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Measurement of Radon Exhalation Rates from the Samples of Soil & Rocks

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ABSTRACT

Radon and its daughter products are the major sources of radiation exposure and recognized as one of the health hazards for human beings. In the present work, I have reviewed 20 papers in which the soil samples are collected to different places of India and other countries. The Effective radium and radon exhalation rates in the samples of soil & rock have been used passive techniques for alpha particles emission with "Closed Can Technique", "RRC passive Technique", "Alpha guard equipment and Gamma tracer". The soil samples belonging to different places of India and other countries. All the values of radium content in soil samples of study area were found to be quite lower than the permissible value of 370 $BqKg^{-1}$ recommended by Organization for Economic Cooperation and development (OECD1979). The highest activity

***KEYWORDS:** Surface exhalation rate, mass exhalation rate, soil, plastic track detector etc*

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1. INTRODUCTION

The radioactive decay of naturally occurring uranium in the earth's crust leads to radon in environment, Soil, Ground water etc (Chauhan,2011) Radium, in the form of Radium chloride was discovered by Marie Curie and Pierre Curie in 1898 (Barooah, 2013). They extracted the radium compound from uraninite and published the discovery at the French Academy of Science after five years. We know that Radon is a decay product of radium, which decays after completing its half life directly into radon. Radon is produced continuously from the decay of naturally occurring radionuclide such as U^{238}, U^{235} and Th^{232}, ^{222}Rn, one of the three isotopes of Radon – ^{219}Rn(action), ^{220}Rn(Thoron), ^{222}Rn(Radon) is an α- emitter that decays with a half life of 3.82 days. Radon beings 8 times heavier than air. Radon is a chemical element with atomicnumber 86. It is a radioactive, colorless, odorless, tasteless noble gas, occurring naturally (B P Singh, 2010).

Soil is the main source of continuous radiation exposure to the human beings. When radium decays in soil grains, the resulting atoms of radon isotopes must first escape from the mineral grains to air filled pores. The rate at which radon escapes or emanates from solid into the surrounding air is known as radon emanation rate or radon exhalation rate of the solid. This may be measured by either per unit mass or per unit surface area of the solid.

The measurement of radon exhalation rates of soil and rocks are helpful to study radon health hazard. When radon inhaled, α- particles emitted by its short lived decay products can damage the cellular DNA mainly (WHO 2009, UNSCEAR 2008)

In present paper, "Closed Can Technique", "RRC passive radiometer", "Alpha guard equipment & Gamma tracer", is used to measure radon exhalation rate from the samples of soil & rock. The main aim of study is the possible health risk to associate with the radon activity.

2. Method and materials Used:

After the study of various literature papers, it has been found that there are various methods available for the measurement of the radon exhalation rate in some soil & rock samples. These various methods such as:

1. Closed Can Technique.
2. RRC passive radiometer.
3. Alpha guard equipment and Gamma Tracer was applied for gamma dose rate measurements.

2.1. Closed Can Technique:

This technique is used to find the radon exhalation rate from the soil samples. The 40 soil samples were collected in clean, dry polyethylene bags. The soil was collected from an auger hole at a depth of ~0.75 m from the ground so as to get the natural soil. After removing the stones and organic materials, samples are crushed into fine powder by using mortar and pestle. Fine quality of the sample is obtained using scientific sieve of 150-µm-mesh size. Then, the samples were dried in an oven at ~ 110 ºC temperatures for 24 h to remove the moisture content. A glass bottle of ~1 L capacity was used as an emanation chamber. The fine powder (0.25 kg) of samples were placed at the bottom of the glass bottles and sealed with thin polyethylene sheets for 1 month so as to attain the equilibrium between radium-radon members of the decay series. After one month, **LR-115 type II plastic track detectors** of size 1.5 cm × 1.5 cm were fixed on the lower side of cork lids, which are then gently pressed against the polyethylene sheets on the glass bottles (acting as emanation chambers) so that the equilibrium is not disturbed or there is minimum possible disturbance, if any. After properly tightening the cork lids, the chambers were sealed with adhesive tape to minimize the leakage and left as such for 3 months so that the detectors can record alpha particles resulting from the decay of radon (Mehra et al., 2006).

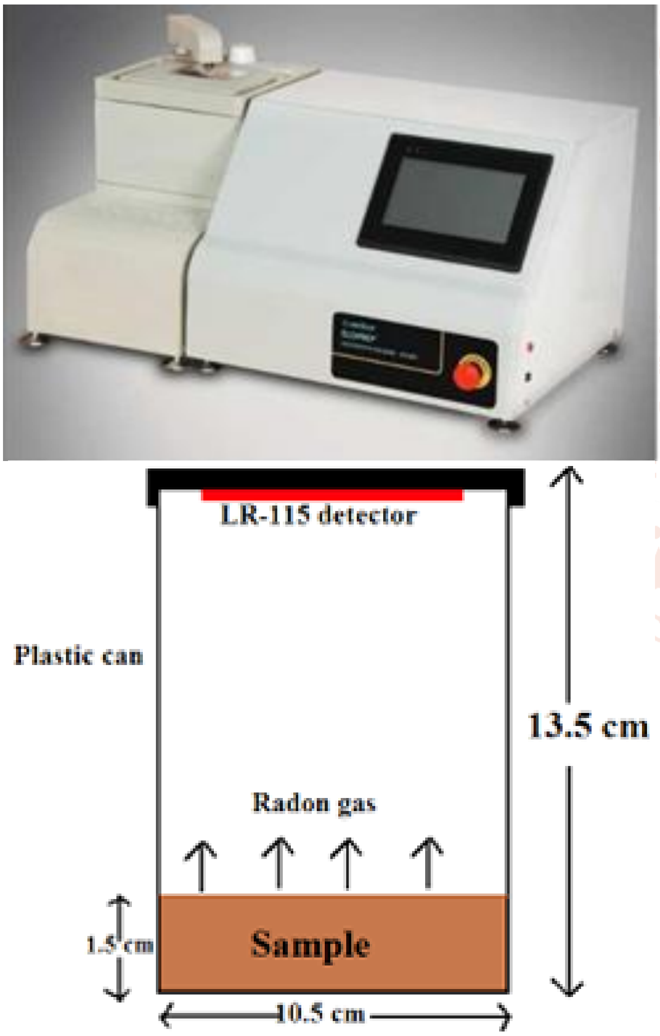

Fig. 2.1: Assembly for the measurement of radon exhalation rate using 'Closed Can Technique' and Etching bath

After exposure, the detectors were removed and etched in 2.5N NaOH solution at 60ºC for 90 min, using a constant temperature bath. The tracks were counted using an optical microscope at a magnification of 400 ×. The resulting track density was then converted into concentration (Bqm^{-3}) by appropriate calibration factor.

Calculation of Radon Exhalation Rate in terms of mass and area

The radon exhalation rate in terms of mass is calculated [Abu- Jarad (1988) and Khan et.al (1992)] from the expression:

$$E_M = \frac{CV\lambda}{[T + 1/\lambda\,(e^{-\lambda t} - 1)]}$$

The radon exhalation rate in terms of area is calculated from the expression:

$$E_A = \frac{CV\lambda}{[T + 1/\lambda\,(e^{-\lambda t} - 1)]}$$

Where E_A is the radon exhalation rate in terms of area ($mBqm^{-2}hr^{-1}$), E_M is the radon exhalation rate in terms of mass ($mBq.kg^{-1}.hr^{-1}$), M is the mass of soil sample (0.25 kg), A is the area covered by the Can or surface area of the sample. C is the integrated radon exposure as measured by LR-115 plastic track detector ($Bqm^{-3}hr$); V is the effective volume of the bottle (m^3), T is the exposure time (h), λ is the decay constant for radon (h^{-1}).

2.2. RRC passive radiometer:

Radon concentration in soil is measured using the RRC passive radiometer (Uvarov and Kulakov, 1995), specially designed to prevent thoron, dust and moisture penetration into the detector chamber. The radiometer has an aluminum cylindrical chamber (inner diameter of 3.2 cm and volume of 17 cm^3) and a cover, with a plastic layer between them. There are twelve holes on the chamber wall for radon entrance, each one 6 mm in diameter, which are covered with a 35 µm thick polyethylene layer. The detector in a drum-type support is fixed to a central holder. The holder is lined with 13 µm thick aluminized Mylar absorber, which decreases α-particle energy and protects the **cellulose nitrate detector** (CND) from spontaneous electrostatic charges. Radon penetrates into the radio meter through a polyethylene layer by diffusion. Decay products plate out on the inner surface of the radiometer. The CND used (Russian-made, K-8 type, thickness of 13 µm, density 1.49 ± 0.01 g/cm^3) enables only detection of α-particles emitted by radon products which are plated out on the Al-Mylar. The radiometer is very convenient for measurement of radon activity in wet soil. Its sensitivity is 0.7 ± 0.2 ($track/cm^2$)/($kBqhm^{-3}$), when chemically etching CND for 100 minutes in 5 N NaOH solution at a temperature of 50 °C. The background of the CN detector is then 70 ± 20 $track/cm^2$. Detectors are read out with a spark counter. Error of radon measurement is about 30 %. Radon concentration in soil gas strongly depends on the depth in soil, increasing with depth (Jonsson, 1995. Detectors are exposed for two weeks in the winter season.

Cellulose Nitrate Plastic Track Detector

The cellulose nitrate (CN) plastic detector, commercially known as LR-115 Type-II with chemical composition $C_6H_8O_9N_2$, manufactured by Kodak Pathé, France has been used to detect the alpha particles emitted by radon, thoron and their progeny. It is a film made of clear polyester base 100 µm thick (etchable) on which a red layer of cellulose nitrate of thickness 11.5 to 12 μm is coated. LR-115 Type II plastic track detector mainly detects the alpha particles having energy ranging from 1.7 to 4.8 MeV (Jonsson, 1981; Abu-Jarad et al., 1980). Therefore, any other alpha particle emitted from radon daughters will not be registered on the surface of LR-115 Type II plastic track detector because of their alpha energies (6.0 and 7.68 MeV from ^{218}Po and ^{214}Po, respectively) being more than its upper threshold energy. Advantages of LR-115 Type II detectors are mostly unaffected by humidity, low temperature, moderate heating and light (Durrani and Ilić, 1997). LR-115 films are not affected by electrons or by radiations in the electromagnetic spectrum (such as gamma rays, X-rays, ultraviolet or infra-red rays).

2.3. Alpha guard equipment and Gamma Tracer was applied for gamma dose rate measurements:

The measurement set-up to analyse radon concentration in soil gas C_{Rn} (Bqm^{-3}) consisted of an Alpha Guard PQ 2000 PRO (AG) radon monitor, a soil-gas probe and an Alpha-Pump (AP) (Genitron, Germany). Soil gas was pumped through the AG ionization chamber at a flow rate of 0.3 dm^{3} min^{-1}. The temporary radon (^{222}Rn) concentrations were registered in one-min intervals over approximately a 20-min period. After initial growth, the concentration became stabilised. The average of the last few stabilised values was taken as the radon concentration in soil gas. At this low flow rate, contribution of thoron (220Rn, half-life 55 s) was negligible (Zuni´c et al., 2006).

Calculation of Radon exhalation from soil: -

The radon exhalation rate E_{Rn} (Bqm^{-2} s^{-1}) from soil was measured using the Exhalation Box (EB) and the same AG monitor and AP pump. The air was circulated in the closed circuit for about 90 min and the concentration of radon accumulated in EB was recorded every 10 min. The exhalation rate was calculated according to the formula:

ERn =B·(V/F)

Where:

B – Slope of the straight line fixed to the increasing radon concentration points in the EB,

V – Volume of the EB (m^{3}),

F – Surface area covered by EB (m^{2}) (Zuni´c et al., 2006)

Soil permeability

The system to measure soil permeability k_{soil} (m^{2}) consisted of a Multi sensor Unit D/D device (Genitron, Germany) and the same AG monitor, AP pump and soil-gas probe. Soil gas was sucked from soil by soil gas probe and pumped through the AG and Multi sensor. The pressure difference between soil air and open air (1P) and flow rate (Q) were measured by the Multi sensor D/D. The soil permeability was calculated using a modified equation of Fick's law of diffusion (Janik, 2005):

ksoil =μ Q /W.ΔP

in which: k_{soil} is soil permeability (m2), μ is dynamic viscosity of air (Pa s), W is shape parameter of the soil-gas probe (m), Q is soil gas flow rate (m3 min−1), and 1P is pressure difference measured (Pa) (Zuni´c et al.,2006).

Gamma dose rate

Gamma dose rate Hγ (nSv h^{-1}) was measured in outdoor air at the height of 1m above the ground using a Gamma Tracer TM Wide Type E probe (Genitron, Germany). The values of gamma dose rate were registered in 5-min intervals. The average value of 12–15 records was taken as a final result.

3. Result and Discussion for these review papers :

In the present study, I have reviewed 20 papers for different authors at different areas to measure the radon exhalation rate from the soil samples. The values of radon activity and radon exhalation rate in soil samples from different areas of National and International are given in Table 1. The worldwide range of activity concentrations of ^{226}Ra, ^{232}Th and ^{40} K reported by UNSCEAR 2000 varied from 17 to 60 Bqkg^{-1}, 11 to 64 Bqkg^{-1}and 140 to 850 Bqkg^{-1}, respectively. The 'Closed Can Technique', 'RRC passive radiometer', 'Alpha guard equipment, & Gamma Tracer was applied for gamma dose rate measurements.' has been used for the measurement of radon exhalation rate in soil & rock samples. The radon exhalation rate varies appreciably from one place to another. This variation may be due to the difference in radium content and porosity of the soil.

Table: 3.1 Different National and International Locations with Radon surface and Mass

Author's Name	Locations	Radon Mass Exhalation Rate (Bqkg^{-1}d^{-1})			Radon Surface Exhalation Rate (Bqm^{-2}d^{-1})		
		Min. Value	Max. Value	Avg. Value	Min. Value	Max. value	Avg. value
M. Shakir Khan	Uttar Pradesh	2.38*10^{-6}	3.86*10^{-6}	2.99*10^{-6}	6.19*10^{-5}	10.03*10^{-5}	7.77*10^{-5}
A.K. Choudhary	Banda district of Uttar Pradesh	2.1	10.6	5.8	84.0	419.8	231.3
Abbas J Al Saadi	Kerbela Iraq	29.715*10^{-3}	70.237*10^{-3}	48.409*10^{-3}	0.9463	2.2369	1.4785
Ajay Kumar	Kapurthala district, Punjab	2.52	7.07	-	98.18	234.37	-
S. SankaranPillai	Tiruchirappalli dist., Tamil Nadu sedimentary rocks)	1.8±0.1	29.2±2.6	6.2	13±0.3	215.4±12	46.2

S. SankaranPillai	Tiruchirappalli district, Tamil Nadu (igneous rock)	13.1	156.2	31.5	94	1127	387.8
AbdElmoniem A. Elzain	Singa&Rabak Towns, Sudan	335±38	370±43	354±36	16.6±1.9	18.4±2.1	17.6±1.8
M.Zubair	Bulandshahar district, U.P.	15.1	30.7	23.1	394.1	798.3	600.7
Naveen Thakur	Jwalamukhi Thrust in Himachal Pradesh	1.0241E-07	7.2414E-08	2.64855	2.03197E-06	4.9388E-07	3.564976E-06
Vikas Duggal	Northern Rajasthan	8.27	23.19	14.96± 3.93	273.80	768.04	495.32±130
R P Chauhan	Aravalli hills in India	23.1	50.5	34.6±5.7	493	1106	746±128
H K Sarma	Noonmati, Guwahati, Assam	3.32	3.99	-	117.48	139.858	125.88

DISCUSSION OF RESULT

Radon mass exhalation rate for the whole of the studied area ranges from 8.27 to 23.19 mBqkg^{-1}h^{-1} with an average value of 14.96 mBqkg^{-1}h^{-1}. The measured values of radon concentration and exhalation rate generally in the recommended limit by (UNSCEAR 2000).

The average values of radon concentration in soil samples are lower than the recommended limit 800 Bqm^{-3} which, reported by (WHO, 1993).

The results of radon concentration, its exhalation rate and radium content in soil samples collected from different locations of refinery area belonging to Noonmati, Guwahati, Assam. The radon concentration varies from 380.95 to 457.14 Bq/m^{3}. The radon exhalation rates in terms of area and in terms of mass vary from 117.48 to 140.97 mBqm^{-2}hr^{-1} and 3.32 to 3.99mBqkg^{-1}hr^{-1}. Twenty-two samples were collected from industrial area of Bulandshahr, Hapur and Meerut districts of Uttar Pradesh (India). The radium concentration ranges from 9.2 to 18.7 Bqkg^{-1}with an average value of 14.1 Bqkg^{-1}. The area exhalation rate for radon ranges from 394.1 to 798.3 mBqm^{-2} h^{-1} with an average value of 600.7 mBqm^{-2} h^{-1}and mass exhalation rate ranges from 15.1 to 30.7mBqkg^{-1}h^{-1}with an average value of 23.1mBqkg^{-1}h^{-1}.

The present work is devoted to measure radium concentration and radon exhalation rates of soil samples collected along Jwalamukhi Thrust in Himachal Pradesh, India. Highest activity was found with radium concentration 0.170×10^{-3} Kg^{-1}h^{-1}.

The radium concentration and radon exhalation rates in soil samples collected from Kerbala Governorate area. The Radium concentration varies from (1.1001- 2.6003) Bqkg^{-1} with an average of 1.7921Bq kg^{-1}. The radon exhalation rate in terms of area varies from (0.9463 – 2.2369) Bqm^{-2}h^{-1} with an average of 1.4785Bqm^{-2}h^{-1}, while radon exhalation rate in terms of mass varies from (29.715– 70.237) ×10-3 Bqkg^{-1}h^{-1}with an average of 48.409×10-3Bqkg^{-1}h^{-1}.

The values of radium concentration in all the soil samples were less than the recommended by Organization for Economic Cooperation and Development (OECD) 1979. Also, the radium concentration and radon exhalation rate in these samples has been found to be well below of 40 Bq/kg and 57.6 Bqm^{-2}. s, respectively.

in the states of Haryana and Himachal Pradesh by Chauhan et al. (2013), Kangra district, Himachal Pradesh by Sharma et al. (2003) and the Bathinda district of Punjab by Singh et al. (2005), but are higher than those reported in Tusham ring complex, Haryana by Singh et al. (2008).

Table 3.2: Values of concentration and exhalation rate of Radon & Radium concentration in different countries as compared with other study.

Country	Area Exhalation + S.D ($Bqm^{-2}h^{-1}$)	Mass Exhalation + S.D ($mBqKg^{-2}h^{-1}$)	Radium Concentration + S.D ($BqKg^{-1}$)	References
India	0.76	2.3	19.6	**B. P. Singh, B Pandit, V N Bhardwaj, Paramjit Singh, Rajesh Kumar**, "*A Study of radium and radon exhalation rate in some solid samples using solid state nuclear track detectors*", **Indian Journal of Pure & Applied Physics, 48, pp. 493-495, 2010**
Saudi Arabia	8.4	251	3.3	**S. M. Farid, "***Indoor Radon in dwellings of Jeddah city, Saudi Arabia and its Correlations with the radium and radon exhalation rates from soil,*" **Indoor and Built Environment, DOI: 10.1177/1420326X14536749, 2014.**
Egypt	-	-	-	**K. A. Korany, A. E. Shata, S. F. Hassam, M. S. E. Nagdy,** "*Depth and seasonal Variations for the soil Radon -Gas Concentration Levels at Wadi Naseib Area, Southwestern Sinai, Egypt,*" **J. Phys. Chem. Biophys, 3, pp. 123, 2013. doi:10.4172/2161-0398.1000123**
Kassala, Sudan	-	-	-	**AbdElmoniem A. Elzain, A. K. Sam, O. M. Mukhtar and M. A. Abbasher** "*Measurements of Radon Gas Concentration in a soil at some Towns in Kassala State*." **Gezira Journal of Engineering & Applied Sciences,4(1), pp.15-42,2009.**
Rabak, Sudan	7.2	145	34	**AbdElmoniem A. Elzain, Y. Sh. Mohammed, Kh. Sh. Mohammed, Sumaia S. M "Radium** *and Radon Exhalation Studies in Some Soil Samples from Singa and Rabak Towns, Sudan using CR-39,*" **International Journal of Science and Research, volume 3 Issue 11, November 2014, ISSN: 2319-7064**
Singa, Sudan	17.6	354	82	**AbdElmoniem A. Elzain, Y. Sh. Mohammed, Kh. Sh. Mohammed, Sumaia S. M** "Radium *and Radon Exhalation Studies in Some Soil Samples from Singa and Rabak Towns, Sudan using CR-39,*" **International Journal of Science and Research, volume 3 Issue 11, November 2014, ISSN: 2319-7064**

4. Conclusion:

The Measurement indicate normal to some higher levels of radon concentration emanated from stone and soil samples collected from Aravalli range of hills of north India. Out of different samples under study the levels are higher in some granite diorite and granite samples compared with other white stones. The exposure of soil to stone dust also increases the activity of soil samples. The value of radon exhalation rate in soil samples observed with in safe limit of the global value of radon exhalation rate from soil which is in the range of 0.02-0.05 Bqm-2h-1. The results reveal that radon gas is chemically unreactive with soil sample. Radium concentration was found with the safe limit. The result also reveals that the area is safe for as the health hazard effects of radium and radon exhalation rate are concerned. Strong positive correlation has been observed between area exhalation rate and mass exhalation rate. The observed values of radium concentration in soil samples in the present study are less than the recommended action level 370 BqKg-1 by OECD (1979) and also less than the world wide average value of 35 BqKg-1 as reported by the UNSCEAR 2000. The high radon activity levels are found in granite type of rocks (igneous). The activity concentration of radon (222Rn) in these building materials analyzed follow a descending order: Granite > sand > cement > brick > sand stone.

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