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Radon Monitoring for geological exploration: A review

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Abstract

Radon is a naturally occurring radioactive alpha particle emitting colorless, odorless and tasteless gas produced by radioactive decay of uranium and thorium. It plays a dual role in man's life, being a fatal health hazard to mine workers and common people living in their homes on one hand and a very useful geological tool on the other hand. As a geological tool radon monitoring technique can be used in uranium and hydrocarbon exploration, earthquake prediction, study of active geological faults and geothermal energy sources. With this technique fault zones have been recognized with fairly good precision worldwide. The technique can also be effectively used in hydrologic research, when studying the interactions between groundwater, streams, and rivers. It has found limited use in geothermal prospecting.

Keywords: Radon gas; Fatal health hazard; Useful exploration tool; Uranium exploration; Earthquake prediction; Study of active geological faults

1. Introduction

Radon is a naturally occurring, radioactive, alpha particles emitting hazardous gas. It is formed in the decay series of both uranium and thorium. In the beginning it was called niton, radium emanation, or just emanation. The radon isotope, ^{222}Rn, was discovered by Dorn in 1901 in the ^{238}U decay series, the radon isotope, ^{220}Rn, (thoron) was discovered by Soddy & Rutherford in 1900 in ^{232}Th decay series, while, the radon isotope , ^{219}Rn, (actinon) was discovered by Giesel in 1902 in ^{235}U decay series (Partington, 1957). Schmidt in 1918 introduced the name radon for the first time and since then the name "radon" has been universally accepted. In the early stages after its discovery, radon was considered as a health giving gas. During the explosive uranium prospecting programs between 1950s and 1960s the importance of ^{222}Rn (radon) as a guide for finding the parent uranium was recognized and a great deal of the information concerning the behavior of the gas shootout from this period (Ball et al., 1991).

Primarily radon was regarded as a quite harmless or even human's friendly part of gases from geological sources. However, in the recent years its significance as the major contributor to the radiation dose received by the human body in the UK has been distinguished. The National Radiological Protection Board (NRPB) has shown that at least 50% of the total dose for an average person in UK is received from combined radon and thoron (Clarke & Southwood, 1989).

Alpha particles emitted by all the three types of isotope of radon gas are comparatively heavy and positively charged ions. They have a very limited penetration limit and if their emitting source is located outside the human body, the particles are easily blocked by the clothes or even by the thin epidermal layer of the human's skin. However, when ingested or inhaled, alpha particles can cause severe damage to stomach or bronchial tissue, because of having restricted penetration limit, thereby, giving-up their entire energy to a relatively small volume of tissue. The major doorway by which alpha activity enters the human body is by the breathing in of radon and, more prominently its immediate daughters which are also alpha particle emitters. Since radon is an inert gas and its half-life is 3.82 days, most of it is breathed out again. The daughter products, however, being solid may remain in the respiratory tract. The major health hazard from radon and its daughters is thought to be an increased risk of lung cancer (O'Riordan et al., 1987).

Although radon by itself is chemically inert gas, its daughters formed due to radioactive decay are radioactive and positively charged and readily attach themselves to small dust particles present throughout an indoor environment. These dust particles are repeatedly inhaled into the lungs or are ingested into the gastrointestinal tract of human beings. The inhaled particles instantly attach to the lung alveoli, whereas, the ingested radon daughters are absorbed into the bloodstream and are finally transported to the lungs. The deposited daughters undergo consequent radioactive decay processes due to the emission of alpha particles which slowly penetrates the inner lung, disorders DNA structure within lung cells, and can potentially provoke lung cancer.

Certain rock types like granite, phosphate, shale, and pitchblende contain high levels of uranium. Radon is constantly being produced in soil and then released to air as a result of the decay of uranium in the soil and rocks near earth surface. Radon in atmosphere is not an issue of health concern because the radon is rapidly diluted to low levels by circulation throughout outdoor air.

The process of transportation of radon gas subsequent to its production at the soil particulate level from a solid to a gas or liquid medium trapped within the pore spaces of soil or rock is termed emanation. This transportation of radon throughout soil is mainly achieved through alpha recoil and the mechanical flow of air and water throughout the soil. Alpha recoil is the process by which a radon nuclide recoils in the opposite direction from the path of alpha particle ejection subsequent to the radioactive decay of its parent atom. Transportation of radon within the soil pores after its release from the mineral grains is then facilitated by the processes of diffusion and convection. The process of the ultimate release of radon from the pore spaces into ambient air is termed as exhalation. Exhalation is the function of the soil porosity, concentration of radon in the soil/gas pores and meteorological factors, including rainfall and atmospheric pressure.

The solubility of radon in water is comparatively low. When groundwater containing dissolved radon reaches at the earth's surface the radon will undoubtedly be out gassed into the atmosphere. Although a large amount of radon present in groundwater will decay before its arrival at the surface, groundwater is, however, still considered to be the second most important source of environmental radon.

In the houses under certain conditions the concentration of radon increases significantly over its usual outdoor level. The majority of houses have a confined air space with restricted air movement and only a slow exchange with external air. Therefore, the concentration of any particulates or gases released into the house environment will tend to increase above the concentration usually found in outside air. The major sources of ^{222}Rn in the indoor air of houses are soil gas emanations from soils and rocks, off-gassing of waterborne ^{222}Rn from taps into indoor air, building materials and outdoor air. Radon entry from the soil inside the house environment occurs principally by the bulk flow of soil-gas motivated by small differences in pressure between the lower part of the house interior and the outdoors. The radon gas penetrates up through porous soils underneath the building and enters through openings and cracks in the foundation or insulation and through transport by means of pipes, drains, walls or other openings. Indoor radon concentration in a house is inversely related to the ventilation rate.

Role of Radon in humans' life is dual. On one hand it causes vital health hazards to not only workers of uranium mines but also to general public living in normal houses and buildings. On the other hand, it helps in mineral exploration, earthquake prediction, study of volcanic activities, and search for geothermal energy sources (Khan et al., 1990; Khan, 1991). It has proved to be a fine friend and an intoxicating enemy at the same time. In this paper the potential benefits of radon and its daughters in the field of geosciences has been briefly outlined.

Several different techniques including scintillation cells, ionization chambers, solid state nuclear track detectors (SSNTDs), solid state surface barrier detectors, thermoluminescent dosimeters and electrostatic precipitation are available for radon measurement (Durrani and Illic, 1997). For developing countries wishing to carry out national survey programs in order to monitor environmental radon levels, the most suitable techniques are those making use of SSNTDs (CR-39 and LR-115) because they are adaptable, simple in handling and processing, low cost and insensitive to beta and gamma radiation. Also these detectors take account of the effects of seasonal and diurnal fluctuation of radon concentrations due to physical and geological factors as well as meteorological conditions [Durrani and Illic, 1997; Gomaa et al., 2006).

2. Radon monitoring for uranium exploration

The conventional exploration methods such as surveys with gamma sensitive instruments are not only costly and time consuming but are also fruitless when targets are deep, mineralization is young or when displacement of an ore body due to remobilization has taken place (Khan et al., 1990). On the other hand, methods based on radon survey provide a potential opportunity to locate uranium deposits buried several hundred metres deep without involving expensive apparatus, time and much finance (Gingrich, 1973; Fleischer and Mogro-Campero, 1981).

Radon monitoring has frequently been successfully applied in exploration for the hidden underground uranium deposits. Since radon gas is a radioactive daughter of uranium, there is a direct relationship between its outflow and presence of a subsurface ore deposits. Qureshi et al. (1988) have carried out an incredible study (Fig. 1) on the uranium ore deposit of Isa Kheil region, Pakistan. In their study the mobilization of uranium ore-body from its original position and its re-deposition at a new nearby location was properly identified on the basis of radon monitoring study at the site of interest. Uranium is usually deposited in a reducing environment below water table. Because of the subsequent lowering of water table, the ore body was mobilized and shifted downward along a southward dipping shale bed. The scintillation counters and borehole logging units indicated a false uranium anomaly at its previous location. However, radon survey with SSNTDs in the suspected as well as the adjoining areas gave two impressions; one with lower radon values corresponding to the previous location of the ore body and one with the high radon anomaly corresponding to its present day location. Mobilization of the ore body was confirmed by exploratory drilling within the region. This method has been very successfully applied in the field during the last couple of decades or so (Fleischer et al., 1975; Gingrich & Fisher, 1976a, b; Miller and Ostle, 1976). Khan (1993) has outlined some successful applications of alpha sensitive plastic film (ASPF) method for U-exploration in the World as below:

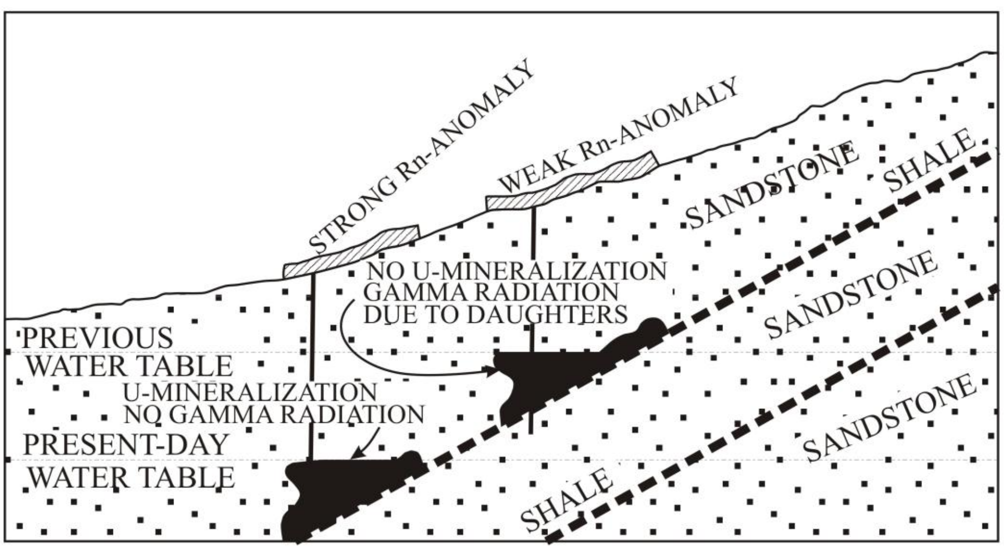

Fig. 1. The mobilization of uranium ore body detected by radon measurements using SSNTDs (after Qureshi et al., 1988).

1. Applied in "Unraven Mineral Belt" of the Colorado Plateau:
   - A number of anomalies were discovered, 20 highly significant anomalies resulted from these investigations.
   - A number of the ore bodies so discovered had nearly the same shapes and sizes as those of radon anomalies.
2. In Northern Wyoming:
   - A number of anomalies were found.
   - Almost every "Good Potential Target Point" intersected uranium mineralization.
   - The depths of these mineralizations ranged between 15-50 meters.
3. At Cloff Lake in Athabasca Basin (Canada):
   - Many ore bodies as deep as 120 metres were discovered. Radon maps defined the "Surface Lithology" of the ore bodies so discovered.
   - He- and Rn- "Emanometry" could not produce any meaningful results in these cases.
4. In about 1000 square metres in basin and range (Arizona):
   - Six major anomalies were discovered.
   - Follow up work helped in locating a 300 metres thick layer of low mineralization in this area.

The radon results achieved at Peralillo area in Chile has yielded favorable results from surveying on three of the four target zones. These results are significant in that they confirm the presence of uranium ore deposits in the region (Radon anomalies identified at Peralillo, 2009).

3. Radon emission as precursor of earthquake

Application of radon monitoring was recognized as a positive tool for prediction of an earthquake, when Okabe studied the correlation between radon content variation and local seismicity in Japan (Okabe, 1956). The strain changes occurring within the earth's surface during an earthquake is expected to enhance the radon concentration in soil gas (Ghosh et al., 2009). The principle seems to be simple: Radon gas which is trapped within the ground, is released through small fractures resulting from many changes taking place in the earth's crust in that region prior to the major physical shock of an earthquake. The stress–strain developed within earth's crust before an earthquake leads to changes in gas transportation and rise of volatiles from the deep earth to the surface (Thomas, 1988; Fleischer, 1997). As a result, remarkable quantities of radon come out of the pores and fractures of the rocks on surface. Thus, due to the seismic activity, changes in underground fluid flow may account for anomalous changes in concentration of radon and its daughters (Steinitz et al., 2003). An increase in radon level occurs in the region of compression and radon concentration decreases in the region of dilation. As small changes in gas flow velocity causes significant change in radon concentration, soil radon monitoring is thus an important way to detect the changes in compression or dilation associated with an earthquake event (Grammakov, 1936; Clements, 1974). Under favorable conditions a careful measurements of radon intensity (at a fixed point) as a function of time should correlate these changes with seismic activity in the area. Research work has given a strong indication that radon/thoron sensitive detectors can be used in the prediction of earthquakes (Fleischer et al., 1975; Campero et al., 1980). The variation of radon concentration has been monitored in soil gas as well as ground water. It is necessary to measure changes of radon before an earthquake reducing the effect of meteorological parameters.

The anomalies associated with an earthquake may disappear after the quake for the following reasons (Ghosh et al., 2009):

- Stress relaxation associated with earthquake
- Healing of fault zone
- Exhaustion of gas supply

If multiple observation stations for monitoring radon gas are established, more accurate earthquake prediction in terms of magnitude and location could possibly be made (Chyi et al., 2005). The new integrated approach in soil gas radon monitoring are superior precursors to delineate the time and place of an earthquake (WIPO IP Services, 2010). Use of multiple units of this system that are placed at different locations, make it possible for the time, place, and even magnitude of earthquakes to be predicted. Housing the detecting system inside a PVC pipe and retrieving the data electronically without removing the PVC pipe cap reduced the influence of environmental factors dramatically. The radon detecting system consists of a silicon photodiode detector, an interface, and a data logger. The system/device 10 of the present invention is capable of recording radon flux changes of less than minute in duration. By connecting the data logger to a modem and telephone line, the data could be retrieved at a remote site at any time, and preferably, in a real time manner.

The radon abnormality 3–8 days prior to earthquakes with magnitudes of 3–3.5 was reported by Antsilevich in 1971. He found that radon anomaly was about 20% above the background value. Afterwards many important studies have been performed worldwide regarding the earthquake prediction by observing radon anomaly in soil gas and ground water. Some of such studies have been briefly mentioned below:

3.1. Pakistan

Khan (1993), using CN-85 track detectors, established radon monitoring stations for earthquake prediction in Islamabad, Pakistan. Two radon measuring stations 0.5 km apart were set up for earthquake prediction experiment. At each station a 50 cm deep hole with diameter of 6 cm was prepared and a PVC pipe was fixed in it so as to avoid the collapse of the hole. The detectors were replaced after every 24 hours. In seven cases of earthquake occurrence, an associated radon level increase was observed either during or immediately after the jolt. In one case the radon level started rising a few days before the jolt and continued to raise a couple of days after it, before coming down to normal level. In two cases there was no radon fluctuation observed. One instance of high radon emanation was not associated with any seismic activity.

3.2. Italy

The radon concentration in well water was monitored in central Italy during 1978–1980 by Allegri et al. (1983). They observed an anomalous increase of about 25% and 17% above background, before the Irpinia earthquake of M=6.5 on 23 November, 1980, 250 km away from the site.

On April 6, 2009 a moderate sized (Mw 6.3) earthquake occurred near the town of L'Aquila in central Italy. This earthquake caused 295 deaths and significant damage of about US$ 16 billion in the local area (Mori, 2010). On March 28, 2009 Giuliani, a technician at the nearby Gran Sasso National Laboratory in Abruzzo, announced an earthquake prediction for the town of Sulmona located about 50 km SE of L'Aquila on the basis of high radon signals. The earthquake was posted on a web page and broadcasted through loudspeakers mounted on vehicles. Local government officials warned Giuliani to stop distributing the information about the prediction. The actual earthquake took place at a location 50 km away from the predicted location. When the earthquake occurred, the perception is that the prediction was correct (Mori, 2010).

3.3. Japan

Hirotaka et al. (1988) observed radon anomaly before the Nagano Prefecture earthquake of M=6.8 on 14 September, 1984 and the measuring site was about 65 km away from the epicenter at the Atotsugawa fault. They observed a gradual increase in radon concentration three months prior to the quake and a remarkable increase 2 weeks before of the shock.

3.4. India

Virk and Singh (1994) recorded spatial and temporal distribution of radon in both soil gas and groundwater. Radon recording stations were set up at one site in Amritsar and four sites in the Kangra valley (Himachal Pradesh) under the Himalayan seismicity project. The Uttarkashi earthquake (mb = 6.5, MS = 7.0) occurred on October 20, 1991 in the Garhwal Himalayas (30.78°N, 78.77°E) about 330 km from the recording stations in the Kangra valley and about 450 km from Amritsar, respectively. Radon anomalies were recorded at all sites in Kangra valley and Amritsar about a week before the Uttarkashi earthquake, which clearly establishes that radon changes can be effectively used for forecasting some earthquakes (Virk and Singh, 1994).

Ghosh et al. (2007) have conducted an experiment on measuring radon concentration in soil gas with the use of CR-39 detector at Kolkata, India. Radon anomalies before the earthquakes that occurred during the period of November 2005 to October 2006 within the range of 1000 km from the measuring site and of M ≥4 were observed 7–11 days prior to the earthquakes.

3.5. China

A major earthquake measuring 7.9 M on the Richter scale struck the highly populated Sichuan Province in SW China on May 12, 2008. This earthquake, with an estimated death toll of over 60,000 in Sichuan alone, was also felt at Beijing (Das et al., 2009). Strong anomalies in the concentration of helium, radon and gamma were observed in gases at the geochemical monitoring station, Bakreswar, West Bengal, India, about two weeks before the earthquake (Das et al., 2009). The distance between the epicenter of the earthquake and the monitoring site is about 1800 km. This long distance preseismic observation indicates that the radius of influence of large magnitude earthquakes may be substantially large and may cut across plate boundaries.

Similar studies have been conducted in several other countries and positive correlations were found in majority of the cases between the radon build-up and earthquake occurrence.

4. Use of radon monitoring in hydrocarbon exploration

Traditionally, exploration of oil and gas reservoirs have principally relied on seismic or geological prospecting and drilling to look for anticlinal traps. Because many of seismically discovered anticlinal traps are usually found to be barren after drilling there has not always been a success in exploring for the anticlinal structures through the traditionally used costly techniques of prospecting. Recently, there have been developed a series of cheap oil and gas prospecting methods based on the isotopic anomalies resulting from the geochemical effects of hydrocarbon seeping from an oil or gas bearing trap. Radon monitoring survey over hydrocarbon reservoirs is one of the several cheap techniques used for locating hidden subsurface oil and gas bearing structures. Radon gas trapped in oil and gas bearing reservoirs can, under favorable circumstances, migrate to the earth's surface (Donovan, 1974; Donovan and Dalziel, 1977; Fleischer and Turne, 1984). Experiments have been carried out in the USA for finding the correlation between radon anomalies and the oil and gas deposits. Investigations based on measurements carried out over a period of two months or so in Oklahoma (USA) indicated a strong correlation between the oil reserves and the intensity of radon signals (Donovan, 1974).

In general, anticlinal traps give halo patterns with low radon values at the centre and high values outside at the boarder (Zuhui et al., 1993; Zhongjun et al., 1995; Sikka and Shives, 2001). Sketches of such halos have been shown in the Fig. 2 (Zuhui et al., 1993) and Fig. 3 (Sikka and Shives, 2001), respectively.

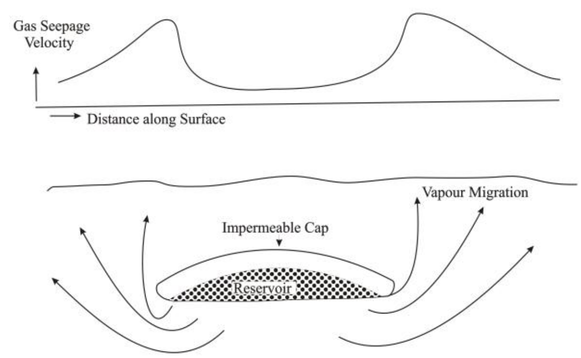

Fig. 2. Sketch of gas seepage from a simplified hydrocarbon Reservoir. The gas emission at the surface of the earth is indicated in the graph. “Reservoir” refers solely to hydrocarbons (modified after Zuhui et al., 1993).

Zuhui et al. (1993) carried out radon survey with CR-39 detectors in both Saihantala Hollow, Inner Mongolia and Weibel Hollow, Shandong and discovered few oil and gas deposits trapped along with their anomalies. From the radon survey results they concluded that this technique can be widely used in oil and gas exploration survey in future and that this method must be combined with the other chemical, physical, and geological prospecting methods to give a correct oil deposit report.

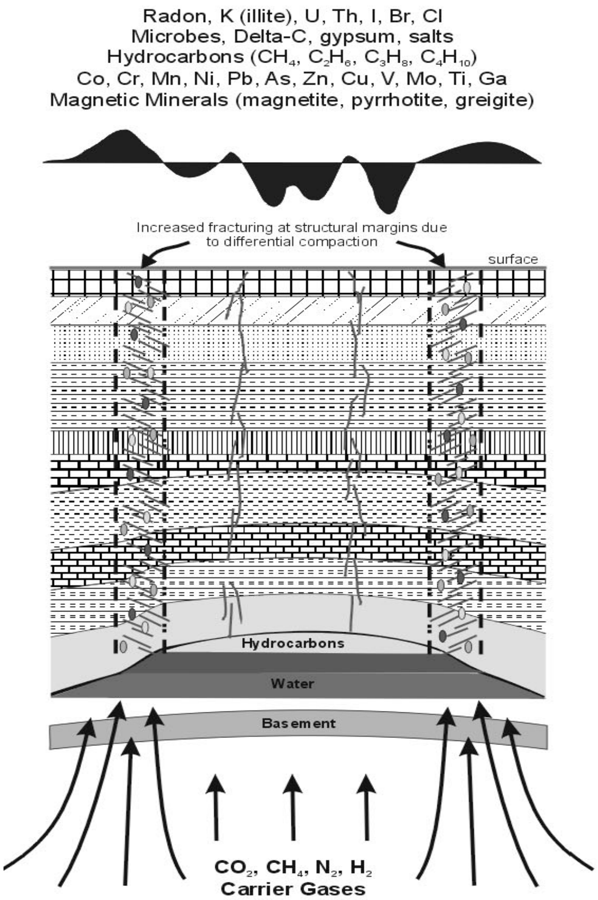

Fig. 3. The proposed working model for the formation of hydrocarbon, radiometric and geochemical anomalies at the earth's surface (after Sikka & Shives, 2001).

5. Application of radon monitoring in locating geological faults

Radon gas generated within the earth crust can travel upward through faults, fractures and other weak zones over long distances and can be detected by the radon measuring techniques. Based on this property of the radon, it is possible to locate subsurface faults and similar structures (Qureshi et al., 1991). Ambron (1921) has probably been the first to document increased levels of radon concentration over geological faults. Since then the technique has been extensively used for locating geological faults in different parts of the world including Pakistan (Qureshi et al., 1991), Canada (Gascoyne et al., 1993), France (Borchiellini et al., 1991), Italy (Ciotoli et al., 1993) and Britain (Duddridge, 1994) amongst many others. Some of the worldwide examples of locating faults with the radon monitoring technique have been mentioned below:

5.1. Pakistan

The Main Boundary Thrust (MBT) is a major fault of Indo-Pakistan which runs all along the foot hills of Himalaya from Assam in the east up to Kurram Agency in the west through north of Islamabad, the capital of Pakistan. The main boundary faults in selected regions were demarcated with alpha sensitive plastic detectors in India by Ramola et al. (1988), and in Pakistan by Qureshi et al. (1991). In Pakistan track detectors were placed in tubes, which were buried at regular intervals along a line crossing the expected fault zone. The detectors closest to fault region showed alpha track density well above the background level indicating well defined radon anomaly (Fig. 4).

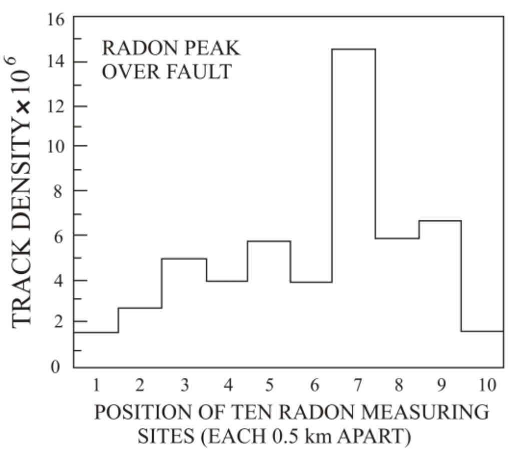

Fig. 4. Diagram showing radon peak over Main Boundary Thrust near Islamabad (after Qureshi et al., 1991).

5.2. India

Extensive work has been conducted to locate subsurface faults and fractures in the different localities of India from time to time by various investigators using the radon monitoring technique.

5.2.1. Dharamsala Area

One recent example of radon survey undertaken for assessing the relationship between soil-gas radon/helium distribution and thrust/neotectonic fault zones is in the vicinity of tectonically active area of Dharamsala in the region of NW Himalayas, India (Walia et al., 2008). Elevated levels of radon and helium in the soil gas were found along a profile of a major fault (MBT-2).

5.2.2. Nadha Area

Nadha area is located NE of Chandigarh city near Panchkula in the neighborhood of the Himalayan Frontal Fault (HFF). The area marks the southernmost edge of the Himalaya, where the un-deformed succession of the Indo-Gangetic Plains is separated from the detached, complex folded-faulted Upper Siwalik Hills comprising molassic sediments of lower Pliocene–early Pleistocene age. The boundary is well-defined by the HFF system. The folding of Siwalik bedrock north of the HFF and the occurrence of large historical earthquakes that apparently have not broken the surface, have been the basis to suggest that the HFF is a blind thrust (Stein and Yeats, 1989; Yeats et al., 1992; Yeats and Thakur, 1998). To determine the relationship between the recently developed tectonic features with soil gas radon/helium variations, a survey was conducted in the study area by Mahajan et al. (2010). The Nadha area shows high values of radon and helium concentrations along/near the Himalayan Frontal Fault as compared to the adjoining areas. This indicates the presence of some buried fault/fault zone running parallel to the HFF, not exposed to the surface and not delineated by satellite data but is geochemically active and might be tectonically active too.

5.3. Jordan

Radon emanation measurement study was carried out in five locations in a limestone quarry area using SSNTDs CR-39 (Al-Tamimi and Abumurad, 2001) in Jordan. Radon levels in the soil air at four different well-known traceable fault planes were measured along a traverse line perpendicular to each of these faults. Radon levels at the fault were higher by a factor of 3–10 than away from the faults (Fig. 5). The method was also applied along a .fifth inferred fault zone. The results show anomalous radon level in the sampled station near the fault zone, which gave a radon value higher by three times than background. This study indicates that in Jordan many cities and villages have been established over an intensively faulted land. Moreover, radon gas is proved to be a good tool for fault zones detection.

5.4. Spain

Soil radon levels have been measured across the Amer fault, which is located near the volcanic region of La Garrotxa, Spain (Font et al., 2008). In this survey 27 measurement points were selected in five lines perpendicular to the Amer fault in the village area of Amer. The averaged results show an influence of the distance to the fault on the mean soil radon values (Fig. 6). The results obtained support the hypothesis that the fault is still active.

6. Prediction of geothermal energy sources with radon monitoring

A geothermal energy source is the natural source of heat inside the earth sufficiently close to the earth's surface to be taken out for utilization inexpensively. This thermal energy contained in the earth; can be used either directly to heat rooms and to provide warm water in bathrooms in cold winter season or can be converted to mechanical or electrical energy. Valuable information determining the potential drilling targets can be obtained by carrying out radon monitoring of the area of interest. In principle, radon buried deep in the earth crust finds geological faults (usually associated with geothermal sources) as easy routes to reach the top surface (Whitehead, 1981). Strong radon anomalies in volcanic regions may indicate the presence of geological faults in geothermal fields (Khan et al., 1990; Hussein, 2008). This method of using radon signal for locating geothermal energy sources has met some success in countries such as New Zealand, Mexico and USA (Fleischer, 1988; Whitehead, 1981). Radon monitoring was carried out in El Salvador (a city on the northeast coast of Brazil) using plastic films which confirmed the existence of active faults and two producing geothermal wells were located (Balcazar et al., 1993).

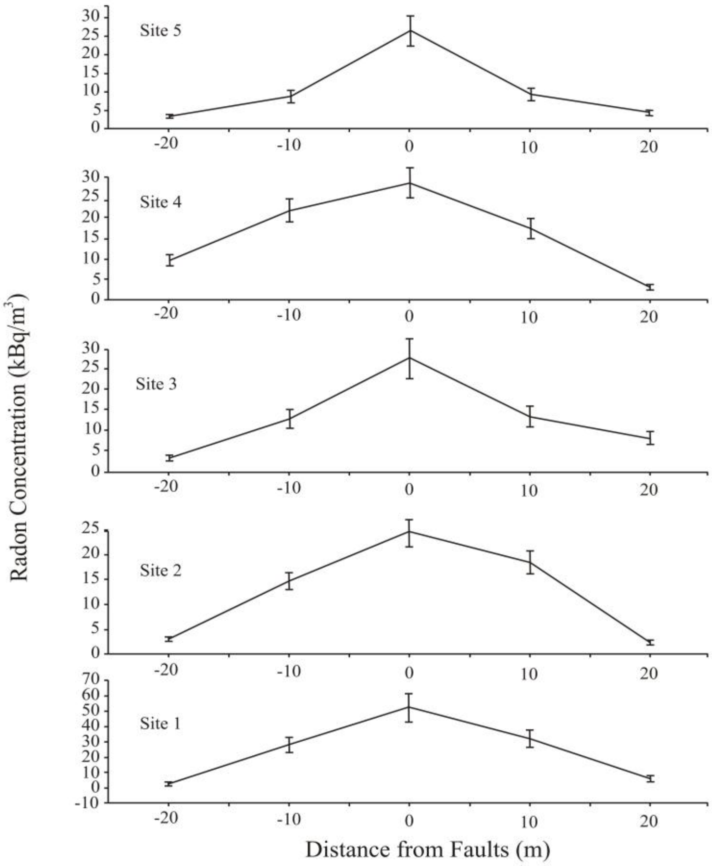

Fig. 5. Measured radon concentration values in soil gas from Jordan (after Al-Tamimi & Abumurad, 2001).

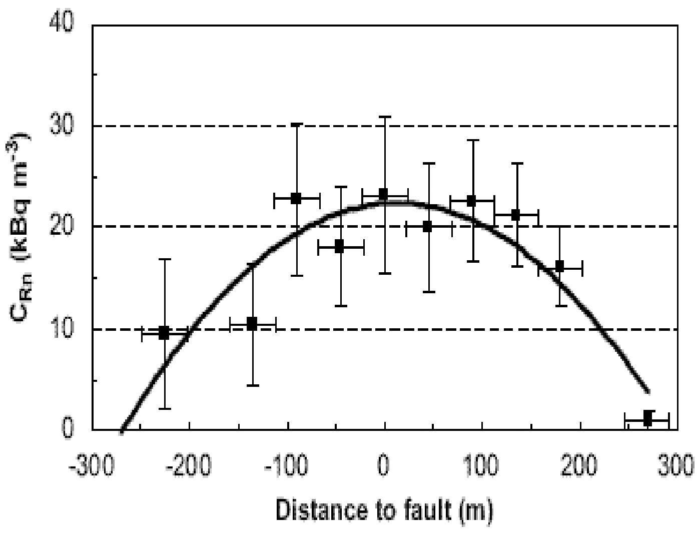

Fig. 6. Mean soil radon concentration versus the distance to the fault obtained with the LR-115 detectors in all transverse lines. The solid line is the second-order polynomial fit (after Font et al., 2008).

7. Hydrologic Studies

Because of its rapid loss to air and comparatively rapid decay, radon is used in hydrologic research that studies the interaction between ground water, streams and rivers. Any significant concentration of radon in a stream or river is a good indicator that there are local inputs of ground water (Radon Applications, 2010).

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