

## **Measurement of radium content and radon exhalation rates in soil samples of Al- Hindiyah City**

**قياس محتوى الراديوم ومعدل انبعاث الرادون في عينات تربة مدينة الهندية**

Abdalsattar Kareem Hashim

Department of physics, College of science, Karbala University, Iraq.

[abdalsattarkareem@yahoo.com](mailto:abdalsattarkareem@yahoo.com)

### **Abstract**

In this work, Effective radium content and radon exhalation rates in soil samples have been measured by using long-term technique for alpha particles emission with solid state nuclear track detectors type CR-39.

The soil samples were collected from twenty different places of Al-Hindiyah city of Karbala government, Iraq. The values of effective radium content were found to vary from (0.383 to 2.696) Bq/kg with a mean value of 1.420 Bq/kg and a standard deviation of 0.668. The values of mass exhalation rates of radon vary from ( $0.308 \times 10^{-7}$  to  $2.168 \times 10^{-7}$ ) Bq/kg.day with a mean value  $1.141 \times 10^{-7}$  Bq/kg.day and standard deviation  $0.537 \times 10^{-7}$ , while the surface exhalation rates of radon vary from ( $0.961 \times 10^{-6}$  to  $6.760 \times 10^{-6}$ ) Bq/m<sup>2</sup>.day with a mean value of  $3.561 \times 10^{-6}$  Bq/m<sup>2</sup>.day and standard deviation  $1.675 \times 10^{-6}$ .

All the values of radium content in soil samples of study area were found to be quite lower than the permissible value of 370 Bq/kg recommended by Organization for Economic Cooperation and Development.

Keywords: Radium content, Radon exhalation rates, CR-39 detectors, Can technique, Soil.

### **الخلاصة**

في هذا العمل تم قياس محتوى الراديوم ومعدل انبعاث الرادون في عينات التربة باستخدام تقنية القياس طويلة الامد لانبعاث جسيمات الفا باستخدام كاشف الاثر النووي الصلب نوع CR-39. وجمعت عينات التربة من مواقع مختلفة من مدينة الهندية في محافظة كربلاء وسط العراق. حيث وجد ان محتوى الراديوم الفعال يتفاوت من (0.383 Bq/Kg الى 2.696 Bq/Kg) وبمعدل 1.420 Bq/Kg وبانحراف معياري 0.668. ووجد ان قيمة معدل انبعاث الرادون الكتلتي تتفاوت من  $0.308 \times 10^{-7}$  الى  $2.168 \times 10^{-7}$  Bq/Kg.d وبمعدل  $1.141 \times 10^{-7}$  Bq/Kg.d وبانحراف معياري  $0.537 \times 10^{-7}$ . فيما كان معدل انبعاث الرادون السطحي يتفاوت من ( $0.961 \times 10^{-6}$  الى  $6.760 \times 10^{-6}$ ) Bq/m<sup>2</sup>.d وبمعدل  $3.561 \times 10^{-6}$  Bq/m<sup>2</sup>.d وبانحراف معياري ( $1.675 \times 10^{-6}$ ). وكانت جميع قيم محتوى الراديوم المقاسة في عينات تربة منطقة الدراسة اقل من القيمة المسموحة 370 Bq/Kg حسب توصيات منظمة التعاون والتنمية الاقتصادية(OECD).

### **Introduction**

Radium (<sup>226</sup>Ra) and radon (<sup>222</sup>Rn) mainly come from naturally occurring uranium (<sup>238</sup>U), which is present in all types of rocks, building materials and soils in parts per million (ppm). The global average of uranium content in the earth's crust is about 4 ppm. Radium, being a member of uranium radioactive series, is present everywhere in the earth's crust; therefore, radon, which is the daughter product of radium, is also found everywhere in varying levels. Radium mainly enters the body through the food chain. Radium, being chemically similar to calcium, tends to follow it in metabolic processes and becomes concentrated in bones. The alpha particles given off by radium and radon bombard the bone marrow and destroy tissues that produce red blood cells. It may cause bone cancer. The radium content of a sample also contributes to the level of environmental radon as radon is produced from <sup>226</sup>Ra through  $\alpha$ -decay. Higher values of <sup>226</sup>Ra in soil contribute significantly to the enhancement of indoor radon [1].

The largest contributor of ionizing radiation to the population is natural radioactivity. It is present everywhere within us and in surrounding environment in varying concentrations because of its natural presence. Soil is the main source of continuous radiation exposure to the human beings. It acts as a medium of migration for the transfer of radio-nuclides into the environment; hence, the soil is the basic indicator of radiological contamination [2].

Radon and its daughter progeny attached to aerosols when present in the ambient air constitute an important radioactive hazard to human lungs. During respiration radon progeny deposit in the lungs and irradiate the tissue thereby damaging the cells and may cause lung cancer [3]. Radon ( $^{222}\text{Rn}$ ) is a naturally occurring  $\alpha$  – emitting radioactive noble gas. It is produced during the natural decay chain of uranium. As radium decays, radon is formed and is released into small air or water containing pores between soil and rock particles. The exhalation of radon from soil involves two mechanisms, the emanation and transport. These mechanisms are affected by many factors including the properties of the soil [4].

The radon isotope  $^{222}\text{Rn}$  has its half-life of 3.82 days that is long enough to allow it to migrate through the soil and enter the atmosphere, thus, reaching the human environment [5].

Radium is present everywhere in the earth's crust so radon is found everywhere in varying quantity. It can move freely from the place of its origin through pores in soil and cracks in walls. Radon transportation is mainly due to diffusion and forced flow.

Radon continuously undergoes radioactive decay spontaneously into four solid short lived radionuclides, viz.,  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Po}$ , in which polonium isotopes are alpha emitters. The dose due to inhaled radon progeny accounts for more than 50% of the total radiation dose to the public from natural sources [6].

In the present study, we have used solid state nuclear track detectors (CR-39) for the analysis of radium content and radon exhalation rates in twenty of soil samples collected from various locations in Al-Hindiyah city. Which is located in the middle of Iraq on the Euphrates River. This city is surrounded by Karbala from west, Babylon from east, Saddat Al-Hindiyah from north and Al-Haidariyah from south. The map of studied area is shown in Fig. (1).

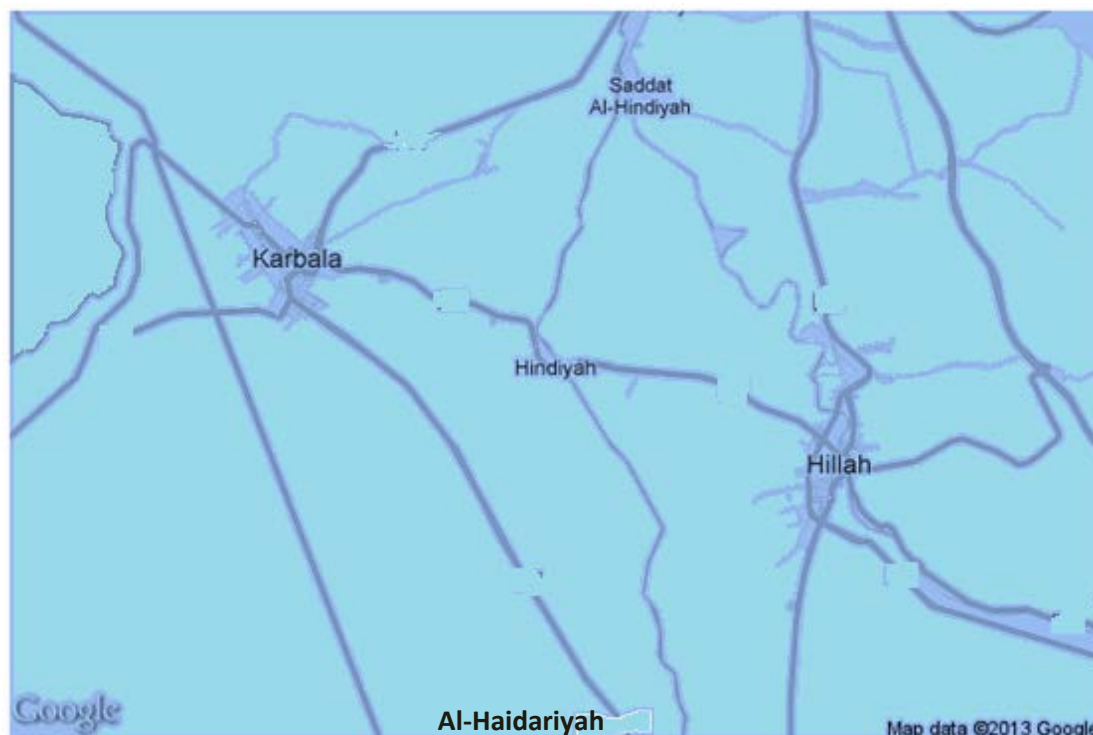


Fig.(1): The map showing Al-Hindiyah city

**Experimental methods**

In the present investigations, ‘‘sealed can technique’’ has been used for the measurement of radium content and radon exhalation rate in soil samples [7]. The experimental set-up is shown in Fig.(2). Solid State Nuclear Track Detectors (SSNTD) with sheet thickness 300 μm were used in this study, which is usually known as CR-39 [8].

The twenty soil samples were collected by the grab sampling method from different places of Al-Hindiyah city is located in the Karbala Governorate. A dried and sieved sample (120 g) was placed at the bottom of a cylindrical sealed can of 7-cm height and 7-cm diameter. The mouth of the cylindrical can was sealed with a cover and fitted with CR-39 plastic track detectors (1 cm ×1 cm) at the top inner surface [9]. The detector records the tracks of α-particles emitted by radon gas produced through the α-decay of radium contents of the samples. The detectors were exposed for a period of about 88 days (from 25-10-2012 to 20-1-2013). After exposure, the detectors were retrieved and etched for six hours in 6.25N KOH solution maintained at a temperature of (70 ± 1) °C in a constant temperature water bath to reveal the tracks. The detectors were washed and dried. Subsequently, α -tracks were counted using an optical microscope (kruss-mbl 2000) at a magnification of 400x (40x objective and 10x eyepiece).

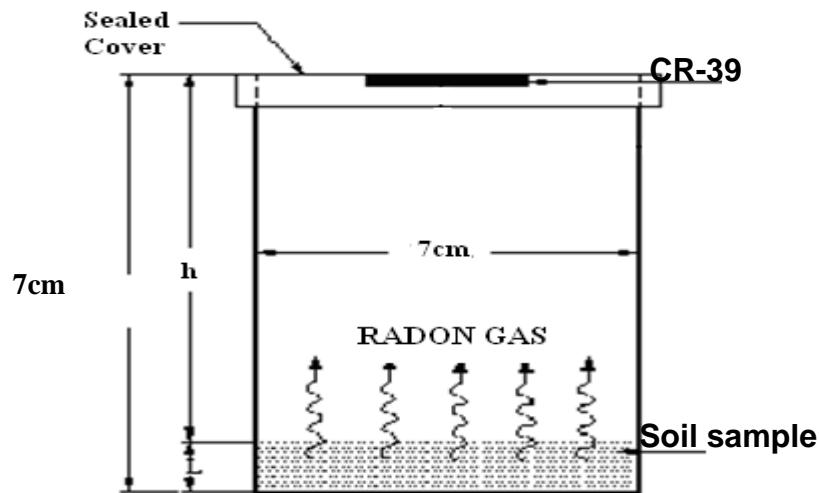


Fig. (2) Experimental setup for measurements of effective radium content and radon exhalation rates in soil samples.

**Theoretical considerations**

The mathematical expression for track density is given by the relation [10]:

$$\rho = \frac{\sum_{i=1}^n N_i}{n \times A} \quad (1)$$

Where  $\rho =$  Track density (tracks per  $cm^2$ ),  $\sum_{i=1}^n N_i =$  Total number of tracks,  $n =$  Total number of field counted,  $A =$  Area of the field of view.

The track density  $\rho$  (in track/ $cm^2$ ) is related to the radon activity concentration  $C_{Rn}$  (in  $Bq/m^3$ ) and the exposure time  $T$  by the formula [11, 12]:

$$\rho = K C_{Rn} T \quad (2)$$

Where  $K$  is the sensitivity factor of CR-39 plastic track detector, the value of  $K$  is  $6.0095 \times 10^{-2} Traks.cm^{-2}.day^{-1}/Bq.m^{-3}$ .

Since the half-life of  $^{226}Ra$  is 1600 years and that of  $^{222}Rn$  is 3.82 days, it is reasonable to assume that an effective equilibrium (about 98%) for radium-radon members of the decay series is reached in about three weeks. Once the radioactive equilibrium is established, one may use the radon alpha analysis for the determination of steady state activity concentration of radium. The activity concentration of radon begins to increase with time  $T$ , after the closing of the can, according to the relation [1]:

$$C_{Rn} = C_{Ra}(1 - e^{-\lambda_{Rn}T}) \quad (3)$$

Where  $C_{Ra}$  is the effective radium content of the sample and  $\lambda_{Rn}$  is the decay constant of  $^{222}\text{Rn}$ . Since a plastic track detector measures the time-integrated value of the above expression i.e. the total number of alpha disintegrations in unit volume of the can with a sensitivity  $K$  during the exposure time  $T$ , hence the track density observed is given by [5]:

$$\rho = K C_{Ra} T_e \quad (4)$$

where  $T_e$  denotes, by definition, the effective exposure time given by [5,13]:

$$T_e = [T - \lambda_{Rn}^{-1}(1 - e^{-\lambda_{Rn}T})] \quad (5)$$

### **Effective radium content**

The effective radium content of the soil sample can be calculated by the formula [8, 14]:

$$C_{Ra}(\text{Bq} \cdot \text{kg}^{-1}) = \left(\frac{\rho}{KT_e}\right) \left(\frac{hA}{M}\right) \quad (6)$$

Where  $M$  is the mass of the soil sample in kg,  $A$  is the area of cross-section of the can in  $\text{m}^2$ ;  $h$  is the distance between the detector and top surface of the soil sample in meter (0.045m).

### **Radon exhalation rates**

The mass exhalation rate of the sample for release of the radon can be calculated by using the expression [14]:

$$E_x(M)(\text{Bq} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}) = C_{Ra} \left(\frac{\lambda_{Ra}}{\lambda_{Rn}}\right) \frac{1}{T_e} \quad (7)$$

The surface exhalation rate of the sample for release of radon can be calculated by using the expression [14]:

$$E_x(S)(\text{Bq} \cdot \text{m}^{-2} \cdot \text{d}^{-1}) = \left[C_{Ra} \left(\frac{\lambda_{Ra}}{\lambda_{Rn}}\right) \frac{1}{T_e}\right] \frac{M}{A} \quad (8)$$

$$E_x(S)(\text{Bq} \cdot \text{m}^{-2} \cdot \text{d}^{-1}) = E_x(M) \left(\frac{M}{A}\right) \quad (9)$$

where  $\lambda_{Ra}$  is the decay constant of  $^{226}\text{Ra}$ .

### **Alpha index (I $\alpha$ )**

Several indexes dealing with the assessment of the excess  $\alpha$ -radiation due to radon inhalation originating from building materials (called ‘‘alpha indexes’’ or ‘‘internal indexes’’) have been developed [15]. Alpha index ( $I\alpha$ ) which is defined as follows [1, 16]:

$$I\alpha = \frac{C_{Ra}}{200\text{Bq/Kg}} \quad (10)$$

The recommended exemption and recommended upper levels of  $^{226}\text{Ra}$  concentrations in building materials are 100Bq/kg and 200 Bq/kg [17]. When the  $^{226}\text{Ra}$  activity concentration of building materials exceeds the value of 200 Bq/kg, it is possible that radon exhalation from this material may cause indoor radon concentration greater than 200 Bq/m<sup>3</sup>. On the other hand, if  $^{226}\text{Ra}$  concentration is less than 100Bq/kg, than resulting indoor radon concentration is less than 200 Bq/m<sup>3</sup>. These considerations are reflected in the alpha index. The recommended limit concentration of  $^{226}\text{Ra}$  is 200 Bq/kg, for which  $I\alpha = 1$  [17, 18].

### **Results and discussion**

Table 1 depicts the values of effective radium content and the mass and surface exhalation rates of radon for soil samples collected from different places of Al-Hindiyah city of Karbala government in mid of Iraq . It is seen that the value of effective radium content in collected samples varies from 0.383 to 2.696 Bq/kg with a mean value of 1.420 Bq/kg and a standard deviation of 0.668.

Radon flux density (i.e.,  $^{222}\text{Rn}$  exhalation rate) depends upon a number of parameters that behave in a stochastic and independent fashion, such as the radioactive disintegration of  $^{226}\text{Ra}$  to produce radon, the direction of recoil of radon in the grain, the interstitial soil moisture condition in the vicinity of the ejected radon atom and its diffusion in the pore space [19]. A series of radon exhalation measurements, including the present one, have demonstrated that radon flux density distribution is better represented by a log-normal distribution than by a normal distribution [20].

This behavior indicates that the random variables involved in radon exhalation interact in a multiplicative rather than an additive manner.

Table 1 also presents the value of mass exhalation and surface exhalation rates of radon of soil samples. The mass exhalation rates has been found to vary from  $(0.308 \times 10^{-7} \text{ to } 2.168 \times 10^{-7})$  Bq/kg .day with a mean value of  $1.141 \times 10^{-7}$  Bq /kg .day and standard deviation of  $0.537 \times 10^{-7}$ . The surface exhalation rates has been found to vary from  $(0.961 \times 10^{-6} \text{ to } 6.760 \times 10^{-6})$  Bq /  $m^2$  .day, with a mean value of  $3.561 \times 10^{-6}$  Bq / $m^2$  day, and standard deviation of  $1.675 \times 10^{-6}$ .

Figure 3 have shown the distribution of radium content in the different soil samples of Al- Hindiyah city. While figures 4 and 5 have shown the distribution values of mass exhalation rates and surface exhalation rates of radon for all soil samples, respectively, These figures and Table 1 show that the value of radium content and radon exhalation rates for soil samples are less than the values reported by many researchers [1, 2, and 5].

The fifth column from Table 1 shows the values of alpha index( $I_\alpha$ ) for soil samples from the various locations in Al-Hindiyah city. The values of  $I_\alpha$  ranged from 0.003 to 0.026 for soil samples with a mean values of 0.013 and standard deviation of 0.006.

There are a number of indexes dealing with the estimation of alpha-radiation due to the inhalation of radon and its short-lived decay products present in the natural environment. Since all the values of alpha indexes for these samples are much less than unity, it means that the soil samples from these locations cannot produce dangerous levels of indoor radon when used as building materials.

The values of effective radium content are less than the permissible value of 370 Bq/kg as recommended by Organization for Economic Cooperation and Development (OECD)[ 21]. Hence, the result shows that this study area is safe as far as the health hazards of radium are concerned.

Table 1: Effective radium content, mass exhalation rates of radon( $E_x(M)$ ), surface exhalation rates of radon( $E_x(S)$ ) and alpha index values for different soil samples.

Samples	$\rho$ trac/cm <sup>2</sup>	$C_{Ra}$ Bq /kg	$(E_x(M) \times 10^{-7})$ Bq/kg .day	$(E_x(S) \times 10^{-6})$ Bq /m <sup>2</sup> .day	$I_\alpha$
1	2061.585	0.600	0.482	1.504	0.006
2	6530.877	1.901	1.528	4.766	0.019
3	1316.703	0.383	0.308	0.961	0.003
4	2558.173	0.745	0.598	1.867	0.007
5	5041.113	1.468	1.180	3.679	0.014
6	6779.171	1.974	1.586	4.948	0.019
7	3303.055	0.962	0.773	2.410	0.009
8	4792.819	1.395	1.121	3.498	0.013
9	1813.291	0.528	0.424	1.323	0.005
10	9262.107	2.696	2.168	6.760	0.026
11	7027.465	2.046	1.644	5.129	0.020
12	6034.289	1.757	1.412	4.404	0.017
13	7524.052	2.190	1.761	5.491	0.021
14	7772.346	2.263	1.819	5.672	0.022
15	4544.525	1.323	1.063	3.316	0.013
16	5537.701	1.612	1.296	4.041	0.016
17	4047.937	1.178	0.947	2.954	0.011
18	3799.643	1.106	0.889	2.773	0.011
19	6282.583	1.829	1.470	4.585	0.018
20	1564.997	0.456	0.366	1.142	0.004
mean	4879.722	1.420	1.141	3.561	0.013
Max.	9262.107	2.696	2.168	6.760	0.026
Min.	1316.703	0.383	0.308	0.961	0.003
SD	2237.854	0.668	0.537	1.675	0.006

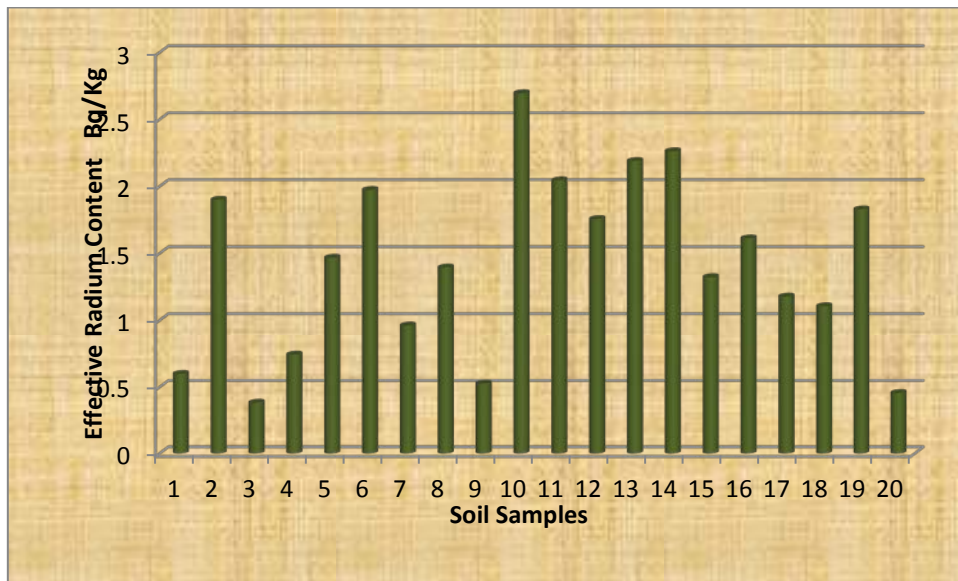


Fig. 3: Effective radium content for different soil samples.

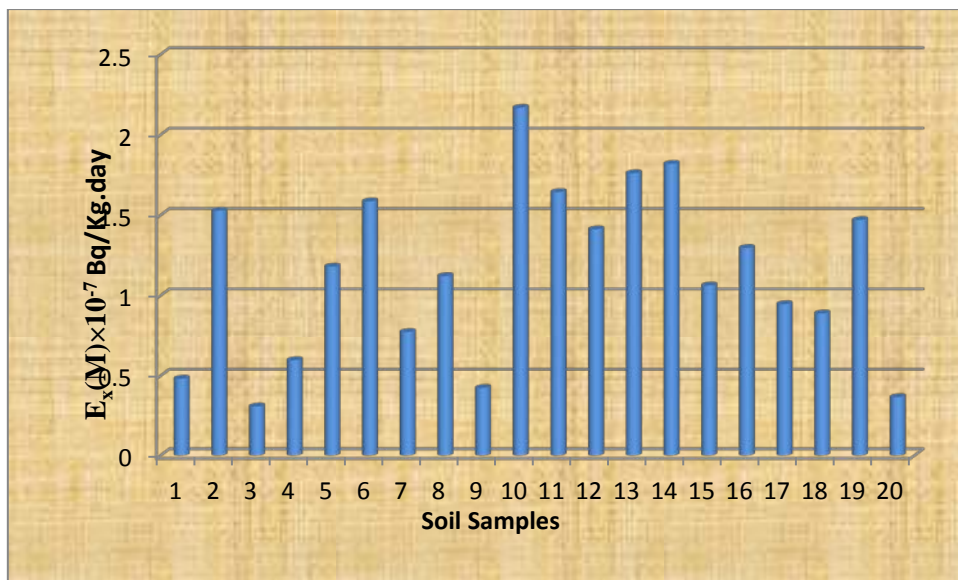


Fig. 4 Mass exhalation rates of radon for different soil samples.

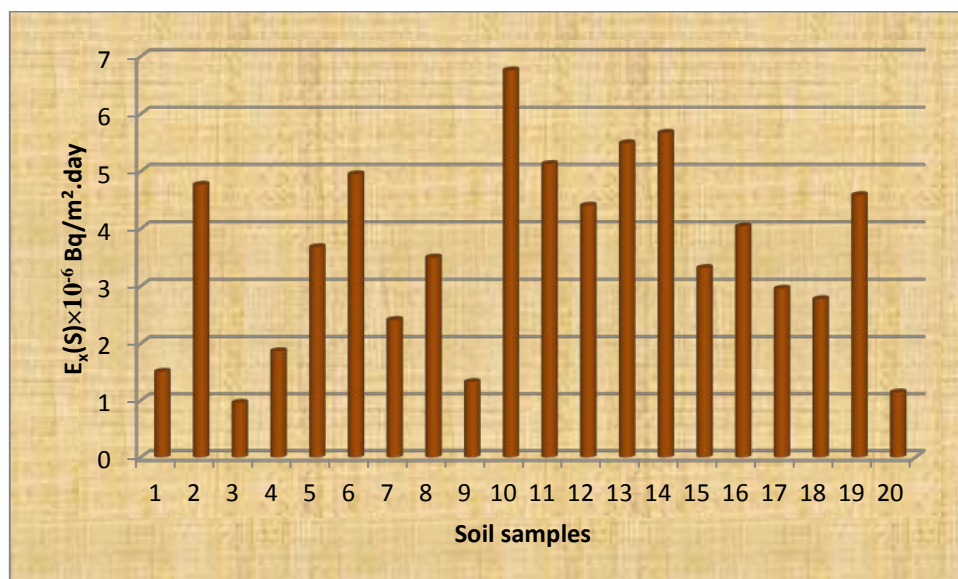


Fig. 5:- Surface exhalation rates of radon for different soil samples.

## Conclusion

Radium content and radon exhalation rates (both the mass and surface exhalation rates) have been measured successfully using CR-39 plastic track detectors by the sealed can technique.

Radon exhalation study is important for understanding the relative contribution of the material to the total radon concentration found inside the dwellings. The values of radon exhalation rate and radium content are found under the safe limit recommended by Organization for Economic Cooperation and Development. Hence it can be concluded that the study area is safe from the health hazard of radium point of view.

The calculated values of alpha indexes for all soil samples are much less than unity in each soil samples. Therefore, it is concluded that the soil samples of this region are quite safe to be used as building materials. They will not produce dangerous radon levels in dwellings.

**References**

- [1] M. Shakir Khan, D. S. Srivastava and Ameer Azam, *Study of radium content and radon exhalation rates in soil samples of northern India*. Environmental Earth Sciences, ISSN 1866-6280, Environ Earth Sci DOI 10.1007/s12665-012-1581-7, (2012).
- [2] Deepak Verma, M. Shakir Khan, Mohd. Zubair, Assessment of effective radium content and radon exhalation rates in soil samples J Radioanal Nucl Chem DOI 10.1007/s10967-012-1694-1. (2012).
- [3] Rohit Mehra, Surinder Singh, Kulwant Singh , A Study of Uranium, Radium, Radon exhalation rate and indoor radon in the environs of some areas of the malwa region, Punjab, International Society of the Built Environment, Indoor Built Environ, 15: 499–505( 2006).
- [4]. Munazza Faheem, Matiullah, Radon exhalation and its dependence on moisture content from samples of soil and building materials, Radiations Measurement 43: 1458-1462(2008).
- [5] M. Shakir Khan, A.H. Naqvi, A. Azam, D.S. Srivastava, Radium and radon exhalation studies of soil, *Iran. J. Radiat. Res.*, 8(4) 207-210(2011).
- [6] Narayanan KK, Krishnan D, Subba Ramu MC, Population exposures from natural and man-made sources of ionizing radiation in India. *Indian Soc of Rad Phys, Report No. ISRP (K)-BR-3*. (1991).
- [7] Kumar, R., Mahur, A. K., Sengupta, D. and Prasad, R., Radon activity and exhalation rates measurements in fly ash from a thermal power plant. *Radiat. Measurements*, 40: 638–641(2005).
- [8] Somogyi G, Track detection methods of radium measurements. Atomki pre print E/25, 1-28(1986).
- [9] Mahur AK, Khan MS, Naqvi AH, Prasad R, Azam A, Measurement of effective radium content of sand samples collected from Chhatrapur beach, Orissa, India using track etch technique. *Radiat Meas* 43(Supplement1) S520–S522 (2008).
- [10] Sen J.K. , Alam M. M., Kabir M. I., Asaduzzaman K. and Rafiuddin M., Measurement of alpha emitter's concentrations in some soft drinks, Bangladesh Journal of physics, 2(1) 35-50 ,(2006).
- [11] Asaad H. Ismail and Mohamad S. Jaafar, Hazards Assessment of Radon Exhalation Rate and Radium Content in the Soil Samples in Iraqi Kurdistan Using Passive and Active Detecting Methods, World Academy of Science, Engineering and Technology 46,700-703,(2010).
- [12] Hamza V. Z., Mohankumar M.N., Cytogenetic damage in human blood lymphocytes exposed in vitro to radon, *Mutation Research*, 661, pp.1-9, (2009).
- [13] Munazza Faheem, Mutiullah, radon exhalation and its dependence on moisture content from samples of soil and building materials, radiation Measurement, 43, 1458-1462, (2008).
- [14] Khan MS, Zubair M, Verma D, Naqvi AH, Azam A, Bhardwaj MK, The study of indoor radon in the urban dwellings using plastic track detectors. *Environ Earth Sci* 63:279–282, (2011a).
- [15] Stoulos S, Manolopoulou M, Papastefanou C, Assessment of natural radiation exposure and radon exhalation from building materials in Greece. *J Environ Radioact* 69:225–240,(2003).
- [16] Righi S and Bruzzi L, Natural radioactivity and radon exhalation in building materials used in Italian dwellings. *J Environ Radioact*, 88: 158-170(2006).
- [17] Reykjavik, The Radiation Protection in Denmark, Iceland, Norway and Sweden, Naturally occurring Radiation in the Nordic Countries- recommendations, The Flag-Book Series (2000).
- [18] Rafique M., Rehman H., Matiullah, Malik F., Rajput M.U., Rahman S.U., Rathore M.H., Assessment of radiological hazards due to soil and building materials used in Mirpur Azad Kashmir; Pakistan, *Iran. J. Radiat. Res.*, 9(2) 77-87(2011).
- [19] Lawrence CE, Akber RA, Bo'llho'fer A, Martin P, Radon-222 exhalation from open ground on and around a uranium mine in the wet-dry tropics. *J Environ Radioact* 100:1–8, (2009).
- [20] Bo'llho'fer A, Storm J, Martin P, Tims S, Geographic variability in radon exhalation at a rehabilitated uranium mine in the Northern Territory, Australia. *Environ Mon Assess* 114:313–330, (2006).
- [21] (OECD) Organization for economic cooperation and development. In: Exposure to radiation from natural radioactivity in building materials. Report by a group of Experts of the OECD Nuclear Energy Agency, OECD, Paris, France. (1979).