

# **Earthquake Prediction Through Radon Gas**

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**Graduating year 2017**



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Earthquakes

Earthquake prediction

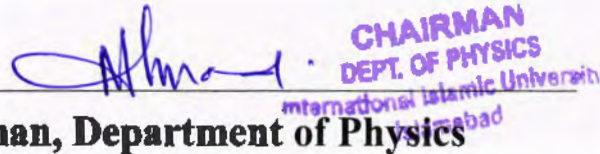
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This Thesis submitted to Department of Physics, International Islamic University  
Islamabad for the award of degree of MS Physics.

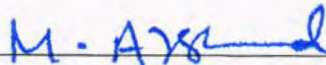


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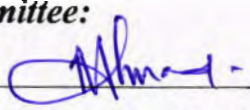
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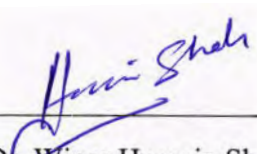
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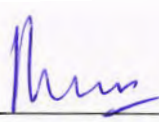
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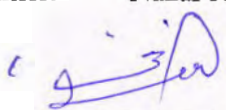
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## **Dedication**

**Dedicated to my beloved father**

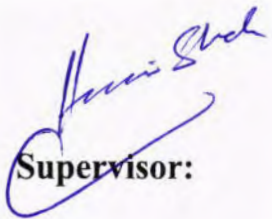
**(may Allah keep his soul in rest), my loving**

**mother and my elder brothers who give me**

**support at every stage of life.**

## Forwarding Sheet by Research Supervisor

Thesis titled “Earthquake Prediction Through Radon Gas” submitted by Zakir Ullah (Registration No 300-FBAS/MSPHY/F14) in partial fulfillment of MS degree in Physics has been completed under my supervision. I am satisfied with the quality of his research work and allow him to submit thesis for further processing to graduate with Master of Science degree from Department of Physics, FBAS, as per IIUI Islamabad rules and regulations.



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Date: 12-10-2017



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## Abstract

Earthquake is a challenging phenomenon for the scientific community due the dependence on several factors. There are many phenomena that may be used for the forecasting purpose these are ionospheric studies (TEC, foF2), vertical electric field (VEF) variation, statistical analysis of seismicity (b-value), thermal infrared variation (TIR), variation of outgoing longwave radiation (OLR) etc: Among these phenomenon radon concentration variations in soil, air and ground water is also a potential source for the forecasting of an earthquake. The aim of the current study is to measure the radon concentration at Islamabad, the capital of Pakistan by the help of two techniques (a) the long-term technique known as passive method and (b) short term technique known as Active method technique. The measurements were taken from 1<sup>st</sup> July, 2016 to 30<sup>th</sup> September, 2016. The passive method was carried out by the help of SSNTDs CN-85 type plastic detector for a duration of 90 days. The detectors were planted in a bore-hole of 65cm deep and 10cm in diameter. After the completion of duration, they were chemically etched in a 6N NaOH solution for a duration of 50 mint at 70°C temperature. The values range from 1554 Bq.m<sup>-3</sup> to 2792 Bq.m<sup>-3</sup> with an average value 2272.7875 Bq.m<sup>-3</sup>. The active method for the measurement was carried out by the help of continuous radon/thorn monitor RTM2200. The instrument measure the meteorological paramcters as well with an interval of 15 mints. The whole data for radon was analyzed hourly in correspond with meteorological paraments. The  $\pm 2\sigma$  upper and lower limit was applied to detect the anomalies in the radon concentration. An abnormal behavior of radon was detected on 31<sup>st</sup> July, 2016 that was followed by an earthquake event of magnitude 4.2 with a depth of 55.9 km. The distance between the epicenter of the earthquake and radon monitoring station was 107 km. The D/R ratio was used to find that the anomaly observed at the station was lying within the earthquake preparation zone. The D/R ratio was 0.6 which is greater than 0.4 recommended by previous author.

# Chapter 01:

## 1.1 Introduction:

Contemporary progress in understating the hierarchical nature of lithosphere and its dynamics help in designing the optimum earthquake forecasting strategy by using multi-precursory analysis of pre- and co-seismic changes prior to geodynamic activity. The multi-precursory strategy of earthquake forecasting includes: ionospheric studies (TEC, foF2), vertical electric field (VEF) variation, statistical analysis of seismicity (b-value), radon concentration variation in soil, air and ground water, water level fluctuations, thermal infrared variation (TIR), variation of outgoing longwave radiation (OLR) etc. along their limitations [12345].

The research community working in the domain of earthquake forecasting primarily focused on fluctuation of radon concentration in different environments i.e. soil, air and ground water prior to seismic activity due to its abnormal behavior. These characteristics include its chemical stability, inner nature, easy detectability, movement along faults zone and nearly linear distribution in the earth crust [6].

The fluctuation in radon concentration have been observed by different researcher and it was detected that radon shows abnormal behavior before seismic events. The abnormal behavior of radon was reported in 1971 by Antsilevich 3 to 8 days before an earthquake of magnitude 3 to 3.5. He found 20 % increase in radon concentration from the background values [7]. In later section, we will discuss a detail overview of radon gas.

## 1.2 Radon Discovery:

Radon was discovered by Friedrich Ernst Dorn a German Chemist in 1900 during the decay process of radium. It was named as Nitron because of its shining nature. The name was derived from Latin word Nitens, after that it was named as radon in 1923 [8]. It's atomic number is 86 and mass numbers 222 [9]. It is a heaviest noble gas having 36 isotopes ranging from  $^{139}\text{Rn}$  to  $^{228}\text{Rn}$ . Among these isotopes only three are present in nature these are  $^{222}\text{Rn}$ ,  $^{219}\text{Rn}$  and  $^{220}\text{Rn}$ . They are obtained from the decay process of  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{232}\text{Th}$  respectively [10]. Only  $^{222}\text{Rn}$  isotope have greater half-life of 3.8 days, while the remaining have half-lives 54.5s and 3.92s respectively [11]. The long half of  $^{222}\text{Rn}$  makes it a good tracer for geophysical, geological and geochemical studies. It is a daughter product of uranium and thorium decay series [12].

It is naturally available in underground sources and diffuses to atmosphere from soil and rocks through underground pathways. It disintegrates through  $\alpha$  decay process. Its concentration in the atmosphere can be measured in ppm level [13]. It plays a central role in the radioactivity of the earth but due to their less availability in nature it cannot be used for the discovery of

inert gas elements [14]. It comes out to the atmosphere where its concentration depends mainly on seismic activities as well as on meteorological processes. Radon production in natural minerals was known from 1920s but its use as an earthquake precursor started few decades before [15].

### 1.3 Physical and Chemical Properties of Radon:

Physical Properties of an element contain its phase, density, melting point, boiling point, heat of fusion, heat of vaporization, oxidation state, molar heat capacity and critical point [11]. Radon is a colorless, odorless and tasteless gas. It is densest gas at room temperature as compared to the other noble gasses. At standard temperature and pressure (STP) its density is  $9.73 \text{ kg/m}^3$  [16]. Melting point of radon is 202 k and boiling point is 211.5 k. The heat of fusion and heat of vaporization of radon are 3.247 kJ/mole and 18.10 kJ/mole respectively. Molar heat capacity and critical point are  $5R/2=20.786 \text{ J/mole. K}$  and 377 K, 6.28 MPa respectively. Radon glow due to the stiff radiations it produces during the condensation process [17]. The chemical properties of an element comprise the ionization energy, electronegativity, solubility and heat of vaporization etc. Ionization energy is the amount of energy required to remove an electron from the outer most shell of an atom. It increases with the decrease in atomic number and decrease with the increase in atomic number therefore moving up in periodic table ionization energy increase and going down it decrease. The first ionization energy of radon is 1.037 KJ/mole [18]. The first ionization energies of noble gases relative to their atomic number are shown in the following Figure 1-1:

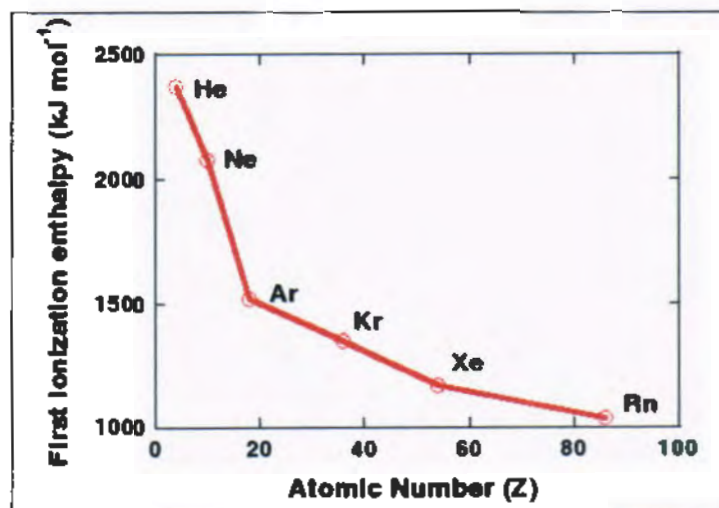


Figure 1-1. Relation between atomic numbers of an element with first ionization energy. Electronegativity of an atom is the ability to attract an electron pair toward its self in a chemical reaction. It decreases with the increase in atomic number therefore, moving down the group in

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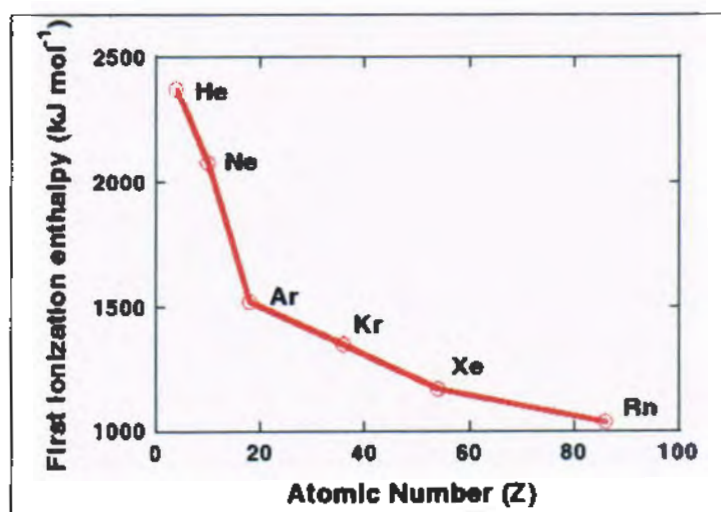


Figure 1-1. Relation between atomic numbers of an element with first ionization energy. Electronegativity of an atom is the ability to attract an electron pair toward its self in a chemical reaction. It decreases with the increase in atomic number therefore, moving down the group in

a periodic table it decreases. Radon has lower electronegativity value which is 2.2 Pauling scale. It is lower than the electronegativity of Xenon (2.60 Pauling scale) and is therefore relatively less reactive than xenon [19]. The solubility in a group also increases with the increase in its atomic number therefore radon is more soluble than Xenon. It is sparingly soluble in water and is more soluble in organic liquids [20]. Heat of vaporization is the amount of heat required to vaporize a certain liquid at a constant temperature. It increases with the increase in atomic number so moving down in a group heat of vaporization also increases. Heat of vaporization of group eight elements corresponding to their atomic number is shown in the following Figure 1-2:

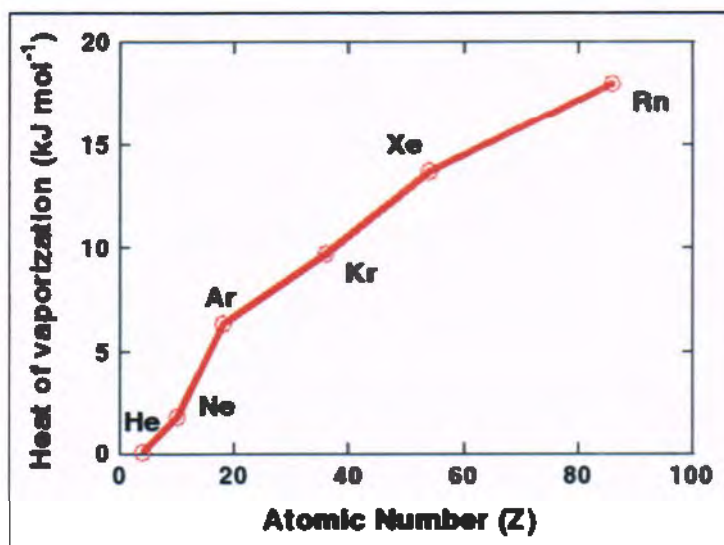


Figure 1-2. Relation between the atomic number of an element and heat of vaporization.

Its position in the periodic table is such that it lies in between metals and non-metals which show both types of properties, therefore it can also be classified as a metalloid element. It can react with fluorine, Halogen fluorides and with oxidizing salts at a temperature of about 25°C or below it. Cationic solutions of radon have been prepared in non-aqueous solution like hydrogen-fluoride and halogen-fluorides, but there are no ionic aqueous solutions of radon compounds. The oxidation state of radon is 0, 2 and 6. Its covalent radius is 0.150 nm and Van-Deer Waals radius is 0.220 nm. Valence shell electronic configuration is 6S<sup>2</sup> 6P<sup>6</sup>. Comparing the properties of Krypton and Xenon fluorides with radon, it is found that it forms radon fluoride, RnF<sub>2</sub> and its by-products RnF+SbF<sub>6</sub><sup>-</sup>, RnF+TaF<sub>6</sub><sup>-</sup>, RnF+BiF<sub>6</sub><sup>-</sup> where RnF<sub>2</sub> produce radon having zero oxidation number on reaction with water while elemental radon has +2 oxidation state. The process is shown by the following chemical reaction:



It produces a nonvolatile difluoride when it reacts with fluorine at 4000oC temperature.



It has the property that it displaces the ions of first group elements making  $H^+$ ,  $Na^+$ ,  $K^+$ ,  $Cs^+$  ions when it is in ionic state [11].

### 1.4 Nuclear Properties of Radon:

There are 86 protons and 136 neutrons in the nucleus of  $^{222}Rn$  therefore, atomic number  $Z=86$ , and atomic weight  $A=222$ . Its nucleus can be represented as  $^{222}Rn_{86}$  [10]. The nuclear properties of  $^{222}Rn$ ,  $^{220}Rn$  and  $^{219}Rn$  are classified in the following Table;

Table 1-1. Nuclear properties of radon stable isotopes.

Parameters	Symbols	$^{222}Rn$	$^{220}Rn$	$^{219}Rn$
Half life	$T_{1/2}$	3.825days	54.5s	3.92s
Decay constant	$\Lambda$	$2.098 \times 10^{-6} s^{-1}$	$1.242 \times 10^{-2} s^{-1}$	$0.174 s^{-1}$
Average recoil energy	$E_{(r)}$	86 Kev	103 Kev	104 Kev
Diffusion coefficient (air)	$D_{(a)}$	$1 \times 10^{-5} m^2 s^{-1}$	---	---
Diffusion coefficient (water)	$D_{(w)}$	$1 \times 10^{-9} m^2 s^{-1}$	---	---

### 1.5 Radon Production through Nuclear Decay Processes:

Natural uranium consists of three isotopes uranium-238, uranium-235, and uranium-234. These isotopes are radioactive. The nuclei of radioactive elements are unstable, meaning they are transformed into other elements, typically by emitting alpha and beta particles. They also emit gamma radiations to become stable. Uranium-238, the most predominant isotope in uranium ore, decays into thorium-234 by the emission of alpha particle, which itself decays by beta emission to protactinium-234. The protactinium decay into uranium-234 by beta decay process, and so on [21]. The three stable isotopes of radon and their decay progenies obtained from the decay process of these three isotopes ( $^{238}U$ ,  $^{235}U$  and  $^{232}Th$ ) are shown in Figures 1-3, 1-4 and 1-5 respectively. The Figure 1-3: show that  $^{238}U$  emits an alpha particle and transformed into  $^{234}Th$ , which further undergoes through two beta decay processes to form  $^{234}U$  Isotope. The  $^{234}U$  decay into  $^{230}Th$  which further decay into radium ( $^{226}Ra$ ). The radium element produce radon ( $^{222}Rn$ ) through alpha decay process [22]. Radon is also a radioactive element therefore, decay into its progeny through alpha and Beta decay processes. The whole process

ends on the production of  $^{206}\text{Pb}$  with several alpha and beta decay process during the nuclear transmutation [23].

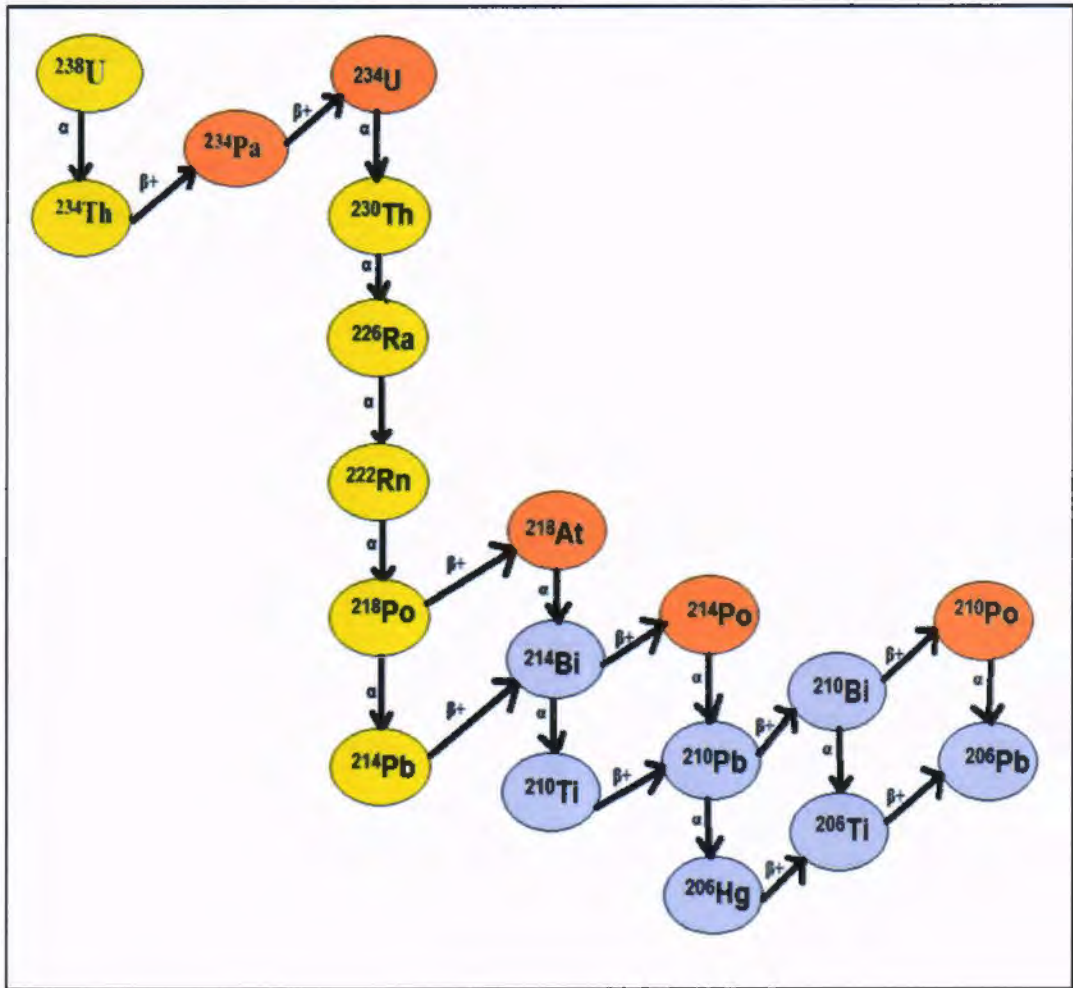


Figure 1-3.  $^{222}\text{Rn}$  production through the decay process of  $^{238}\text{U}$ .

Several radiation protection contemplations deal with the products of fission. A number of elements undergo fission, but the most important ones are  $^{235}\text{U}$ ,  $^{238}\text{U}$ , and  $^{239}\text{Pu}$ . The decayed nucleus produces energy, high-speed neutrons, speedy gamma rays, and extremely charged fission fragments. Absorption of a neutron by a heavy nucleus cause the decay of the parent element. For a neutron added to  $^{235}\text{U}$  have energy 5.2 Mev. Now the adding a single thermal neutron make the energy 6.4 Mev. This excitation energy must be relieved, for this purpose the nucleus under go through decay process [24]. Figure 1-4: Show the decay process of  $^{235}\text{U}$ . It decays into  $^{231}\text{Th}$  through alpha decay process which further decay into  $^{231}\text{Pa}$  following beta decay mechanism.  $^{231}\text{Pa}$  then decay into  $^{227}\text{Ac}$  by alpha decay process. Now there are two possibilities for the decay process of  $^{227}\text{Ac}$ , either it decays into  $^{223}\text{Fr}$  through alpha decay process and then transform into  $^{223}\text{Ra}$  through beta decay

process or first it transforms into  $^{227}\text{Th}$  by beta decay process and  $^{227}\text{Th}$  than decay into  $^{223}\text{Ra}$  through alpha decay process.  $^{223}\text{Ra}$  will produce  $^{219}\text{Rn}$  which further decay into its progeny and finely the process end on the production of  $^{207}\text{Pb}$  [23].

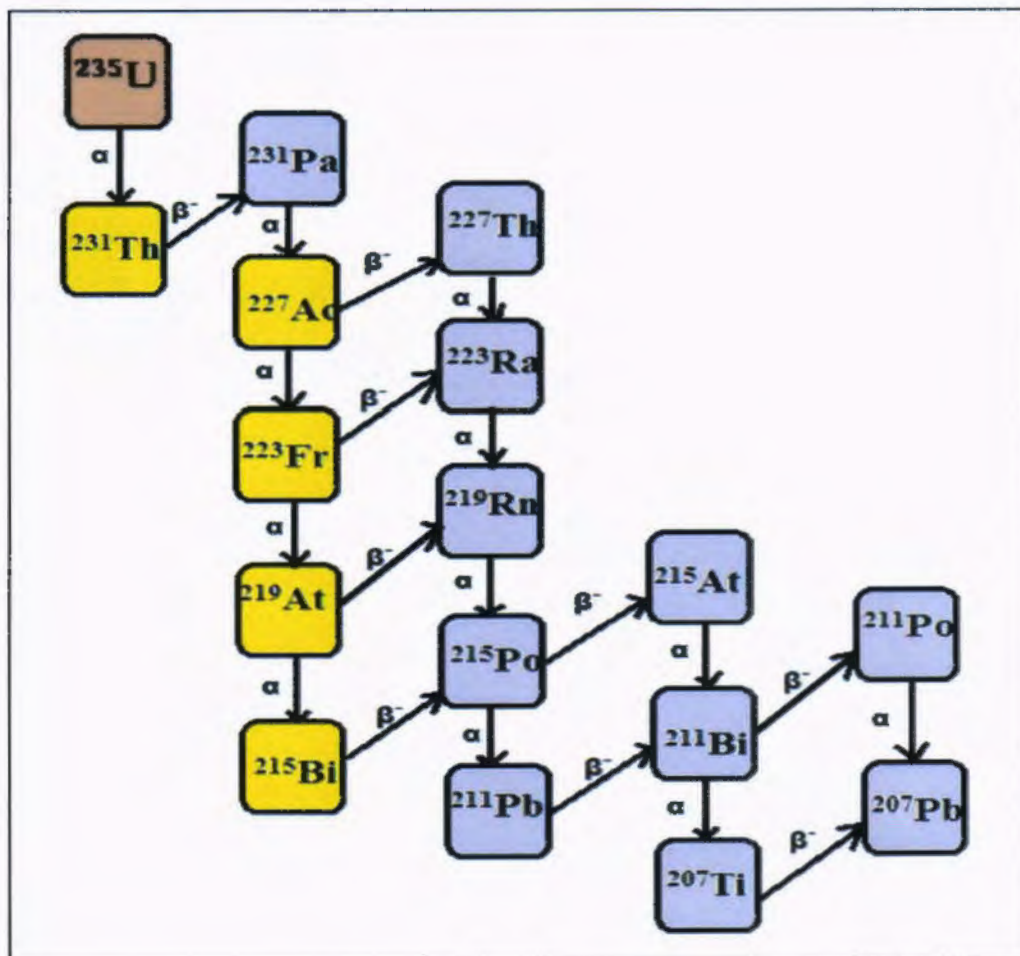


Figure 1-4.  $^{219}\text{Rn}$  production through the decay process of  $^{235}\text{U}$ .

Natural radioactivity has played a crucial role in the advance of the earth to the form and shape. The decay-series of thorium cascade to produce radioactive progenies such as Ra, Rn, and Po, but the eventual destiny of all members of these series is to end as a stable isotope of Pb [25]. Figure 1-5: Shows the decay process of  $^{232}\text{Th}$ . The decay process starts with naturally occurring thorium element. It decays into  $^{228}\text{Ra}$  through alpha decay process.  $^{228}\text{Ra}$  further decay into  $^{228}\text{Ac}$  through beta decay process. The decay products of  $^{228}\text{Ac}$  include  $^{228}\text{Th}$  and  $^{224}\text{Ra}$ . They are obtained through beta and alpha decay processes respectively. Now  $^{224}\text{Ra}$  undergo through

alpha decay process to produce  $^{220}\text{Rn}$  isotope which further decay into its progenies and finally the process ends by the production of  $^{208}\text{Pb}$  [23].

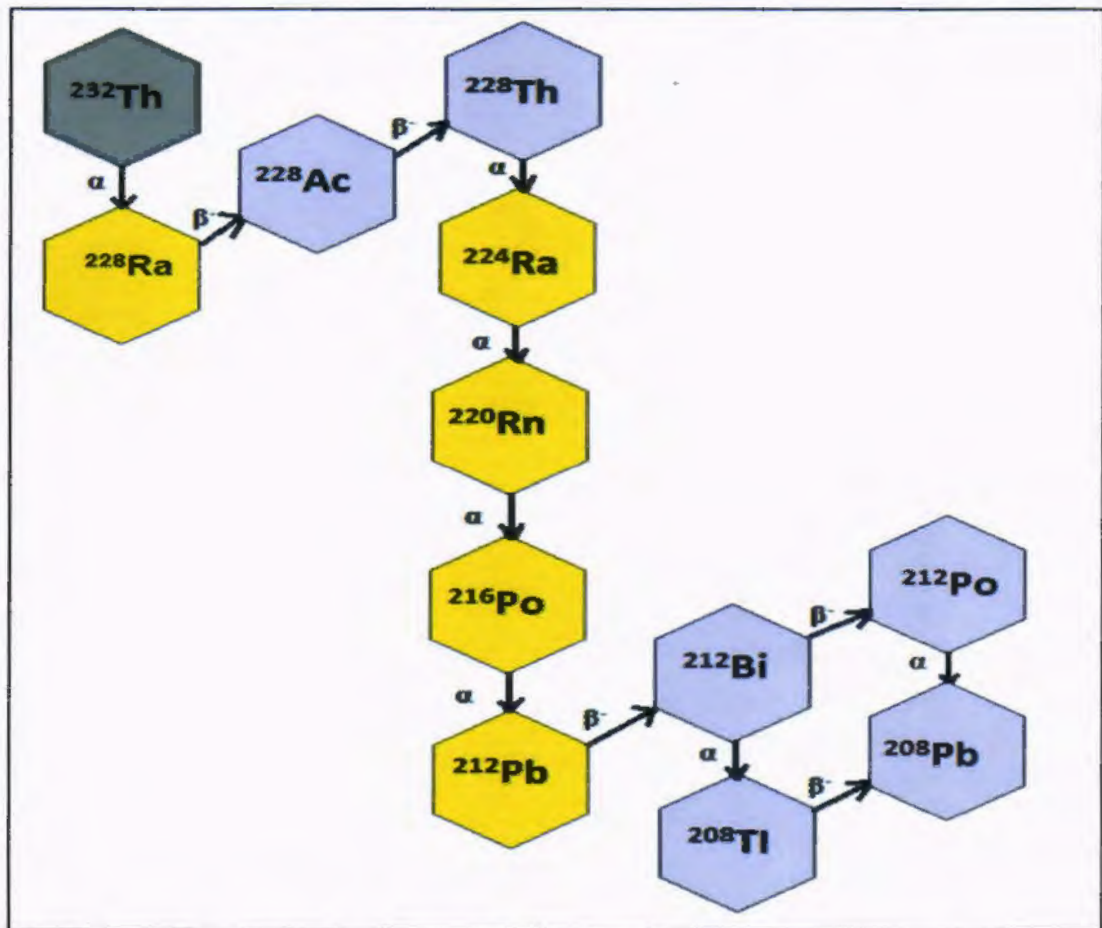


Figure 1-5.  $^{220}\text{Rn}$  production through decay process of  $^{232}\text{Th}$ .

### 1.6 Radon Decay Chain:

As declared before, the noble gas  $^{222}\text{Rn}$  produced in the uranium series can become airborne before decaying. Soil and rocks under houses are generally the main contributors to indoor radon which is typically four or five times more concentrated than radon outdoors, where greater air dilution occurs. Other contributions to indoor radon come from outside air, building materials, and the use of water and natural gas [26]. The radon gas obtained in the decay process of Uranium, also undergoes through two decay process forming short lived daughters  $^{218}\text{Po}$  with half-life of about 3.1 mint and  $^{214}\text{Pb}$  and so on, finally reaching to  $^{206}\text{Pb}$ . During the whole process, alpha and beta decay mechanisms are followed for the transformation process. The decay products from  $^{222}\text{Rn}$  to  $^{210}\text{Pb}$  are short lived. These short-lived decay products may be used for the measurement of radon concentration [27].

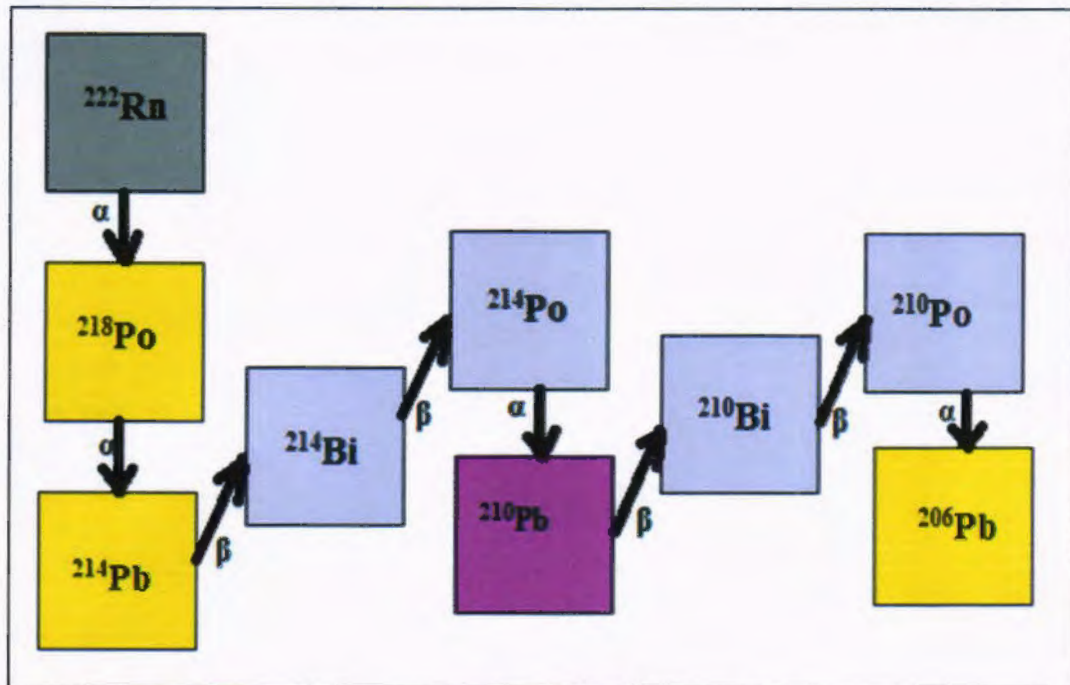


Figure 1-6. Decay chain of Radon and their products.

### 1.7 Radon as an Earthquake Precursor:

There are many factors causing an earthquake. They are volcanic activities, tectonic faults etc. These factors produce changes in the physical state of the earth crust. The changes produced in the physical properties of earth crust help in some precursory phenomenon for earthquake happening. The successful prediction of the earthquake with exact magnitude, time and location is still a challenging task for scientific community. There are many geophysical and geochemical phenomenon the scientists are using it as a precursory toll in an earthquake prediction. By precursory phenomenon we mean some geological, geophysical and geochemical phenomenon that happened before an earthquake [28]. Radon anomalies detection is also a potential method for the earthquake prediction. These anomalies are connected to geological faults produced by earthquakes [29]. Anomalies in radon concentration means sudden changes from the normal values before an earthquake [30]. Radon anomalies have been observed in earth crust before nearly happening earthquakes reported by seismically active regions including USSR, USA, Japan and China. The Kobe earthquake occurred on 17 January 1995, of magnitude 7.2 distracted badly this area. The population of this area is about 10 million. Five thousand peoples were died and 300,000 have lasted their homes. The radon concentration in a 17 m deep well and 30 m away from the hypocenter was reported to be 20 Bq. L<sup>-1</sup> and was stable at the end of 1993. This stable value started increase in the beginning of October 1994 and reached to 60 Bq. L<sup>-1</sup> at the end of November which is 3 times greater than

the previous year report. On 8<sup>th</sup> January 1995, a sudden increase was observed 9 days before the earthquake and a sudden decrease was observed on 10<sup>th</sup> January 1995, 7 days before the earthquake. These changes in concentration clearly indicate the precursory signal of radon preceding to an earthquake [11]. It was observed that it emanates from the minerals grain containing radium which is the daughter product of uranium <sup>238</sup>U series [31].

## 1.8 Radon Transport to surface from Earth Crust:

Radon emission from the grain is because of alpha recoil. Alpha recoil process conserved the law of momentum. During the emission of alpha particle of 86keV energy is emitted which pushes the radon nucleus and comes out from the soil grain. The recoiled radon nucleus transport from the inner surface of the earth to the outer surface follow two mechanism:

(a) Molecular diffusion process. (b) Advection of liquid [32].

Molecular diffusion process is simple process in which the molecules of a gas moves from high concentration region to the lower concentration regions the process is governed by Fick's first Law and is applicable for the radon transportation from less deep regions of the earth crust, while advection process takes place for the transportation from deeper regions of the earth crust [15]. Fick's first Law states that "The diffusion flux of a gas from high concentration to low concentration area is directly proportional to the concentration gradient" [33]. Mathematically Fick's law is given by the following equation:

$$J = -D \frac{\partial C}{\partial x} \quad (3)$$

Where J show the diffusion flux, D is the diffusion coefficient and  $\frac{\partial C}{\partial x}$  shows the concentration gradient of the gas.

Darcy's Law is a numerical equation which describe the flow of a fluid through a porous media. It was given by Henry Darcy and is totally based on experimental results. The experiments were performed for water through beds of sand [34]. Mathematically Darcy's Law is given by the following equation:

$$Q = -KA \frac{\Delta p}{\mu L} \quad (4)$$

Here Q show total discharge in m<sup>3</sup>/s, K is soil coefficient permeability in m<sup>2</sup>, A is cross-sectional area in m<sup>2</sup>, Δp is the change in pressure μ is the viscosity and L is the length in meter. After reaching to surface it diffuses to atmosphere with mutable ranges which can be monitored through different techniques [35,36]. The variations in the concentration depend mainly on earthquakes and meteorological parameters therefore it becomes difficult to say whether the anomaly was caused by seismic activity or by metrological parameters [37, 39]. For this



purpose, we must find methods for detecting the anomalies with the removal of metrological effects and before that we must see a relation between radon concentration and different parameters [15].

### **1.9 Metrological Effects on Radon Anomalies:**

Many researchers are dealing with the measurement of radon gas concentration in soil which gives a positive signal to volcanic activities [30]. Its transportation from its main source to the earth crust surface depend on many factors, such as distribution of uranium in soil and bedrocks, soil porosity and humidity, micro-cracks of bed rocks, rainfall, temperature pressure and surface wind [31]. The changes in these parameters cause a change in its concentration because they change the properties of the soil and rocks. Barometric pressure has been observed to have an inverse relation with the radon concentration in soil gas. A decrease in barometric pressure cause an increase in radon emission from the earth keeping the remaining parameters constant, while increases in barometric pressure cause decrease in the concentration because barometric pressure push the low radon content air into the ground [39,40,41].

Environmental temperature effects on radon have also been studied and it is found that environmental temperature effect its concentration in soil gas. Klusman and Jacks were the founder of this relation in 1987. They found that there is an inverse relationship between the environmental temperature and radon concentration. Its concentration decrease with the increase in temperature and increases with the decrease in temperature, this is because that when environmental temperature is high as in summer season then the soil temperature is lower and when environmental temperature is lower as in winter season than soil temperature is high. We know that heat flows from high temperature regions to lower temperature regions, therefore in summer season heat flow from soil to the environment is very low which cause a decrease in the its concentration. While in winter season soil temperature is higher than the environmental temperature therefor more heat flow from soil to the environment occurs causing an increase in its concentration. The transportation of radon from inner earth crust to outer surfacc occurs by diffusion and advection methods. These two processes occur through the pores and cracks in the earth crust, therefore soil moisture and rain-fall will also affect the concentration value as they effect the porosity of the earth crust. From these observed phenomena, it is clear, that making any prediction for earthquake through radon anomalies it is necessary to include all metrological parameters [42].

## 1.10 Detection Methods for Radon Anomalies:

The changes in radon concentration from an average value is defined as the anomaly. These anomalies mostly depend upon meteorological parameters, hydrological parameters and seismicity. Therefore, it is difficult to say that, either the anomalies are produced by a seismic event and meteorological parameters. For this purpose, statistical methods can be used for the data evaluation. Different researcher has used different methods to detect the anomalies produced in radon concentration [43]. The following section define different methods used for finding these anomalies.

### 1.10.1 Standard Deviation Method:

Standard deviation method is a common method used for finding the anomalies in radon concentration. In this method, the concentration can be measured in soil gas, groundwater and air, for the four seasons yearly in correspondence with the meteorological parameters. A straight line should be taken for the average value of radon and two other lines  $\pm 2\sigma$  above and below the standard line. These two lines can be taken as standard deviation for the average concentration. The concentration variation between the two lines show its relationship with the seasonal effects while the deviation from these lines may be due to the earthquake events. This type of radon anomalies was observed ten days before three earthquakes with magnitude 1.8, 3.2 and 2.2 at Krsko basin Slavonia [42].

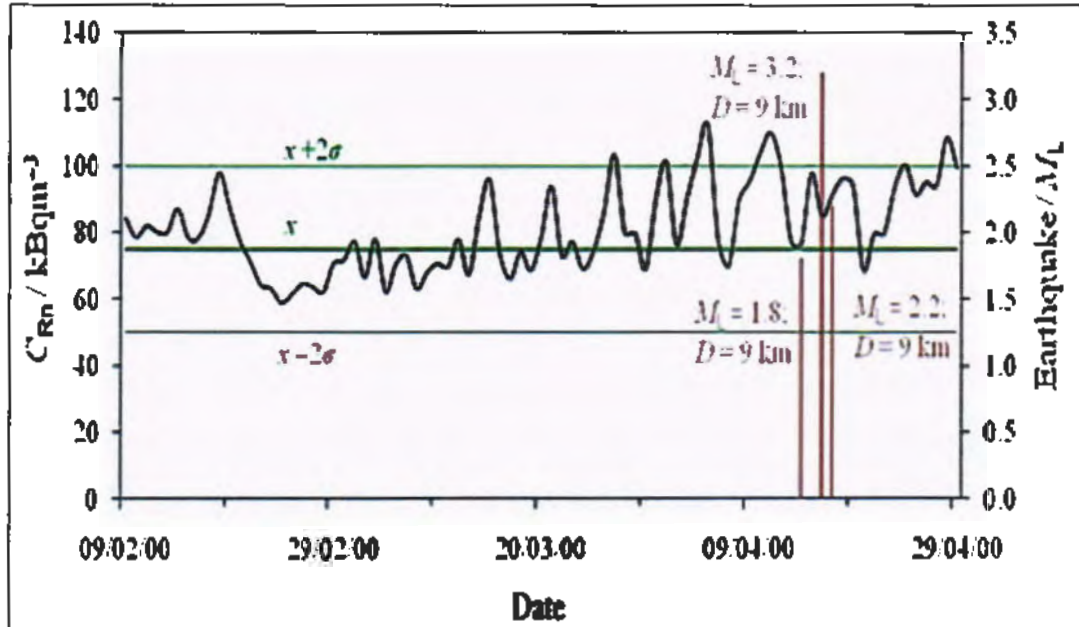


Figure 1-7. Statistical method for identification of radon anomaly for three earthquakes at Krsko Basin.

### 1.10.2 Relationship between Radon exhalation and barometric pressure:

The studies relating meteorological effects with radon concentration have shown that environmental pressure and temperature have an inverse relation with radon concentration. As environmental pressure decrease, the pressure in the earth crust will be greater so gases inside the earth crust will be pushed to the external environment and when the environmental pressure increases then air which contain low radon value will be pushed into the earth surface. If some where this phenomenon is violated, means the inverse relationship between pressure and radon concentration is disturbed this is known as anomaly in radon concentration. This phenomenon was observed 14 days before an earthquake of magnitude 2.6 at Krsko Basin Slavonia [44].

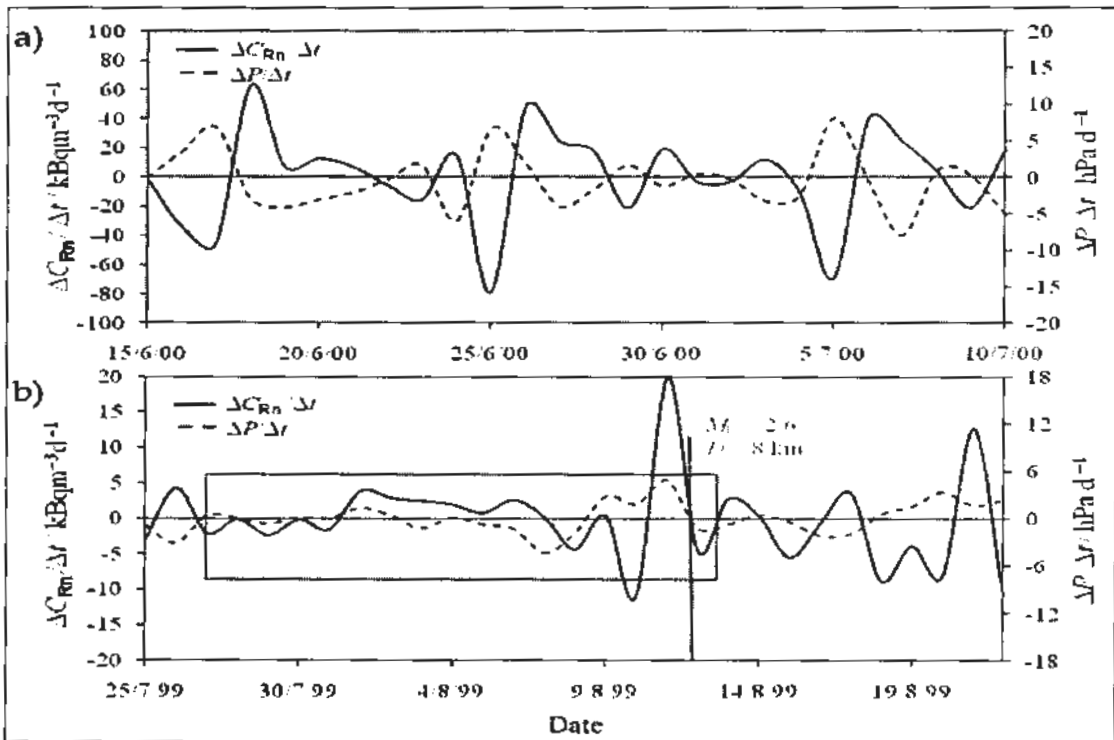


Figure 1-8. Time gradient of radon concentration with time gradient of barometric pressure without seismic events and during seismic event.

### 1.11 Radon as Health Hazard:

At least 445 million peoples are living in radon prone areas. These areas include those regions which are volcanically active. Volcanically active areas and geothermal areas are associated with the emission of gasses. These gasses include Carbon-dioxide ( $\text{CO}_2$ ), sulfur-dioxide ( $\text{SO}_2$ ), hydrochloric Acid (HCL), Hydrogen-fluoride (HF), hydrogen-sulfide ( $\text{HS}_2$ ), Carbon-monoxide (CO), radon and heavy metals like lead and mercury. Volcanic Eruption causes the emission of gasses due to which radon and its progenies present in the earth crust also comes out to the outer surface [45]. Around the whole world 50% of radiation received by people from natural source are due to radon therefore, it is a cause of lung cancer death after smoking which become 90-95 % in radon prone area [46]. The contribution of radon to death rate is about 0.8-12 %,

which is high then other sources like fires, transport accidents or flood [47]. As it is a radioactive gas, when undergoes through decay process it emits alpha radiations and transform into its daughter nuclei. The daughter nuclei also decay and emit alpha and gamma radiations of high energies. These radiations produce ionization in human cells in such a way that, they strike the biological cells in the lung which produce abnormality in the activity of human cell and cause damage in the DNA of cell [27]. Radon exist in air and water so when we breathe, this heaviest gas is inhaled and is therefore considered to be a major source of lung cancer after smoking. Studies related to indoor radon concentration and lung cancer around the universe have shown many cases of radon gas which produce lung cancer in public. A rough estimation for lung cancer due to radon is about 3-14 %. This is because of the major contribution of radon to natural radioactivity [11]. Human's lung cells receive beta radiation and alpha radiation from radon decay products. The receiving amount of beta radiations are smaller than alpha radiation [27].

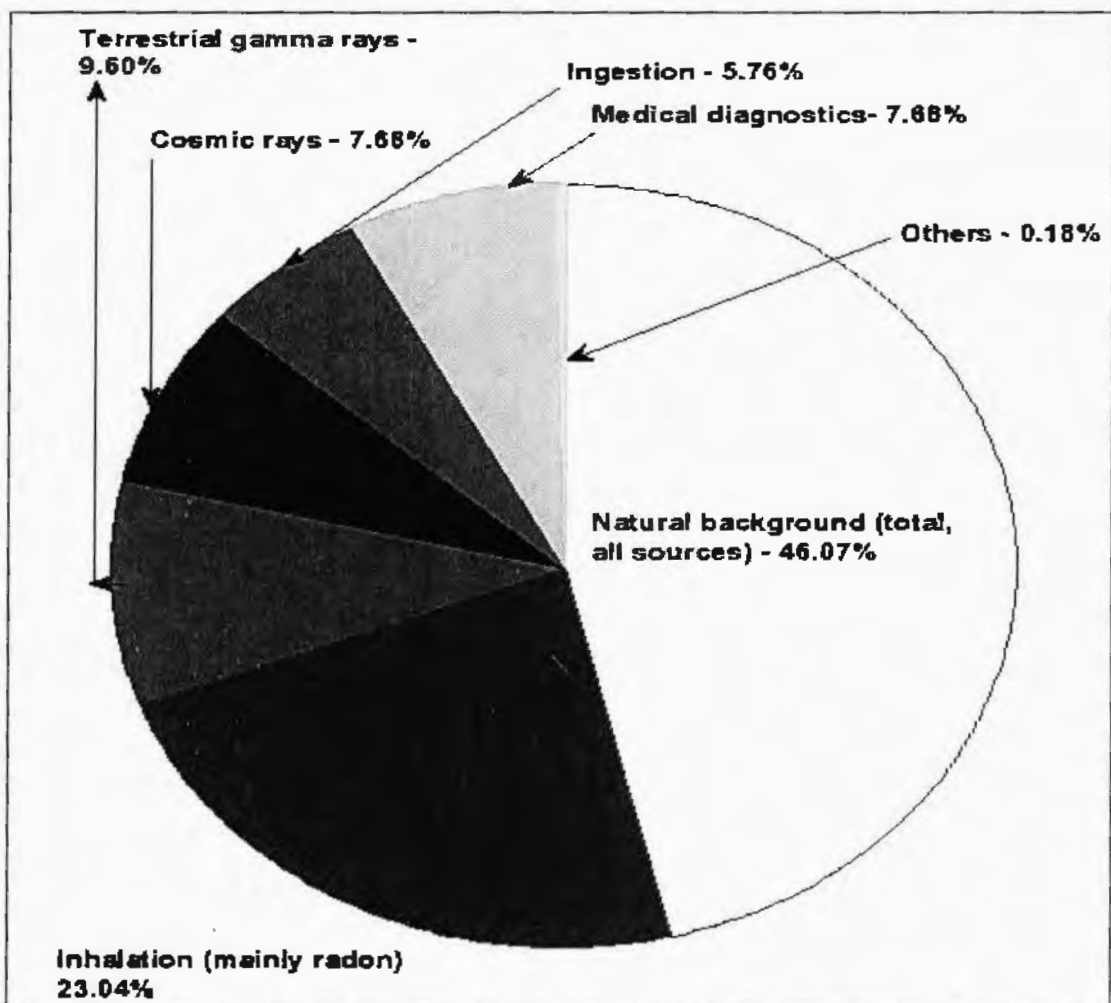


Figure 1-9. Radon dose received by human from different source.

## Chapter 02:

### 2.1 Literature review:

Radon is a heaviest radioactive element that is present everywhere in the environment therefore, effect human life in two different way.

- (a) Bad effects.
- (b) Good effects.

The study related to radon gas is for two purposes. First to find its bad effects and to find its good effects on human life. During the inhalation process it enters to the human lungs where it decays with the emission of alpha radiations. These alpha radiations when fall on cells produce lung cancer. On the other-hand measurement of radon gas which is a decay product of Uranium present in earth crust help in the determination of uranium prone areas and active tectonic zones, a precursor for the soil degassing and seismic events and an indicator for the geo-hydrological changes. This chapter include the previous studies related to radon as precursory tool. Many authors around the world have reported radon anomaly observed before earthquakes and is therefore considered to be a good tracer for the upcoming earthquakes. Here are some cases which will represent the desired work.

Radon gas have been used in Pakistan as precursory tool in geological studies and as a explorative tool for uranium sources [48]. Due to its radioactive nature, different areas have been investigated for its fatal effects on human life [49]. There are three regions of Pakistan and its surrounding that are seismically active; (a) Northern Pakistan (b) Hindu Kush (c) Southern Pakistan [50].

Jilani et al., (2017) reported the fluctuation of soil radon concentration at Sobra city of Khyber Pakhtun-Khuwa province of Pakistan. Sobra city is situated in the northern area of Pakistan. In 2005 an earthquake of magnitude 7.6 struck the northern areas of Pakistan in which at least 68,000 peoples were died and 69,000 were injured. He uses active method for the measurement purpose by the help of continuous radon/thoron monitor RTM2200. It consists of an internal vacuum pump which is connected to a PVC pipe 3 cm in diameter and 3feet lengthy. The vacuum pump suck the air from pipe and send it to the ionization chamber. Ionization chamber detect the polonium ion produced in the decay process of radon. The number of detected polonium ion is proportional to radon concentration [51]. The monitor provides meteorological parameters as well therefore, the resultant values may be correlated to these parameters [52]. The observed values of radon vary from 1052 kBq.m<sup>-3</sup> to 22,874 kBq.m<sup>-3</sup>. Most values were near to 3252 kBq.m<sup>-3</sup>. Important time-based variations have been observed in radon

concentration which show positive relation to air pressure and relative humidity and negative relation to environmental temperature.  $2\sigma$  upper control limit was used to detect the radon anomaly on monthly basis. The overall data was observed with increasing trend. Now five earthquakes were selected from the earthquake catalog for the same period where only two of them were related with the radon anomalies. The anomalies depend on the magnitude of earthquake and distance between earthquake preparation zone and monitoring station. The events occurred on 13<sup>th</sup> September and 14<sup>th</sup> October 2014 respectively with equal magnitudes. Some other large peaks were also observed in last two months of the study period which were not related to any earthquake but may be due to some other geological and environmental events [53].

Iqbal et al., (2010) measure indoor radon concentration and outdoor gamma dose rate in Dhirkot area of Azad Jammu and Kashmir Pakistan. The selected area was splintered by a strong earthquake in 2005. This splintered structure provide path to the movement of radon gas to outer surface of the earth. The radon concentration in dwelling ranges from  $36\pm 14$  Bq.m<sup>-3</sup> to  $195\pm 27$  Bq.m<sup>-3</sup> with mean value of  $121\pm 26$  Bq.m<sup>-3</sup>. The mean radon concentration in pukka, semi-kucha and kucha houses were  $124\pm 25$  Bq.m<sup>-3</sup>,  $143\pm 25$  Bq.m<sup>-3</sup> and  $136\pm 26$  Bq.m<sup>-3</sup> respectively. The outdoor gamma dose rates were  $74.0\pm 1.4$  to  $113.1\pm 2.4$  nGy/h and  $69.6\pm 2.2$  to  $108.8\pm 1.4$  nGy/h respectively. The mean dose rate  $91.3\pm 14.3$  nGy/h. These measurements were taken in dwellings and basements. The gamma dose rate was lower and higher at lower and higher altitudes respectively. The higher mean values of radon concentration were due to the dearth of suitable ventilation system in houses, humidity and temperature, cracks formed in houses, the geology of the area and aggregates. The gamma dose rate depends on sand stone and clay lithology, altitude and radionuclide contribution. The study mention that mean radon concentration was in acceptable range ( $200$  Bq.m<sup>-3</sup>, ICRP,1993; UNSCEAR, 1993) [54].

Winkler et al., (2000) study the relation of seasonal variation and spatial heterogeneity with temporal variability of radon concentration from 1996 to 1999 at nine different regions with a tested field of area  $20\text{m}\times 20\text{m}$  and four plots with area  $1\text{m}\times 1\text{m}$  at Neugherberg Germany. At both regions, the sample was collected from 0.5m and 1m depth through evacuated scintillation cells weekly. The spatial variability was described by coefficient of variation (CV). It was observed that the coefficient of variation was different for samples taken from different depth. There was 26 % coefficient of variation for 0.5 m depth and 13 % for 1 m depth for  $20\text{m}\times 20\text{m}$  region and for  $1\text{m}\times 1\text{m}$  region the coefficient of variation was 4 % for 0.5 m depth and 2 % for 1 m depth. Time series analysis was applied for soil radon data and seasonal variations in this

data were observed. The data was maximum in winter season with range from 6 kBq.m<sup>-3</sup> to 50 kBq.m<sup>-3</sup> for 0.5 m and for 1 m depth the values range from 8 kBq.m<sup>-3</sup> to 3 kBq.m<sup>-3</sup>. These observations show that the concentration was greater at 0.5 m depth as compared to 1m depth [55]. Active and passive methods were used for monitoring purpose of radon concentration. Samples collected by evacuated scintillation cell was taken as reference. Different methods were used for the collection of samples for the same region and was related to the first observations that was similar with a little uncertainty. Another observation was taken at another field for greater impermeable soil and the results were compare with the first one. A variation about twice was observed from the first observations which the dependence of radon concentration on spatial heterogeneity and seasonal variations [56]. It has been shown in several studies that meteorological parameters influence soil radon concentration [57]. Interesting results have been observed for radon concentration in borehole air and water with meteorological parameters [58]. Correlation between soil radon concentration and atmospheric temperature, pressure, rainfall and wind have also been investigated with meaningful results [59].

Iveta et al., (2006) selected Mudra site of Hungary for radon concentration measurement in borehole air. Short term and seasonal variation in radon activity concentration in bore-hole air was observed. Seasonal changes in radon activity concentration in bore-hole air were associated with atmospheric temperature changes, as well as with soil air temperature difference. Short term radon variations in bore-hole air were negatively correlated with atmospheric pressure. Seasonal radon changes in bore-hole water was not unambiguously confirmed. In comparison with bore-hole air, the radon activity concentration measured in bore-hole water was three to four times lower and it was positively correlated with the state of water level in bore-hole. Rainfall was found to be important parameter influencing radon concentration in both studied environments. Short term variations in radon activity concentration in bore-hole water appeared to be more related with rainfall as in bore-hole air. Intensive rainfall preceded every radon concentration increase in bore-hole water, while in bore-hole air this effect appeared in spring and summer seasons exclusively. As the results indicate, during spring and summer months both bore-hole air and water were not influenced by additional interaction with surrounding environment. After an increase in spring and summer month from April to September, radon activity concentration in both environment decreased in accordance with radioactive decay law. From October to March, in bore-hole water the calculated radioactivity decay was much faster than the actual observed decrease.

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The higher radon concentration in bore-hole water maintained for longer time due to the horizontal movement of water. On the contrary, in bore-hole air the measured radon concentration decreased faster in comparison with calculated radon course, probably also as a result of bore-hole ventilation caused by the meteorological condition changes [60].

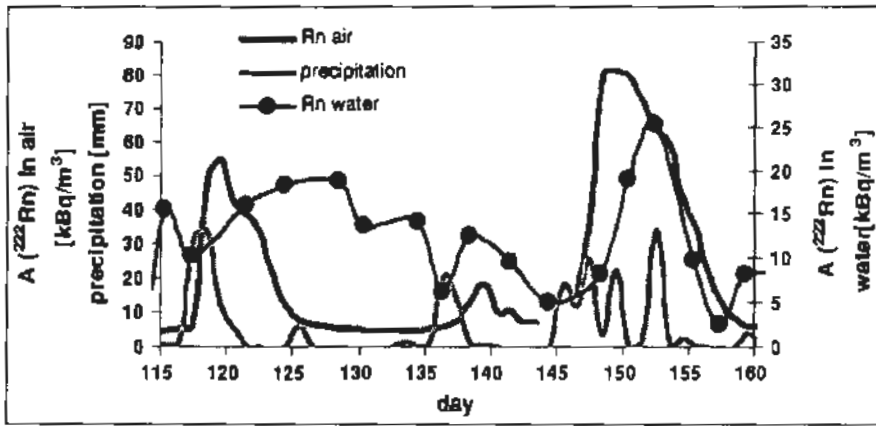


Figure 2-1. Comparison between radon concentration in bore-hole air and water.

Radon concentration in groundwater depend upon the uranium contain in earth crust [61]. The groundwater radon concentration depends upon many factors like seismic activities, hydrological and meteorological condition.

Yalim et al., (2012) investigated radon concentration in groundwater from August 2009 to December 2010 for finding a correlation between the radon anomalies and the earthquake. The water samples were taken from 4 wells on the Akasehir-Samav fault system (ASFS). They were collected from mid-west Anatolia region that is 10 km to 30 km wide and 550 km long. Through 250 mL bottles that was tightly capped to prevent radon leakage. Alpha GURAD PQ2000PRO detector was used for measurement purpose which an ionizing chamber and use alpha spectrometric technique for the measurement of radon data. The obtained data was then analyzed through expert software. A decrease in radon concentration was observed from the mean value that was followed by two future happing earthquakes of magnitude 2.6 m and 3.9 m. It was suggested that by doing continuous radon monitoring in ground water can help-out in finding the possible large earthquake [62].

Table 2-1. Specifications of water well with mean and deviated values of radon concentration.

Well no	Latitude	longitude	Altitude(m)	Depth(m)	Mean Value (Bq/L)	Mean Deviation (Bq/L)
1	38.7110°	30.6055°	1008	47	18.42	4.61
2	38.5373°	30.9186°	988	50	13.46	1.97
3	38.6020°	30.9385°	998	85	8.58	1.96
4	38.5799°	31.2251°	891	200	4.59	0.85

Turkey is famous for its high volcanic activities because it is situated at Mediterranean earthquake plate which is generated by the collision of African and Eurasian plate. There are three elements that generate the tectonics of Turkey. (a) The Aegean- Cyprian Arc, a convergent plate to south. (b) The dextral North Anatolian fault system plate to the North. (c) The sinistral East Anatolian fault system [63, 64,65]. North Anatolian fault system (NAFS) is known for strike-slip fault due to volcanic activities [65, 66]. East Anatolian fault system (EAFS) is known for series of parallel to subparallel fault zones. Between 1939 and 1999 along (NAFS) a series of nine large earthquakes occurred, but along (EAFS) for about 130 years no big earthquake occurred therefore, this site will cause a serious of destructive large earthquake [66]. Inceon et al., (2006) measured soil radon concentration along North Anatolian fault system (NAFS) and East Anatolian fault system (EAFS) by the help of plastic detector CR-39 type. The detectors were planted for a duration of three weeks. The detectors were 2 cm<sup>2</sup> in size. They were placed at 5-10 m away from each other. The concentration near the NAFS and EAFS varies from 4.3 kBq.m<sup>-3</sup> to 9.8 kBq.m<sup>-3</sup>. The mean values were 8.4 kBq.m<sup>-3</sup> and 6.5 kBq.m<sup>-3</sup> along NAFS and EAFS respectively. This difference was due to the main rocks and bedrocks sites that were under observation [67]. Greater values of radon concentration along NAFS was observed and after six months of the observation four earthquakes of magnitudes 5.7-5.9 occurred at this region. Which show greater seismic activity along NAFS. The changes along fault lines were correlated to the variation in the characteristics of rocks along fault lines. The mean value changed from 6.36±1.10 kBq.m<sup>-3</sup> and 6.29± 1.04 kBq.m<sup>-3</sup> at EASF and long NAFS the mean value change was 7.73±1.24 kBq.m<sup>-3</sup>. This show that radon concentration level become higher near fault lines which can help in finding active fault zones [68]. Fujiyoshi et al., (2006) measured variability in radon concentration from November 2004 to March, 2005 also from December, 2005 to March, 2006 in correspondence with meteorological parameters at Hokkaido University Sapporo by the help of continuous radon monitor Barasol detector [69]. A sudden increase was observed in soil radon with a nearly happened large earthquake on 26 September 2003 therefore the present study was carried out to find its relation with the other factors as well. It was observed that temperature was active toward the radon concentration variability. One degree rise in soil temperature causes 740 Bq.m<sup>-3</sup> increase in the activity. The concentration level was greater in day time and was lower at night. After that a sudden decrease in the level was observed due to the change in the barometric pressure, due to an irregular rain. The radon concentration start falling from November 2004 to December 2004 and the level became very low when Sapporo city was completely covered with snow. Some Peaks were

observed on time series chart in the winter season and spring season that were considered to be due to the seismic activities. These values were observed on 7<sup>th</sup>-9<sup>th</sup> December and 22<sup>th</sup>-25<sup>th</sup> December 2004. The values changed from 2000 Bq.m<sup>-3</sup> to 6000 Bq.m<sup>-3</sup>. The same value decreased to 2500 Bq.m<sup>-3</sup> on next day. In some cases, increase was observed due to increase in pressure. This increase in pressure was considered, to be due to the volcanic activities. The measured values changed from zero to 140 mBq.m<sup>-2</sup>.s<sup>-1</sup> with an average value 37±22 mBq.m<sup>-2</sup>.s<sup>-1</sup>. This lower value was in winter as the earth surfaces was covered by snow [69].

Japan is located at a region of high volcanic activities that is way mostly earthquakes happen over there. A large earthquake of magnitude 7.2 was happened on 17<sup>th</sup> January 1995. Igarashi et al., (1995) reported that radon concentration was normal in the last days of 1993 it was 20 kBq.L<sup>-1</sup>. The data was taken from 17 m deep well which was located at 30 km north to the hypocenter of the earthquake. In the start of October, 1994 the concentration started increase and reached to 60 kBq.L<sup>-1</sup>, that was about three times the normal value. After that a sudden increase in the concentration was observed before nine days of the earthquake. This increase in concentration fall 7 days before the earthquake event on 22<sup>th</sup> January, 1995 the radon level again became to the same level observed in October 1994. This anomaly was observed 3 months before the earthquake. It was correlated to the data obtained for the water level of the Ozu-Ohima earthquake and was suggested to have the ability to use it as a precursory phenomenon [70].

Al-Hilal et al., (1998) observed radon gas concentration in ground water for two years at monthly period in western Syria during 1993 and 1994. They selected the monitoring station along Dead Sea Fault zone because of the occurrences of large earthquake ( $M \geq 6.5$ ) [71]. Second reason was some distinct geo-morphological and geological indication of dynamic neotectonics of this area [72]. The mean values of radon were 115 PCi.L<sup>-1</sup> and 145 PCi.L<sup>-1</sup> for Serghaya and Bouqeah station respectively. Mean value plus two standard deviation method ( $X+2SD$ ) was used for the normal values and any value crossing this limit was considered as the anomaly. In the Figure2-7 two to three times limit crossing peaks were observed. At Serghaya station radon increase was observed on 5<sup>th</sup> July 1993 with a peak value of 110% above the mean value crossing the limit ( $X+2SD$ ). This increase in concentration was followed by two earthquakes of  $M=2.2$  and  $M=4.5$  occurred on 23<sup>th</sup> July 1993 and 30<sup>th</sup> July 1993. The epicentral distance of these events were 67 km and 47 km respectively. Another increase was observed on 8<sup>th</sup> November 1993 with a peak value of 126 % followed by an earthquake of magnitude 2.4 on 12<sup>th</sup> November 1993. The epicentral distance of the event was 82 km.

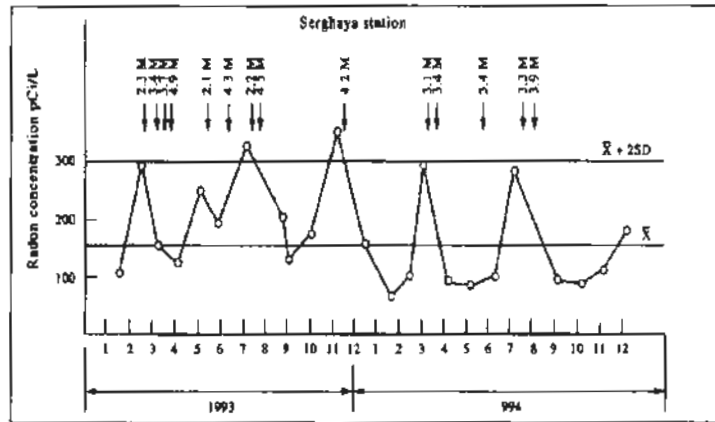


Figure 2-2. Groundwater radon concentration obtained at Serghaya.

Sharp radon concentration values at Bouqeah station were also detected on 4<sup>th</sup> March 1993 and it was followed by two earthquakes of  $M=4.1$  and  $M=4.9$  occurred on 10<sup>th</sup> and 22<sup>th</sup> March 1993. The epicentral distance of the events were 34 km and 160 km. Another increasing trend in radon concentration was observed in August 1994, followed by two earthquakes of  $M=3.0$  and  $M=2.2$  occurred on 11<sup>th</sup> and 13<sup>th</sup> September 1994. The epicenters were  $35.83^\circ$  N,  $36.08^\circ$  E and  $33.86^\circ$  N,  $36.05^\circ$  E respectively.

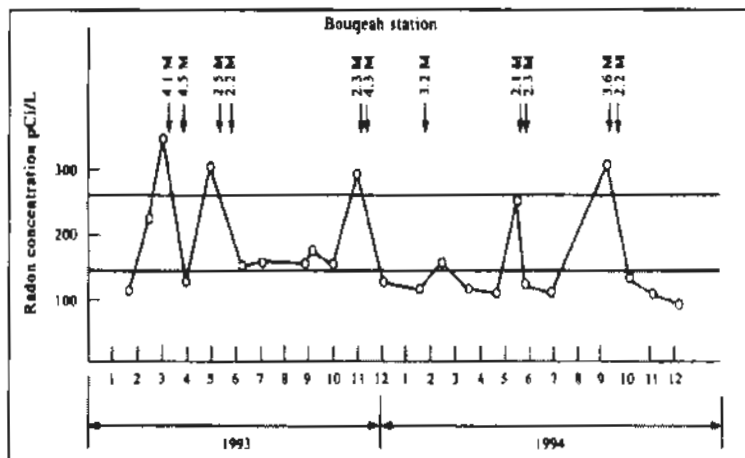


Figure 2-3. Groundwater radon concentration obtained at Bouqeah

These anomalies may be due to the strain produced in the earth crust showing probable relation with the seismic activities. Thus, the abnormal increase in radon can be used as possible indicator for the tectonic disturbance [7].

King, (1978) performed continuous radon monitoring along two seismically active regions, the California and San-Andreas. An area of  $200,000 \text{ km}^2$  was instrumented for the measurement purpose. He started observations from 7<sup>th</sup> May 1975 along San-Andreas and Calaveras fault of California. During the whole period of two years it was observed that the mean values changed weekly from  $0.19 \text{ Bq.L}^{-1}$  to  $85 \text{ Bq.L}^{-1}$  ( $5\text{-}2 \text{ pCi.L}^{-1}$ ) and two earthquakes of  $M=4.3$  and  $M=4$  occurred near to the time when radon got its highest value. The samples were collected from 0.7 m depth of the earth crust. Several peaks of radon that were reported

before an earthquake of magnitude 4.3, were assigned to be due to the force inside the earth crust build due to strain. The whole data was taken from 20 superficial holes [73].

Elsayed et al., (2009) observed the behavior of radon concentration for finding a correlation with the earthquake magnitude, time and location by comparing radon anomalies that rise due to tectonic events. The method used by the author was to observe the variation in the ratio of radon daughter concentration  $[^{219}\text{Rn}]_x$  and radon concentration  $[^{222}\text{Rn}]_y$ . The subscript x and y was used for  $^{219}\text{Rn}$  and  $^{222}\text{Rn}$  respectively. It was observed that during normal situations the ratio between  $[\text{Rn}]_x$  and  $[\text{Rn}]_y$  was approximately constant and is independent of temperature, pressure, wind speed, humidity etc. The life time of  $^{219}\text{Rn}$  is smaller than the life time of  $^{222}\text{Rn}$  therefore, only  $^{222}\text{Rn}$  escape from fracture zones. This ratio decreases rapidly when fault occurs in the earth crust. This decrease is due to the increase in the concentration of  $^{222}\text{Rn}$  and this variation in radon concentration can be used as radon anomaly. The emission of radon gas depends upon the magnitude of the earthquake, so the change in the ratio of  $[^{219}\text{Rn}]$  and  $[^{222}\text{Rn}]$  is due to the earthquake which can be taken as measure of the earthquake power. Thus, it is clear that continuous monitoring of radon gas concentration and its daughter is useful in earthquake prediction [74].

Miklavcic et al., (2008) investigated two different sites of Croatia the Osijek and Kasina for Radon concentration for different periods by different techniques. The Osijek area was investigated for a period of four years by the help of passive method. The second site Kasina was investigated for a period of two years by the help of Barasol semiconductor detector which is an active method. The active measurement technique was carried out together with meteorological parameters. During a period of four years at Osijek 49 % of the earthquake or 39 earthquakes were indicated by radon anomalies. These earthquakes were of magnitude ( $M \geq 2.6$ ) with epicenter distance  $R < 200$  km. At Kasina, the two years' period of measurement of radon help-out the detection of 33 earthquakes or 30 % of the earthquakes of magnitude ( $M \geq 2.5$ ) with epicenter distance  $R < 200$  km [75].

Planinic et al., (2001) measured radon concentration through passive method technique by the help of plastic detector LR-115 type at Osijek Croatia. They observed changes in radon concentration in correspondence with barometric pressure, environmental temperature and precipitation. The measurements were made for a period of two years. Standard deviation method was used for the detection of anomaly. In May 1999 radon concentration was observed that was  $26.7 \text{ Bq.m}^{-3}$  these values were compared with the calculated value  $20.4 \text{ Bq.m}^{-3}$ . The difference between the values was 6.3 which is greater than the value of  $2s$ . Where  $s$  is a scale

on the deviation graph for radon concentration and was equal to  $3.1 \text{ Bq.m}^{-3}$  in July 1999, a decrease in the radon concentration was observed. The observed value was  $8.6 \text{ Bq.m}^{-3}$  and calculated value was  $22.3 \text{ Bq.m}^{-3}$  and this difference was  $13.7 \text{ Bq.m}^{-3}$  which was also greater than  $2s$  value. This decrease was not related to any earthquake but have a negative relation with temperature and no relation was observed between radon anomaly and barometric pressure during the study period. During two-year period six earthquakes of  $M=2.7$  to  $M=3.8$  was detected in relation with the variation in radon concentration in soil [6].

Virk and Baljinder., (1993) measured the radon concentration anomalies in ground water and soil gas due to earthquake phenomena near Kangra valley, Himachal Pradesh, India. The selected site is located near the main boundary fault of Himalayas. The measurement was done by both active and passive methods. Active method was carried out by the help of  $\text{ZnS(Ag)}$  scintillator and passive method was done by LR-115 type SSNTDs [76]. Eleven earthquake related anomalies have been recorded since August 1989 in both soil gas and ground water. These anomalies were also correlated to meteorological parameters to found the genuine anomalies due to earthquake. The most anomalies were detected to be due to the earthquake rather than the changes in meteorological parameters [77].

Radon concentration measurement along active fault show a positive signal to be a tracer for soil degassing. This is because of high porosity of fault regions. As the active fault lines are greater porous media than the surrounding, therefore the gasses inside earth crust comes out to the surface level with greater amount and this brings up radon and thoron from sub-surface to surface level causing an increase in radon concentration [78]. Burton et al., (2004) started radon observation in 2002 along fault lines and observed different values of radon at different places. For example, at 3000 m at Pernicana fault the observed radon concentration reached to  $10,000 \text{ Bq.m}^{-3}$ . At Torr del Filosofo at 2920 meter  $300 \text{ Bq.m}^{-3}$  to  $9000 \text{ Bq.m}^{-3}$ . These measurements were done by continuous radon/thoron monitor RAD7. The measurements were taken for two different periods. First period started from 1<sup>st</sup> October 2002 to 2003. This measurement was disturbed by an earthquake of  $M=4.5$  occurred on 29<sup>th</sup> October 2002. This earthquake was registered along three faults at NW-SE and Pernicana fault. Second period started from 6<sup>th</sup> February 2003 after the Etean eruption by a lower spatial resolution over an area of  $16 \text{ km}^2$ , the observed radon concentration was greater than the measurements before eruption [79].

Jaishi et al., (2014) studied radon release from the soil gas using passive method by the help of plastic detector LR-115 type film. The study was carried out from March 2012 to February

2013 at Mat Bridge (23°18' N, 92°48' E) along Mat Fault in Serchhip district, Mizoram (India). Variations in radon concentrations have been detected. Effects of meteorological parameters on radon release were also calculated. The measured radon data shows a reasonable optimistic relationship with relative humidity but no precise relation with air temperature and rainfall. Data obtained have been correlated to the earthquakes that occurred around the measuring sites. The mean measured value at Mat fault was 243.2 Tracks.cm<sup>-2</sup> with a standard deviation of 152.2 Tracks.cm<sup>-2</sup>. The first increase was observed in between 135<sup>th</sup> and 250<sup>th</sup> days of the measuring period before an earthquake of M=6.0 occurred on 151<sup>th</sup> day at 147 km away from the monitoring site. Second anomalous increase in radon concentration was observed in between 195<sup>th</sup> and 210<sup>th</sup> days of the measurement. This anomalous change may be due to an earthquake of M=6.7 occurred at 328 km distance on 256<sup>th</sup> day of the measurement. A third increase in the concentration was detected between 300<sup>th</sup> and 315<sup>th</sup> day of the measurement period. This increase occurred on the same day when an earthquake of M=5.9 occurred at 315 km away from the monitoring site. Another earthquake of M=5.2 was also recorded on 367<sup>th</sup> day 174 km away from the monitoring site. The third anomaly was concerned to be due to the combine effect of these two events without any meteorological effect. These anomalies are connected to crustal deformation of the region and therefore increased before the earthquake [80].

Swakon et al., (2005) correlate radon concentration with the geological fault to find the radon risk area. The geological data of Krakow region of Poland was available therefore, it was selected for the current study. He uses active and passive methods for the measurement of the concentration of radon and thoron. Active method was carried out by the help of Alpha GUARD PQ2000 PRO radon monitor and passive method was done by the help of diffusion chamber containing CR-39 detectors [81]. The mean value of radon from active method was 39 kBq.m<sup>-3</sup> with median value of 35.5 kBq.m<sup>-3</sup> and the mean value of radon from passive method was 35.8 kBq.m<sup>-3</sup> with median value of 333.8 kBq.m<sup>-3</sup>. The highest value reached to 89 kBq.m<sup>-3</sup>. For the same region, the thoron concentration was also determined that was 10.8 kBq.m<sup>-3</sup> with median value of 11.8 kBq.m<sup>-3</sup>. Now four other experiments were performed by the author in different geological structure area. The results were 8 kBq.m<sup>-3</sup> for radon and 3 kBq.m<sup>-3</sup> for thoron. Thus, it was obtained that the results from the study area were three times greater than the results of different sites. Therefore, correlation between radon concentration and geological structure of bed rock help in finding radon risk areas [82].

Peceaneaga-Cemena fault system (PSF) is seismically active fault on Romanian territory along NW-SE and show the northern limit of Moesian plat-form [83]. Cosma et al., (2014) made the first indoor radon concentration measurement for Stei-Baita area by the help of CR-39 and compared with annual values of other areas [84]. The etching condition for CR-39 was 6.25 molar NaOH aqueous solution and etching temperature was 90 °C the time taken for this process was 4.5 to 6 hours [85]. These measurements were done at 335 dwelling areas at a different height from the floor [86]. Average value for Peceaneaga-Cemena fault was 247 Bq.m<sup>-3</sup> which is higher than 124 Bq.m<sup>-3</sup>, the annual average value of Transylvanian and 129 Bq.m<sup>-3</sup> the annual average value of Bihor area, the reason for this higher value is the use of radioactive materials in the construction of buildings and dwelling [87]. . The average value of indoor radon concentration in this area varies from 10 Bq.m<sup>-3</sup> to 3653 Bq.m<sup>-3</sup>. After seasonal corrections, the average value was 343.5 Bq.m<sup>-3</sup> which is four times than the average value of indoor radon concentration at Transylvania. A reported value of average indoor radon concentration for Transylvania, is 82.5 Bq.m<sup>-3</sup> [84, 88]. Due to this high concentration 25 % of lung cancer death for nonsmoker and 50 % for smoker were estimated [89]. Second measurement of radon was made for finding the active fault. This measurement was taken at a village known as Fantana Mare [90, 91]. LUK3C Radon detector was used for the soil gas samples. The total time for every measurement was 10 mints and 50 measurements were recorded in which the values vary from 8 Bq.m<sup>-3</sup> to 50.3 Bq.m<sup>-3</sup>. The values above 50 was taken as radon anomaly. The data was distributed in three groups PA, PB and PC with maximum value. For PA, the maximum concentration value was 50.2 kBq.m<sup>-3</sup> for PB group the maximum value was 48.49 kBq.m<sup>-3</sup> and for PC group the data was 31.5 kBq.m<sup>-3</sup>. The maximum concentration for PA, PB and PC ranges between 20 kBq.m<sup>-3</sup> to 50 kBq.m<sup>-3</sup> therefor, it was taken to be the rang of radon anomaly in soil along a fault lines [92].

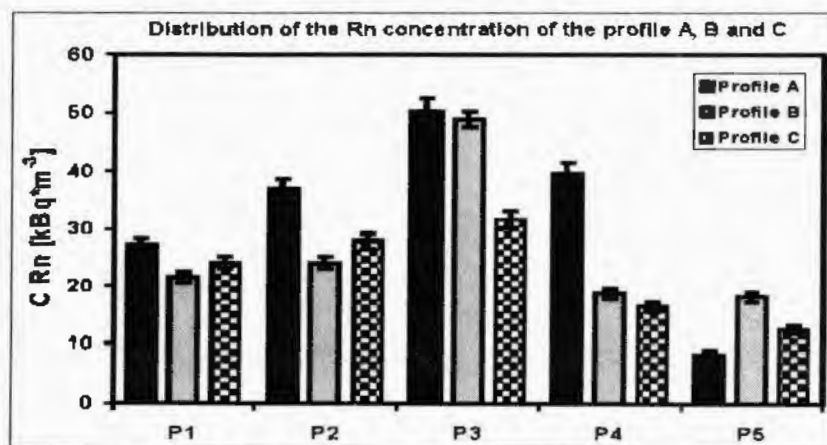


Figure 2-4. Radon concentration taken at three different sites at Romania.



## **Chapter 03:**

### **3.1 Measurement Techniques:**

Radon measurement in different environments show optimistic signal in geological, geophysical, geochemical, geo-hydrological studies and can be used for the Uranium exploration. therefore, it is important to make precise measurement techniques for radon concentration. There are two types of techniques that can be used for the measurement of radon gas concentration. These techniques are:

- (a) Active technique
- (b) Passive technique.

Active technique of measurement includes continuous radon/thoron monitors RTM2200, RTM2100 RTM1688-2, RAD-7 and Barasol detector etc. While Passive method of measurement includes, the uses of solid state nuclear tracks detectors (SSNTDs), Luca cell and high purity germanium detector etc. This chapter include the experimental techniques and measuring methods used for radon concentration and its decay products for the current study.

### **3.2 Selection of sites and with Geology:**

Most earthquakes are concentrated near the plate boundaries and that earthquake epicenters within the plate are scarce. This theory is also known as elastic rebound theory and successfully explains the formation of large geological features, mountain ranges and faults resulting from plate movement. The earth crust contained uranium that is the source of radon gas. It comes out from the inner surface of the earth to outer surface at different rate. Those areas that are lying along or near a fault zone are observed to have high emission rate as compared to those regions that are away from fault lines. Pakistan is situated on north-west section of the Indian plate that pushes into the Eurasian plate. The selected area, Islamabad lie along the fault line known as Main Boundary Thrust (MBT). It is the capital of Pakistan and is located at a lower latitude. It consists of plain areas and is situated at relatively low altitude (33° 38' N, 73° 09' E,) [93].

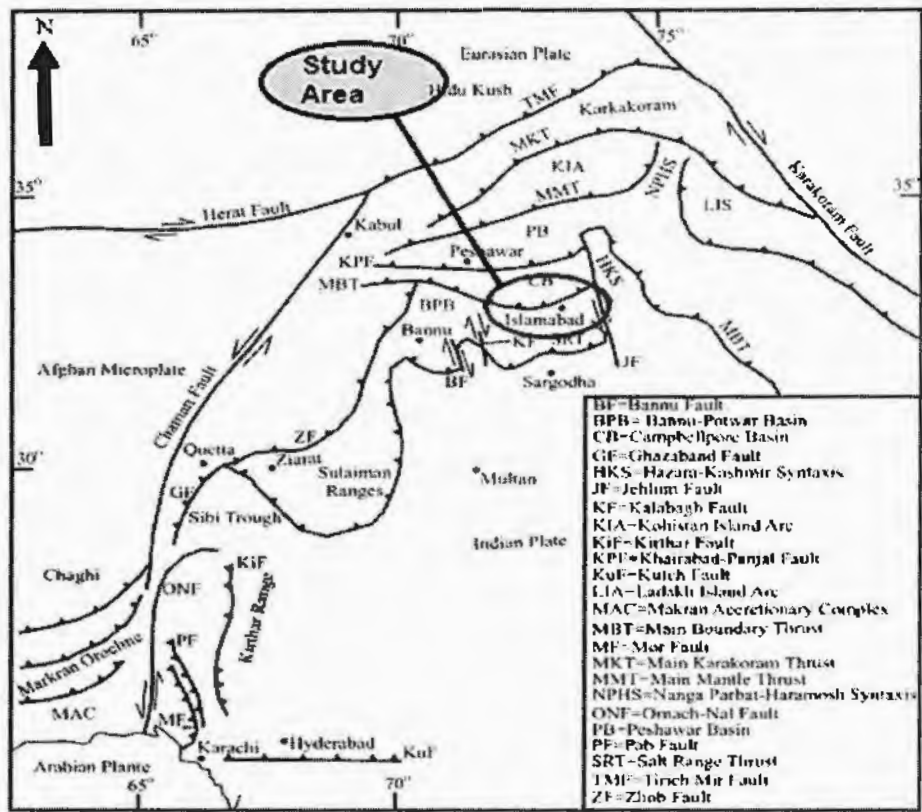


Figure 3-1. Selected Sites for Radon Measurement on tectonic map.

### 3.3 Radon Measurement Methods:

Two methods are usually used for the measurement of radon gas concentration and its decay progenies in different environments.

(A) Active Method. (B) Passive Method (Plastic detectors, HPGc etc.).

#### 3.3.1 Active Method:

Active method of radon concentration consists of different types of electronic detectors. They are RTM2200, RPM2200, EQF3200, RAD7, Pylon detector etc. Each detector is connected to a spectrometer. These detectors contain semiconductor detectors inside and detect either the alpha particles emitted by radon in the decay process or its other decay products through which measurement can be made for radon concentration.

##### (A) Radon Monitor (RTM2200):

RTM-2200 is consummate for radon/thoron concentration measurement. Using RTM-2200 a multi-parameter station may be made easily because of its ability to attach to further sensor. The RTM-2200 used DCMA (data acquisition and control module) for the data processing and control the tools which are essential for the sampling. It can be coupled to extra radon chambers which makes it capable to measure the radon concentration at different place at the same time by the help of a single monitor. Radon transportation processes depend upon pressure and temperature so that RTM-2200 use additional sensor for these two factors. It is also appropriate

in Geophysical instrumentation when we are interested to examine the soil gases and water analytics.

RTM-2200 has chamber for the measurement of radon gas based on high voltage collection principle, therefore using a little inner volume it gives greater sensitivity. This sensitivity is helpful for thoron and  $^{210}\text{Po}$  because the measurement of  $^{210}\text{Po}$  concentration with the help of other principle is rejected. RTM-2200 chamber is not sensitive to variation in ambient humidity therefore, drying units are not necessary. The chamber of RTM-2200 consist of high quality silicon detectors which detect nuclear radiations and it progeny inside the chamber by alpha spectroscopy process.

RTM2200 store a complete alpha spectrum of each data which have been recorded and gives error free measurement. The results can be seen on a large display with touch buttons and for data storing purpose an SD memory card is also allowed [94]. The records will be realized by GPS coordinates in case GPS option is chosen and for data transfer USB port and serial interface (RS232) is recognized. RS232 is used for GSM modems connection. We can use integrated wireless network adapter instead of serial port.



Figure 3-2. Continuous radon thoron monitor (RTM 2200).

#### **(B) Measuring Principal of RTM2200:**

Radon is a radioactive element therefore counting experiments must use for the measurement of its concentration. This type of experiment consists of errors which may done by the experiment performer which must be removed for good results. The collected radon gas will decay into its progeny inside the detector chamber. These progenies consist of short living daughter products like  $^{218}\text{Po}$ . Radon usually under goes through alpha decay process. The

emitted alpha particle made a collision with the shell electron of the  $^{218}\text{Po}$  nuclei making it positively charge. Internal chamber of the detector consists of semiconductor. These ions may be collected by detector and are proportional to radon concentration [51]. The half-life of  $^{218}\text{Po}$  is 3.05mint therefore, after 3.05mint 50% of the decayed daughter nuclei will be recorded on detector. After five half-lives of  $^{218}\text{Po}$  which is a duration of about 15mints an equilibrium begins between detected  $^{218}\text{Po}$  ion and  $^{222}\text{Rn}$  decay rate. Another decay product  $^{214}\text{Po}$  nuclei are obtained after 3hours during in the decay chain with a different energy from  $^{218}\text{Po}$  which can be differentiate from each other by alpha spectroscopy. The spectroscopic calculations provide two mode of calculation, slow mode and fast mode. The slow mode of calculation gives more accurate calculation then fast mode because slow mode of calculation gives the concentration of both  $^{218}\text{Po}$  and  $^{214}\text{Po}$ . Fast mode gives only  $^{218}\text{Po}$  in a very short time therefore there will be greater possibility of statistical errors. For slow mode, the statistical errors are very low. For the measurement of thoron concentration, the instantaneous detection of  $^{216}\text{Po}$  nuclei can be measure directly due to is very short half-life. The half-life of  $^{216}\text{Po}$  is only one second which is responsible for the quick equilibrium state between radon gas concentration and detected activity.  $^{216}\text{Po}$  further decay into  $^{212}\text{Pb}$  and  $^{212}\text{Bi}$  by alpha and beta decay process respectively [51].

### (C) Measurement of Radon Concentration in water Sample:

The radon gas concentration measurement in water sample depends upon the equilibrium state of radon gas and water activity concentration in a closed system after certain time. The ratio between these concentrations depends on the temperature of water which can be represented by Oswald coefficient. The Oswald coefficient state that the solubility of radon gas in water sample decrease with the increase of water temperature. The Oswald coefficient is given below;

$$K_{(oswald)} = \frac{C_{Rn(w)}}{C_{Rn(a)}} \quad (7)$$

If the temperature lies in between 0 °C and 40 °C, then Oswald coefficient becomes as

$$K_{(oswald)} = 0.425 * \exp(0.05 - \text{temprature in } ^\circ\text{C}) \quad (8)$$

Now the total concentration of radon obtained from water sample is the sum of concentration before degassing of water and after degassing of water therefore we have the following relation;

$$A_{(w)} = A_{1(w)} + A_{(a)} \quad (9)$$

Where  $A_{(w)}$  is the total activity of water sample before degassing it  $A_{1(w)}$  is the water activity after degassing and  $A_{(a)}$  is the activity in air. As we know that activity concentration of a sample, the activity and volume of the sample are related by the following relation;

$$C_{(A)} = \frac{A}{V} \quad (10)$$

From here we can have;

$$C_{(A)}V = A \quad (11)$$

For water, this equation becomes

$$C_{A(w)} \times V_{(w)} = A_{(w)} \quad (12)$$

Or 
$$C_{(w)} \times V_{(w)} = A_{1(w)} + A_{(a)} \quad (13)$$

Or 
$$C_{A(w)} \times V_{(w)} = C_{A1(w)} \times V_{(w)} + C_{A(a)} \times V_{(a)} \quad (14)$$

In equation (14)  $C_{A(w)}$  is the concentration activity of water before degassing,  $V_{(w)}$  is the volume of water,  $C_{A1(w)}$  is the activity concentration after degassing,  $C_{A(a)}$  is the activity concentration in air and  $V_{(a)}$  is the volume of the air inside the system. now putting equation (7) in equation (14) we get;

$$C_{A(w)} \times V_{(w)} = K_{(oswald)} \times C_{A(a)} \times V_{(w)} + C_{A(a)} \times V_{(a)} \quad (15)$$

$$C_{A(w)} \times V_{(w)} = C_{A(a)} [K_{(oswald)} \times V_{(w)} + V_{(a)}] \quad (16)$$

$$C_{A(a)} = C_{A(w)} \times [K_{(oswald)} \times V_{(w)} + V_{(a)}] / V_{(w)} \quad [95]. \quad (17)$$

#### (a) Sample Collection Setup:

A degassing process for radon gas dissolved in water sample will be made first by making air bubbles in flask. To collect the entire gas produced by degassing process a closed loop can be made between the bubbling flask and radon monitor. An internal pump inside the radon monitor circulate the desired air volume in a closed loop. Radon will be transfer through small bubbles very efficiently because of a large resulting surface of junction between water and air. Before starting any measurement, it is necessary to have zero concentration of radon in the air loop. The reason is that no preceding measurement is in process and the monitor is not activated. For this purpose, all tubes and radon monitor will be clean for at least 15mints. And the measurement should be start after 30mints when equilibrium establish between air and water activity [95].

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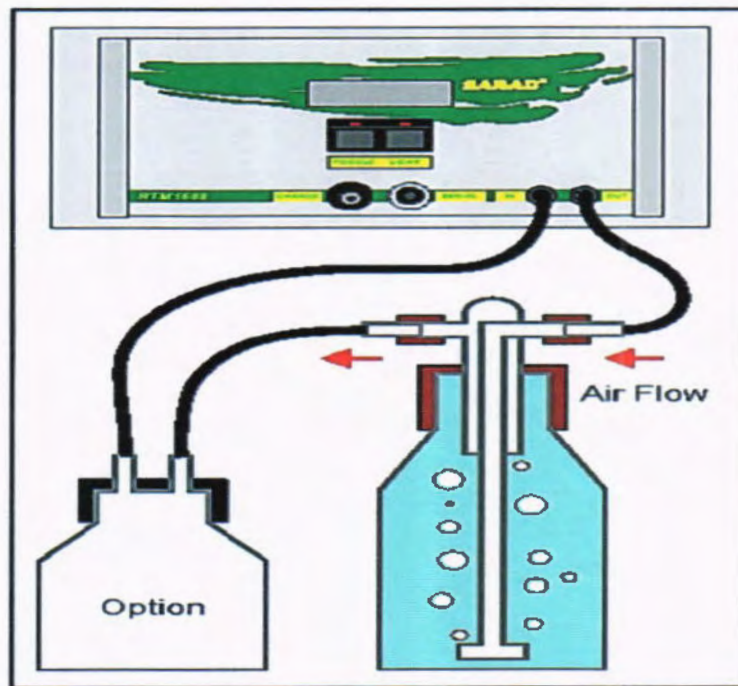


Figure 3-3. Water degassing system for radon gas of water sample.

**(b) Suitable Radon Monitors:**

Such types of radon monitor are required which can provide internal pressure tight air loop, small internal volume, sensitive to detect alpha radiations and spectroscopic technique for short time sample collection. The above conditions are necessary because internal pump provide low leakage and small volume and sensitivity are the parameters that provide information about the detection limits of sampling method. RTM2200 is recommended radon/thoron monitors in water analysis process. The device obtained 1.5 cpm (kBq.m<sup>-3</sup>) from 370 ml, work with an internal pump and alpha spectroscopic technique [95].

**(c) Standard Operating Procedure:**

Radon monitor and air loop may be flush for 15 mints before starting any measurement. Water samples should be taken in bubbling flask. The bubbling flask may be filled completely to avoid any type of turbulence and must be close properly. Now connect bubbling flask to radon monitor and turn on the monitor to pump air volume through loop. After 30 mints concentration equilibrium, will be established between water and air then stop the measurement and discard readings. Now start the monitor again for the next measurement with specific interval and read radon and temperature from the display of monitor. "Radon in water calculator" software can use for radon concentration of water probe [95].

**(d) Condensation:**

The condensation processes of water vapor during water analysis is expected to happen which cause a leakage current due to which a voltage drop occurs. This effect the measuring principle

of the detector but do not damage the detector. So before starting any measurement the humidity of the detector must be checked wither it is in the limit that will do not disturb the alpha spectrum [95].

**(e) Detection limits:**

The detection limit depends on three conditions. Volume of sample, radon monitor and period for sample analysis. The sampling time must be two hours because increasing this interval cause diffusion of radon through the tube walls. The detection limit of radon monitor must be fixed in “Radon in water calculator” software. The radon concentration in water will be equal to the detection limit of water measurement [95].

**(D) Radon Concentration measurement in Soil Gas:**

Continuous and instantaneous measurements can be made for radon activity concentration in soil gas. The samples can be collected from bore hole through a probe. The monitor used internal pump for continuous sampling. In many cases, external pump may also be used. They are used for radon sucking process. The unit used for radon concentration activity in soil gas is  $Bq.m^{-3}$  and is given by the following equation.

$$C_{(Rn)} = A[Bq/m^3]/V[m^3] \quad (18)$$

Radon concentration in soil samples vary from  $1,000 Bq.m^{-3}$  to  $1,000,000 Bq.m^{-3}$  [96].

**(a) Procedure of Measurement:**

A bore hole of 1-2 m depth with a diameter of 7 cm can be made by the help of machine, hand operated drilling system for silty/sandy ground. A packer probe can be inserted into bore hole as deep as possible. For taking soil gas samples, the packer probe metallic tube connected to special hand operated pump or directly to radon monitor be a flexible PVC tube. Special hand operated pump connected to packer probe by a flexible PVC collect two or three samples of gas and release it for cleaning purpose from ambient air. After that measuring chamber of radon monitor can be filled with soil gas per measurement [96].

**(b) Appropriate Radon Monitors:**

There are some requirements for the radon monitor which must be taken in account. The monitor must provide an internal pressure tight air loop. An internal pump is advantage. Membrane pump are more suitable then rotary pumps because membrane pumps offer lower leakage rate. For short sample period, alpha spectroscopic monitor is required. The internal volume of the monitor should as small as possible because small volume and instrument sensitivity of the instrument are the parameters that define the detection limit of sampling method. The sensitivity of RTM2200 is  $1.5 cpm/(Bq.m^{-3})$  for an internal volume of 370 ml.

This instrument is suitable for radon soil gas measurement. Both units work with Alpha spectroscopy and are equipped with membrane pumps [96].

**(c) Standard Operating Procedure:**

Make a bore hole and carefully seal the packer probe in the bore hole. Connect the probe with pump and radon monitor. Release first two or three soil gas samples. Determine the maximal radon concentration of the soil gas by taking several samples. Determine falling times  $T_0$  and  $T_1$ . Calculate permeability and radon availability according for the following equations.

$$R = C \times K [KBq/m] \text{ [96].} \tag{19}$$

**(d) Detection limit:**

Detection limit depends upon sampling volumes, the sensitivity of radon monitor and the period of radon measurement in air loop. Detection limit and statistical error of the measurement corresponds to the number of detected decays within the sampling period. The configuration can be optimized by the following rules.

(1) Use of high sensitive radon monitor. (2) Choose sampling interval possible [96].

**(E) Radon Emanation measurement on Surface:**

Radon emanation on surface is obtained by relating the change in its concentration to another well-known gas volume. The radon flux is denoted by  $E$  and is defined as the 'The ratio of radon gas activity in Bq to the radon emanation area in  $m^2$  and time in s'. The flux is measurement in  $Bq.m^{-2}.s^{-1}$ .

$$E = A/F \times t \tag{20}$$

Here  $F$  represent the area in  $m^2$  and  $t$  show the time in seconds. If a rectangular shape solid having an area  $F$  and height  $h$  is taken for the emanation of radon, then we have:

$$V = F \times h \tag{21}$$

Where  $V$  show the volume of the sample taken.

As activity concentration is defined as

$$C = A/V \tag{22}$$

$$C \times V = A \tag{22}$$

$$C(F \times h) = A \tag{23}$$

Putting equation (23) in equation (19) becomes

$$E = C(F \times h)/F \times t \tag{24}$$

By cancelling  $F$ , we obtain the following equation

$$E = C \times h/t \tag{25}$$

If the increase in concentration is represented by  $C(\text{Diff})$  then equation (25) can be written as

$$E = C_{\text{Diff}} \times h/t \tag{26}$$

This equation gives the emanation of radon on surface of a rectangle shape sample [97].



**(a) Procedure of Measurement:**

The radon gas concentration  $C_1$  in ambient air can be measured through a radon monitor with the help of an internal or external pump and then emanation of radon  $C_2$  from the rectangular shape should be calculated. A closed loop is made between the sample and radon monitor through PVC tubes for air circulation. From these two values of concentration we find the value of  $C(\text{Diff})$  by the following relation.

$$C_{\text{Diff}} = C_2 - C_1 \quad (27)$$

Using the value of  $C(\text{Diff})$ ,  $h$  and  $t$  in (25) we can find the value of radon flux  $E$  [96].

**(b) Operating Procedure:**

First radon monitor and air loop will be clean for at least 15 mins in fresh air and then measurement will be made for ambient air. The entire setting may be sealed carefully and should be connect with radon monitor. The maximum radon concentration inside the circuit and the time taken will be noted and concentration should be find by the help of equation (25) [97].

**(c) Recommended Radon Monitor:**

RTM 1688-2 is recommended monitor for the radon gas analysis in water samples, soil gas and surface exhalation of radon. Since it provides an internal pump which offer lower leakage rate. Internal volume and sensitivity are the parameters which can be used in observation for the detection limit and RTM 1688-2 provide these two parameters. Its sensitivity is more than 3 cpm ( $\text{kBq.m}^{-3}$ ) for a very small internal volume of about 130 ml. RTM 2200 used an internal volume of 370 ml and give a sensitivity of about 1.5 cpm ( $\text{kBq.m}^{-3}$ ). Therefore, it is also suitable for the water sample measurements. RTM 1688-2 and RTM 2200 both work with alpha spectroscopy and both are equipped with membrane pumps [97].



Figure 3-4. Continuous radon thoron monitor (RTM1688-2).

### 3.3.2 Passive Method:

In passive method, solid state nuclear track detector (SSNTDs) and HPGe detector were used for the measurement purpose of radon gas concentration and elemental analysis of soil samples for radionuclides. HPGe detectors are two types one is positive HPGe detector and second one is negative HPGe detector while SSNTDs include many types that are LR-115, CR-39, CN-85, Lexan etc. The used detector for the current study was CN-85.

#### (A) Plastic Detectors SSNTDs:

Solid State Nuclear Track Detectors (SSNTDs) are insulator solids present naturally and man-made forms [98]. They are in numerous types including inorganic crystals, glasses and plastics. When a heavily ionizing charged particle passes through such insulator solids, it produces a thin track of about 50 Å in diameter along its path. This is called 'Latent Track' as it cannot be seen with the naked eye therefore, an electron microscope may be used to view this thin track. The precise nature of the physical and chemical changes occurring at the damage site depends on the charge ( $Z$ ) and velocity ( $\beta = v/c$ , where  $v$  is the particle velocity and  $c$  is the velocity of light) of the particle, chemical structure of the detector material. These thin tracks can be enlarged so that they can be observed under an optical-microscope by chemical etching with sodium-hydroxide and hydrofluoric-acid. Cellulose-nitrate was the first detector used for detecting alpha tracks. It is less sensitive and is now substituted by a more sensitive detector known as CR-39 [99]. Poly-carbonate detectors such as Lexan are commonly used for detecting fission fragment tracks.

**(B) SSNTDs planting Method:**

Solid state nuclear tracks detectors (SSNTDs) are reliable and convenient detectors for the investigation of long term monitoring of radon in different environments. They are unaffected by meteorological parameters and are low cost therefore they are widely used in radon measurements [100]. CN-85 solid state nuclear track detector (SSNTDs) were used systematically for the measurement of soil radon concentration for a period of 90 days.

**(C) SSNTDs planting method at Islamabad:**

Eight detectors were planted at the studied area. The detectors were divided into two groups every one containing four detectors. The first group detectors were inserted one by one in box type casing and was represented by B while the second group detectors were inserted one by one in cup type dosimeter and was represented by C. Now one box type and one cup type casing were Combinly hanged in a plastic bottele by the help of wire at same level. This method was used to compaire the values of radon determind from both type of casing. Now every bottel were planted in a 60 cm deep bore-hole. The diameter of the bore-whole was 10 cm. the Alpha particles emitted by radon gas during the decay process when fall on these detectors, produced tracks on it. These traks will not be visible clearlly for this purpose the detectros were then recolected after the desired duration for chemically etching process. These detector were etched in 6N NaOH solution for 70mint at a temperature of 50 °C. These nuclear tracks becomes visible upon chemical reaction with NaOH solution. These tracks may be counted by the help of an electrical microscope. The number of tracks recoded on a detector is directly proportional to the radon concentration.

## Chapter 04:

### Results and Discussions:

Islamabad the capital of Pakistan was selected for the measurement of soil radon concentration by the help of two different techniques.

- I. Active technique.
- II. Passive technique.

Active technique was done by the help of continuous radon thoron monitor RTM2200 while passive technique was done by the help of solid state nuclear track detectors (SSNTDs).

CN-85 SSNTDs plastic type detectors were used in passive technique. These detectors were mounted from 1<sup>st</sup> July 2016 to 30<sup>th</sup> September 2016 at Islamabad station. After the duration of 90 days they were collected for chemical etching process and then were subjected to automatic nuclear track counter. The obtained results of radon concentration for the selected site were higher than the normal values published in the literature. Therefore, this site was selected for continuous monitoring of radon through RTH2200. The monitor was planted at Islamabad stations. The instrument is developed by a German company SARAD. It provides wide range of gas sensors. The instrument measure radon, thoron with barometric pressure, environmental temperature and relative humidity. The sampling rate of the instrument is 15 mints.

#### 4.1 Results of passive technique for the selected area:

Radon concentration was measured along Islamabad fault line by the help of CN-85 type solid state nuclear tracks detector (SSNTDs). Figure3-2 show the plantation area of solid state nuclear track detectors (SSNTDS) at Islamabad. The measurements were done for 90 days along the Islamabad fault line. During the measurements four points were selected along the fault line. At every point two SSNTDs were planted in different casing (Box type, Cup type) to compare their measuring ability as well. The maximum and minimum radon concentration were found to be 2792 Bq.m<sup>-3</sup>/hr to 1554 Bq.m<sup>-3</sup>/hr respectively. The average values at Islamabad was 2272.7875 Bq.m<sup>-3</sup>/hr.

The cup type of casing showed good ability in the measurement as compared to box type. The efficiency of the cup type casing is due to the smaller calibration factor value that is 0.0027 and for box type casing it is 0.009. The individual vales of box type casing and cup type dosimeter recorded at studied area are given in the following table.

Table 4-1. Average track density with average radon concentration for Box and Cup type dosimeters at Islamabad.

Site(A)	Dosimeter	Average of (25 values) Tracks/graticulate/90 days	Average of (25 values) Tracks/graticulate.h <sup>-1</sup>	Av. Track Density.h <sup>-1</sup>	Radon (Bq.m <sup>-3</sup> .h <sup>-1</sup> )
A	Box type	22.2	0.0102	13.99	1554.00
	Cup Type	21.48	0.0099	13.58	2235.00
B	Box type	32.92	0.0152	20.85	2316.00
	Cup Type	16.12	0.0074	10.15	2792.00
C	Box type	27.12	0.0125	17.14	1904.40
	Cup Type	9.25	0.0044	5.62	2233.30
D	Box type	36.4	0.0168	23.04	2560.00
	Cup Type	11.08	0.0051	6.99	2588.00

Snapshot of the solid state nuclear tracks detector planted at Islamabad taken by automatic counts detector are given below.

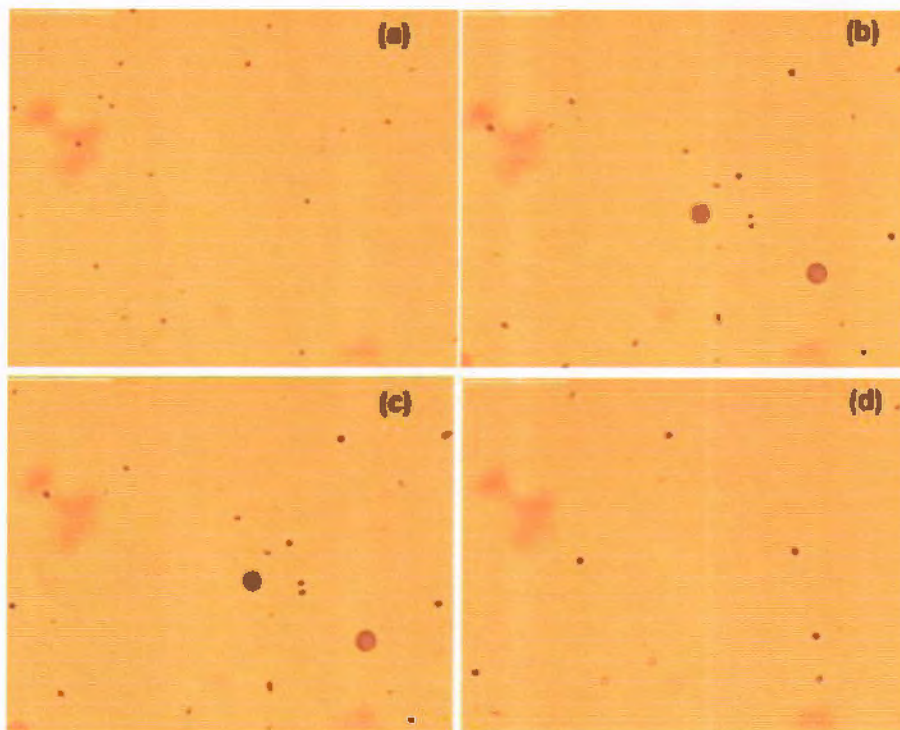


Figure 4-1. SSNTDs planted at Islamabad snapshots taken from automatic track counter. The average hourly observed value for soil radon was very high as compared to the previous data published by N. Ali et al., (2010) at Islamabad. Now to investigate the reason of these higher values at our study area. We have monitored the radon concentration through a continuous radon/thoron monitor RTM2200 for the same duration. The instrument provides radon concentration profile with meteorological parameters for 15 mints period. The resultant data were analyzed on hourly base and monthly base to observe the temporal variation in radon

concentration for the three months from July 2016 to September 2016. The data were correlated to the variations in meteorological parameters. Some special peak values were obtained in monthly data while an anomalous increase was obtained in the combined data during the study period which were investigated for their cause.

## **4.2 Continuous monitoring of radon fluctuation results from Islamabad.**

The radon concentration profile for the whole month of July 2016 is shown in the Figure4-3(a). For the same month, the meteorological parameters (barometric pressure, environmental temperature and relative humidity) profile have been recorded and their variation with time have been shown in Figure4-3(b), Figure4-3(c) and Figure4-3(d) respectively.

Figure4-3(a) show an increasing and decreasing trend in radon concentration profile with normal values. In the beginning of the month a drop is detected after which the values became to normal situation for two days and on fifth day 16<sup>th</sup> July, 2016 a sudden decrease from the normal values was detected. On next day 17<sup>th</sup> July, 2016 an increasing peak was detected. During these two anomalies, barometric pressure was fluctuating with normal values but environmental temperature and relative humidity were showing increasing and decreasing trend respectively. This the increasing peak remained only for a single day. On 18<sup>th</sup> July, 2016. The concentration started variations with a large value. These changes remained till to 24<sup>th</sup> July, 2016 on the same day the value drop suddenly and on next day 25<sup>th</sup> July, 2016 an increase was observed.

During this period the barometric pressure, environmental temperature and relative humidity were also observed with large variations. The values were showing the same behavior for 26<sup>th</sup> July, 2016 to 30<sup>th</sup> July, 2016 as were recorded for the previous five days. At the last day on 31<sup>th</sup> July, 2016 a significant large peak was recorded. At the same day, it was observed that the barometric pressure, environmental temperature and relative humidity were fluctuating with a systematic minor. The environmental temperature and barometric pressure were observed to fluctuate normally during the whole month while relative humidity mostly showed decreasing values. This may be due to the weather going toward warmer. The radon fluctuations for the whole month was observed to have no meaningful relation with the fluctuations in environmental temperature, barometric pressure and relative humidity.

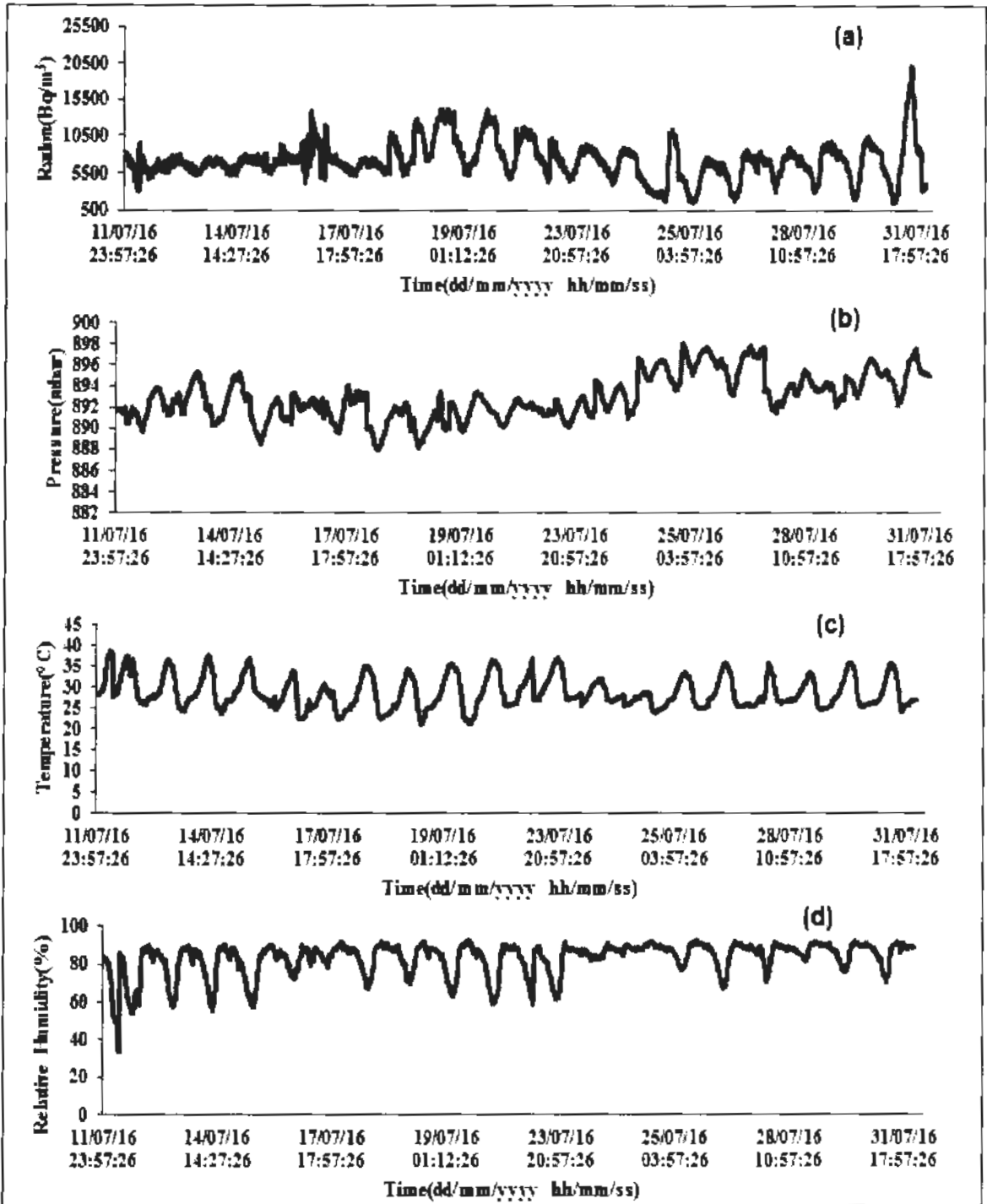


Figure 4-2. Radon fluctuation with meteorological parameters for the whole month of July, 2016. Table 4-2 give the statistic of the parameters. From here we have a total of 1808 data for every variable in which the missing points for radon, barometric pressure, environmental temperature and relative humidity were 1, 3, 1 and 1 respectively. The radon concentration was observed to vary in the range from 1460 Bq.m<sup>-3</sup> to 20067 Bq.m<sup>-3</sup> while thoron range from 50 Bq.m<sup>-3</sup> to 1841 Bq.m<sup>-3</sup> during the whole month. Barometric pressure ranged from 887.92 to 898.00 mbar. The environmental temperature ranged from 21.28 to 38.46°C and relative humidity ranged

from 33.79 to 92.20%. The average value of radon for the whole month was 7158.36 Bq.m<sup>-3</sup>. The standard deviation for radon was 2610.36 with % standard deviation of 0.3646. Average pressure, temperature and relative humidity for the whole month were 892.94 mbar, 28.88°C and 82.50% respectively.

Table 4-2. Statistic of radon concentration, pressure, temperature and relative humidity for the whole month of July, 2016.

Parameters	Radon (Bq/m <sup>3</sup> )	Pressure (mbar)	Temperature (°C)	Humidity (%)
Average	7158.36	892.94	28.88	82.50
SD	2610.63	2.144729	3.804737265	8.957542
%SD	0.3646			
Man	1460	887.92	21.28	33.79
Max	20067	898.00	38.46	92.20
Median	6991	892.63	27.846	86.11
Number	1807	1805	1807	1807
Total	1808	1808	1808	1808
Missing points	1	3	1	1
%age data	0.999447	0.998341	0.999446903	0.999447

Now to find a correlation between the radon concentration and the meteorological, we plotted scattered plots for these variables as shown in Figure 4-4.

The Figure 4-4(a) represent the correlation-ship of radon with barometric pressure through a trend-line representation which shows a positive trend-line relation therefore, we can say that radon concentration decreases with the decrease in barometric pressure.

Figure 4-4(b) represent the correlation-ship of radon concentration with environmental temperature with the same technique. But this time we find a negative trend line relation between temperature and radon concentration therefore, we say that radon concentration decreases with the increase in environmental.

Figure 4-4(c) represent the correlation-ship of radon concentration with relative humidity. The humidity and radon concentration trend line are positively related with each other this show that radon concentration in soil increase with the increase in relative humidity.



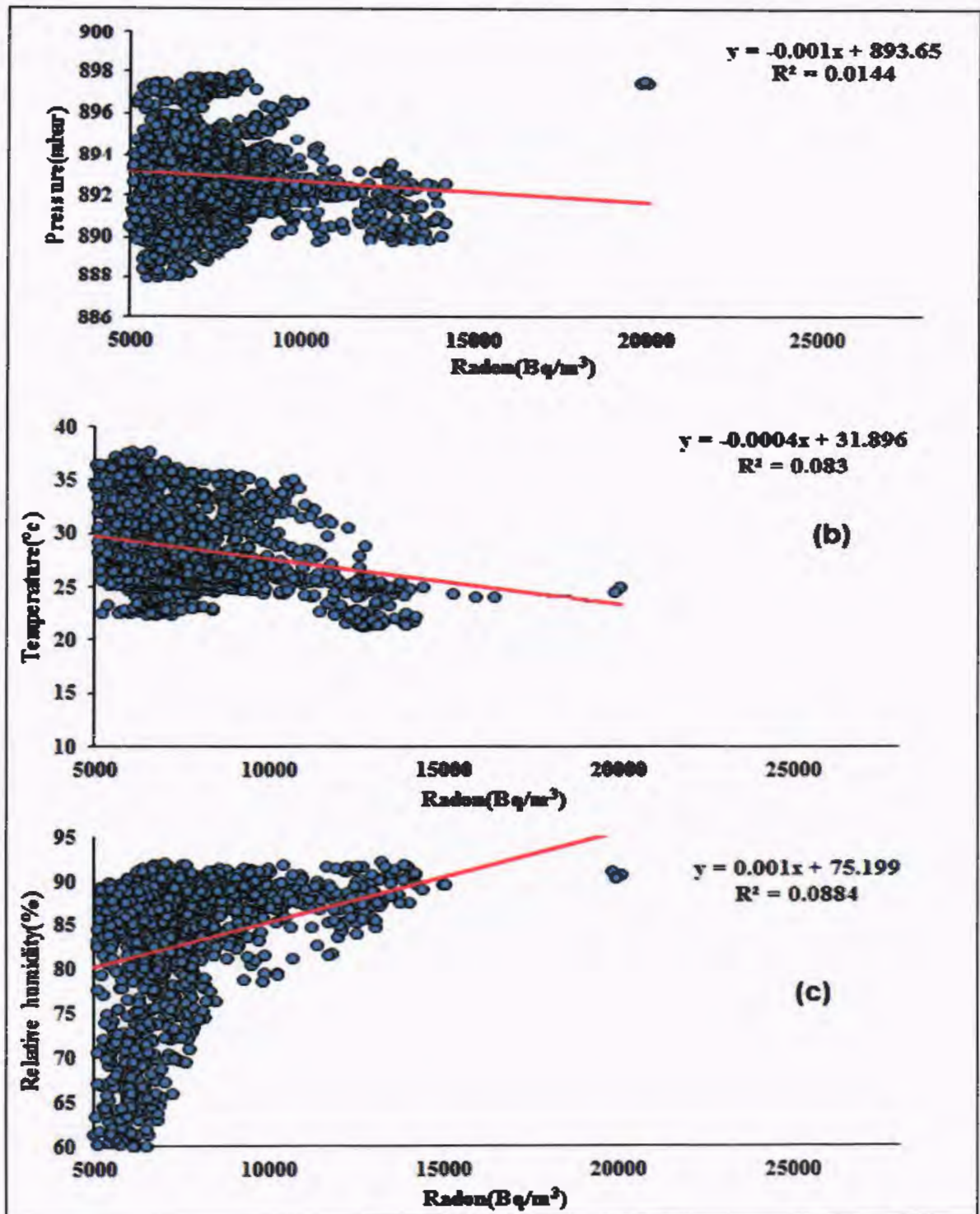


Figure 4-3. Correlation of radon concentration with meteorological parameters for July 2016.

The Table4-3 show numerically the correlation-ship of radon concentration with its self and with barometric pressure, environmental temperature and relative humidity for the whole month of July 2016. It is clear from the table that radon overall is negatively related to pressure and temperature but is positively correlated to relative humidity of the soil. The Table4-3 also show that on the whole thoron is negatively correlated to barometric pressure and environmental temperature and is positively related to relative humidity of the soil. The

correlation-ship of barometric pressure to environmental temperature is negative which show that the barometric pressure decreases with the increase in environmental temperature and increase with the decrease in it. Barometric Pressure and humidity are negatively related to each other that means increase in humidity causes a decrease in barometric pressure. The relative humidity and environmental temperature are again also negatively correlated to each other means the increase in environmental temperature cause a decrease in the relative humidity of soil. From this correlation, we obtained a linear equation that represent the correlation of these parameters for this month. The equation is given as:

$$C_{Rn} = (-0.1198)P + (-0.2881)T + (0.2972)H + C. \tag{1}$$

Where P represent barometric pressure, T represent the environmental temperature and H represent the relative humidity and C any obituary constant. That may be due any other factor.

Table 4-3. Correlation table of radon and meteorological parameters for the whole month of July, 2016.

	Radon (Bq.m <sup>-3</sup> )	Pressure (mbar)	Temperature (°C)	Humidity (%)
Radon	1			
Pressure	-0.1198	1		
Temperature	-0.2881	-0.1749	1	
Humidity	0.2972	0.4441	-0.7592	1

The radon concentration profile for the whole month of August 2016 is shown in the Figure4-5(a). For the same month, the meteorological parameters (barometric pressure, environmental temperature and relative humidity) profile have been studied which are represented in Figure4-5(b), Figure4-5(c) and Figure4-1(d) respectively.

Figure4-5(a) show increasing and decreasing trends in radon concentration with only one specific increasing value in the mid of the month. The barometric pressurc and relative humidity was behaving normally during this increase but a temperature dropped was observed on the same day. The temporal variations in barometric pressurc, environmental temperature and relative humidity are also observed during the whole month of August 2016. Due to summer season, the temperature for the whole month was mostly high and barometric pressure and relative humidity was observed with less variations.

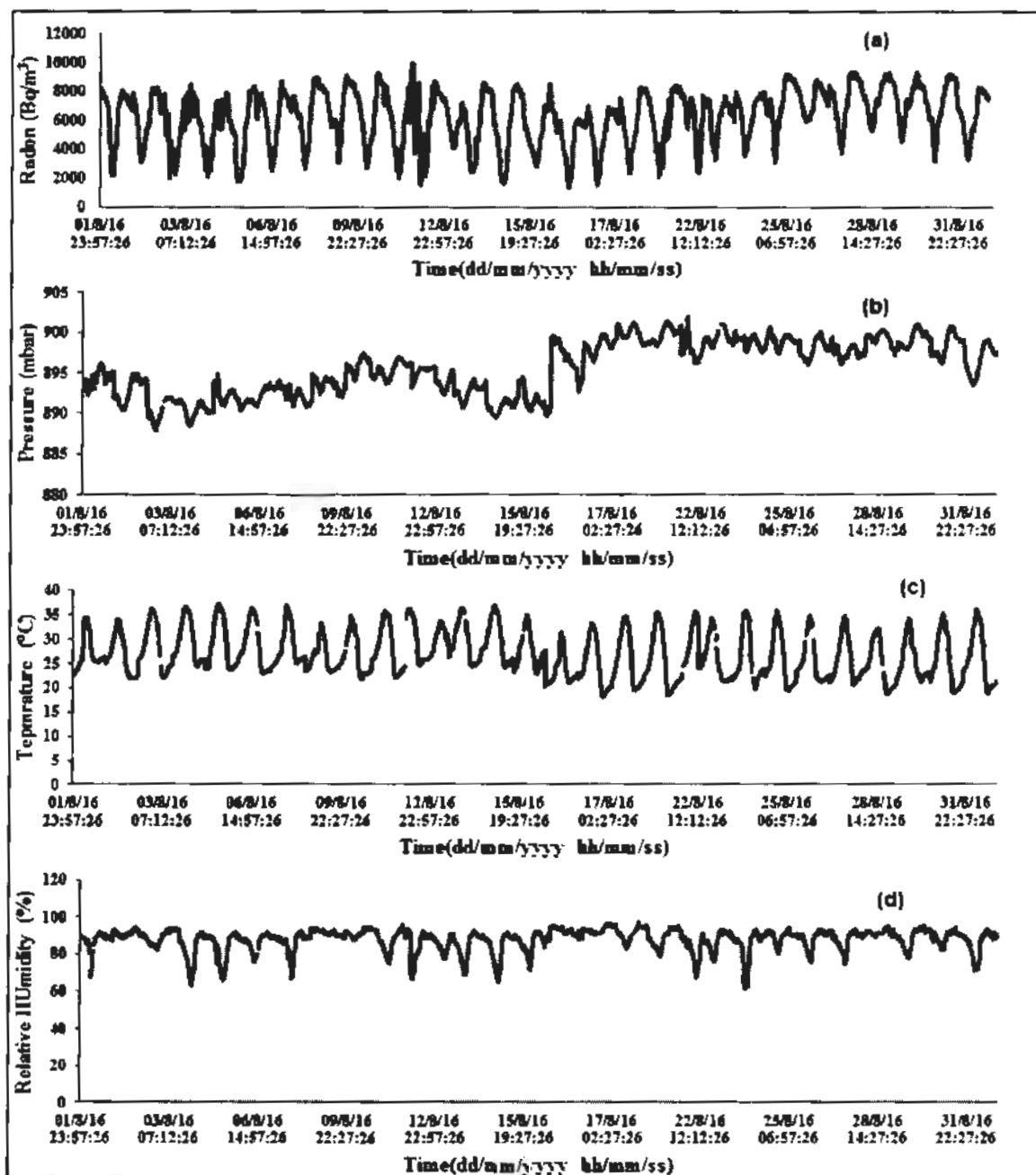


Figure 4-4. Radon fluctuation with meteorological parameters for the month of August, 2016. From Table 4-4 we have a total of 2650 data for every variable in which the missing points for radon, barometric pressure, environmental temperature and relative humidity were 61, 63, 59, and 58 respectively. The radon was observed to vary in the range from 1123 Bq.m<sup>-3</sup> to 10350 Bq.m<sup>-3</sup>. Barometric pressure ranged from 888.038 to 902.062 mbar. The environmental temperature ranged from 18.184 to 37.189°C and relative humidity ranged from 60.848 to 97.529%. The average value of radon concentration for the month was 6243.3279 Bq.m<sup>-3</sup> with standard deviation for radon was 2045.6292 and % standard deviation of 0.3276.

Average pressure, temperature and relative humidity for the whole month were 895.5923.5 mbar, 27.0211 °C and 87.7076 % respectively.

Table 4-4. Statistic of radon concentration, pressure, temperature and relative humidity for the whole month of August, 2016.

Parameters	Radon (Bq/m <sup>3</sup> )	Pressure (mbar)	Temperature (°C)	Humidity (%)
Average	6243.3279	895.5923	27.0211	87.7076
SD	2045.6292	3.37142	4.9110	6.1831
%SD	0.3276	0.0037	0.1817	0.07049
Min	1123	888.038	18.184	60.848
Max	10350	902.062	37.189	97.529
Number	2589	2587	2581	2592
Total	2650	2650	2650	2650
Missing points	61	63	69	58
%Age Data	0.97698	0.97622	0.97396	0.97811

Now to find a correlation between the radon concentration and the meteorological parameters, pressure, temperature and relative humidity we plotted scattered plots for these variables as shown in Figure4-5.

The Figure4-5(a) represent a correlation-ship of radon with barometric pressure through trend-line relationship which show that radon concentration have a positive relation with barometric pressure. The increase in barometric pressure cause increase in radon concentration.

Figure4-5(b) represent the correlation-ship of radon concentration with environmental temperature again by trend-line relation, which show a negative relation between them. This show that the increase in temperature cause a decrease the radon concentration.

Figure4-6(c) represent the correlation-ship of radon concentration with relative humidity by the same way which show a positive relation between radon concentration and relative humidity. This show that radon concentration in soil increase with the increase in humidity of the soil.

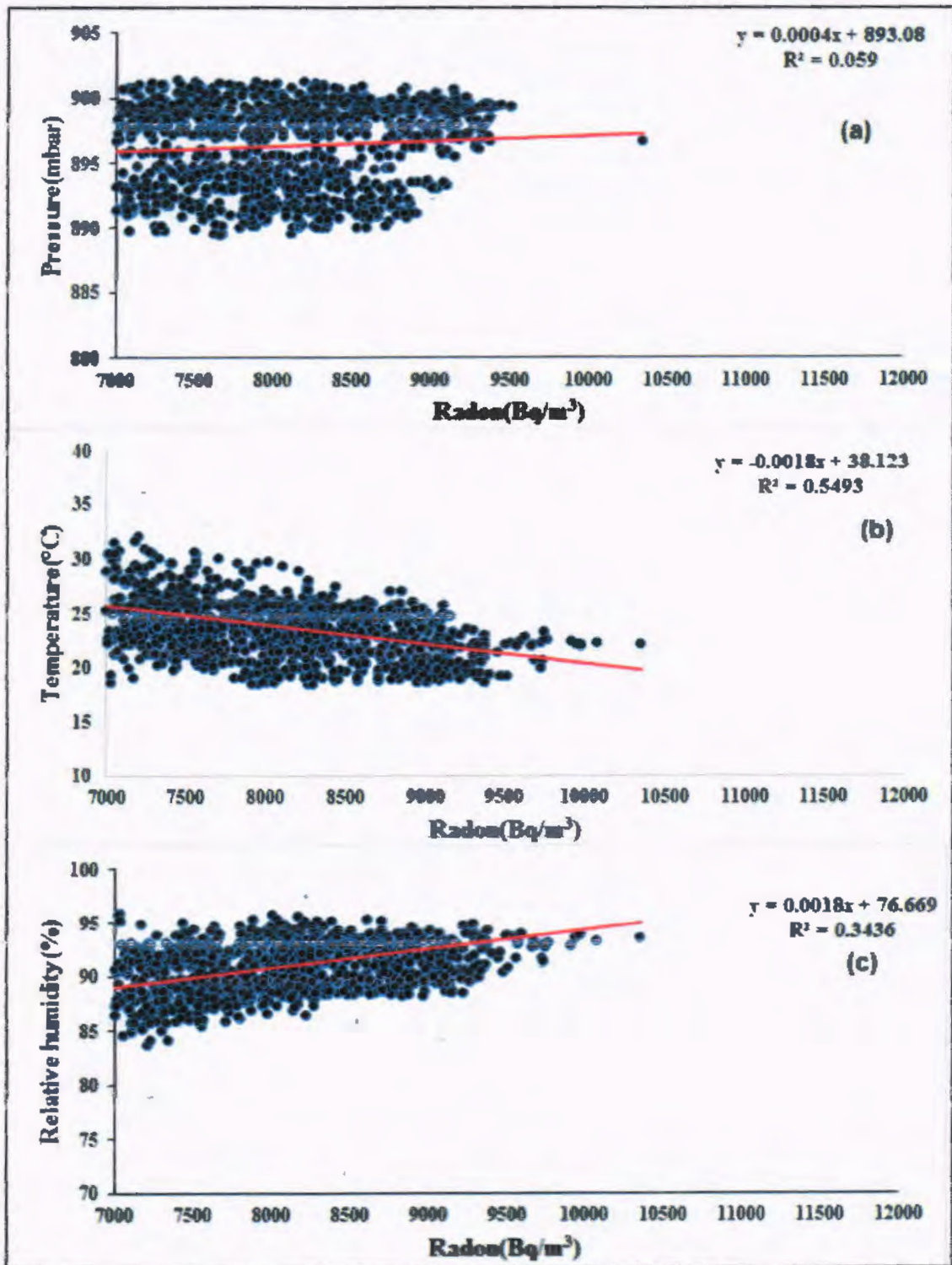


Figure 4-5. Correlation of radon concentration with meteorological parameters for August, 2016. The Table4-5 show the correlation-ship of radon concentration with pressure, temperature and relative humidity. The table also show correlation-ship between these parameters as well for the whole month of August 2016. The table show that radon to radon correlation is positive, radon to pressure is positive and radon to temperature is negative but again it is positive to the

humidity in soil. The correlation-ship of pressure to temperature is also negative which show that pressure decrease with the increase in temperature and increase with the decrease in it. Pressure and humidity are positively related to each other that means increase in humidity increase the value of pressure while humidity and temperature are again negatively correlated to each other. The increase in temperature cause a decrease in the relative humidity of soil. From these correlation-ship between radon concentration and the other parameters we have find a mathematical formula that may represent the relation between them.

$$C_{Rn} = (0.2426)P + (-0.7411)T + (0.5861)H + C \quad (2)$$

This is a statistical relation between these parameters. Here again P represent the barometric pressure, T represent the temperature and H represent the relative humidity while C is any obituary constant.

Table 4-5. Correlation table of radon and meteorological parameters for the whole month of August, 2016.

	Radon (Bq.m <sup>-3</sup> )	Pressure (mbar)	Temperature (°C)	Humidity (%)
Radon	1			
Pressure	0.2426	1		
Temperature	-0.7411	-0.3457	1	
Humidity	0.5861	0.3442	-0.6636	1

Finally, the radon concentration was recorded for the whole month of September, 2016. The whole month concentration profile is shown in the Figure4-6(a). The meteorological parameters (barometric pressure, environmental temperature and relative humidity) for the same month are also represented in Figure4-6(b), Figure4-6(c) and Figure4-6(d) respectively. The Figure4-6 show that the flection in radon concentration is normal in the beginning. The radon concentration rise on 5<sup>th</sup> September, 2016 and fall-down on 6<sup>th</sup> September, 2016. The concentration again rises on next day 7<sup>th</sup> September, 2016 and on 8<sup>th</sup> September, 2016 again fall-down. The concentration has shown the same behavior for the next two days. During these six days, the barometric pressure and environmental temperature were also higher and have shown fluctuated nature while relative humidity was getting decrease in these days. Another increase was recorded on 20<sup>th</sup> September, 2016, on the same day barometric pressure and environmental temperature were also high but relative humidity was lower. On next day, the anomaly come down suddenly but on the same day the barometric pressure and environmental temperature were observed to have lower values and relative humidity was higher. Two other peaks have also been observed on 23<sup>rd</sup> September, 2016 and 24<sup>th</sup> September, 2016. These were also related to the change in barometric pressure, environmental temperature and relative humidity. For the whole month, the radon concentration has shown positive response to the

variations in barometric pressure and environmental temperature while negative response to relative humidity.

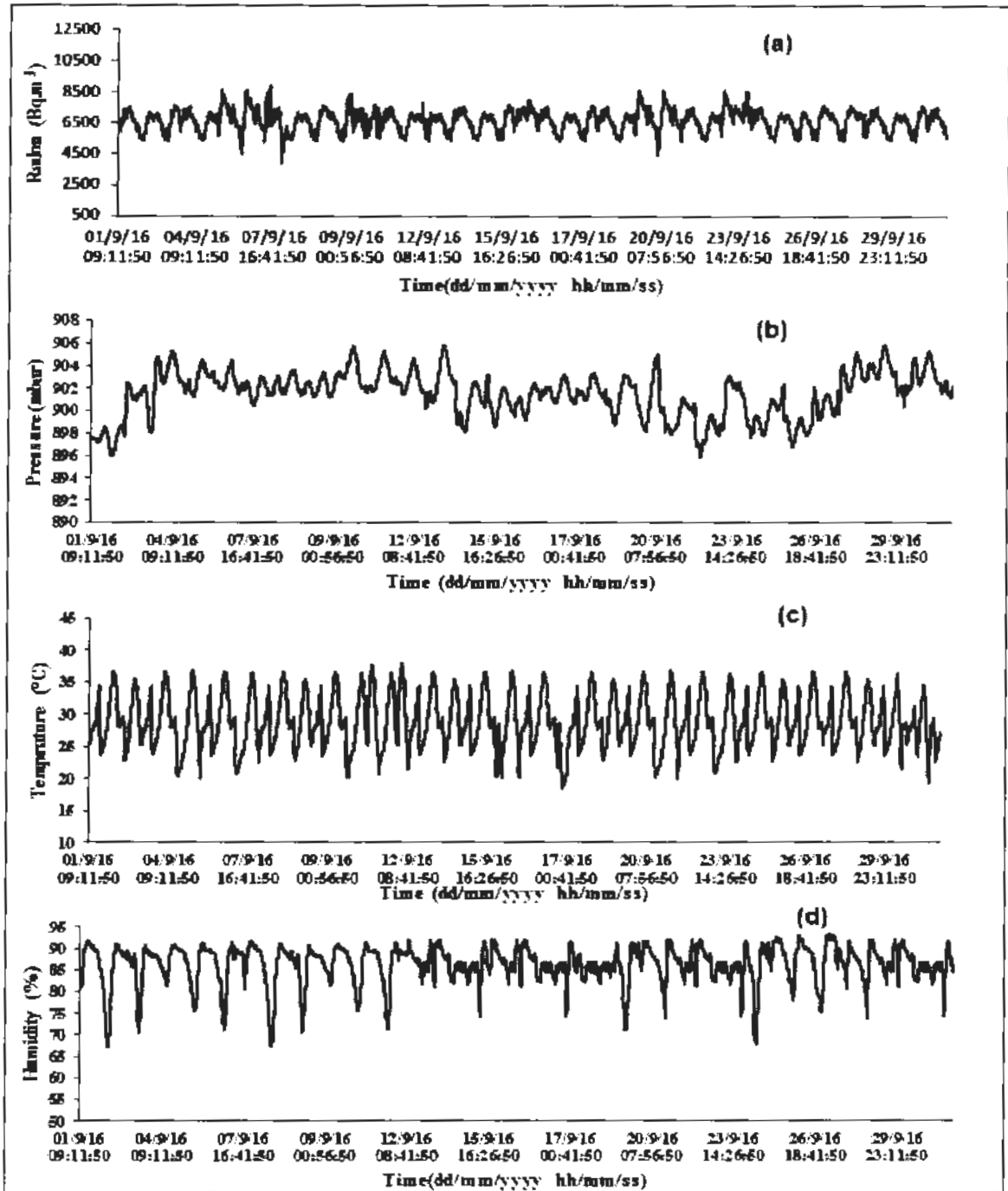


Figure 4-6. Radon fluctuation with meteorological parameters for the whole month of September, 2016. From Table 4-6 we have a total of 2727 data for every variable in which the missing points for radon, barometric pressure, environmental temperature and relative humidity were 5, 83, 30, and 18 respectively. The radon was observed to vary in the range from 884 Bq.m<sup>-3</sup> to 9970 Bq.m<sup>-3</sup>. Barometric pressure ranged from 895.648 to 905.904 mbar. The environmental temperature ranged from 18.203 to 38.085°C and relative humidity ranged from 66.001 to

93.466%. The average value of radon concentration for the month was 6717.301 Bq.m<sup>-3</sup> with standard deviation for radon was 808.025 and % standard deviation of 0.120. Average pressure, temperature and relative humidity for the whole month were 901.512 mbar, 29.059 °C and 86.285% respectively.

Table 4-6. Statistic of radon concentration, pressure, temperature and relative humidity for the whole month of September, 2016.

Parameters	Radon (Bq/m <sup>3</sup> )	Pressure (mbar)	Temperature (°C)	Humidity (%)
Average	6717.301	901.512	29.059	86.286
SD	808.025	2.018	4.296	4.912
%SD	0.120	0.0022	0.1478	0.0569
Min	884	895.648	18.203	66.001
Max	9970	905.904	38.085	93.466
Median	6835	901.8065	28.569	87.884
Number	2722	2644	2697	2709
Total	2727	2727	2727	2727
Missing points	5	83	30	18
%Age Data	0.9981	0.9695	0.9889	0.9933

Now to find a correlation between the radon concentration and the meteorological parameters, pressure, temperature and relative humidity we plotted scattered plots for these variables as shown in Figure4-7.

The Figure4-7(a) represent a correlation-ship of radon with barometric pressure through trend-line relationship which show that radon concentration have a positive relation with barometric pressure. The increase in barometric pressure cause increase in radon concentration.

Figure4-7(b) represent the correlation-ship of radon concentration with environmental temperature again by trend-line relation, which also show a positive relation between them. This show that the decrease in temperature cause a decrease in radon concentration.

Figure4-7(c) represent the correlation-ship of radon concentration with relative humidity by the same way which show a negative relation between radon concentration and relative humidity. This show that radon concentration in soil decrease with the increase in humidity of the soil.



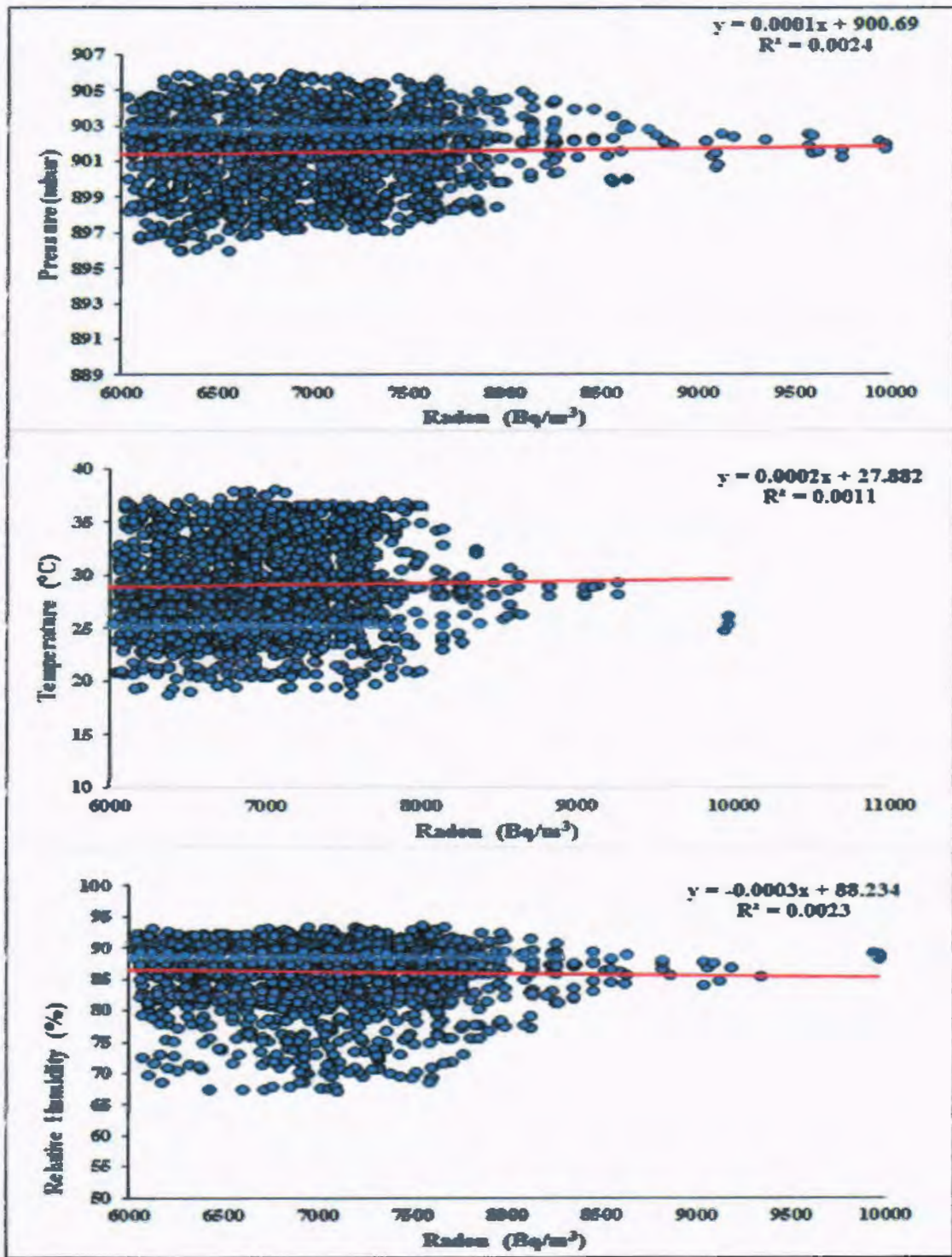


Figure 4-7. Correlation of radon concentration with meteorological parameters for September, 2016.

The Table 4-5 show the correlation-ship of radon concentration with pressure, temperature and relative humidity. The table show that radon to radon correlation is positive, radon to pressure and temperature is also positive but it is negative to the humidity in soil. The correlation-ship of pressure to temperature is also positive which show that pressure decrease with the decrease

in temperature and increase. Pressure and humidity are positively related to each other that means increase in humidity increase the value of pressure while humidity and temperature are negatively correlated to each other so, the increase in temperature cause a decrease in the relative humidity of soil. From these correlation-ship between radon concentration and the other parameters we have find a mathematical formula that may represent the relation between them.

$$C_{Rn} = (0.0486)P + (0.0328)T + (-0.0478)H + C \quad (3)$$

This is a statistical relation between these parameters. Here again P represent the barometric pressure, T represent the temperature and H represent the relative humidity while C is any obituary constant.

Table 4-7. Correlation table of radon and meteorological parameters for the whole month of September, 2016.

Parameters	Radon (Bq.m <sup>-3</sup> )	Pressure (mbar)	Temperature (°C)	Humidity (%)
Radon	1			
Pressure	0.0486	1		
Temperature	0.0328	0.009366	1	
Humidity	-0.0478	0.033340	-0.05586	1

Now radon concentration anomalies are also related to the earthquake phenomena therefore, the whole data was also combinedly investigated with the meteorological parameters to observe that the anomalies recorded in the concentration were either due to the variations in these parameters or it is due to some nearly occurred earthquake event. For this purpose, a linear regression relation was applied to the data and it was observed that radon concentration was positively related to barometric pressure and relative humidity while it was negatively related to environmental temperature. The relation is given by the following linear equation;

$$C_{Rn} = (0.0485)P + (-0.0366)T + (0.2883)H + C \quad (4)$$

This is a statistical relation between these parameters. Here P represent the barometric pressure, T represent the environmental temperature and H represent the relative humidity of soil while C is any obituary constant. The equation show that radon concentration shows variable nature due to the variation in meteorological parameters.

Table 4-8. Correlation table of radon and meteorological parameters from July to September, 2016.

	Radon (Bq.m <sup>-3</sup> )	Pressure (mbar)	Temperature (°C)	Humidity (%)
Radon	1			
Pressure	0.0485	1		
Temperature	-0.3665	-0.0563	1	
Humidity	0.2883	0.2700	-0.5001	1

On 31<sup>st</sup> July, 2016 a special increasing anomaly was recorded in the concentration. To separate this anomaly from the fluctuations that were observed due to the meteorological parameters we have find the average of the whole data with the standard deviation and then we have applied  $\pm 2\sigma$  upper and lower control limit technique to the whole data with an average value of  $\bar{X}$  as shown in Figure4-8:

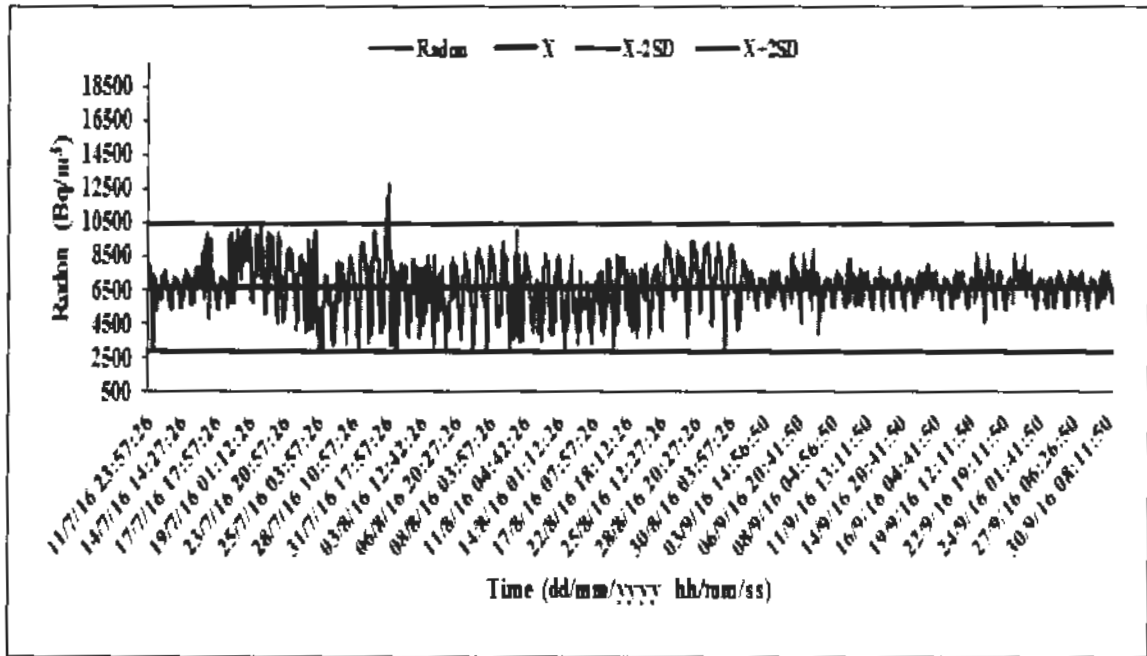


Figure 4-8. Radon concentration variations from July, 2016 to September, 2016 with average value and  $\pm 2\sigma$  upper and lower control limit.

The anomaly that is crossing the upper limit of the  $\pm 2\sigma$  was investigated for the earthquake occurred at Topi town in Swabi the district of Khyber Pakhtoon-Khuwa province of Pakistan. The magnitude of the earthquake was 4.2 with 72.710°N latitude and longitude 33.937°E. The depth of the event was 55.9 km. The earthquake data was taken from USGS (United States Geological Survey). This event happened after two days of this recorded anomaly. According to Surinder et al., 2009 a significant earthquake that can produced the anomaly in radon concentration may follow the following three conditions:

- (A) For earthquake of  $M \geq 2$  the distance may be in the range  $10 < D \leq 100$  km.
- (B) For  $M \geq 3$  the distance may be in the range  $100 < D \leq 200$  km.
- (C) For  $M \geq 4$  the distance may be greater than 200 km [101].

For the current study, the third condition was satisfied as the distance between the monitoring station and epicenter was 107 km. M. Namvaran, A. Negarastani 2013 used the D/R ratio for the detected anomaly that may be in the range  $D/R \geq 0.4$ . where D represent the earthquake preparation zone and R represent the distance between the monitoring station and epicenter of

the earthquake. The earthquake preparation zone was find by the following mathematical relation;

$$D = 10^{0.43M} \quad (5)$$

The earthquake preparation zone for the current study was 63.9734 km with 107 km epicentral distance. The calculated D/R ratio was 0.68 which is greater than 0.4 therefore, the selected event satisfies the desire conditions so the anomaly was concentrated to be due to the selected earthquake event.



Figure 4-9. The earthquake event occurred at Topi Swabi on 31<sup>st</sup> July, 2016.

Hence it is recommended that radon gas concentration show fluctuated nature due to the change in meteorological parameters but show anomalous behavior before a nearly happening earthquake therefore, it can be used as a good tool for the earthquake prediction.

### **4.3 Conclusion:**

The radon concentration variation has been observed for the three-month duration from 1<sup>th</sup> July, 2016 to 30 September, 2016. These variations were analyzed for meteorological parameters and for the earthquake phenomena. The concentration has shown temporal variation for the whole duration due to meteorological but an abnormal variation was detected before an earthquake event of  $M=4.2$ . The detection of this anomaly due to the earthquake phenomena was obtained by the help of  $\pm 2\sigma$  upper and lower control limit, and it was observed that the anomaly was crossing the limit therefore, this anomaly was recommended to be due to the earthquake phenomena. Thus, it is recommended that radon gas showing abnormal behavior before a nearly happening earthquake event, can be used as a good tracer for the same earthquake. Hence it is possible to detect an earthquake event by the help of continuous radon monitoring and with the removal of the remaining factors that influence the radon concentration.

### **4.4 Future Recommendations:**

The continuous radon monitoring with meteorological help in the forecasting phenomena for earthquake events. The removal of meteorological parameters dependence on radon concentration and increasing the monitoring stations enable us to predict earthquakes and to prevent the humanity from this hazard. As the fault line is more porous media therefore, the radon gas exhalation with a greater rate at these areas so the long-term measurements for the variation in radon concentration can also help in finding the hidden fault lines.

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