and cellulose nitrate (12 μm) of effective thickness 37 μm is used as the absorber. This combination of absorber detects mainly 7.67 MeV alpha particles emitted from ^{214}Po. Fig 3.9 gives the cross section of the DRPS and DTPS detectors.

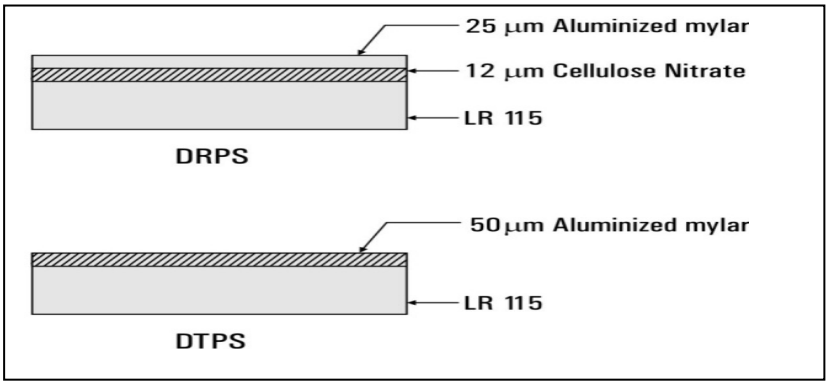

Fig: 3.9 Crossection of DRPS and DTPS

LR-115 films detect the alpha particles emitted from the deposited progeny atoms. The tracks can be converted to Equilibrium Equivalent Progeny Concentration (EEC) in air using the Sensitivity factor (S) as [98]

$$EEC = \frac{T}{S} \quad (3.11)$$

Here EEC is in Bqm^{-3}, T is the number of tracks per cm^{-2}. d^{-1} and S is the sentivity in (Tracks. cm^{-2}. d^{-1} / Bqm^{-3}). The sensitivity factor for the thoron progeny in DTPS is taken as 0.94 Tr. cm^{-2}. d^{-1} / Bqm^{-3}. For radon progeny the sensitivity factor for DRPS is 0.09 Tr $cm^{-2}.d^{-1}$/Bq m^{-3}. Since DRPS has interference from thoron progeny as well a sensitivity factor of 0.09 Tr. cm^{-2}. d^{-1} / Bqm^{-3} is taken for the thoron progeny which should be subtracted while calculating EERC from DRPS. The minimum detection limit of DTPS and DRPS are 0.1 Bq m^{-3} and 1 Bq m^{-3} respectively.

This sensor has no interference from the radon and thoron gas. Hence it gives a direct estimation of the progeny concentration in the air. The measurements were made by suspending the detector at least 30 cm away from the adjacent wall and the deposition surface faces the indoor environment.

The detectors were exposed for a period of 3 months and then replaced with new ones. After removal of the exposed films the general procedure for etching of the films and counting of tracks is performed.

3.4.3 Equilibrium factor for radon and thoron

Radon and thoron decay constantly giving rise to their progenies. These progenies have a short half-life and decay further till a stable isotope of lead is produced. It is well known that the inhalation dose is not due to exposure to radon and thoron but due to their progeny. Hence the estimation of the progeny and its concentration with respect to the parent is necessary to estimate the inhalation dose due to radon and thoron. The equilibrium factor (F) describes the ratio between the parent (radon or thoron) and its progeny.

UNSCEAR proposed an average value of 0.4 and 0.1 as the equilibrium factor for radon and thoron respectively [18]. However it is now known that equilibrium factor depends significantly on environmental factors like humidity, ventilation rate etc [99, 100]. The equilibrium factor of thoron varies more as compared to radon as it has a very short half-life. The equilibrium factor was calculated as

$$F_R = \frac{EERC}{C_R} \tag{3.12}$$

$$F_T = \frac{EETC}{C_T} \tag{3.13}$$

Here F_R denotes the equilibrium factor for radon and F_T denotes the equilibrium factor for thoron. EERC and EETC are the Equilibrium Equivalent Radon Concentration and Equilibrium Equivalent Thoron Concentration respectively. C_R and C_T are the concentrations of radon and thoron respectively.

3.5 ANNUAL EFFECTIVE DOSE

The study of radon thoron and progeny was done in two phases. In the first phase indoor radon and thoron concentration was estimated for the residents of Bathery Taluk. In this phase the progeny concentration was not estimated. The health effect due to radon and thoron exposure was quantified by using the concept of Annual Effective Dose (AED). AED was calculated using the following formula

$$\mathrm{AED} = \mathrm{C} \times \mathrm{Q} \times \mathrm{F} \times \mathrm{t} \tag{3.14}$$

Here C (Bq m^{-3}) is the concentration of the isotope in study, Q (nSv/Bq h m^{-3}) is the dose conversion factor (9 nSv/Bq h m^{-3} for radon and 40 nSv/Bq h m^{-3} for thoron) and F is the equilibrium factor. In the first phase UNSCEAR [101] provided value of F

was taken (0.4 for radon and 0.02 for thoron). The total time of exposure, *t,* was calculated using the indoor occupancy factor of 0.8 (t = 7000 h/y).

In the second phase intensive study was done in Ambalavayal Panchayat in 19 houses. In this study concentration of both parent as well as progeny was estimated. Hence in this study the AED was estimated using the F value derived from the study itself.

3.6 SOURCE TERM STUDIES

The main factors affecting indoor radon and thoron concentration and their progeny are the entry or production rate from various sources, the ventilation rate and the rates of physical and chemical transformations. Out of these three the source of radon entry into the indoor environment namely the soil beneath the building and the construction material of the building play the major role in increasing the concentration. Hence a study of the source term is necessary to understand the reason for the indoor radon thoron and progeny concentration. Radon and thoron emission from the soil is controlled by the flux. It is defined as the emission per unit area of the porous matrix like soil, wall etc. The SI unit of radon/thoron flux is Bq m^{-2} s^{-1}. Along with the radon flux, radon exhalation per unit mass of the sample is also estimated. Radon mass exhalation rate is measured in Bq kg^{-1} s^{-1}. In the case of thoron, surface exhalation rate is estimated as thoron has a very small half-life and hence those available at depths in the sample container may not be available for study. All these measurements are done using Continuous Radon Monitors.

Continuous Radon Monitors are of several kinds. Nearly all of these detect alpha radiation. This is because it is difficult to build a portable detector for gamma and beta

radiation with both low background and high sensitivity. The three main types of electronic radon monitors for alpha particle detection are

1. Scintillation cells
2. Ion chambers
3. Solid state alpha detectors.

The soil exhalation studies were done using RAD 7 fig 3.10. Durridge RAD 7 uses solid state detector to convert alpha radiation directly to electrical signal.

Fig: 3.10 RAD 7 detector

The RAD7 possesses a periodic-fill cell. The cell is filled with air by means of a small pump that draws air into the cell once during each pre-selected time interval. In this defined cell, radon or the ^{218}Po may decay, and the decays are counted and the cycle repeated. RAD7 has a solid state detector which is silicon ion-implanted detector. The semiconductor material in the detector converts the alpha radiation from the decay of the radionuclide (e.g. ^{218}Po or ^{214}Po) into an electrical signal. The detector can electronically determine the energy associated with the incoming alpha particle. Thus the specific radionuclide can be identified. RAD7 possesses an internal sample cell of about 0.7 litres. It has a hemispherical shape, inside of which is coated with an electrical

conductor. A high voltage power supply charges the inside of the conductor to a potential of about 2000-2500 Volts relative to the detector. This creates an electrical field throughout the cell. The positively charged particles are propelled onto the detector in the periodic-fill cell by this electric field. ^{218}Po, which is the daughter product of ^{222}Rn is accelerated onto the detector and sticks to it. The electrical signal recorded from the decay of the radionuclide is then amplified, filtered and then sorted according to its strength. The detector can also determine the thoron (^{220}Rn) concentration from the ^{216}Po signal which has energy of 6.78 MeV. RAD7 device is almost completely insensitive to beta decay. Hence there is no interference from daughter products which decay by beta emission.

For the measurement of Radon and thoron exhalation from the soil samples, the samples were enclosed in a closed container. A pump in the RAD7 continually cycles air from the sampling environment (e.g., closed chamber) through the detector to distinguish between different energy levels so that only Radon is measured. A pump flow rate of 800 mL/min and a sampling frequency (cycle time) of one hour were used for all tests reported in this study. Measured counts of Radon during build up in the sampling chamber are then calculated into a corresponding concentration.

Radon emission potential from soil sample is governed by radon mass exhalation rate (J_m). The radon concentration C(t) at time *t* since closing the chamber builds up according to the formula

$$C(t) = C_0\, e^{-\lambda t} + \frac{J_m\, M}{V\, \lambda}\, [\, 1 - e^{-\lambda t}] \qquad (3.15)$$

Here C_0 is the ^{222}Rn concentration (Bq m^{-3}) present in the chamber volume at time t = 0. M is the total mass of the dry sample (kg), V is the effective volume (residual air volume of exhalation chamber r+ Porous Volume of Sample + internal

volume of RAD7 (m^3), λ is the effective decay constant for ^{222}Rn, which is sum of the leak rate (if existing) and the radioactive decay constant of ^{222}Rn (h^{-1}). *t* is the measurement time (h).

By linear approximation Eq 3.15 can be converted to

$$C(t) = \frac{J_m M}{V} t + C \qquad (3.16)$$

Upon least square fitting of the data to the above equation we get J_m from the fitted slope value with the information of the mass M of the sample and residual air volume, V of the set up.

Thoron measurement is also done by keeping the sample in a closed chamber. The thoron surface exhalation rate is calculated using the following formula [102]

$$J_s = \frac{C_T.V.\lambda}{A} \qquad (3.17)$$

Here C_T is the equilibrium thoron concentration which is attained in a short time period. V is the residual air volume of the set up in m^3, λ is the thoron decay constant (0.0126 s^{-1}) and A is the surface area (m^2) of the sample.

3.6.1 Portable Radon Monitor – SRM for indoor active measurement of Indoor Radon and thoron

The active measurements were done using the state of the art equipment, the Portable Radon Monitor (SMART RnDuo), developed by BARC (Bhaba Atomic Research Center, Mumbai) as shown in fig.3.11. The detector works on the principle of scintillation counter by scintillation with ZnS(Ag). The detection limit of Portable Radon Monitor (SMART RnDuo) is in between 8 Bq/m^3 and 50 MBq/m^3 (Operational Manual of Portable Radon Monitor—SMART RnDuo, March 2015). The SMART RnDuo is a

portable continuous activity monitor. It samples radon (^{222}Rn), thoron (^{220}Rn) and gross alpha in the sampled air. The device has a cylindrical geometry with a volume of 153$cm^{3.}$ The scintillations generated in ZnS(Ag) are converted to electrical pulses. The counts obtained from electrical pulses for each interval are converted to activity concentration of the respective species using an algorithm installed in the micro-controller. This advanced algorithm was developed by Bhabha Atomic Research Centre, Mumbai. One of the key features of the algorithm is that it accounts for the alpha counts obtained from the decay products generated during the previous intervals. The device doesn't have any interference from charge neutralizing species such as humidity, methane, carbon dioxide etc. Air is sampled into the scintillation cell by diffusion through a glass fibre filter paper and a pin-hole arrangement in the radon diffusion model. The filter paper blocks the entry of any decay products which is already in the air into the detector volume. The pin-hole arrangement restricts the entry of short-lived (half-life 55.6 sec) thoron into the scintillation cell by creating a diffusion- time delay. The scintillations due to the alpha particles emitted by radon and its decay products formed inside the cell are constantly counted for a user-programmable counting interval by PMT and the associated counting electronics. These counts are converted to the radon concentration in Bqm^{-3} by the algorithm.

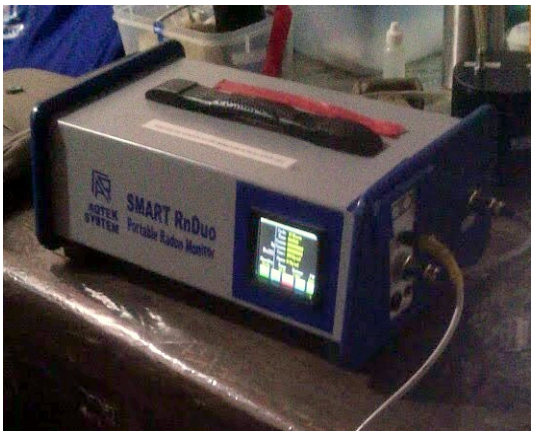

Fig:3.11 Smart RnDuo

Program based sampling is carried out for thoron monitoring, using a flow mode sampler connected to the pump inlet of the monitor. Each measurement cycle consists of 15 minutes. Within this the pump is kept ON for the first 5 minutes. This measures the thoron and the background. A delay of 5 minutes is maintained after this to ensure the complete decay of thoron. The last 5 minutes gives a measure of the background count for the cycle. The background count is subtracted from the total count of the initial 5 minutes to get the thoron concentration. Schematic diagram of Smart RnDuo is as given in figure 3.12.

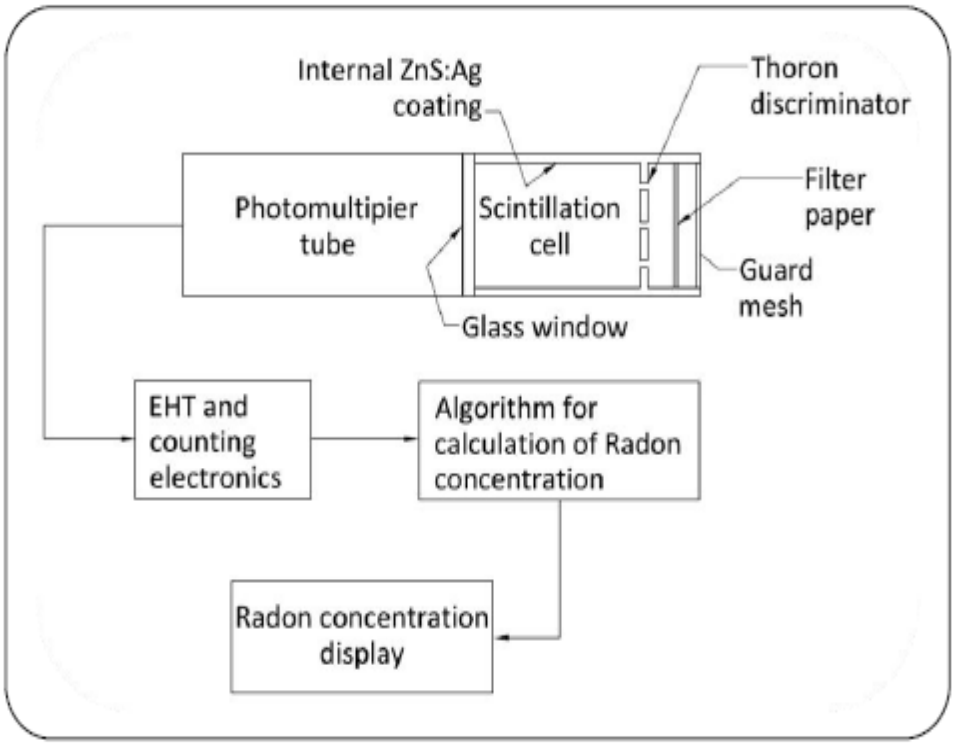

Fig: 3.12 Schematic diagram of Smart RnDuo

The soil gas radon activity measurements were done to study the contribution of the radon gas in the sub soil to the indoor radon concentration. The soil gas measurement were done at a depth of 0.8m as it is already known from the literature that the activity concentration of radon in the soil increases with increasing depth up to around 1 m from

the surface and then it reaches saturation [102,103]. Soil gas radon activity measurements were done in ten houses (selected based on indoor radon and thoron concentration, availability of power point and safety of the equipment) where the active measurements were conducted using Portable Radon Monitor Smart RnDuo. A steel probe was inserted into the soil for measurement. The measurement cycle was set at 15 minutes. Soil gas was sampled for 2 minutes using a pump before starting the measurement Fig. 3.13. The measurement is continued for 1 hour to have a stabilised reading of radon concentration.

Fig: 3.13 Picture of soil gas radon measurement

RESHMA BHASKARAN “ ENVIRONMENTAL RADIOACTIVITY STUDIES IN WAYANAD, KERALA, INDIA”. THESIS. DEPARTMENT OF PHYSICS, UNIVERSITY OF CALICUT, 2018.

CHAPTER 4

GAMMARAY SPECTROMETRY

Terrestrial radioactivity and inhalation dose due to radon and thoron form the major part of exposure due to NORM. The main terrestrial radionuclides to be accounted for are ^{238}U series, ^{232}Th series and ^{40}K. Average natural radiation exposure to the Indian population has been estimated to be 2.3 mSv y^{-1} [105]. The same has been estimated to be 2.4 mSv y^{-1} for the world population [18]. Since Wayanad is a hitherto unstudied part of India, this work involved the study of terrestrial radioactivity, indoor radon, thoron and progeny measurements in this district. Study of terrestrial radioactivity is essential to understand the behaviour of radioactivity in an ecosystem, the major pathways of exposure of radiation to man being ingestion, inhalation and external exposure. Radioisotopes present in the soil enter the food chain through plants. Hence a study of terrestrial radioactivity is essential to estimate the dose received by the inhabitants of a particular place due to the flora and fauna there. Present work involved three stages. First was the general radiation survey to estimate the dose in air due to naturally occurring radioactive materials. In the second stage soil samples were collected from 58 random locations. Concentration of terrestrial radioisotopes, namely, ^{40}K, ^{226}Ra and ^{232}Th were estimated using gamma ray spectrometry. In the third stage indoor radon, thoron and their progeny concentration were studied. Radon and thoron exhalation studies were also done.

4.1 CONCENTRATION OF TERRESTRIAL RADIONUCLIDES IN SOIL

The equipment used for Gammaray spectrometry was NaI(Tl) scintillation detector available at Indira Gandhi Center for Atomic Research (IGCAR), Kalpakkam, Chennai. Samples were collected from 58 locations in different parts of Wayanad district. Peaks corresponding to ^{40}K, ^{226}Ra and ^{232}Th were studied in the soil and rock samples. Sample spectrum of soil samples obtained using the detector is as shown in Fig. 4.1 and 4.2.

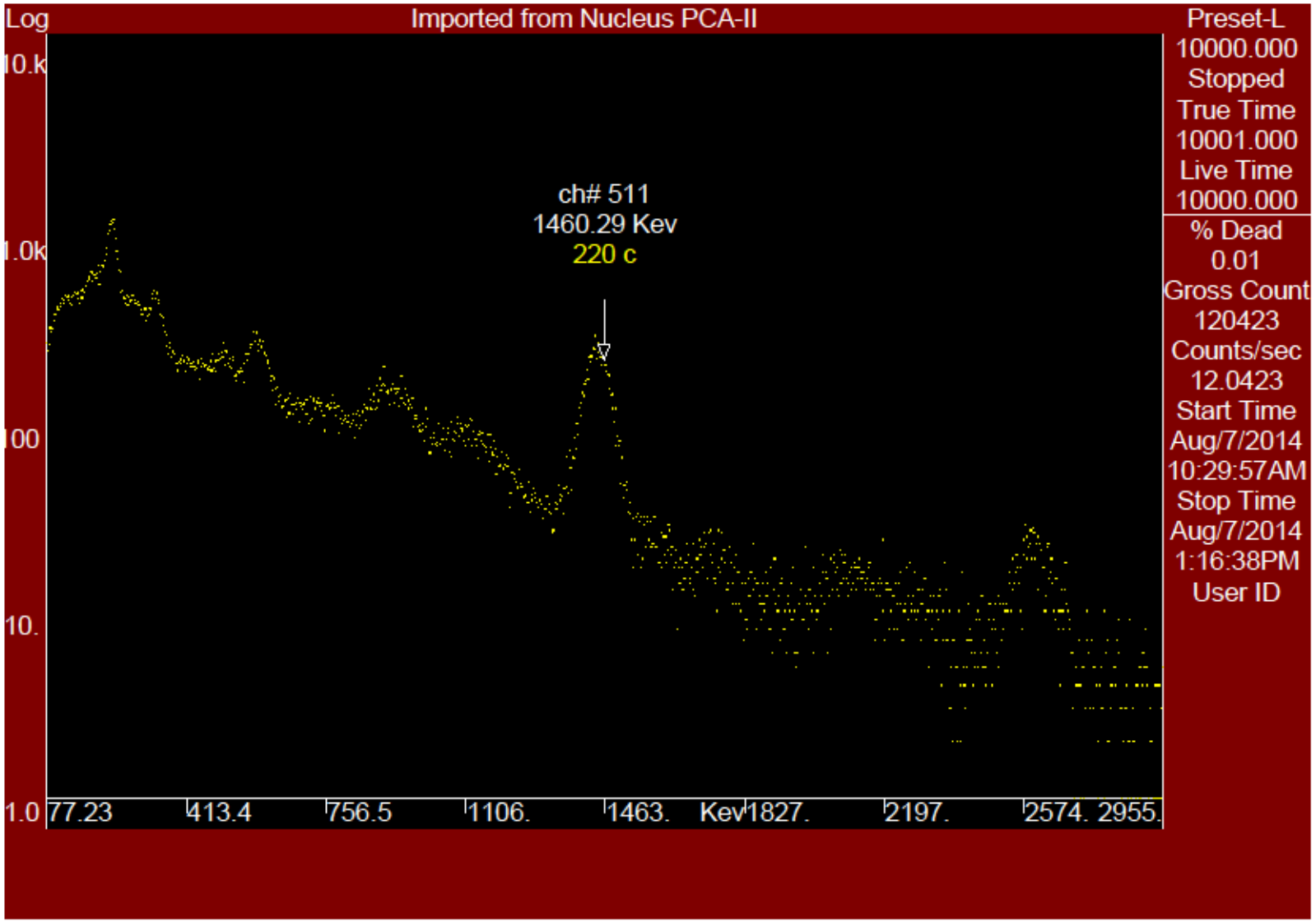

Fig 4.1Spectrum of Soil sample (1)

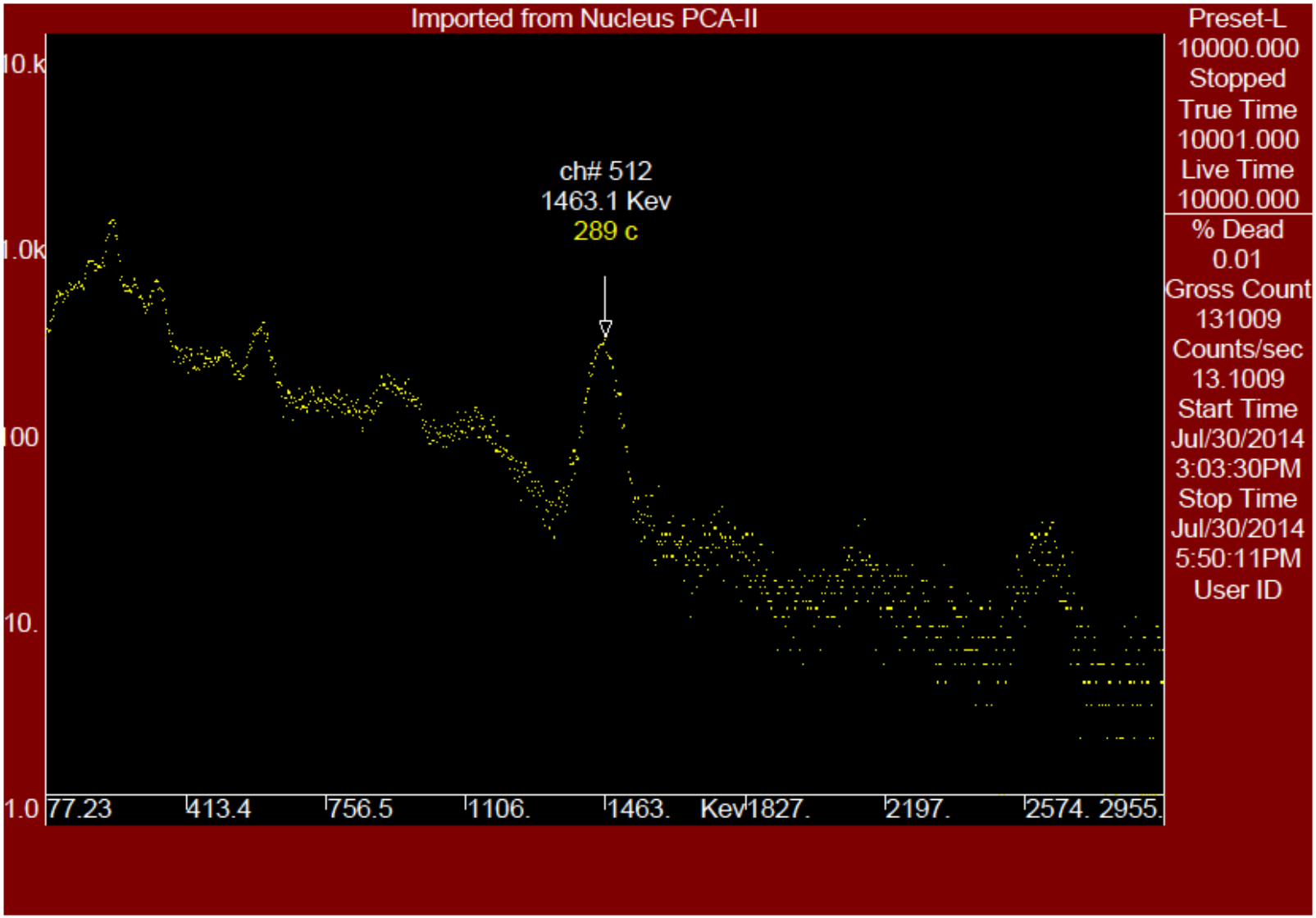

Fig 4.1Spectrum of Soil sample (2)

4.1.1 Potassium

Out of 58 soil samples, 27 had ^{40}K activity concentration Below Detectable Limit (BDL). Rest of the samples showed activities in the range 37.6- 1180 Bq/kg. The average ^{40}K activity concentration was 265.1 Bq/kg. Maximum values were found for soil samples taken from Nandankavala, Ambalavayal Panchayat. Figure 4.3 shows the activity concentration for all the soil samples. As per UNSCEAR [18], world average of ^{40}K concentration in terrestrial radioactivity is 420 Bq/kg. The activity concentration of ^{40}K was found to be less than the world and the Indian averages [3]. There were only 6 samples that showed activity concentration higher than both the world and the Indian average.

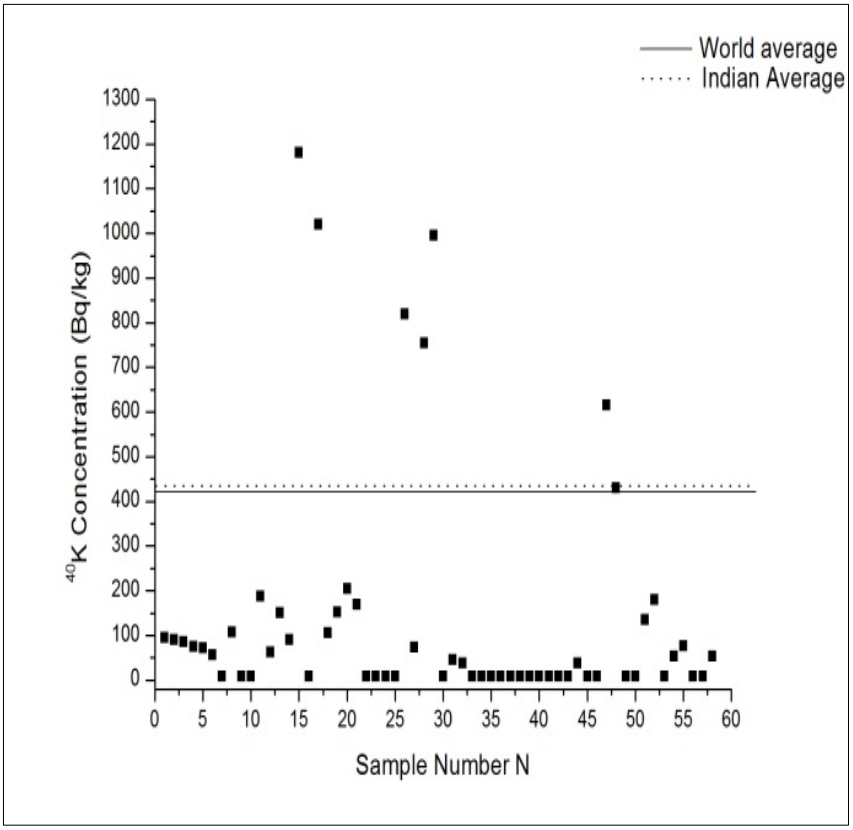

Fig 4.3 Concentration of ^{40}K in the soil of Wayanad, Kerala. (World and Indian averages were taken from UNSCEAR 2000)

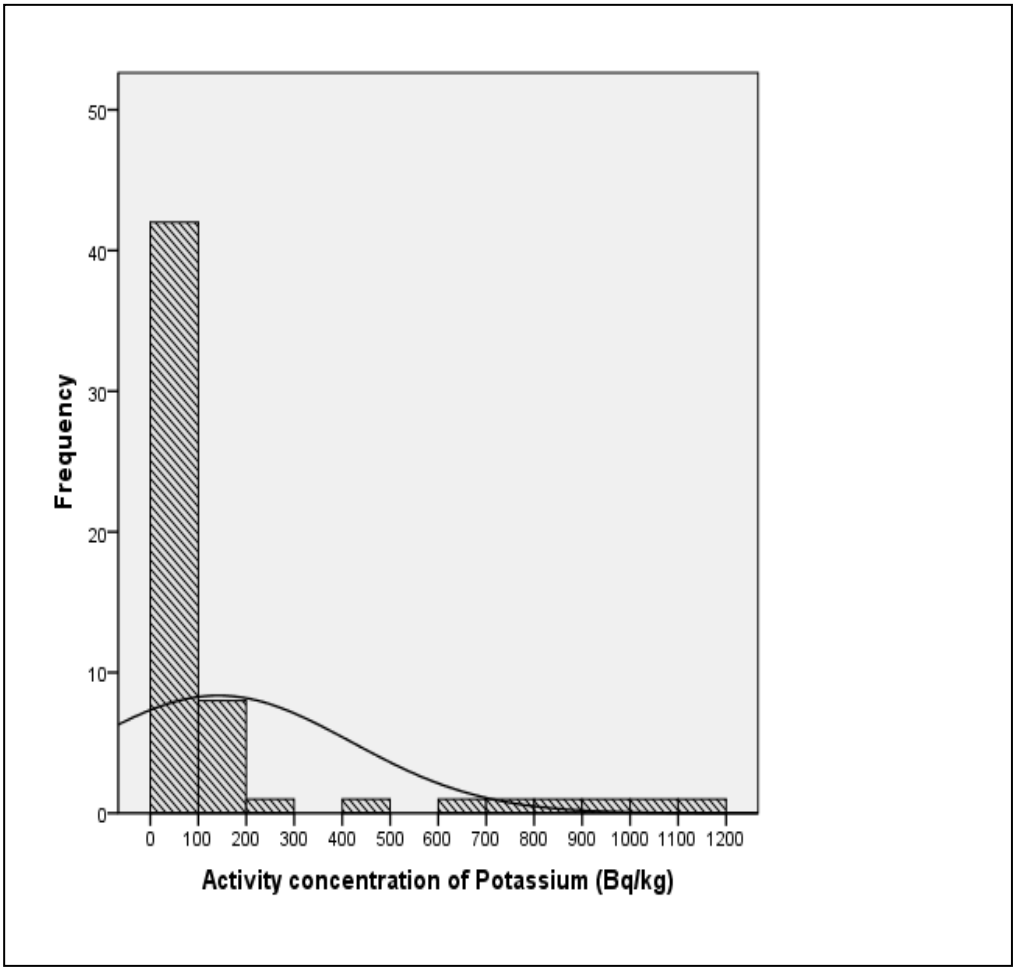

Fig. 4.4 Histogram for activity concentration of ^{40}K

4.1.2 Radium

Out of the 58 samples, activity concentration of ^{226}Ra for 16 soil samples was BDL. Activity of rest of the samples was in the range of 9.9 - 95.6 Bq/kg with an average of 20.8 Bq/kg. Soil sample from Nandankavala in Ambalavayal Panchayat showed the maximum activity of ^{226}Ra. Fig.4.5 shows ^{226}Ra concentration against sample numbers along with world and Indian averages.

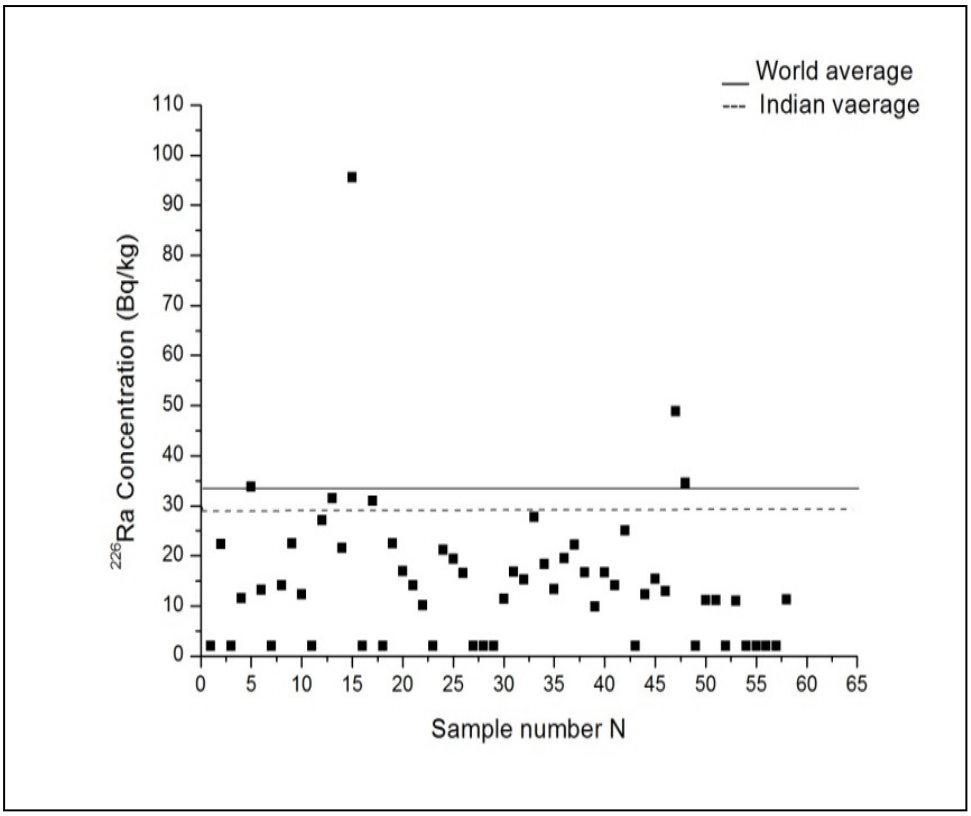

Fig 4.5 Concentration of ^{226}Ra in the soil of Wayanad, Kerala.(World and Indian averages were taken from UNSCEAR 2000)

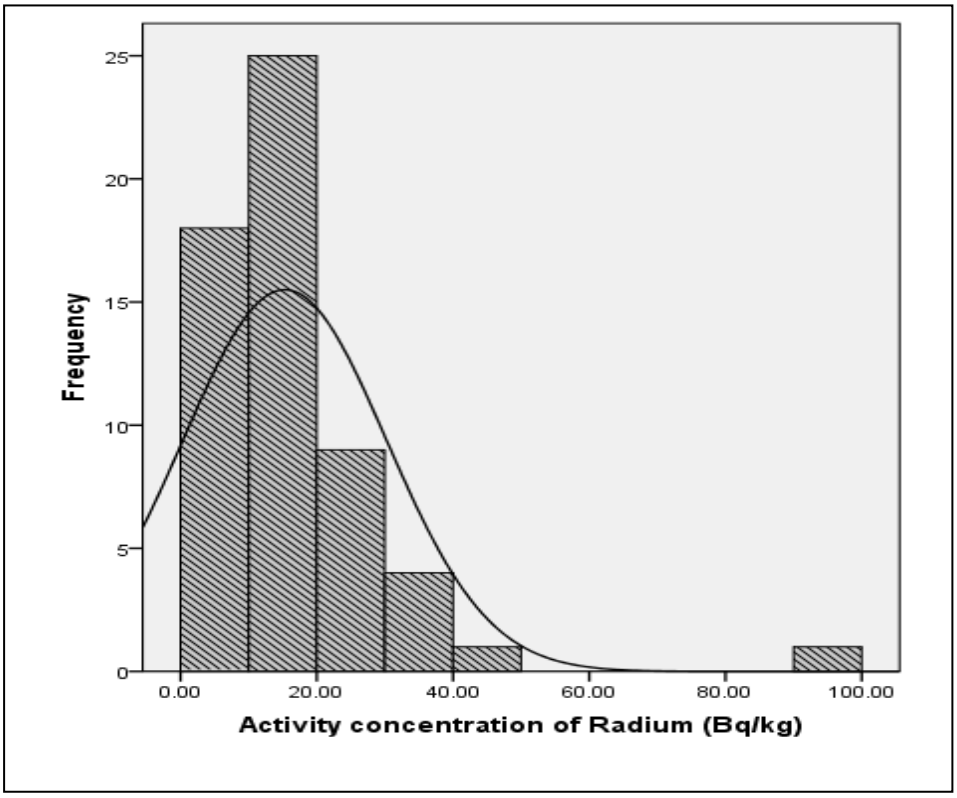

Fig 4.6 Histogram for activity concentration of ^{226}Ra.

4.1.3 Thorium

For ^{232}Th, 10 soil samples showed BDL activity concentration. For the remaining samples activity concentration ranged from 7.21- 209.03 Bq/kg with an average of 39.09 Bq/kg. The maximum value was found for Ponkuzhi and the second highest value of 155.24 Bq/kg was found in Nandankavala. The mean value of ^{232}Th is comparable to the world average of 45 Bq/kg [1] and the Indian average of 30.9 Bq/kg [3] (Fig.4.7). The soil radioactivity concentration for ^{40}K, ^{226}Ra and ^{232}Th are given in Appendix B

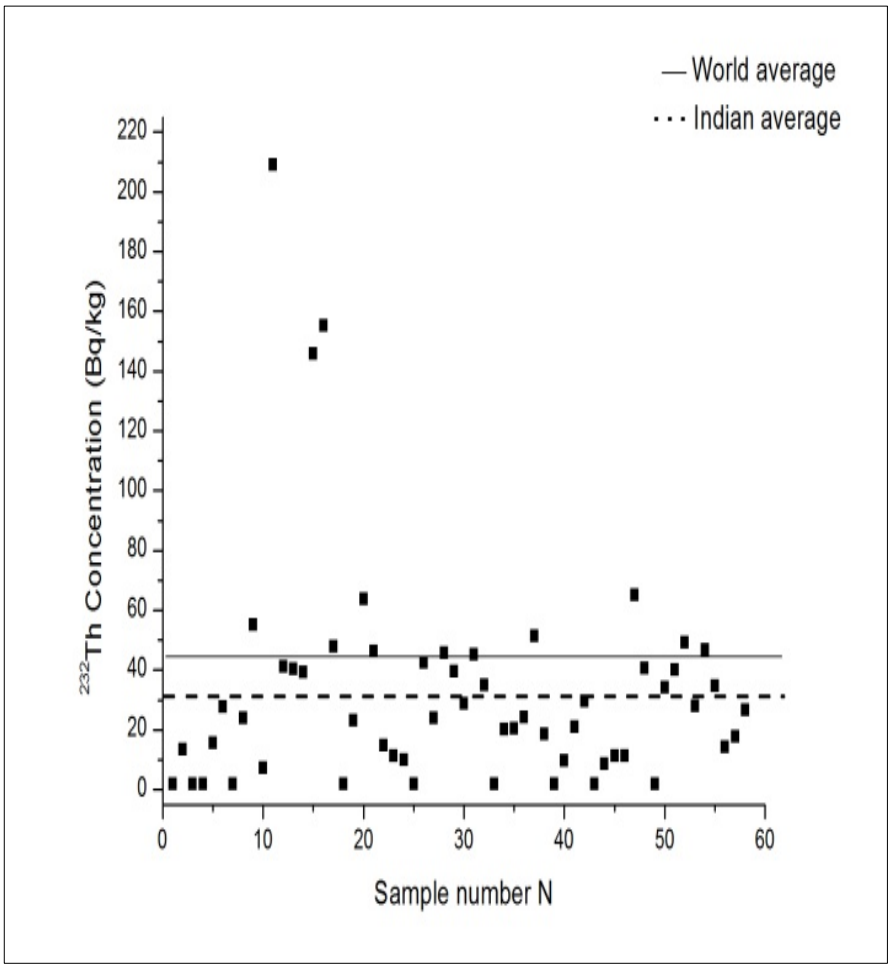

Fig 4.7 Concentration of ^{232}Th is the soil of Wayanad, Kerala. (World and Indian averages were taken from UNSCEAR 2000)

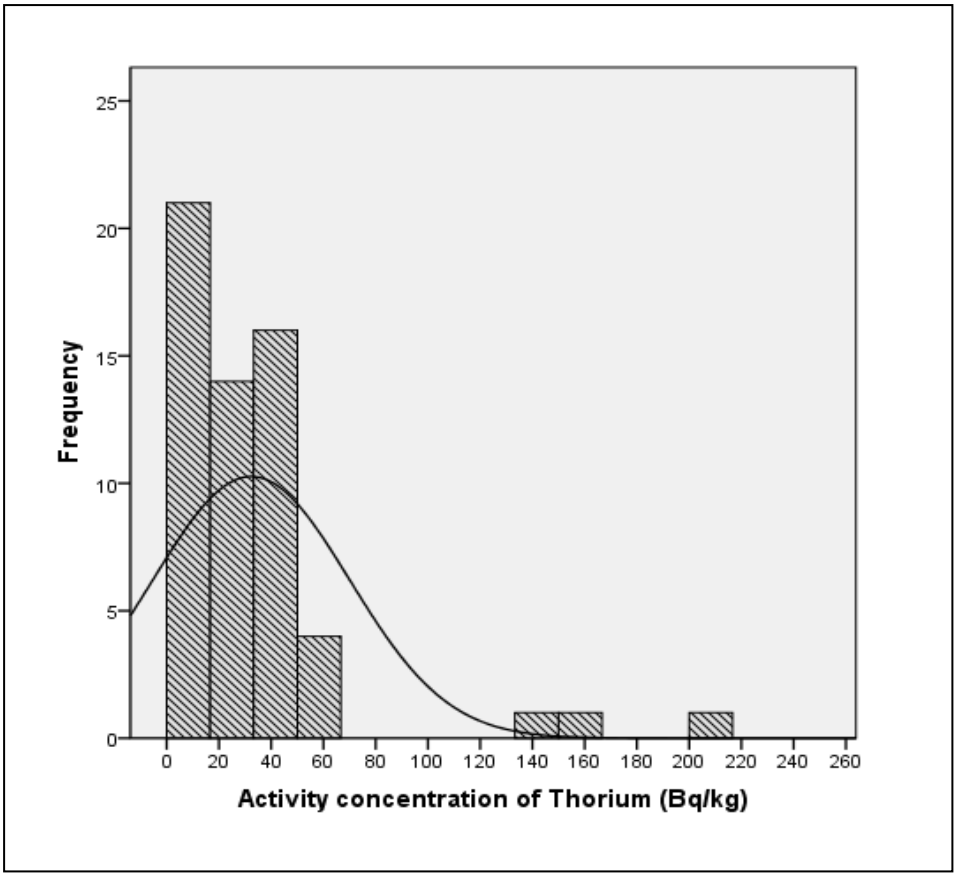

Fig 4.8 Histogram for activity concentration of ^{232}Th.

From the above results it is evident that more number of samples show BDL values for Potassium and Uranium as compared to Thorium. The concentration of natural potassium is highly non uniform in the soil of Wayanad district. According to the report presented by Kerala State Planning Board[106], 15% of the soil samples collected by them show very low potassium levels. In the report, researchers explain this as due to the intensive leaching by irrigation and very strong acidic condition of the soil. Radioactive potassium ^{40}K is only 0.0118% of naturally occurring potassium. This may be the reason that some samples show the activity concentration of ^{40}K as BDL.

On comparing ^{226}Ra. and ^{232}Th it is seen that more number of soil sample show BDL for ^{226}Ra as compared to ^{232}Th. It is well known that ^{226}Ra is more soluble in water as compared to Thorium. Since Wayanad experiences very heavy rainfall, radium

concentration may be washed off from the soil at some places. However, thorium is less soluble as compared to radium and hence its retention will be more in the soil samples. From Fig 4.4, Fig 4.6 and Fig 4.8 it is evident that the radionuclides show lognormal distribution in the region of study. The concentration of ^{40}K, ^{238}U and ^{232}Th are given in Appendix C.

4.2 CONCENTRATION OF TERRESTRIAL RADIONUCLIDES IN ROCK

Study of rock samples is necessary while analysing the background radioactivity of a region. This not only gives us an idea of the external hazard due to the terrestrial component of NORM but also is useful to assess the dose due to indoor exposures since these rocks are used for the construction of buildings. Gammaray spectrometry, as done for soil samples, was done for the 13 rock samples also. Result of the activity concentration of the rock samples is as given in Appendix D.

From the results it is seen that the concentration of the studied isotopes are higher in the rock samples as compared to the soil ones, with a similar trend (activity concentration of $^{40}K > ^{232}Th > ^{226}Ra$). Heavy rain fall along with the presence of enormous water bodies in Wayanad could result in leaching of radioisotopes from soil. In high range, the isotopes may be washed out from soil more than that from rocks. Similar results were reported from the neighbouring state of Karnataka where the rocks showed higher activity concentration of the isotopes as compared to the soil [107]. Statistics of the soil and rock samples are presented in Table 4.1.

Table 4.1

Statistics of soil and rock samples from Wayanad, Kerala.

	SOIL			ROCK		
	^{40}K	^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th
Mean (Bq/Kg)	265	21	39	669	65	100
Median (Bq/Kg)	106	17	29	853	38	65
Std. Deviation	334	15	38	458	54	79
Minimum (Bq/Kg)	38	10	7	107	6	41
Maximum (Bq/Kg)	1180	96	209	1195	146	253
Skewness	1.70	3.72	3.01	-0.12	0.54	1.98
Kurtosis	1.59	17.62	10.22	-2.10	-1.66	4.01

The ratio of ^{232}Th to ^{226}Ra ranged from 0.46 to 3.77 and the average was 1.67. Out of the 36 soil samples which were considered to calculate the ratio, only 8 showed higher ^{226}Ra activity concentration. Rest of the samples had higher ^{232}Th activity concentration. Table 4.2 compares the observed activity concentrations for ^{40}K, ^{226}Ra and ^{232}Th in soil samples with many studies conducted in other parts of the world. Many studies conducted in various countries [108, 109] showed higher Thorium concentration compared to Uranium.

Table 4.2
Comaprison of the concentration of terrestrial radionuclides in Wayanad with other places in India, available in literature.

Location	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	Reference
World Average	33	45	420	[18]
All India	28.60	30.96	432.70	[110]
Gudalore, TN	37.70	75.30	195.20	[48]
Ullal, Karnataka	546	2971	268	[111]
Mysore, Karnataka	14	19.85	183	[112]
Kalpakkam	16	119	406	[113]
Kavery River Sediment	5.31	34.04	401.11	[114]
Nilgiri, Western Ghats	26.26	53.61	231.93	[115]
Chavara-Neendakara Belt	2355	14000	1116	[24]
Kurdistan, Iraq	83.33	19.14	284.86	[116]
Ptolemias, Greek	42	36	496	[117]
Guangdong, China	140	187	680	[108]
Wayanad, Kerala	**21**	**39**	**265**	**Present Study**

Rock samples also showed higher ^{232}Th concentration. Concentration of ^{226}Ra and ^{232}Th in Ambalavayal granites was higher as compared to the Kalpetta granites. Table 4.3 gives the comparison of the activity concentration of rock samples with a few studies across the world. Studies of the rock samples show that the results are similar to that in other parts of the world.

Table 4.3

Comparison of the concentration of rock samples in Wayanad, Kerala in Bq/Kg with available literature

Location	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	References
India	43.7	39.5	530.3	[110]
Bangalore Rural	93	306	1074	[118]
Tamilnadu	10-649	BDL-542	533-1484	([119])
Kerala	45.1	47.6	558	[110]
Egypt	15.6	14.5	406	[120]
Greece	83.5	76.8	989	[40]
Yemen	57.09	80.26	846.21	[121]
Wayanad	65	100	669	Present study

4.3 DOSIMETRIC PARAMETERS

4.3.1 Dose rate

In the present study dose rate in air due to soil, calculated from the activity concentration of soil, ranged from 3.58 - 181.50 nGy/h. Average dose rate for the residents of Wayanad was found to be 34.01nGy/h. This is less than the world average of

59 nGy/h. The values reported from the high background region of Kerala, which has an average of 958.7 nGy/h [28], are higher than the average dose rate due to terrestrial radioactivity as found in the present study.

The dose rate in air measured using the survey meters was found to be higher than that measured from the soil samples. Positive correlation was found between them and is shown in Fig.4.9. The increase in the measured dose rate may be due to the additional contribution from the cosmic rays.

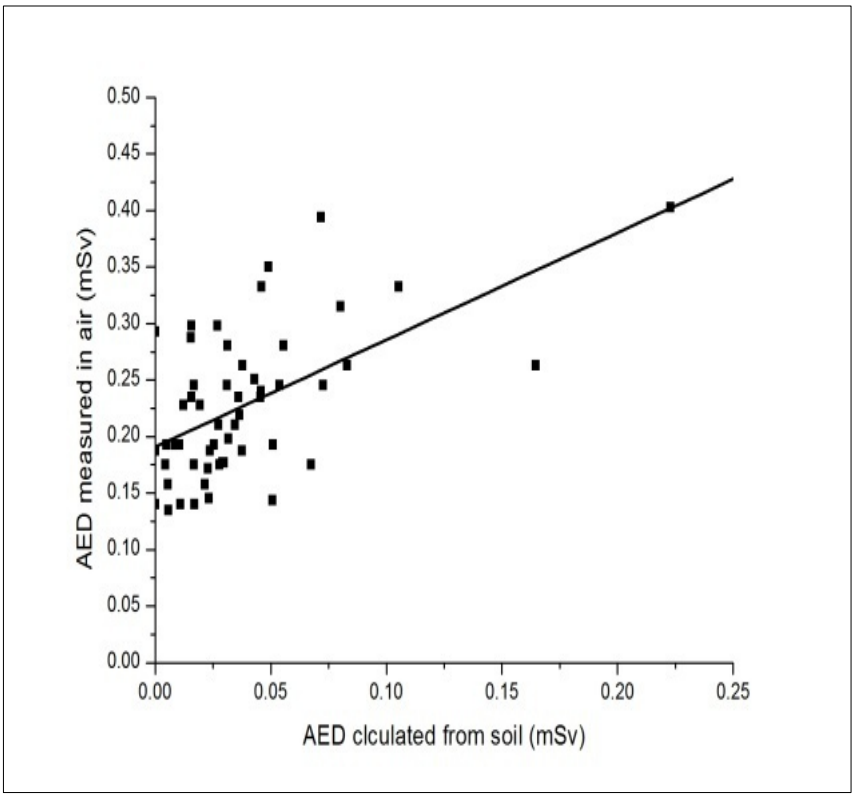

Fig.4.9 Correlation between AED from soil and survey meter

A map of Wayanad district depicting the distribution of external gamma ray dose rate is given below (Fig. 4.10). The map has been plotted using software ArcGIS version 10.3.

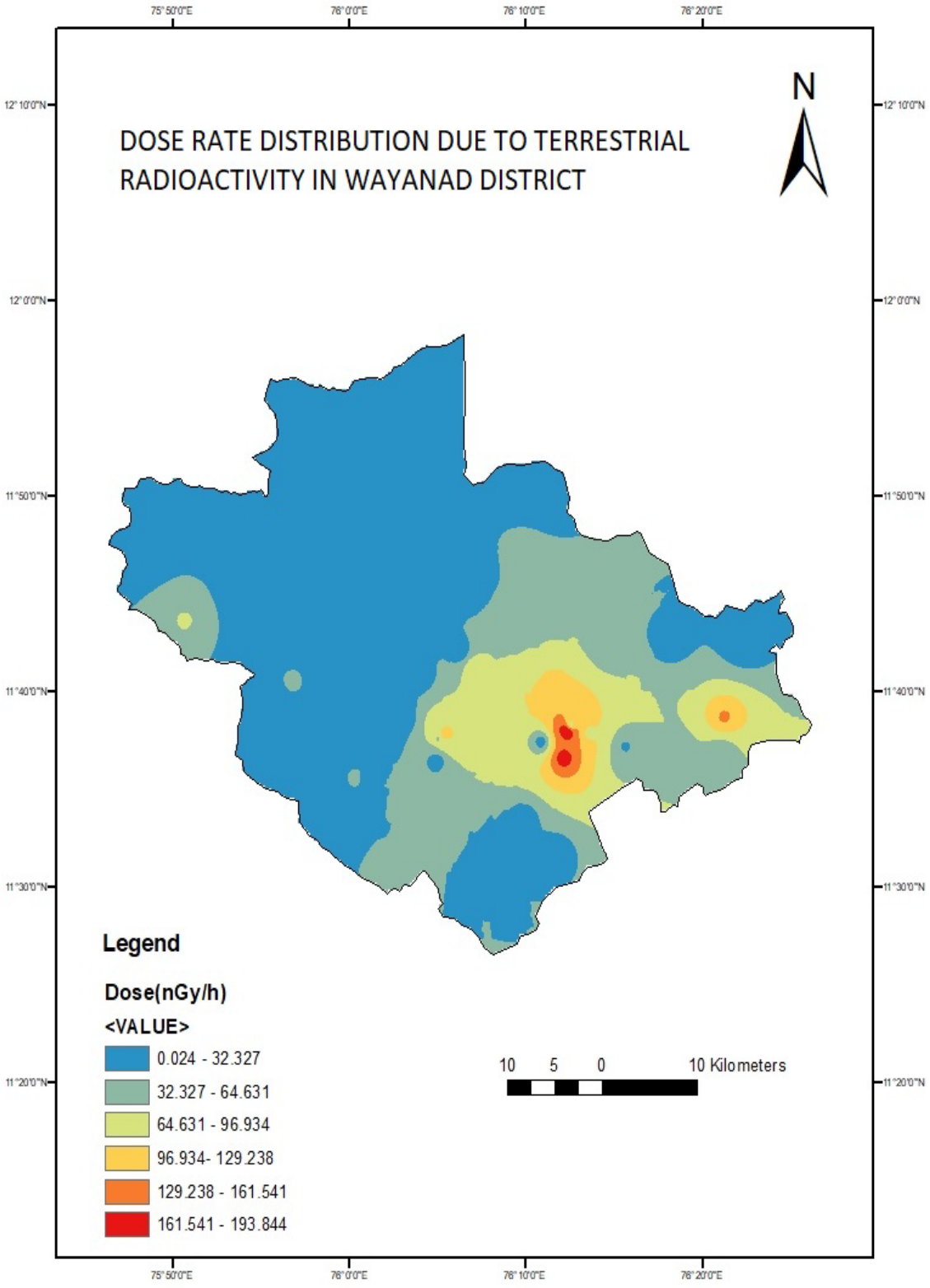

Fig. 4.10 Map of Wayanad district depicting the distribution of Gammaray dose rate.

4.3.2 Annual Effective Dose

The outdoor AED due to soil ranged from 0.01 - 0.22 mSv with an average of 0.04 mSv. This value is comparable to world average outdoor AED of 0.07 mSv. The indoor AED was found to be in between 0.02 mSv and 0.89 mSv with an average of 0.17 mSv. The total average AED (AED indoor + AED outdoor) is 0.21 mSv. The world average annual effective dose is 0.48mSv and Indian average is 0.71 mSv (including the cosmic ray component of 0.287 mGy/yr) [110]. The dosimetric data of the soil and rock samples are given in Appendix E.

4.3.3 Hazard Indices

Hazard indices were calculated for the collected soil and rock samples to estimate the hazards due to the building materials. Majority of the building materials are made out of soil and rocks available in the district. The results are as shown in Table 4.4.

Table 4.4.

Dose rate, AED and Hazard Indices for soil and Rock samples at Wayanad, Kerala

		Minimum	Maximum	Mean	Std. Deviation
SOIL	Dose Rate (nGy/h)	3.58	181.55	34.01	32.64
	AED outdoor (mSv)	0.01	.22	0.04	0.04
	AED indoor (mSv)	0.02	0.89	0.17	0.16
	Hex	0.02	1.07	0.20	0.19
	Hin	0.02	1.33	0.24	0.22
ROCK	Dose Rate (nGy/h)	4.46	254.8	72.09	79.67
	AED outdoor (mSv)	0.01	0.31	0.09	0.09
	AED indoor (mSv)	0.02	1.25	0.35	0.39
	Hex	0.02	1.53	0.41	0.47
	Hin	0.02	1.86	0.51	0.61

There was only one sample (Nandankavala in Ambalavayal Panchayat) that showed hazard indices greater than the limits prescribed for the soil samples. In the case of rock samples, the Ambalavayal granites showed hazard index greater than one. However, all the samples in the Kalpetta granites had hazard indices less than the limit prescribed by ICRP (Hex and Hin < 1).

4.4 DISCUSSION

Activity concentration of Potassium, Uranium and Thorium is similar to the values found for the neighbouring district of Udagamandalam and Gudalore in Tamilnadu [46, 48]. Similar to the case of Wayanad district, both Udagamandalam and Gudalore form a part of the Western Ghats. In both the districts activity concentration of ^{232}Th is found to be more as compared to ^{226}Ra, similar to the present study (Table 4.2). Reports of the work done in Mysore district [112], which is a part of Karnataka and a neighbouring district of Wayanad, also has a similar trend.

It is seen that in igneous rocks, ^{232}Th is higher as compared to ^{226}Ra [1]. Results in this study also show the same trend. Higher concentration of ^{40}K in the rocks as compared to ^{226}Ra and ^{232}Th may be due to the presence of K-Feldspar in these rocks. In earlier studies also the pink granites have been reported to have higher radioactivity content as compared to the other types of granites [122, 123]. In Nandankavala region, the granites are mainly pink granites. The higher thorium content seen in these granites may be due to the monazite minerals in these rocks [124].

From the Gammaray spectrometry analysis, it was found that the dose rate due to soil and rock (Fig. 4.11) shows significant correlation (R=0.87 with P < 0.05). Activity concentration for ^{40}K and ^{232}Th also show good correlation for soil and rock with an R value of 0.86 and 0.90 respectively (Fig 4.12 and Fig 4.13). Activity concentration of ^{226}Ra for soil and rock has a week positive correlation with R = 0.67(Fig. 4.14). The

correlation found between the ^{226}Ra and ^{232}Th content of the rocks was good with R= 0.90 (Fig. 4.15). No such correlation was found between the ^{226}Ra and ^{232}Th content of the soil (R= 0.27). Correlation of ^{226}Ra in soil and rock was found to be 0.84 with a P value less than 0.05. It is known that ^{226}Ra is more soluble in water as compared to ^{232}Th. Hence continuous study of the Thorium to Radium ratio may throw some light on the weathering of rocks in this region.

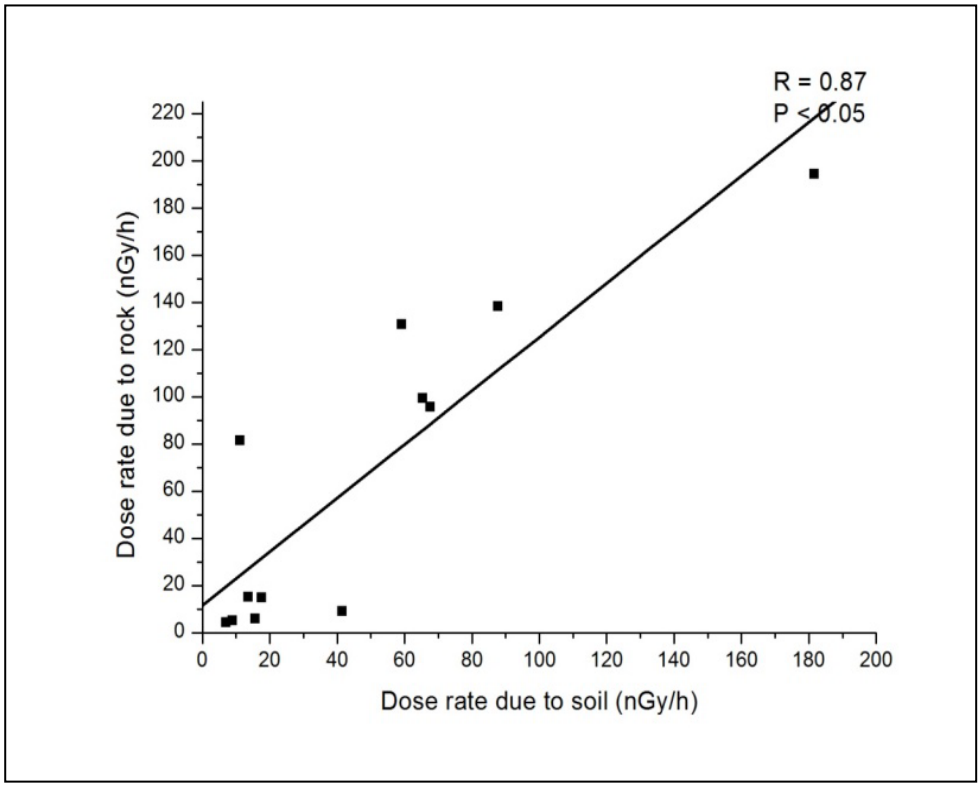

Fig 4.11 Dose rate due to soil and rock samples from Wayanad, Kerala.

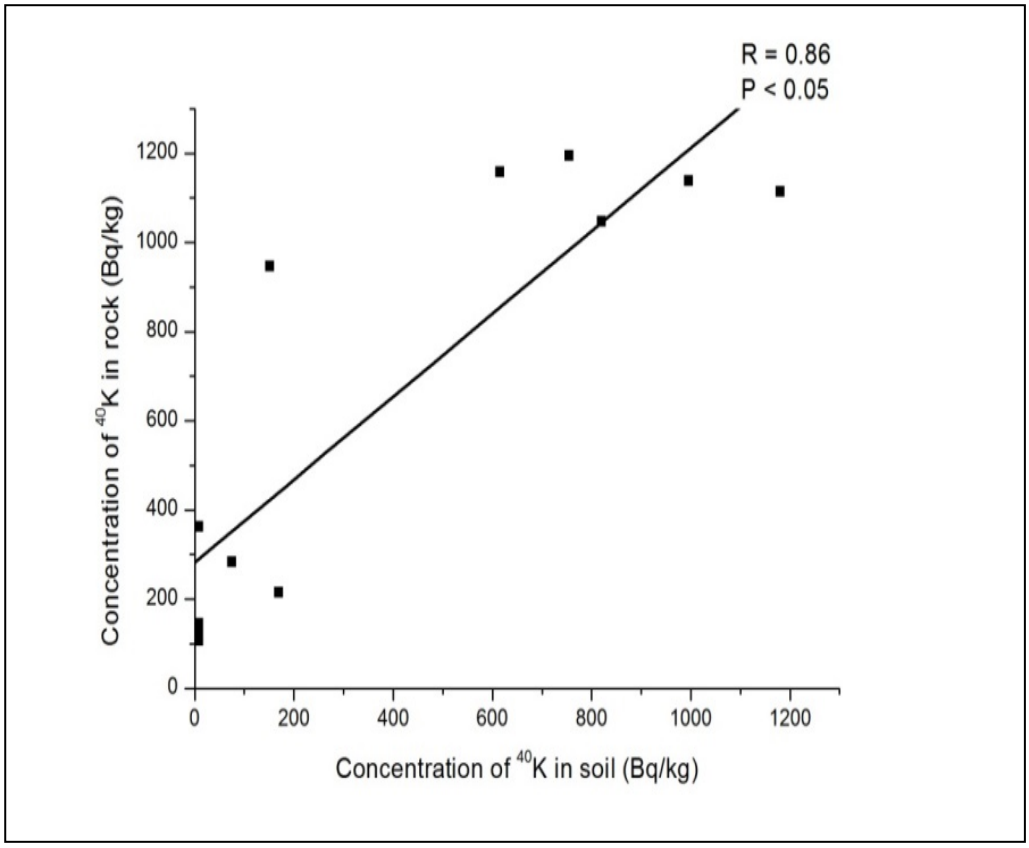

Fig 4.12 Correlation of ^{40}K concentration in rock and soil

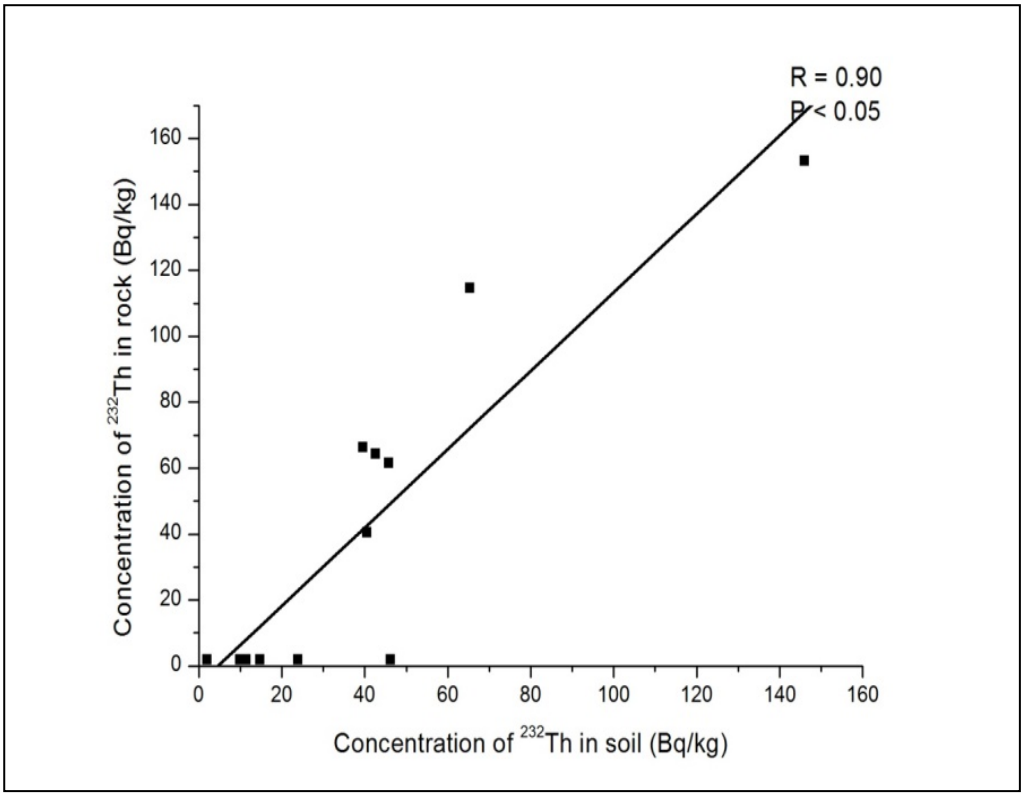

Fig 4.13 Correlation of ^{232}Th concentration between rock and soil

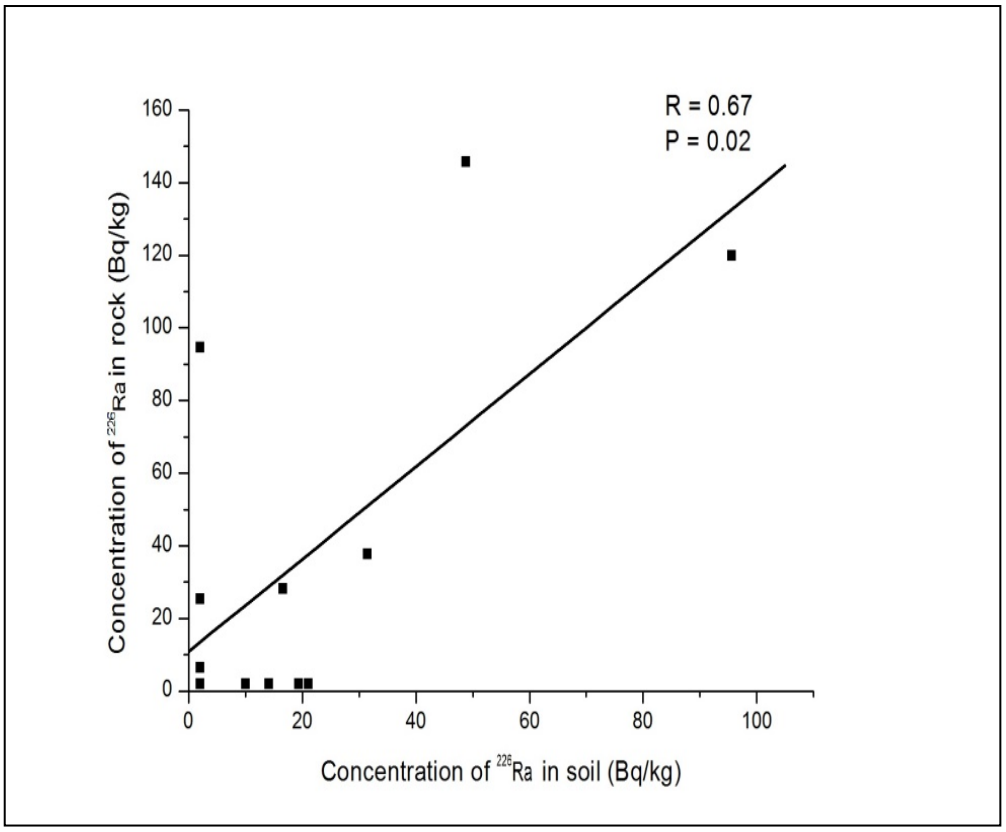

Fig 4.14 Correlation of ^{226}Ra in soil and rock

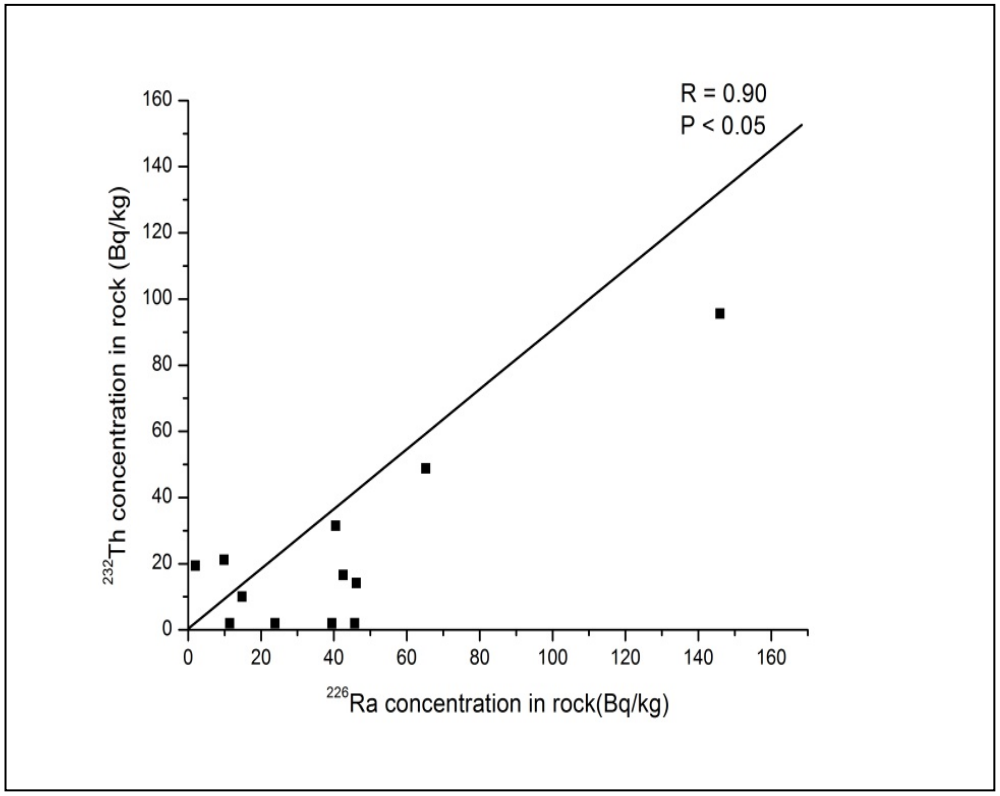

Fig 4. 15 Correlation of ^{226}Ra and ^{232}Th in rock

RESHMA BHASKARAN “ ENVIRONMENTAL RADIOACTIVITY STUDIES IN WAYANAD, KERALA, INDIA”. THESIS. DEPARTMENT OF PHYSICS, UNIVERSITY OF CALICUT, 2018.

Chapter 5
RADON THORON MEASUREMENTS

Indoor radiation exposure to man is mainly due to the exposure to radon, thoron and their progenies. These gases are the normal constituents of soil gas and enter the indoor atmosphere either from the cracks in floor or from the building material used. Human technological and industrial activities may influence the pathways. Besides this natural factors like temperature, humidity, seasons, rainfall etc may affect the indoor concentration of radon and thoron. The importance of radon and thoron progeny lies in the fact that the major contribution to the inhalation dose is not from the parent radon and thoron but from the progeny.

From Gammaray spectrometry of Wayanad district, it was seen that the soil and rock samples of Bathery taluk, especially Ambalavayal Panchayat, showed relatively higher concentration of terrestrial radioactivity in both soil and rock samples. Since radon and thoron are daughter products of ^{226}Ra and ^{232}Th, their concentration was studied in this region. The study was divided into two phases. In the first phase radon and thoron concentration was measured in Bathery for one complete year in four quarters. Four quarters were studied to understand the dependence of radon and thoron concentration on the changes in climatic conditions. To assess the indoor radiation exposure, 25 houses were studied using pin hole dosimeters with LR 115 films. Soil samples were also collected from the vicinity of these houses to study the dependence of terrestrial radioactivity on indoor radon thoron concentration. Soil exhalation studies were also done in these soil samples to analyse the correlation between the concentration of terrestrial radionuclides and the radon thoron concentration.

After this, second phase included intensive study of Ambalavayal region. 19 houses were selected from this region, many of which were in the vicinity of the granite quarries. Indoor radon, thoron and their progeny were studied using the pinhole dosimeters and the

DPS. Active measurements were done to assess the diurnal variation of radon as also the variation of radon with ventilation conditions. Floor and wall exhalation studies were done in the houses showing highest radon and thoron concentration to arrive at an idea about the possible source of indoor radon and thoron. Concentration of soil gas was also measured.

5.1 PHASE I

Result of the indoor radon and thoron measurements in the 25 houses of Bathery taluk are as given in Appendix F and Appendix G. Average annual concentration of radon in Bathery ranged from 13 to 108 Bq m^{-3} with a geometric mean of 27 Bq m^{-3}. Average annual thoron concentration varied from 12 to 621 Bq m^{-3} with a geometric mean of 63 Bq m^{-3}. From Appendix F we can see that the concentration of Radon is highest during the months from November to January. These months are the coldest in Wayanad. The minimum was found during the months from February to April. In the case of thoron, the maximum was found during the months of February to April and the minimum during the months of August to October. No significant correlation (Fig.5.1) was found between indoor Radon and Thoron gas concentrations (R = 0.28)

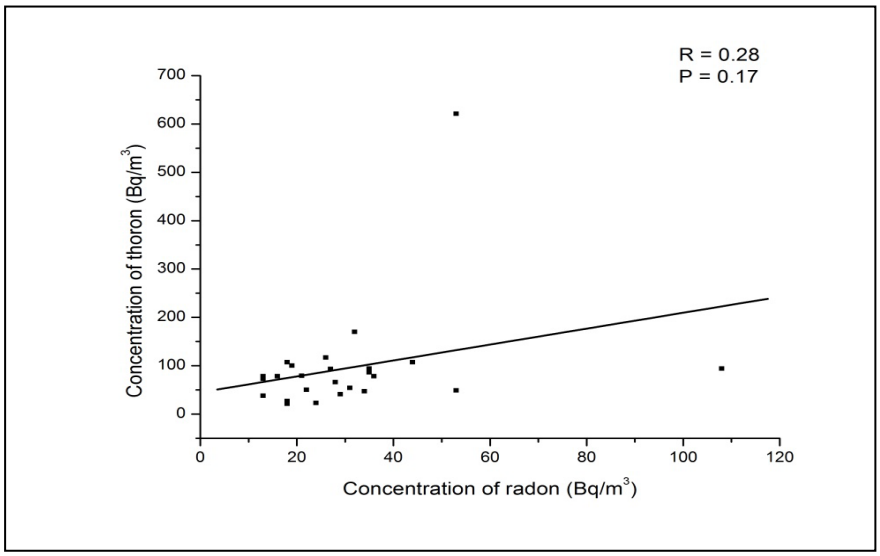

Fig. 5.1 Correlation between indoor Radon and Thoron in 25 samples of Bathery, Wayanad

The average ratio of Thoron to Radon is 3.3. It is seen that the Thoron concentration in one house is highly elevated (annual average Thoron concentration = 621 Bq m^{-3}) as compared to all the other houses. As this house is very close to the granite quarries, intensive study was planned in houses in the vicinity of this house and those near other granite quarries. The frequency distribution of both Radon and Thoron has a positive skewness as shown in Fig.5.2 and Fig.5.3. The Q-Q plot of the logarithm of radon concentration for the 25 samples is shown in Fig.5. 4. The same is repeated for thoron excluding the outlier and given in Fig.5.5. The linearity of the plots (Fig.5.4 and Fig.5.5) confirms the lognormal distribution of radon and thoron concentrations. A similar observation of lognormal distribution of radon and thoron concentration was made in the HBNRA of Kerala and some other studies [10, 25].

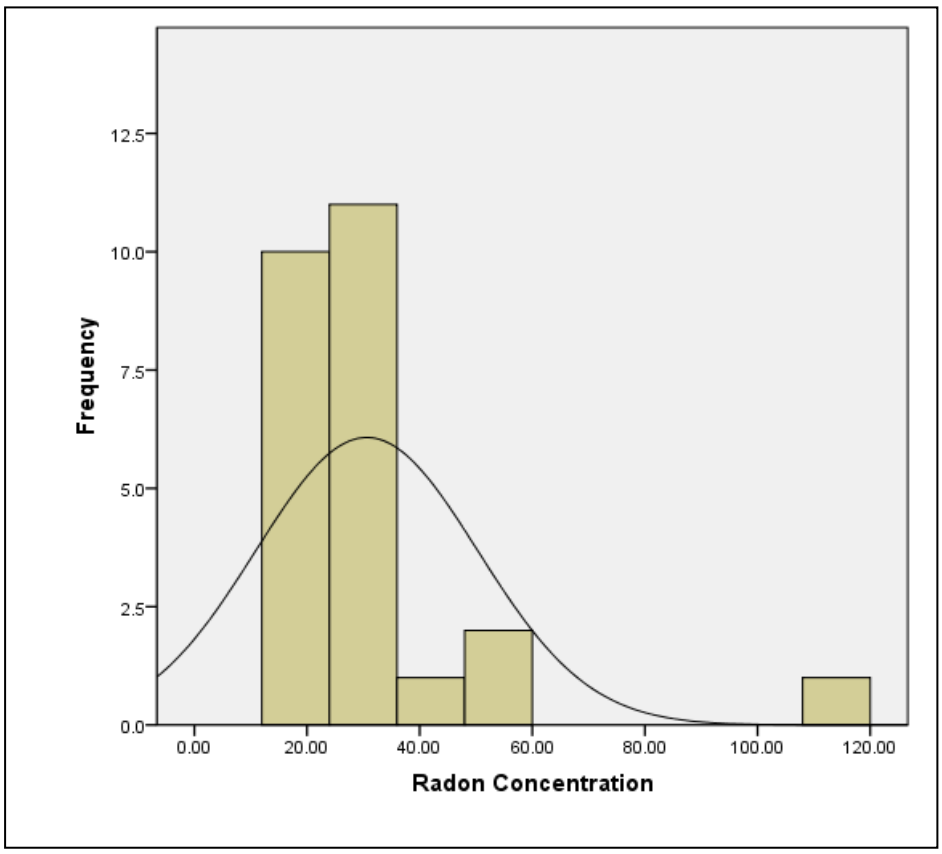

Fig 5.2 Distribution of Radon concentration in phase 1 study

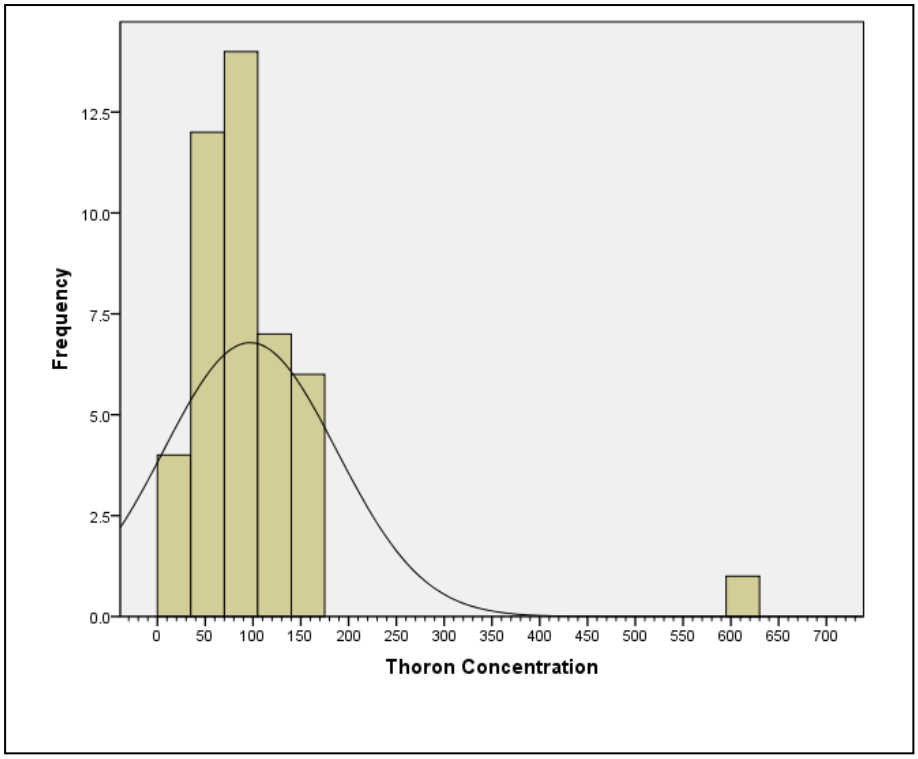

Fig 5.3 Distribution of Thoron concentration in phase 1 study

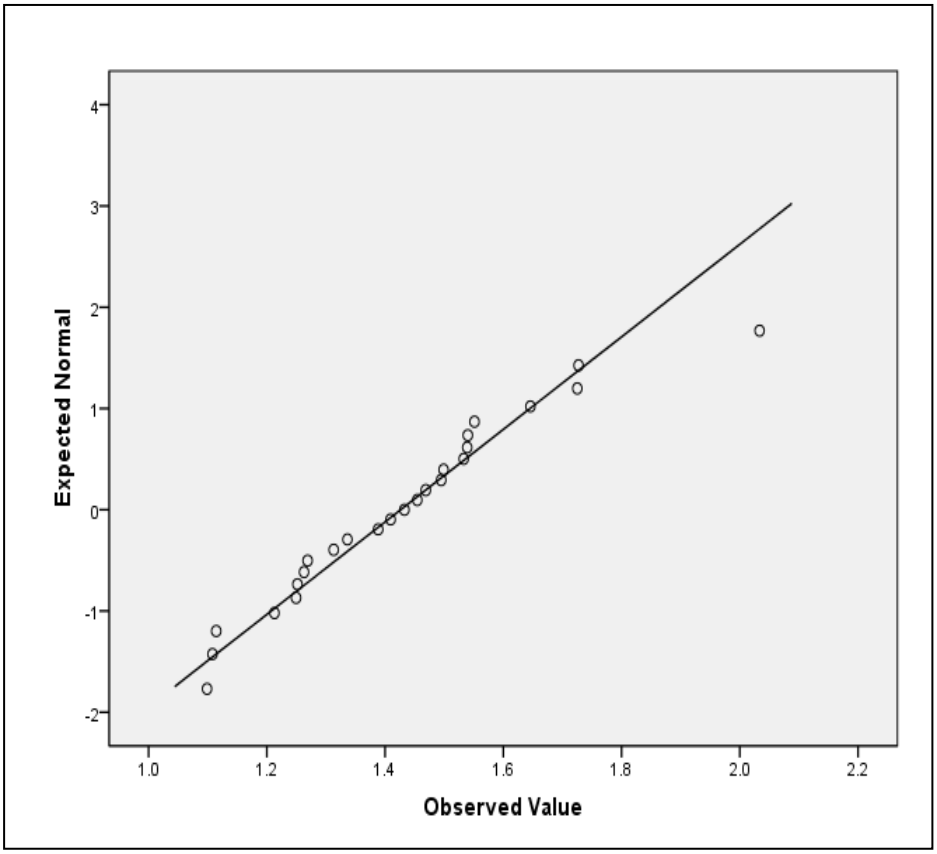

Fig.5. 4 Q-Q plot for radon concentration

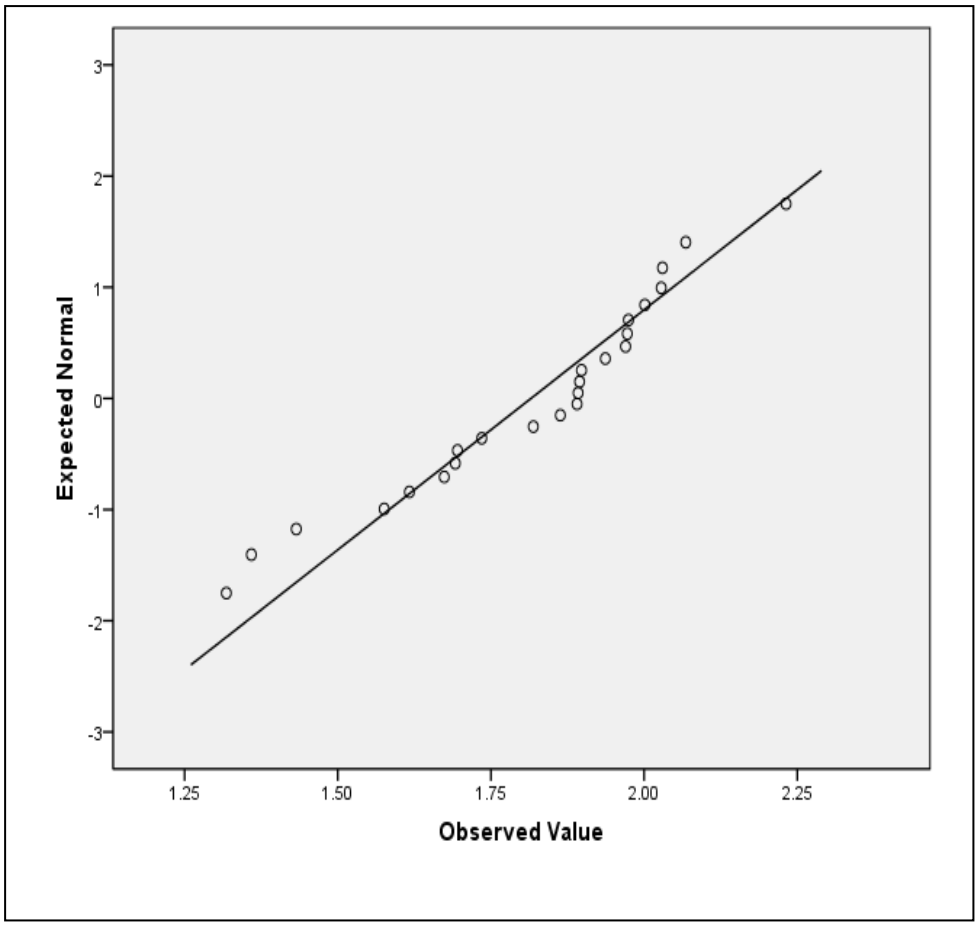

Fig.5. 5 Q-Q plot for thoron concentration

The Annual Effective Dose (AED) for radon and thoron calculated using the equilibrium factor (F_R = 0.4 and F_T = 0.02) given in UNSCEAR [101] is as given in Appendix H. Average AED for radon ranged from 0.32 to 2.72 mSv with a geometric mean of 0.77 mSv. Average AED for thoron varied from 0.12 to 3.48 mSv. The total average AED varied from 0.54 mSv to 4.82 mSv with a geometric mean of 1.31 mSv. From Annexure H it is seen that none of the samples have average AED for radon greater than the reference level prescribed by ICRP. Average concentration of radon and thoron in phase 1 study is given in Table 5.1.

Table 5.1

Average concentration of radon and thoron in phase 1 study

	Radon [Bq/m^3]	Thoron [Bq/m^3]
Minimum	13	21
Maximum	108	621
Average	31	96
Standard deviation	20	115
Geometric mean	27	72
Geometric Standard deviation	2	2

Gammaray Spectrometry of the 25 soil samples (Appendix I) obtained from the vicinity of the houses in which the indoor radon thoron measurements were done showed that the ^{226}Ra concentration in the Bathery taluk varies from below detectable limit (BDL) to 68 Bq/kg with an average of 18 ± 15 Bq/kg. The concentration of ^{232}Th ranges from BDL to 112 Bq/kg with an average of 40 ± 29 Bq/kg. Correlation analysis was done between ^{226}Ra concentration in soil and the indoor radon concentration and ^{232}Th and the indoor thoron concentration. No correlation was found in either case.

5.2 PHASE 2

From the results of the initial soil Gammaray spectrometric studies we found that the soil and rock samples from Ambalavayal Panchayat showed higher concentration of ^{226}Ra and ^{232}Th. Again in the studies of indoor radon and thoron, the concentration of thoron was found to be higher in house near the granite quarry. Second phase of the study comprised of measurements in the vicinity of the house showing high concentration of Thoron as well as few houses in the vicinity of other quarries. The concentration of Radon and Thoron for the phase 2 study is as given in Appendix J and Appendix K. Since the dose due to inhalation of radon and thoron is primarily due to

their progeny, concentration of progeny was also assessed. Result of the progeny concentration as well as the equilibrium factor for each house is given in Appendix L The average equilibrium factor determined for the region using the EERC and EETC is 0.69 ± 0.13 Bq/m^3 for Radon and 0.03 ± 0.03 Bq/m^3 for Thoron. The EF for Radon and thoron is higher as compared to the UNSCEAR value of 0.4 and 0.02 respectively[101]. The average values of radon, thoron and progeny concentration is as given in Table 5.2. The AED for the residents of the region was calculated using the equilibrium factors derived for the respective houses. The average annual effective dose due to radon and thoron and the total average annual effective dose are given in Appendix M.

Table 5.2
Average concentration of radon, thoron and progeny in Wayanad, Kerala, India. Result of the phase 2 study

	Maximum	Minimum	Average ± SD	GM (GSD)
Radon Concentration [Bq/m3]	379	20	68 ± 74	54 (2)
Thoron Concentration [Bq/m3]	576	16	123± 115	95 (2)
EERC [Bq/m3]	160	10	43 ± 30	37 (2)
EETC [Bq/m3]	8	1	3 ± 2	3 (2)
F_{Rn}	0.89	0.43	0.69 ± 0.14	0.68 (1.24)
F_{Tn}	0.14	0.01	0.04 ± 0.03	0.03 (1.83)
Annual Effective dose [mSv]	0.98	12.06	3.42 ± 2.39	2.91 (1.66)

From the study of the Radon – Thoron concentration it is seen that out of the 44 houses studied for Radon–Thoron, only one house [2.3%] showed Radon concentration greater than the action limit (300 Bq/m^3) prescribed by ICRP[14].

5.2.1 Active measurements

It has been earlier discussed by several researchers that the passive measurements using pin hole dosimeters may be influenced by factors like the bulk etching rate, leakage from the cup etc [92, 125]. Hence to ascertain and validate the concentration obtained using pin hole dosimeters, active measurements were done at ten houses including the houses showing higher Radon and Thoron concentrations. Measurements were done for five days in every house to find the indoor radon concentration. Result of the active measurement and its comparison to the passive measurement is as given in Table 5.3.

Table 5.3

Comparison of indoor radon concentration measured using active and passive methods

Sample	Radon concentration from active measurement (Bq/m^3)	Radon concentration from passive measurement (Bq/m^3)
P6	38.38	42.05
P8	22.47	22.01
P9	34.2	39.75
P10	46.28	52.25
P12	60.96	59.17
P14	55.88	60.68
P16	31.31	37.30
P17	81.74	78.71
P18	386.5	377.29
P19	25.18	34.15

It is seen that there is good correlation between the active and the passive measurements as depicted in Fig. 5.6

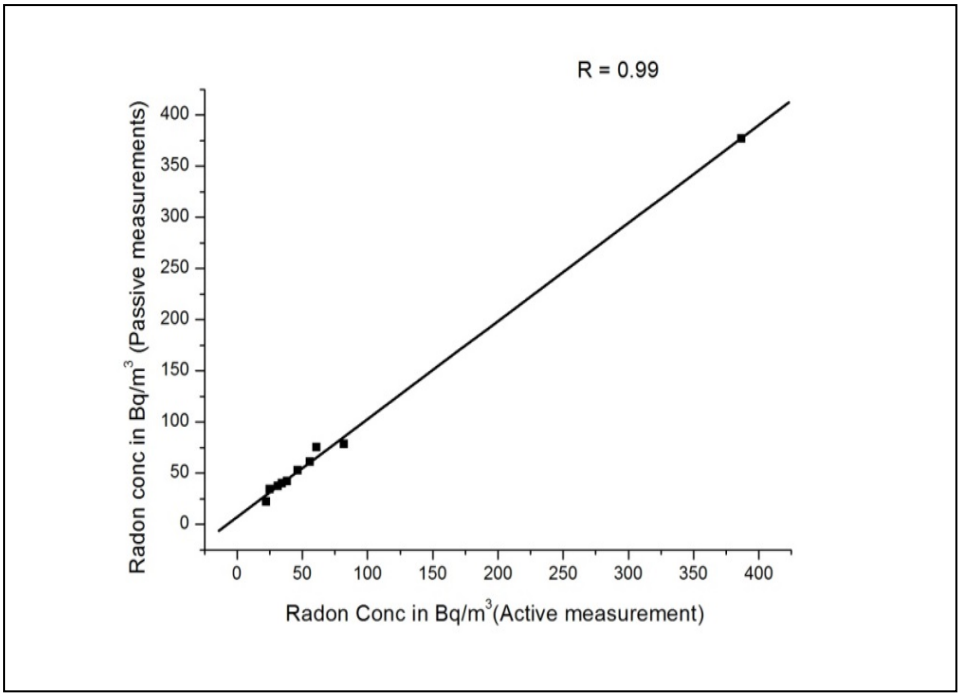

Fig 5.6 Correlation between Active and Passive measurements

Active measurements were done for a period of five days in every house to study the diurnal variation. Plot of the diurnal variation is as given in Fig 5.7 and Fig. 5.8

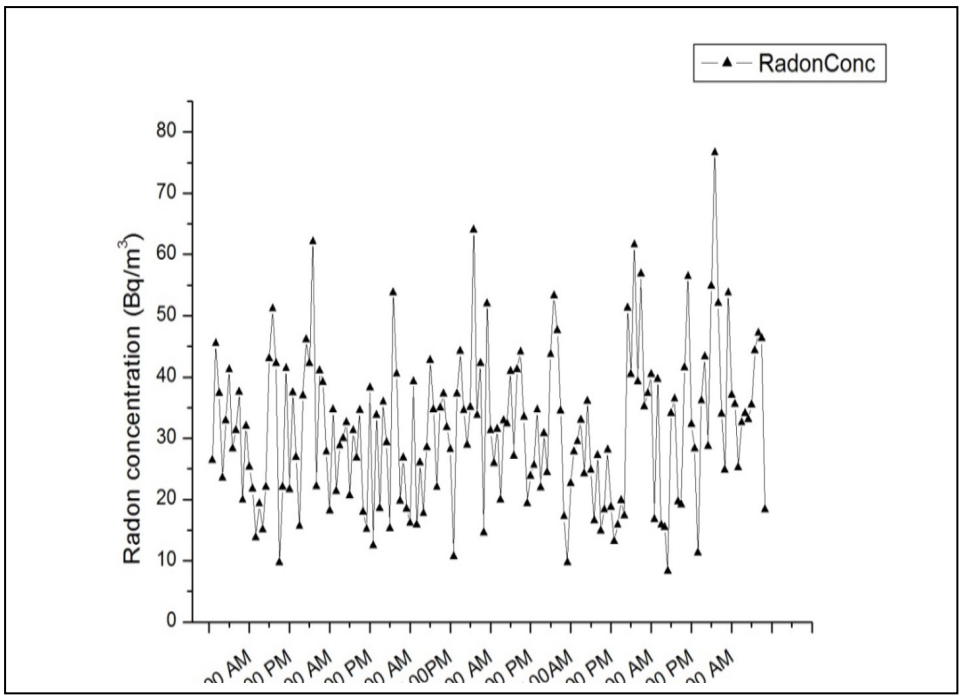

Fig 5.7 Diurnal variation of indoor Radon concentration

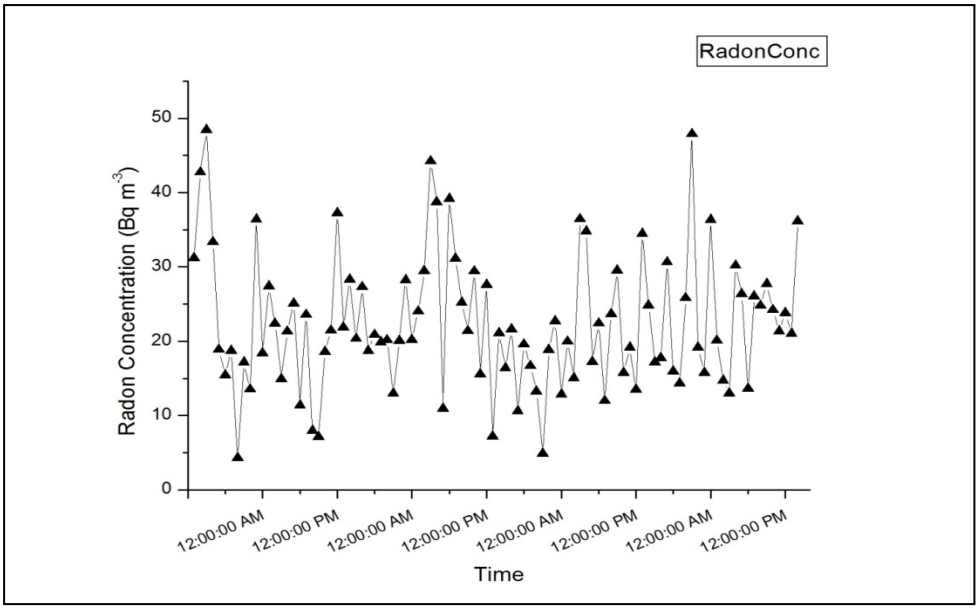

Figure 5.8 Diurnal variation of indoor Radon concentration

From Fig.5.7 and Fig 5.8 we see that the concentration of Radon reduces during day time when the houses are more ventilated as compared to night. There is no particular pattern and the concentration keeps fluctuating. The maximum concentration of Radon is found after midnight and in the early morning hours. In the house showing the highest concentration of Radon, the value increased to as high as 1533 Bq/m^3 at night (Fig 5.9).

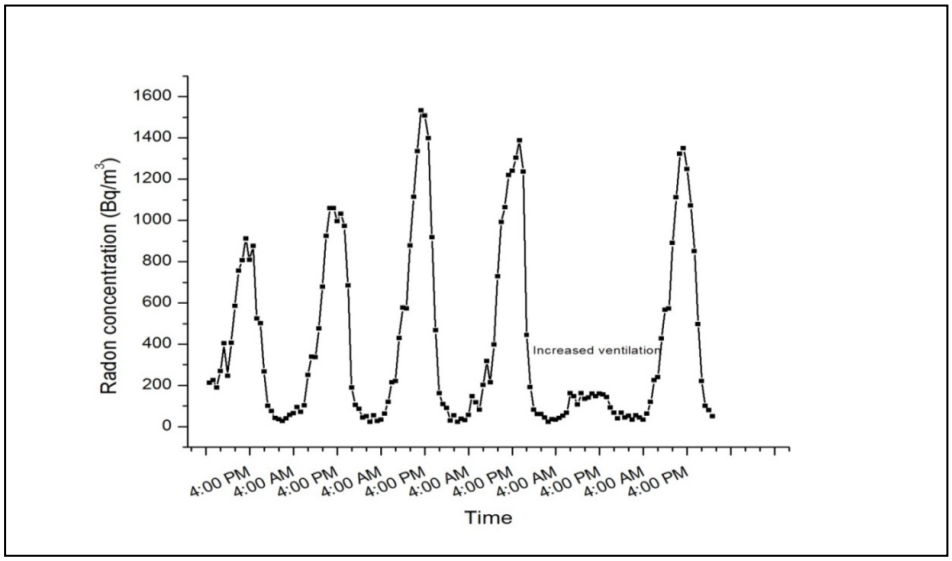

Fig 5.9 Diurnal variation of Radon in the house showing maximum Radon concentration.

By keeping the door of the room open overnight this radon concentration dropped to levels similar to that shown during the day time when the room had increased ventilation. Fig.5.9 shows the significant variation which ventilation can make to the concentration of Radon. Due to the increase in ventilation overnight the maximum concentration of Radon decreased by a factor of 5.

5.2.2 Exhalation measurements

During the phase 1 study, soil samples were collected from the vicinity of the houses. Majority of the houses in this area are made up of mud blocks procured from the site of construction itself. Along with the estimation of terrestrial radioactivity concentration, the aim of collecting soil samples was also to study the correlation, if any, between indoor radon thoron concentration and the radon thoron exhalation rate from soil (Table 5.4).

Out of the 25 samples used for exhalation studies, 14 samples had radon exhalation rate as BDL. 12 samples showed BDL values for thoron exhalation rate too. The radon exhalation rate varied from BDL to a maximum of 80 mBq/kg/h. The thoron exhalation rate ranges from BDL to 17470 Bq/m^2/h. No correlation was found between both the indoor radon concentration and radon mass exhalation rate from soil or the indoor thoron concentration and the thoron surface exhalation rate. No correlation was found between radon exhalation rate from soil and ^{226}Ra concentration in the soil. However, there is very good correlation between thoron exhalation rate and ^{232}Th concentration in soil as shown in Fig. 5.10

Table 5.4

Comparison of soil exhalation studies, passive measurement of indoor radon thoron concentration and terrestrial radioactivity content in soil.

Sample number	Thoron exhalation $Bq/h/m^2$	Thoron concentration (Bq/m^3)	^{232}Th Concentration (Bq/kg)	Radon exhalation Bq/h/kg	Radon concentration (Bq/m^3)	^{226}Ra concentration (Bq/kg)
S1	9788.38	41.38	87	0.029	29.43	21
S2	BDL	621.44	24	BDL	53.11	BDL
S3	11724.97	116.93	112	BDL	25.66	69
S4	8012.71	65.95	66	0.048	28.47	52
S5	6603.27	93.27	47	0.077	27.08	29
S6	3171.04	77.70	29	0.026	35.58	21
S7	BDL	22.85	7	BDL	24.45	12
S8	3953.62	49.17	38	0.027	53.36	32
S9	BDL	54.32	11	BDL	31.26	15
S10	BDL	47.18	75	BDL	34.11	14
S11	1310.05	78.47	10	0.025	12.82	17
S12	5881.91	20.80	53	BDL	18.32	22
S13	1936.46	86.43	20	0.023	34.58	18
S14	BDL	37.65	17	BDL	13.01	15
S15	BDL	78.03	66	BDL	16.33	14
S16	BDL	79.02	10	BDL	20.56	BDL
S17	BDL	72.98	22	BDL	12.55	BDL
S18	BDL	27.05	BDL	BDL	17.85	BDL
S19	BDL	93.89	50	BDL	34.67	BDL
S20	17470.60	170.49	92	0.081	31.54	19
S21	2988.06	49.59	35	0.022	21.69	21
S22	BDL	100.25	32	BDL	18.57	9
S23	BDL	106.62	60	BDL	17.76	14
S24	3919.30	107.14	26	0.014	44.23	17
S25	3455.90	94.29	17	0.047	108.02	20

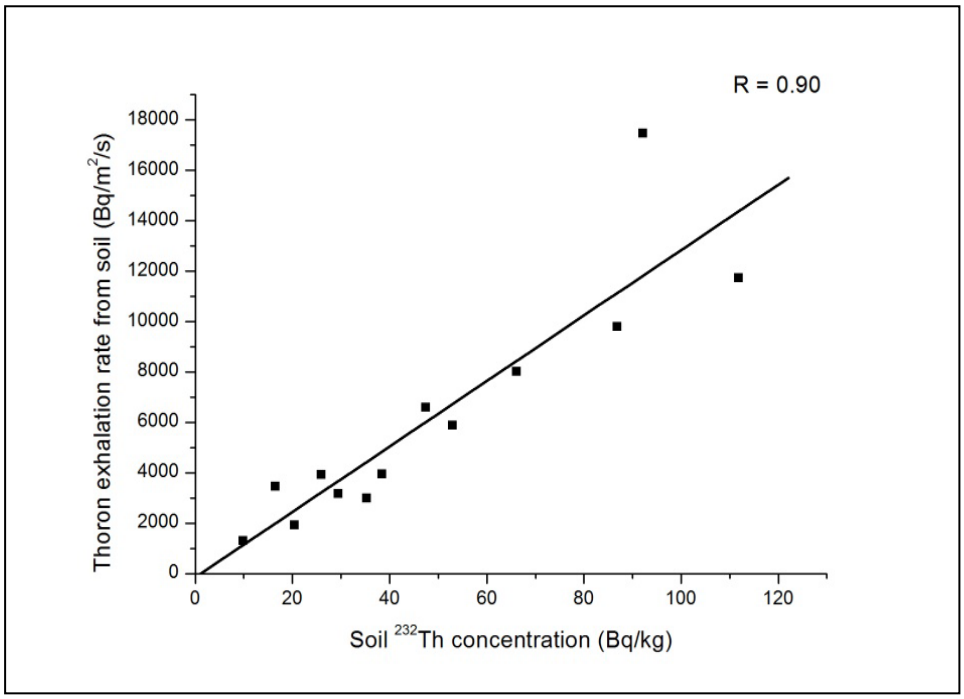

Fig 5.10 ^{232}Th versus thoron exhalation rate in soil

Source term for the relatively higher concentration of Radon and Thoron were analysed by doing exhalation studies for the walls and floor in the houses showing highest concentration of Radon and Thoron. The results show that exhalation rate for radon is more from floor (4.39±0.19 Bq/m^2/hr) as compared to the walls (1.34 ± 0.12 Bq/m^2/hr). The house showing high concentration of Thoron was also studied by wall and floor exhalation. It is found that walls are the major source of Thoron as compared to the floor (mean walls and floor contribution being 5.81±0.20 and 2.53±0.18 Bq/m^2/s, respectively).

Radon from soil gas is considered as the main reason for the spatial variation in Radon concentration. To analyse the cause of spatial variation in the region, Radon exhalation studies were done for soil gas in the vicinity of the houses at a depth of 0.8m. This depth was selected as it is known that the Radon from a depth of up to 1m below the floor can contribute to the indoor Radon concentration [126]. As shown in Table 5.5, the Radon concentration in the soil gas was found to be a maximum of 28 kBq/m^3.

Only marginal variation was found in the Radon concentration in the soil gas at various sites. In Sweden, 2.5% of the houses have Radon concentration greater than the prescribed limit of 400 Bq/m^3[127]. The reason for this has been specified as the soil radon gas. Based on this study Sweden has established criteria under which soil gas radon concentration less than 10 kBq/m^3 is classified as low risk, between 10 kBq/m^3 and 50 kBq/m^3 as normal risk where radon protective construction may be undertaken and above 50 kBq/m^3 as high risk which require radon safe construction [128].The value in the present study falls in the category of normal risk as per the Swedish criteria. Hence detailed analysis of the area is required to arrive at whether protective measures are required in this region for house constructions.

Table 5.5

Comparison of active and passive indoor radon concentration with soil gas radon concentration.

Sample	Radon concentration from active measurement (Bq/m^3)	Radon concentration from passive measurement (Bq/m^3)	Soil gas radon concentration (Bq/m^3)
P6	38.38	42.05	25528
P8	22.47	22.01	22350
P9	34.2	39.75	24230
P10	46.28	52.25	25000
P12	60.96	59.17	26260
P14	55.88	60.68	24520
P16	31.31	37.30	22500
P17	81.74	78.71	25650
P18	386.5	377.29	28000
P19	25.18	34.15	23530

Week correlation was obtained between indoor radon concentration using active measurements and soil gas radon concentration as shown in Fig. 5.11. Similarly, week positive correlation was obtained between indoor radon concentration using passive measurements and soil gas radon concentration (Fig.5.12) For the estimation of the correlation coefficient the sample showing the highest value of indoor radon concentration has been considered as an outlier and avoided.

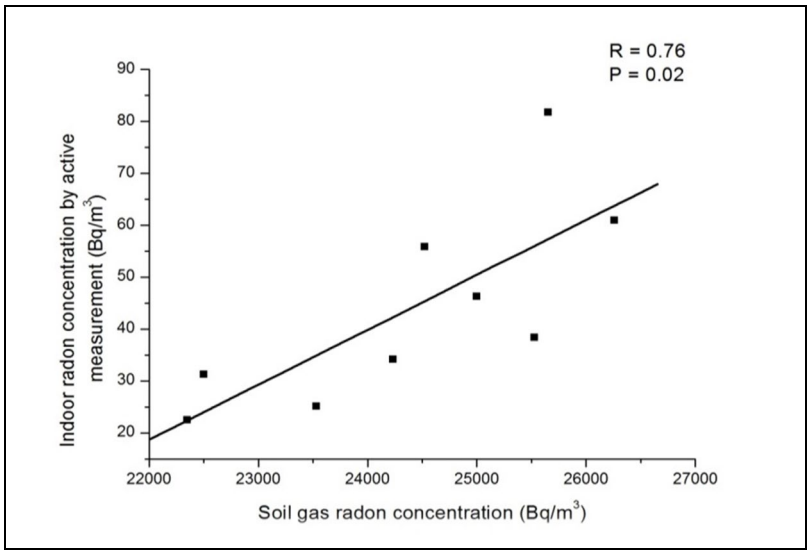

Fig 5.11. Correlation between soil gas radon concentration and indoor radon concentration using active measurements

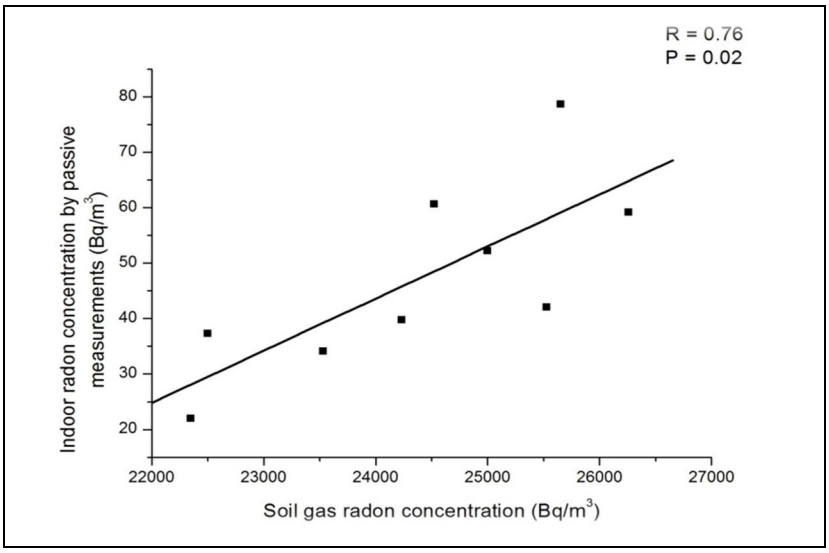

Fig 5.12 Correlation between soil gas radon concentration and indoor radon concentration using passive measurements

A study of the concentration of Radon and Thoron in comparison to the construction material shows that mud houses have the highest concentration of Radon (Fig.5.13). Houses which showed highest Radon and Thoron levels are mud houses with very poor ventilation. Ventilation estimation was made by taking into consideration the number of doors and windows in the room where the dosimeters were kept. Rooms with only one door and no window were called poorly ventilated. Most of such houses had the door of the room facing another room or the corridor. Thus the exchange of air from the outside environment was comparatively less. Rooms with one door and one window, which were kept open for the major part of the day, were considered to be moderately ventilated. Rooms with more than one door and more than one window which were kept open for the major part of the day were assumed to be well ventilated. Concentration of indoor radon with regard to the different wall materials used is presented in Table 5.6.

Table 5.6

Concentration of indoor Radon (in Bq/m^3) with reference to the various wall materials

MUD BLOCK	CEMENT BLOCK	BRICK	LATERITE
29.43	18.32	24.45	25.66
`12.82	17.76	31.26	35.58
16.33	42.05	34.11	18.57
39.75	35.70	12.55	44.23
	34.15	17.86	47.27
		40.07	
		48.21	
		36.55	

53.11	31.54	28.47	27.08
13.01	49.86	53.36	21.69
20.56		34.58	
108.02		34.67	
54.33			
57.01			
47.74			
52.25			
59.17			
60.68			
37.30			
78.71			
377.29			

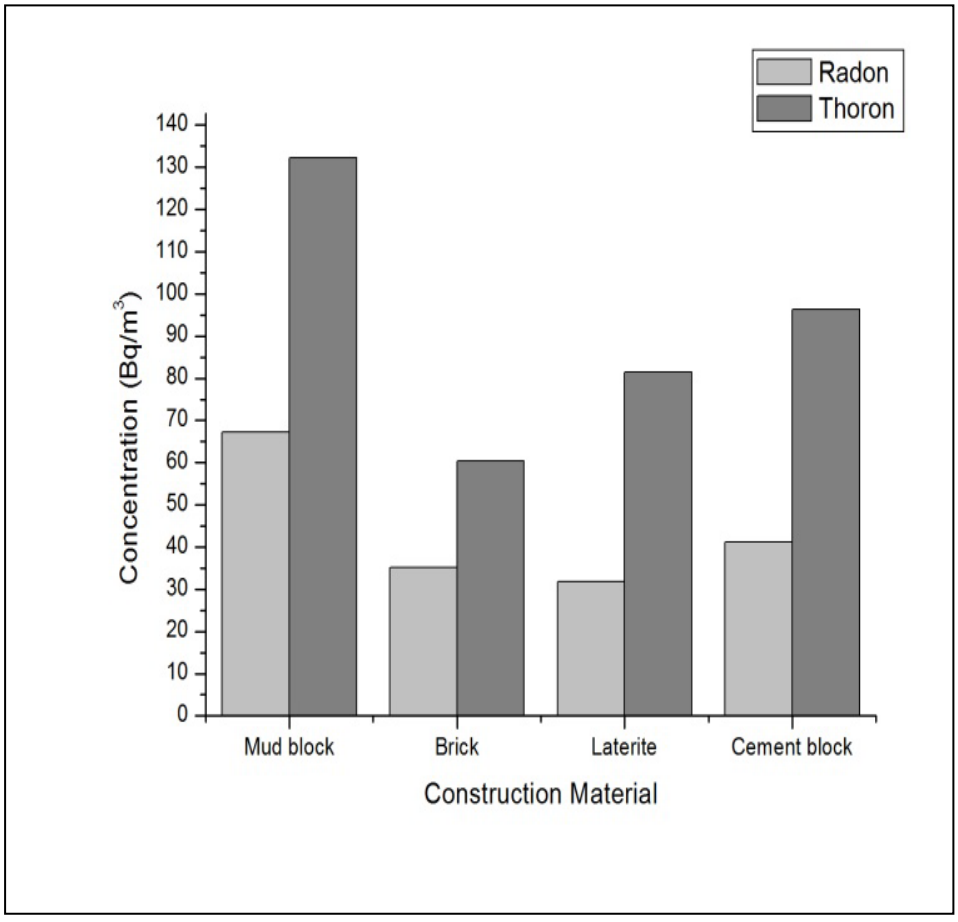

Fig 5.13 Comparison of Radon and Thoron concentration with type of construction material

5.3 DISCUSSION

In the phase 1 study we see that Thoron concentration in the region is higher as compared to the Radon concentration. This may be due to the fact that majority of houses in this region are made up of building materials which are locally available. A major portion of the granite and mechanised sand made out of crushed granite to Malabar as well as the neighbouring districts of Tamilnadu and Karnataka states are supplied from the Wayanad district. It is known that the concentration of ^{232}Th is higher in granite due to the presence of monazite [107]. In the study of soil samples of Wayanad district, in this work, this result has been proved. Hence higher concentration of Thoron in the indoor environment may be attributed to the presence of the higher concentration of thorium in the building materials. Many researchers [22, 27] have studied indoor Radon and Thoron concentration in the Neendakara-Chavara region, which is one of the highest background radioactivity regions of the world. Their results show a median of 23 Bq/m^3 for Radon and 24 Bq/m^3 for Thoron [27]. Even though Neendakara-Chavara region has very high terrestrial radioactivity due to the presence of monazite sand, the indoor Radon/Thoron levels are less as compared to the present study. This result is in agreement with the discussion by R. Mishra and B. K Sapra in the BARC newsletter [129]. They suggest that even though the HBNRA of Kerala experiences 10 times more external gamma effective dose but the inhalation dose due to indoor Radon Thoron concentrations are similar to the Normal Background Radiation Areas (NBRA). The reasons provided by them are the excessive ventilation and the type of flooring which shields Radon from reaching the indoor environment in the HBNRA. Our study proves that along with this the construction material of the houses in the HBNRA may also be contributing to the reduction in Radon Thoron concentrations. Unlike Wayanad, the construction material for the coastal region of Neendakara-Chavara, mainly consisting of

sand, may be coming from outside the region which may not be part of HBNRA. Hence radioactivity content in these construction materials may not be as high as the sand in HBNRA. In the present study too the average value of Radon Thoron concentration increases when the region of interest nears the granite quarries. The workers in the quarries mainly reside in this area. Houses are made of mud blocks with very little ventilation. Temperature in the southern part of Kerala is higher (average of 30^{o}C to 35^{o}C) as compared to the Wayanad district. Due to the cold climate people here opt for lesser ventilation as compared to houses in the warmer regions of Kerala. Hence lack of ventilation may also be one of the reasons for the increase in indoor Radon Thoron concentration as compared to the HBNRA of Kerala. Studies from the Tehri – Garwal regions of India which are also high altitude regions and hence colder places, showed similar results. The average Radon concentration in these places is reported to be 100 Bq/m^3 in winter and that of Thoron is 126.4 Bq/m^3 [130]. The average EF for Radon, found in our study, is more as compared to the value given for the world average in UNSCEAR [101]. The EECR values are effected by both natural and mechanical ventilation [131]. The houses in the present study were selected based on their type of construction. Most of the houses had poor ventilation for energy efficiency. The life style of people in the area does not involve artificial ventilation or air conditioners. Besides this, since the study was done near the areas of the granite quarries, dust loading may also contribute to the accumulation of the progeny in the houses. This may be the reason for the higher EECR and hence higher EF of Radon. Thoron EF is also greater than the value mentioned in UNSCEAR for the world average thoron concentration [101]. The concentration of Thoron is less at the centre of the rooms as compared to the walls due to its very short half-life [8].Since the pin hole dosimeters were hung in the centre of the

room, the concentration of Thoron would have been less and non-uniform over the period of three months.

As per the study only one house has Radon concentration greater than the limit prescribed by ICRP [132]. No such limit is prescribed for Thoron so far. Hence to estimate the hazard due to thoron, the concept of equivalent Radon concentration for Thoron is used as suggested by Jing Chen and Deborah Moir [133]. The equivalent Radon concentration, R_{Tn}, is the Radon concentration that delivers the same annual effective dose as resulted from the Thoron concentration C_{Tn}:

$$\mathbf{R_{Tn}} = \frac{\mathbf{C_{Tn} \times F_{Tn} \times 7000 \times 40}}{\mathbf{F_{Rn} \times 7000 \times 9}} \qquad [5.1]$$

The total indoor exposure to Radon and Thoron may be regarded as the sum of Radon concentration and the equivalent Radon concentration for Thoron. This total exposure can then be compared to the Radon reference value. The equivalent Radon concentration as given by Jing Chen and Deborah Moir [133] was calculated for all the samples. Out of the 44 samples only two samples showed values for total exposure to radon and thoron greater than the limits prescribed by EPA (150 Bq/m^3). Only one sample showed values greater than the limits for indoor Radon concentration (300 Bq/m^3) prescribed by ICRP (Fig.5.14).

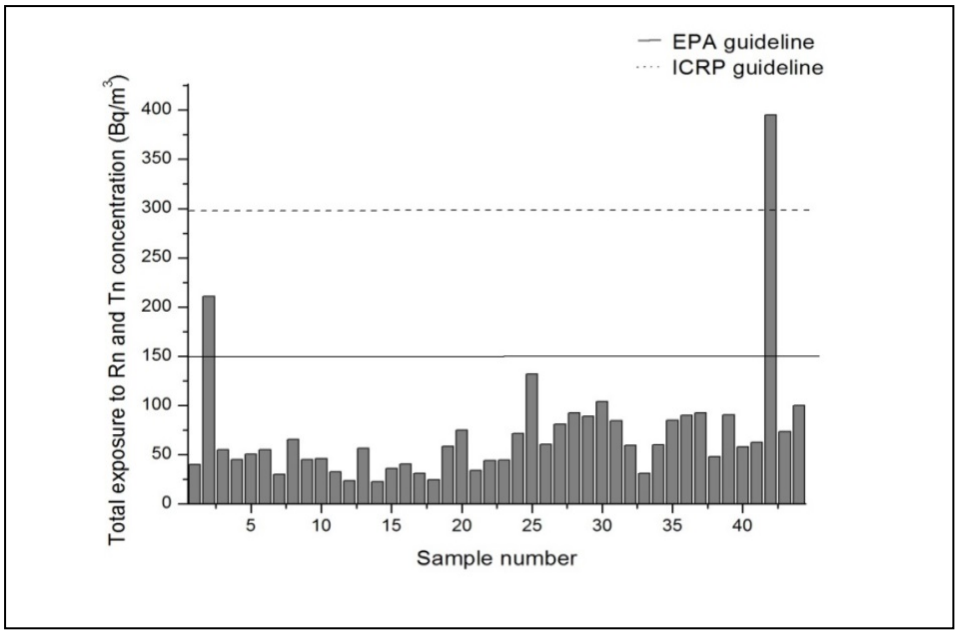

Fig.5.14 Distribution of exposure to total radon concentration (radon concentration + equivalent radon concentration for thoron concentration)

5.3.1 Active measurements and exhalation studies

From the Gammaray spectrometry of soil samples from the vicinity of the houses it is observed that the average thorium concentration is more as compared to the average radium concentration. However, no significant correlation was found between the indoor radon concentration and the exhalation rate of radon from soil. Similarly, no significant correlation was found between the exhalation rate of thoron and the indoor thoron concentration. The reason for this may be the variation in ventilation condition of the houses. Since the concentration of radon varies significantly with the ventilation conditions hence even in houses with high exhalation rate from the source of radon the concentration of indoor radon may not be very high or may be even low if well ventilated. Besides this the variation in the type of building material and flooring may also effect the indoor radon concentration and hence the correlation may be lost. In the case of thoron along with the factors mentioned for radon the short half-life may also contribute to the lack of correlation.

The diurnal variation shows that the concentration of Radon builds up at night when the ventilation in the houses is significantly reduced. The maximum concentration was found in the early mornings. In the house showing highest concentration of Radon, the concentration increased during the early morning hours. Concentration starts dropping once the residents wake up and open the door to the room (which happens to be the only ventilation in the room). As the average Radon concentration is higher than the limits prescribed by ICRP [134], residents of the house were advised to increase the ventilation in the room by affixing an exhaust fan. Another room in the house was also studied using active measurements to check whether the concentration is the same in all the rooms. However, this room showed lesser concentration (average 161.98 Bq/m^3). The room showing the highest concentration of Radon had very large cracks in the floor. The exhalation studies of the walls and floor showed that the major contribution to the Radon concentration came from the floors. Due to the closed room, at night temperature inside the room will be higher as compared to the outside environment. This results in stack effect, drawing the radon inside the room. The large cracks on the floor further assists in the flow of radon from soil gas to the indoor atmosphere due to the pressure variation between the indoor areas and the soil gas. Besides this the room has only one door giving entry to another room and not the outside environment. Hence lack of air circulation from the outdoor environment may also be contributing to accumulation of Radon at night when the door was kept closed. As the second room has intact floors and the door of the room faces the outside environment, the concentration of Radon was not as high as seen in the first room. This shows that in spite of the source (concentration of Radon in the soil gas) being the same, cracks and openings in the floor and walls contribute significantly to increase the Radon concentration and hence the inhalation dose to the residents. A similar effect was seen in the case of Thoron. The exhalation

study of the walls and floors of the house showing higher Thoron concentration showed that the contribution from the walls was more as compared to the contribution from the floor. In this case also the cracks in the walls may be one of the reasons for the excessive exhalation of Thoron. In this study nearly all the houses had tiled roof or roof made of asbestos. Hence the contribution from roof was not taken into consideration.

Comparison of the Radon and Thoron concentration with respect to the construction material used showed that the maximum concentration of Radon and Thoron was found in the houses made of mud blocks. In most of the houses, mud required for the mud blocks were taken from the site of construction itself. Wayanad district has a major population of tribal community who are below poverty line and reside in houses made of mud. The architecture involves very less ventilation to conserve energy. Hence a detailed study of the Radon and Thoron in the tribal villages may lead to better understanding of the inhalation dose to the residents due to Radon and Thoron. The modern houses are all well ventilated and hence the Radon/Thoron levels are within the permissible limits.

RESHMA BHASKARAN “ ENVIRONMENTAL RADIOACTIVITY STUDIES IN WAYANAD, KERALA, INDIA”. THESIS. DEPARTMENT OF PHYSICS, UNIVERSITY OF CALICUT, 2018.

Chapter 6
CONCLUSIONS

The present work dealt with the generation of baseline data of background radioactivity for Wayanad district of Kerala. Even though Kerala has been studied a lot in terms of environmental radioactivity analysis, this study is one of its kind since no data is available for the high altitude regions of Kerala so far. The study has been useful in understanding the distribution of terrestrial radioactivity in the district. Since raw material for several districts for the construction of buildings are supplied from the quarries in Wayanad, the study also gave a glimpse of the indoor radon thoron concentration that may be expected in other regions utilising the raw materials from the quarries here.

6.1 IMPORTANT RESULTS

In air survey of radiation at a distance of 1 m from the soil and rock surfaces gave a very clear indication that higher radioactivity was concentrated in the Bathery taluk with the regions of the granite quarry namely the Ambalavayal Panchayat showing relatively higher dose rate in air.

Gammaray spectrometry for the soil samples of the region showed that the concentration of Potassium is higher in soil than the other two radionuclides. Potassium ranged from 37.6 – 1180 Bq kg^{-1} with a mean of 265.1 Bq kg^{-1}. The value of ^{226}Ra ranged from 9.9 – 95.6 Bq kg^{-1} with an average of 20.8 Bq kg^{-1} and that of ^{232}Th ranged from 7.21 – 209.03 Bq kg^{-1} with an average of 155.24 Bq kg^{-1}. The highest concentration of all three radioisotopes was found in Ambalavayal Panchayat.

The average radioactivity concentration of ^{40}K, ^{226}Ra and ^{232}Th was found to be 669 Bq kg^{-1}, 65 Bq kg^{-1} and 100 Bq kg^{-1} respectively for rock samples. Rock samples showed higher radioactivity concentration as compared to the soil samples. The rock

samples from Ambalavayal had higher concentration of thorium as compared to the Kalpetta granites.

Dose rate in air due to soil was found to be from 3.58 – 181.50 nGy h^{-1} with an average of 34.01 nGy h^{-1}. Average annual effective dose due to both indoor and outdoor exposures due to the soil sample was found to be 0.21 mSv. The average annual effective dose due to rock samples of Wayanad district was 0.44 mSv.

Hazard indices were calculated due to the soil and rock samples. Soil had an average external hazard index of 0.20 and average internal hazard index of 0.24. Rock samples showed average external hazard index of 0.41 and average internal hazard index of 0.51. The rock samples from Nandankavala quarry of Ambalavayal Panchayat showed hazard indices greater than the prescribed limits for the soil samples and rock samples.

The study of radon, thoron and their progeny in the regions of Bathery taluk showed geometric mean radon concentration of 27 Bq m^{-3} with geometric standard deviation of 2. The geometric mean concentration of thoron was found to be 72 Bq m^{-3} with GSD of 2.

Average ratio of thoron to radon in Bathery Taluk was found to be 3.3. High elevated thoron concentration of 621 Bq m^{-3} was found in one house which was very close to the granite quarries. Total indoor average annual effective dose due to radon and thoron was found to be 1.31 mSv.

Intensive study of indoor radon and thoron in the Ambalavayal region which has the major number of granite quarries showed the average radon concentration to be double of that in Bathery taluk. The average radon concentration was found to be 54 Bq/m^3 with GSD of 2 Bq/m^3. Average thoron concentration was also relatively higher at

95 Bq/m^3 with GSD 2 Bq/m^3. Study of the progeny concentration resulted in an average equilibrium factor of 0.7 for radon and an average equilibrium factor of 0.04 for thoron, both values being higher than the world average. Hence the average annual effective dose indoors was found to be 3.42 ± 2.39 mSv using the equilibrium factors derived for the region.

Higher radon exhalation rate from the floor as compared to the walls was observed in the house showing higher radon concentration. This needs to be studied further on a larger number of samples to arrive at the exact reason for elevated levels of indoor radon concentration.

6.2 SCOPE FOR FUTURE STUDIES

Taking the average indoor annual effective dose for Wayanad district due to terrestrial radioactivity as 0.17 mSv the total average annual effective dose due to both terrestrial radionuclides and indoor radon thoron concentration was found to be 2.25 mSv in Bathery taluk. The same was found to be 3.59 mSv for Ambalavayal region. This value includes only the external exposure due to terrestrial radionuclides and the inhalation dose due to radon and thoron. However, since this value is higher than both the Indian average and the world average, detailed study of indoor radon thoron concentration needs to be conducted in the whole of Wayanad district to arrive at the exact representative values. Since Wayanad is a major farming district of Kerala with many tea and coffee plantation detailed study of the various types of plants may also be undertaken to understand the soil to plant transfer factor for the various radionuclides. The number of houses covered for indoor radon thoron measurements were limited due to time and infrastructure constraints. Larger number of sample studies may be conducted in the whole of Wayanad district to arrive at an average value for the

Wayanad district. This study has not elaborated on the variation in radioactivity with respect to type of rocks and soil in detail. Since Wayanad is host to different kinds of rocks, detailed study of radioactivity of the region with regard to the geology of the region may lead to a wealth of knowledge about the variation in radioactivity with respect to the type and the age of various rocks. Long term study of terrestrial radioactivity of the region to analyse the effectiveness of this kind of study in understanding the weathering of rocks and soil in the region can also be undertaken.

RESHMA BHASKARAN “ ENVIRONMENTAL RADIOACTIVITY STUDIES IN WAYANAD, KERALA, INDIA”. THESIS. DEPARTMENT OF PHYSICS, UNIVERSITY OF CALICUT, 2018.

Appendix A

List of sampling sites for soil Gammaray spectrometry

Sample No	*Name*	*Latitude*	*Longitude*
1	*Chanthanathode*	*11.8466*	*75.8092*
2	*Periya*	*11.8326*	*75.8521*
3	*Thavinjal*	*11.848*	*75.952*
4	*Kuttimoola*	*11.8328*	*75.9913*
5	*Cheruvoor*	*11.9048*	*76.0650*
6	*Tholpetty*	*11.9519*	*76.0571*
7	*Nallurnad*	*11.7699*	*76.0081*
8	*Cherukatoor*	*11.7733*	*76.0623*
9	*Kenichira*	*11.7280*	*76.1471*
10	*Odapallam*	*11.6922*	*76.2875*
11	*Ponkuzhi*	*11.6887*	*76.3907*
12	*Thoduvetty*	*11.6553*	*76.2867*
13	*Noolpuzha*	*11.6071*	*76.3426*
14	*Cheeral*	*11.6114*	*76.3135*
15	*Nandankavala*	*11.6315*	*76.2074*
16	*Meenangadi*	*11.6580*	*76.1626*
17	*Kalpetta near quarry*	*11.6251*	*76.0883*
18	*Kalpetta*	*11.6251*	*76.0883*
19	*Vythiri*	*11.544*	*76.0402*
20	*Lakkidi*	*11.5134*	*76.0224*
21	*Patham mile Kalpetta*	*11.6078*	*76.0821*
22	*Arambattakunnu*	*11.6677*	*76.0130*

23	*Vellalykotathara*	*11.6582*	*76.0274*
24	*Manjoora*	*11.6531*	*75.9772*
25	*Vengapally*	*11.6313*	*76.0349*
26	*Puliyarmala*	*11.6399*	*76.0842*
27	*Chennalode*	*11.6592*	*75.9957*
28	*Civil station Kalpetta*	*11.6291*	*76.0904*
29	*Mandad*	*11.6276*	*76.1371*
30	*Narikodu Mukku*	*11.5434*	*76.0329*
31	*Pozhuthana*	*11.5869*	*76.0203*
32	*Aramangalam*	*11.5934*	*76.0130*
33	*Thariode*	*11.6444*	*76.0056*
34	*Edavaka*	*11.6829*	*76.0254*
35	*Banasura Dam*	*11.6700*	*75.95778*
36	*Padinjarethara*	*11.6854*	*75.9748*
37	*Panthipoyil*	*11.6747*	*75.9463*
38	*Kuppadithara*	*11.6975*	*75.9841*
39	*Vellamunda*	*11.7341*	*75.9376*
40	*Kanjirangad*	*11.7575*	*75.9057*
41	*Thondernad*	*11.7694*	*75.8376*
42	*Valatt*	*11.7966*	*75.8979*
43	*Tharuvanna*	*11.7349*	*75.9826*
44	*Anchukunnu*	*11.7514*	*76.0348*
45	*Kolagappara*	*11.6544*	*76.2116*
46	*Aayirankolli*	*11.6191*	*76.2606*

47	*Edakkal caves*	*11.6314*	*76.2265*
48	*Chulliyode*	*11.6053*	*76.2566*
49	*Kuppady*	*11.6791*	*76.2625*
50	*Aaram Mile (Pulpally Sulthan Bathery Highway*	*11.7438*	*76.2406*
51	*Irulam*	*11.7413*	*76.2485*
52	*Pulpally*	*11.7923*	*76.1663*
53	*Padichira*	*11.8396*	*76.1789*
54	*Kuruva Island*	*11.8266*	*76.0929*
55	*Neervaram*	*11.7785*	*76.0801*
56	*Kaniyambatta*	*11.7005*	*76.1041*
57	*Kambalakad*	*11.6754*	*76.0767*
58	*Meppadi*	*11.5738*	*76.0578*

Appendix B

Result of soil radioactivity analysis

Sample Code	C_K *(Bq kg^{-1})*	C_{Ra} *(Bq kg^{-1})*	C_{Th} *(Bq kg^{-1})*	*External gamma ray dose rate (nGyh^{-1})*
1	*95 ± 37*	*BDL*	*BDL*	*3.96*
2	*90 ± 36*	*22 ± 8*	*14 ± 11*	*22.28*
3	*86 ± 42*	*BDL*	*BDL*	*3.58*
4	*76 ± 34*	*12 ± 7*	*BDL*	*8.49*
5	*73 ± 36*	*34 ± 9*	*16 ± 11*	*28.14*
6	*57±27*	*13 ± 6*	*28 ± 9*	*25.22*
7	*BDL*	*BDL*	*BDL*	*BDL*
8	*108 ± 44*	*14 ± 9*	*24 ± 14*	*25.49*

9	*BDL*	*21 ± 8*	*55 ±13*	*43.76*
10	*BDL*	*12 ± 6*	*7 ± 10*	*10.02*
11	*188 ± 34*	*BDL*	*209 ± 16*	*134.10*
12	*62 ± 39*	*27 ± 9*	*41 ± 14*	*39.91*
13	*151 ± 34*	*32 ± 8*	*40 ± 11*	*45.26*
14	*90 ± 37*	*22 ± 8*	*39 ± 12*	*37.46*
15	*1180 ± 47*	*96 ± 11*	*146 ± 13*	*181.55*
16	*BDL*	*BDL*	*155 ± 17*	*93.76*
17	*1020 ± 54*	*31 ± 8*	*48 ± 12*	*85.81*
18	*106 ± 36*	*BDL*	*BDL*	*4.42*
19	*153 ± 40*	*22.± 8*	*23 ± 12*	*30.71*
20	*205 ± 41*	*17 ± 8*	*64 ± 14*	*54.89*
21	*169 ± 34*	*14 ± 8*	*46 ± 7*	*41.48*
22	*BDL*	*10 ± 7*	*15 ± 6*	*13.57*
23	*BDL*	*BDL*	*11 ± 6*	*6.91*
24	*BDL*	*21 ± 8*	*10 ± 6*	*15.74*
25	*BDL*	*19 ± 7*	*BDL*	*8.95*
26	*820 ± 37*	*17 ± 7*	*43 ± 6*	*67.56*
27	*74 ± 32*	*BDL*	*24 ± 7*	*17.51*
28	*755 ± 40*	*BDL*	*46 ± 7*	*59.09*
29	*995 ± 40*	*BDL*	*39 ± 6*	*65.36*
30	*BDL*	*11 ± 8*	*29 ± 7*	*22.68*
31	*466 ± 34*	*17 ± 8*	*45 ± 8*	*37.05*
32	*38 ± 31*	*15 ± 8*	*35 ± 7*	*29.73*
33	*BDL*	*28 ± 8*	*BDL*	*12.83*
34	*BDL*	*18 ± 8*	*20 ± 7*	*20.65*
35	*BDL*	*13 ± 7*	*21 ± 7*	*18.53*
36	*BDL*	*20 ± 8*	*24 ± 7*	*23.66*
37	*BDL*	*22 ± 8*	*51 ± 8*	*41.33*
38	*BDL*	*17 ±7*	*18 ± 6*	*18.86*
39	*BDL*	*10 ± 6*	*BDL*	*4.57*
40	*BDL*	*17 ± 7*	*10 ± 6*	*13.63*

41	*BDL*	*14 ± 8*	*21 ± 7*	*19.30*
42	*BDL*	*25 ± 8*	*30 ± 6*	*29.40*
43	*BDL*	*BDL*	*BDL*	*BDL*
44	*38 ± 29*	*12 ± 7*	*9 ± 6*	*12.58*
45	*BDL*	*15 ± 8*	*11 ± 6*	*13.87*
46	*BDL*	*13 ± 8*	*11 ± 7*	*12.84*
47	*615 ± 40*	*49 ± 9*	*65 ± 7*	*87.61*
48	*430 ± 36*	*34 ± 8*	*41 ± 7*	*58.44*
49	*BDL*	*BDL*	*BDL*	*BDL*
50	*BDL*	*11 ± 8*	*34 ± 7*	*25.74*
51	*136 ± 31*	*11 ± 7*	*40 ± 7*	*35.02*
52	*180 ± 40*	*BDL*	*49 ± 8*	*37.32*
53	*BDL*	*11 ± 7*	*28 ± 6*	*21.99*
54	*53 ± 30*	*BDL*	*47 ± 7*	*30.56*
55	*77 ± 29*	*BDL*	*35 ± 6*	*24.20*
56	*BDL*	*BDL*	*14 ± 7*	*8.64*
57	*BDL*	*BDL*	*18 ± 7*	*10.67*
58	*53 ± 31*	*11 ± 7*	*27 ± 17*	*23.56*

Appendix C

Result of rock radioactivity analysis

Sample Code	C_K *(Bq kg^{-1})*	C_{Ra} *(Bq kg^{-1})*	C_{Th} *(Bq kg^{-1})*	*External gamma ray dose (nGyh^{-1})*
59	*947 ± 38*	*38 ± 7*	*41 ± 8*	*81.42*
60	*853 ± 40*	*BDL*	*BDL*	*35.58*
61	*1114 ± 42*	*120 ± 11*	*253 ± 12*	*254.81*
62	*1158 ± 43*	*146 ± 12*	*115 ± 11*	*184.83*
63	*107 ± 22*	*BDL*	*BDL*	*0.93*

64	*127 ± 21*	*BDL*	*BDL*	*089*
65	*1195 ± 41*	*95 ± 7*	*62 ± 6*	*5.42*
66	*145 ± 24*	*BDL*	*BDL*	*1.01*
67	*363 ± 26*	*BDL*	*BDL*	*1.08*
68	*1138 ± 36*	*25 ± 6*	*66 ± 6*	*4.80*
69	*216 ± 27*	*BDL*	*BDL*	*1.11*
70	*284 ± 24*	*6 ± 5*	*BDL*	*2.42*
71	*1048 ± 39*	*28 ± 7*	*64 ± 7*	*5.37*

Appendix D

Dosimetric parameters of Soil

Sample Number	*Radium Equivalent Activity*	*External Hazard Index*	*Internal Hazard Index*	*Gamma Index*	*Annual Effective Dose*	
					Outdoor (mSv)	*Indoor (mSv)*
1	*23.35*	*0.02*	*0.02*	*0.06*	*0.01*	*0.02*
2	*48.7*	*0.13*	*0.19*	*0.34*	*0.03*	*0.11*
3	*11.46*	*0.02*	*0.02*	*0.06*	*0.01*	*0.02*
4	*17.37*	*0.05*	*0.08*	*0.13*	*0.01*	*0.04*
5	*61.87*	*0.17*	*0.26*	*0.43*	*0.03*	*0.14*
6	*57.24*	*0.15*	*0.19*	*0.40*	*0.03*	*0.12*
7	*6.6*	*BDL*	*BDL*	*BDL*	*BDL*	*BDL*
8	*56.69*	*0.15*	*0.19*	*0.41*	*0.03*	*0.13*
9	*101.49*	*0.27*	*0.33*	*0.70*	*0.05*	*0.21*
10	*22.57*	*0.06*	*0.09*	*0.15*	*0.01*	*0.05*
11	*321.13*	*0.85*	*0.85*	*2.22*	*0.16*	*0.66*

12	*90.59*	*0.24*	*0.31*	*0.63*	*0.05*	*0.20*
13	*101*	*0.27*	*0.35*	*0.72*	*0.06*	*0.22*
14	*84.73*	*0.23*	*0.29*	*0.60*	*0.05*	*0.18*
15	*395.21*	*1.07*	*1.33*	*2.88*	*0.22*	*0.89*
16	*224.53*	*0.60*	*0.60*	*1.55*	*0.12*	*0.46*
17	*178.09*	*0.48*	*0.56*	*1.37*	*0.11*	*0.42*
18	*16.21*	*0.02*	*0.02*	*0.07*	*0.01*	*0.02*
19	*67.29*	*0.18*	*0.24*	*0.48*	*0.04*	*0.15*
20	*123.7*	*0.33*	*0.38*	*0.89*	*0.07*	*0.27*
21	*93.19*	*0.25*	*0.29*	*0.67*	*0.05*	*0.20*
22	*31.19*	*0.08*	*0.11*	*0.21*	*0.02*	*0.07*
23	*21.62*	*0.04*	*0.04*	*0.11*	*0.01*	*0.03*
24	*35.27*	*0.10*	*0.15*	*0.24*	*0.02*	*0.08*
25	*24.11*	*0.05*	*0.10*	*0.13*	*0.01*	*0.04*
26	*140.57*	*0.37*	*0.42*	*1.08*	*0.08*	*0.33*
27	*43.46*	*0.11*	*0.11*	*0.29*	*0.02*	*0.09*
28	*132.05*	*0.33*	*0.33*	*0.96*	*0.07*	*0.29*
29	*156.93*	*0.35*	*0.36*	*1.06*	*0.08*	*0.32*
30	*52.63*	*0.14*	*0.17*	*0.36*	*0.03*	*0.11*
31	*85.15*	*0.23*	*0.28*	*0.60*	*0.05*	*0.18*
32	*68.13*	*0.18*	*0.23*	*0.48*	*0.04*	*0.15*
33	*35.45*	*0.08*	*0.15*	*0.19*	*0.02*	*0.06*
34	*47.17*	*0.13*	*0.18*	*0.32*	*0.03*	*0.10*
35	*42.62*	*0.12*	*0.15*	*0.29*	*0.02*	*0.09*

36	*54.19*	*0.15*	*0.20*	*0.37*	*0.03*	*0.12*
37	*95.76*	*0.26*	*0.32*	*0.66*	*0.05*	*0.20*
38	*43.09*	*0.12*	*0.16*	*0.30*	*0.02*	*0.09*
39	*9.9*	*0.03*	*0.05*	*0.07*	*0.01*	*0.02*
40	*30.67*	*0.08*	*0.13*	*0.21*	*0.02*	*0.07*
41	*44.36*	*0.12*	*0.16*	*0.31*	*0.02*	*0.09*
42	*67.26*	*0.18*	*0.25*	*0.46*	*0.04*	*0.14*
43	*25.58*	*BDL*	*BDL*	*BDL*	*BDL*	*BDL*
44	*27.78*	*0.08*	*0.11*	*0.20*	*0.02*	*0.06*
45	*31.59*	*0.08*	*0.13*	*0.21*	*0.02*	*0.07*
46	*30.7*	*0.08*	*0.11*	*0.20*	*0.02*	*0.06*
47	*189.48*	*0.51*	*0.64*	*1.39*	*0.11*	*0.43*
48	*125.79*	*0.34*	*0.43*	*0.92*	*0.08*	*0.29*
49	*20.19*	*BDL*	*BDL*	*BDL*	*BDL*	*BDL*
50	*62.06*	*0.16*	*0.19*	*0.42*	*0.03*	*0.13*
51	*78.93*	*0.21*	*0.24*	*0.57*	*0.04*	*0.17*
52	*94.4*	*0.23*	*0.23*	*0.61*	*0.05*	*0.18*
53	*52.84*	*0.14*	*0.17*	*0.35*	*0.03*	*0.11*
54	*76.59*	*0.19*	*0.19*	*0.50*	*0.04*	*0.15*
55	*64.8*	*0.15*	*0.15*	*0.40*	*0.03*	*0.12*
56	*26.98*	*0.06*	*0.06*	*0.14*	*0.01*	*0.04*
57	*33.85*	*0.07*	*0.07*	*0.18*	*0.01*	*0.05*
58	*53.57*	*0.14*	*0.18*	*0.38*	*0.03*	*0.12*

Appendix E

Dosimetric parameters of rocks

Sample Number	*Radium Equivalent Activity (Bq/kg)*	*External Hazard Index*	*Internal Hazard Index*	*Gamma Index*	*Annual Effective Dose*	
					Outdoor (mSv)	*Indoor (mSv)*
59	*168.66*	*0.46*	*0.56*	*1.29*	*0.10*	*0.40*
60	*70.67*	*0.18*	*0.18*	*0.57*	*0.04*	*0.17*
61	*424.83*	*1.15*	*1.47*	*3.07*	*0.24*	*0.95*
62	*398.75*	*1.08*	*1.47*	*2.89*	*0.23*	*0.91*
363	*14.94*	*0.02*	*0.02*	*0.07*	*0.01*	*0.02*
64	*13.65*	*0.03*	*0.03*	*0.08*	*0.01*	*0.03*
65	*229.82*	*0.74*	*1.00*	*2.04*	*0.16*	*0.64*
66	*14*	*0.03*	*0.03*	*0.10*	*0.01*	*0.03*
67	*27.97*	*0.08*	*0.08*	*0.24*	*0.02*	*0.07*
68	*208.02*	*0.56*	*0.63*	*1.59*	*0.12*	*0.49*
69	*22.15*	*0.04*	*0.04*	*0.14*	*0.01*	*0.04*
70	*28.25*	*0.08*	*0.09*	*0.23*	*0.02*	*0.07*
71	*201.05*	*0.54*	*0.62*	*1.53*	*0.12*	*0.47*

Appendix F

Concentration of Radon in phase 1 study

Sample number	*Radon concentration (Bq/m3)*				*Average (Bq/m3)*
	AUG – OCT	*NOV – JAN*	*FEB – APR*	*MAY – JUL*	
S1	*15*	*26*	*27*	*50*	*29*
S2	*62*	*50*	*58*	*42*	*53*
S3	*24*	*34*	*28*	*16*	*26*
S4	*20*	*41*	*35*	*18*	*28*
S5	*40*	*19*	*26*	*23*	*27*
S6	*44*	*45*	*25*	*27*	*36*
S7	*29*	*32*	*19*	*18*	*24*
S8	*55*	*58*	*44*	*56*	*53*
S9	*22*	*41*	*24*	*38*	*31*
S10	*49*	*24*	*27*	*37*	*34*
S11	*14*	*15*	*11*	*11*	*13*
S12	*19*	*10*	*21*	*23*	*18*
S13	*22*	*43*	*40*	*33*	*35*
S14	*27*	*12*	*6*	*7*	*13*
S15	*12*	*22*	*10*	*21*	*16*
S16	*30*	*31*	*11*	*10*	*21*
S17	*15*	*17*	*10*	*7*	*13*
S18	*21*	*23*	*13*	*15*	*18*
S19	*45*	*23*	*27*	*44*	*35*
S20	*31*	*48*	*17*	*30*	*32*
S21	*58*	*12*	*9*	*8*	*22*

S22	*12*	*29*	*6*	*27*	*19*
S23	*18*	*20*	*17*	*16*	*18*
S24	*33*	*56*	*37*	*52*	*44*
S25	*139*	*176*	*67*	*50*	*108*
Average ± Std dev	*34 ± 26*	*36 ± 32*	*25 ± 16*	*27 ± 15*	*37 ± 20*
GM (GSD)	*28 (2)*	*29(2)*	*20(2)*	*22(2)*	*27(2)*

Appendix G

Concentration of thoron in phase 1 study

Sample number	*Thoron concentration (Bq/m3)*				*Average (Bq/m3)*
	AUG – OCT	*NOV – JAN*	*FEB – APR*	*MAY – JUL*	
S1	*47*	*54*	*37*	*27*	*41*
S2	*667*	*787*	*507*	*525*	*621*
S3	*59*	*101*	*200*	*108*	*117*
S4	*57*	*54*	*35*	*117*	*66*
S5	*81*	*13*	*220*	*59*	*93*
S6	*44*	*51*	*86*	*130*	*78*
S7	*14*	*38*	*29*	*10*	*23*
S8	*94*	*37*	*35*	*31*	*49*
S9	*82*	*15*	*30*	*90*	*54*
S10	*25*	*78*	*78*	*8*	*47*
S11	*107*	*36*	*124*	*47*	*78*
S12	*47*	*12*	*16*	*7*	*21*
S13	*42*	*59*	*131*	*113*	*86*

S14	*19*	*11*	*51*	*70*	*38*
S15	*90*	*39*	*97*	*86*	*78*
S16	*48*	*38*	*121*	*109*	*79*
S17	*79*	*34*	*67*	*112*	*73*
S18	*36*	*32*	*19*	*22*	*27*
S19	*140*	*49*	*95*	*91*	*94*
S20	*59*	*265*	*230*	*128*	*170*
S21	*47*	*24*	*59*	*68*	*50*
S22	*121*	*46*	*138*	*97*	*100*
S23	*24*	*31*	*186*	*185*	*107*
S24	*83*	*152*	*119*	*74*	*107*
S25	*51*	*149*	*96*	*82*	*94*
Average ± SD	*87 ± 125*	*88 ± 156*	*112 ± 103*	*96 ± 100*	*96 ± 115*
GM (GSD)	*60(2)*	*48(3)*	*81(2)*	*64(3)*	*72(2)*

Appendix H

Average Annual Effective Dose due to Radon and Thoron

Sample no	*Average AED radon (mSv)*	*Average AED Thoron (mSv)*	*Total AED (mSv)*
S1	*0.74*	*0.23*	*0.97*
S2	*1.34*	*3.48*	*4.82*
S3	*0.65*	*0.65*	*1.30*
S4	*0.72*	*0.37*	*1.09*
S5	*0.68*	*0.52*	*1.20*

S6	*0.90*	*0.44*	*1.33*
S7	*0.62*	*0.13*	*0.74*
S8	*1.34*	*0.28*	*1.62*
S9	*0.79*	*0.30*	*1.09*
S10	*0.86*	*0.26*	*1.12*
S11	*0.32*	*0.44*	*0.76*
S12	*0.46*	*0.12*	*0.58*
S13	*0.87*	*0.48*	*1.36*
S14	*0.33*	*0.21*	*0.54*
S15	*0.41*	*0.44*	*0.85*
S16	*0.52*	*0.44*	*0.96*
S17	*0.32*	*0.41*	*0.73*
S18	*0.45*	*0.15*	*0.60*
S19	*0.87*	*0.53*	*1.40*
S20	*0.79*	*0.95*	*1.75*
S21	*0.55*	*0.28*	*0.82*
S22	*0.47*	*0.56*	*1.03*
S23	*0.45*	*0.60*	*1.04*
S24	*1.11*	*0.60*	*1.71*
S25	*2.72*	*0.53*	*3.25*
Average ± SD	*0.77 ± 0.50*	*0.54 ± 0.64*	*1.31 ± 0.92*
GM(GSD)	*0.67(1.7)*	*0.40(1.9)*	*1.13(1.67)*

Appendix I

The result of the soil Gammaray spectrometry and indoor Radon, Thoron concentration of phase 1 study

Sample	*^{40}K (Bq/kg)*	*^{226}Ra (Bq/kg)*	*^{232}Th (Bq/kg)*	*Rn conc (Bq/m^3)*	*Tn conc (Bq/m^3)*	*AED indoor from soil (mSv)*	*AED from Rn+Tn (mSv)*
S1	*675*	*21*	*87*	*29.43*	*41.38*	*0.44*	*1.76*
S2	*1096*	*BDL*	*24*	*53.11*	*621.44*	*0.30*	*9.30*
S3	*1218*	*69*	*112*	*25.66*	*116.93*	*0.74*	*2.44*
S4	*787*	*52*	*66*	*28.47*	*65.95*	*0.48*	*1.99*
S5	*400*	*29*	*47*	*27.08*	*93.27*	*0.29*	*2.24*
S6	*116*	*21*	*29*	*35.58*	*77.70*	*0.16*	*2.44*
S7	*BDL*	*12*	*7*	*24.45*	*22.85*	*0.05*	*1.33*
S8	*41*	*32*	*38*	*53.36*	*49.17*	*0.19*	*2.90*
S9	*BDL*	*15*	*11*	*31.26*	*54.32*	*0.07*	*1.99*
S10	*1612*	*14*	*75*	*34.11*	*47.18*	*0.58*	*2.03*
S11	*BDL*	*17*	*10*	*12.82*	*78.47*	*0.07*	*1.44*
S12	*95*	*22*	*53*	*18.32*	*20.80*	*0.23*	*1.04*
S13	*BDL*	*18*	*20*	*34.58*	*86.43*	*0.10*	*2.49*
S14	*38*	*14*	*17*	*13.01*	*37.65*	*0.09*	*1.00*
S15	*BDL*	*14*	*66*	*16.33*	*78.03*	*0.23*	*1.59*
S16	*BDL*	*BDL*	*10*	*20.56*	*79.02*	*0.03*	*1.79*
S17	*181*	*BDL*	*22*	*12.55*	*72.98*	*0.10*	*1.37*
S18	*212*	*BDL*	*BDL*	*17.86*	*27.05*	*0.04*	*1.09*
S19	*313*	*BDL*	*50*	*34.67*	*93.89*	*0.21*	*2.58*
S20	*937*	*19*	*92*	*31.54*	*170.49*	*0.51*	*3.30*

S21	*112*	*21*	*35*	*21.69*	*49.59*	*0.17*	*1.51*
S22	*150*	*8.55*	*32*	*18.57*	*100.25*	*0.14*	*1.94*
S23	*59*	*14*	*60*	*17.76*	*106.62*	*0.22*	*1.98*
S24	*64*	*17*	*26*	*44.23*	*107.14*	*0.13*	*3.15*
S25	*56*	*20*	*16*	*108.02*	*94.29*	*0.10*	*5.82*

Appendix J

Radon Concentration for the phase 2 study

SAMPLE NO	*SEP-NOV (Bq/m^3)*	*DEC-FEB (Bq/m^3)*	*MAR-MAY (Bq/m^3)*	*JUN-AUG (Bq/m^3)*	*AVERAGE (Bq/m^3)*
P1	*52*	*61*	*56*	*49*	*54*
P2	*55*	*67*	*54*	*52*	*57*
P3	*50*	*55*	*45*	*41*	*48*
P4	*49*	*56*	*45*	*40*	*47*
P5	*33*	*87*	*39*	*41*	*50*
P6	*32*	*60*	*37*	*39*	*42*
P7	*30*	*47*	*42*	*41*	*40*
P8	*17*	*23*	*25*	*23*	*22*
P9	*30*	*60*	*41*	*28*	*40*
P10	*71*	*36*	*59*	*43*	*52*
P11	*44*	*58*	*46*	*44*	*48*
P12	*45*	*77*	*56*	*58*	*59*
P13	*31*	*42*	*36*	*34*	*36*
P14	*42*	*80*	*61*	*60*	*61*
P15	*28*	*49*	*40*	*30*	*37*

P16	*38*	*41*	*36*	*34*	*37*
P17	*73*	*81*	*89*	*71*	*79*
P18	*367*	*390*	*394*	*358*	*377*
P19	*31*	*31*	*40*	*36*	*34*
Average ± SD	*59 ± 76*	*74 ± 79*	*65 ± 81*	*59 ± 73*	*64 ± 77*
GM (GSD)	*44(1.86)*	*59(1.77)*	*51(1.76)*	*46(1.77)*	*50(1.75)*

Appendix K

Thoron Concentration for the phase 2 study

SAMPLE NO	*SEP-NOV (Bq/m^3)*	*DEC-FEB (Bq/m^3)*	*MAR-MAY (Bq/m^3)*	*JUN-AUG (Bq/m^3)*	*AVERAGE (Bq/m^3)*
P1	*12*	*19*	*25*	*23*	*20*
P2	*78*	*78*	*72*	*76*	*76*
P3	*161*	*156*	*171*	*149*	*159*
P4	*165*	*123*	*147*	*134*	*142*
P5	*182*	*167*	*157*	*168*	*168*
P6	*139*	*161*	*146*	*140*	*147*
P7	*103*	*62*	*93*	*50*	*77*
P8	*48*	*38*	*49*	*43*	*44*
P9	*48*	*70*	*51*	*44*	*53*
P10	*81*	*169*	*91*	*89*	*108*
P11	*159*	*146*	*134*	*137*	*144*
P12	*137*	*112*	*117*	*120*	*121*
P13	*42*	*49*	*46*	*45*	*45*
P14	*124*	*111*	*112*	*116*	*116*

P15	*113*	*42*	*49*	*50*	*63*
P16	*86*	*92*	*83*	*73*	*83*
P17	*505*	*648*	*612*	*522*	*572*
P18	*63*	*67*	*72*	*60*	*66*
P19	*48*	*42*	*49*	*46*	*46*
Average ± SD	*121 ± 105*	*124 ± 136*	*120 ± 127*	*109± 109*	*118± 118*
GM (GSD)	*92(2.19)*	*90(2.16)*	*91(2.01)*	*83(2.03)*	*90(2.05)*

Appendix L

EERC, EETC, F_R and F_T for phase 2 study

Sample Number	*EERC (Bq/m^3)*	*EETC (Bq/m^3)*	*FR*	*FT*
P1	*28.54*	*2.32*	*0.52*	*0.12*
P2	*29.62*	*2.42*	*0.52*	*0.03*
P3	*37.83*	*2.24*	*0.79*	*0.01*
P4	*35.42*	*2.86*	*0.75*	*0.02*
P5	*38.19*	*3.12*	*0.77*	*0.02*
P6	*31.38*	*3.64*	*0.77*	*0.03*
P7	*31.48*	*2.61*	*0.79*	*0.04*
P8	*11.51*	*0.92*	*0.53*	*0.02*
P9	*26.93*	*2.27*	*0.69*	*0.05*
P10	*32.47*	*2.90*	*0.65*	*0.03*
P11	*41.26*	*3.61*	*0.85*	*0.02*
P12	*47.45*	*4.26*	*0.80*	*0.04*

P13	*20.29*	*1.82*	*0.56*	*0.04*
P14	*32.33*	*2.45*	*0.56*	*0.02*
P15	*29.27*	*2.68*	*0.79*	*0.05*
P16	*24.09*	*2.19*	*0.64*	*0.02*
P17	*69.20*	*8.04*	*0.88*	*0.01*
P18	*167.61*	*5.36*	*0.44*	*0.08*
P19	*23.37*	*2.05*	*0.63*	*0.04*
Average ± SD	*40.54 ± 33.12*	*3.08 ± 1.55*	*0.69 ± 0.13*	*0.03 ± 0.03*
GM (GSD)	*33.69(1.70)*	*2.75(1.57)*	*0.67(1.22)*	*0.03(1.76)*

Appendix M

Average Annual Effective Dose for Ambalavayal region of Bathery, Wayanad.

Sample Number	*AED Radon (mSv)*	*AED Thoron (mSv)*	*Total AED (mSv)*
P1	*1.80*	*0.65*	*2.45*
P2	*1.87*	*0.68*	*2.54*
P3	*2.38*	*0.63*	*3.01*
P4	*2.23*	*0.80*	*3.03*
P5	*2.41*	*0.87*	*3.28*
P6	*1.98*	*1.02*	*3.00*
P7	*1.98*	*0.73*	*2.71*
P8	*0.73*	*0.26*	*0.98*
P9	*1.70*	*0.63*	*2.33*
P10	*2.05*	*0.81*	*2.86*

P11	*2.60*	*1.01*	*3.61*
P12	*2.99*	*1.19*	*4.18*
P13	*1.28*	*0.51*	*1.79*
P14	*2.04*	*0.69*	*2.72*
P15	*1.84*	*0.75*	*2.59*
P16	*1.52*	*0.61*	*2.13*
P17	*4.36*	*2.25*	*6.61*
P18	*10.56*	*1.50*	*12.06*
P19	*1.47*	*0.57*	*2.05*
Average ± SD	*2.55 ± 2.09*	*0.86 ± 0.43*	*3.42 ± 2.39*
GM (GSD)	*2.15(1.70)*	*0.77(1.57)*	*2.91(1.66)*

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