The Effect of Grain Size on Radon Exhalation Rate in Soil Samples of Dera Ismail Khan in Pakistan

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Abstract: Radon concentration and its exhalation rate has been measured from twenty five soil samples collected from seven different locations of Dera Ismail Khan, in the north west of Pakistan. These samples were crushed after drying and passed through sieves with different sizes of pores to get three grain sizes, ≤0.595 mm, 0.595-2.00 mm and 2.00-4.00 mm of soil. CR-39 based NRPB (National Radiological Protection Board) radon dosimeters have been used to measure the radon concentration and exhalation rates. The maximum average value of radon exhalation rate has found to be: (3.57±0.38) Bq m^{-2} hr^{-1} in the samples having grain size 2.00-4.00 mm and the minimum as: (2.21±0.26) Bq m^{-2} hr^{-1}, in the samples with grain size ≤ 0.0595 mm. This indicates that the radon exhalation rate increases with the grain size. The maximum average value of radon exhalation rate has been found in samples collected from river side where soil texture is silty clay loam. The obtained values of radon exhalation rate for all the samples are well below the world average value of 57.60 Bq m^{-2} hr^{-1}.

Keywords: Radon concentration, Radon exhalation rate, grain size, soil texture.

1. INTRODUCTION

High concentration of radon (^{222}Rn) and its decay products are widely known to be dangerous to human health. It is commonly associated with different types of cancer, and especially with lung cancer [1]. The emission percentage of radon into the atmosphere coming from the top few meters of ground is at least 80% [2]. According to an estimate, 2,400 million curies of radon are released form soil worldwide annually [3].

The radon concentration from soil, according to the various researches carried out so far, may vary over a wide range depending on weather conditions, climatic factors and soil types [4]. Closely spaced areas with the same soil type show variation in the radon concentration [5, 6]. Additionally, the geological structure can strongly effect the ^{222}Rn concentration in soil (faults and volcanoes) [7, 8]. The radioactivity of the soil also depends on grain size; it decreases with sand contents in the soil and increases with clay content [9].

Radon exhalation is the passage of radon from the soil/building material to the indoor environment. Exhalation rate is the number of atoms leaving the soil per unit surface area per unit time. The parameters like atmospheric pressure, temperature and wind force greatly influence the radon exhalation rate [10]. Grain size is one of important factors that control radon concentration and exhalation rate in soil. To study the radon exhalation from soil into the human houses for health consideration, the measurement of radon concentration in soil gas is helpful [11]. Soil gas measurement is also helpful in geological studies for potential faults and precursors of earthquakes [12].

The objective of this study was to analyze the effect of grain size on radon concentrations in soil and radon exhalation from soil surface.

The etched track detection technique is the most important technique used for time integrated radiation monitoring. This technique has been used in a number of studies for the measurement of radon exhalation rate from soil and building materials [13-15]. For the present study CR-39 Track Detectors have been used to measure radon concentration and radon exhalation rates in the soil of Dera Ismail Khan city of Pakistan.

Dera Ismail Khan is the southernmost District of the Khyber Pakhtunkhwa province of Pakistan. It has its location on the world map as $70^{0}55'$ E longitude and $31^{0}49'$N latitude. The total area of D. I. Khan is 7, 3226 km^2 (2, 8286 sq mile). The Dera Ismail Khan climate for most part of the year is dry and hot. The winters are normally quite mild and temperature rarely falls below 32 ^{0}F. The summers are hot with the maximum mean temperature of 116 ^{0}F in the month of May and June and minimum mean temperature of 81 ^{0}F. The location ranges from 600 ft. along the Indus River to 11,000 ft in Suleman range. It has all types of soil textures but most of soil is sandy/loamy sand, loam/silty loam and silty clay loam [15].

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Figure **1** is the map showing soil texture of Dera Ismail Khan city.

2. EXPERIMENTAL PROCEDURE

Dera Ismail Khan is situated at the bank of river Indus. The riverside soil and sand is used for the construction of dwellings in the city. Radon can be expected in soils and rocks which contain uranium and radium. Loose soil is transported, sorted and redeposited by moving water, waves and wind, often with different means of transport combining. Examples of soils formed by water transport and sorting are river deposits of gravel, sand and clay. Throughout the process of soil formation changes take place in the original uranium and radium concentrations. Moreover, uranium and radium are easily leached [4]. So there is possibility of higher radon concentration in riverside soil. In order to observe any variation in radon concentration of riverside soil, six samples were collected along the bank of river while nineteen samples were collected from different locations of the city. The site locations of all the investigated twenty five

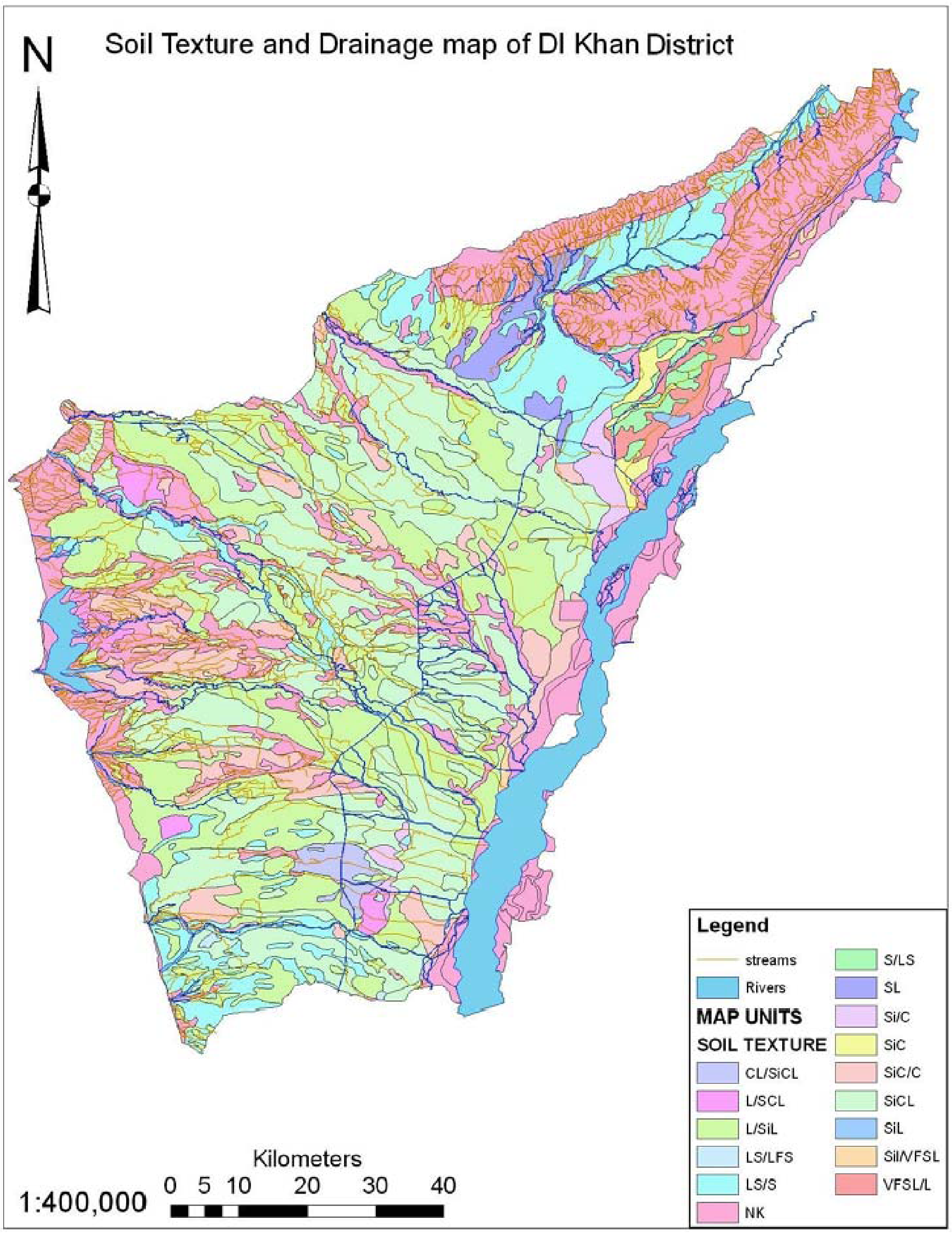


Figure 1: Soils texture map of Dera Ismail Khan (Soil Survey of Pakistan, 2005). Taken from reference [16].

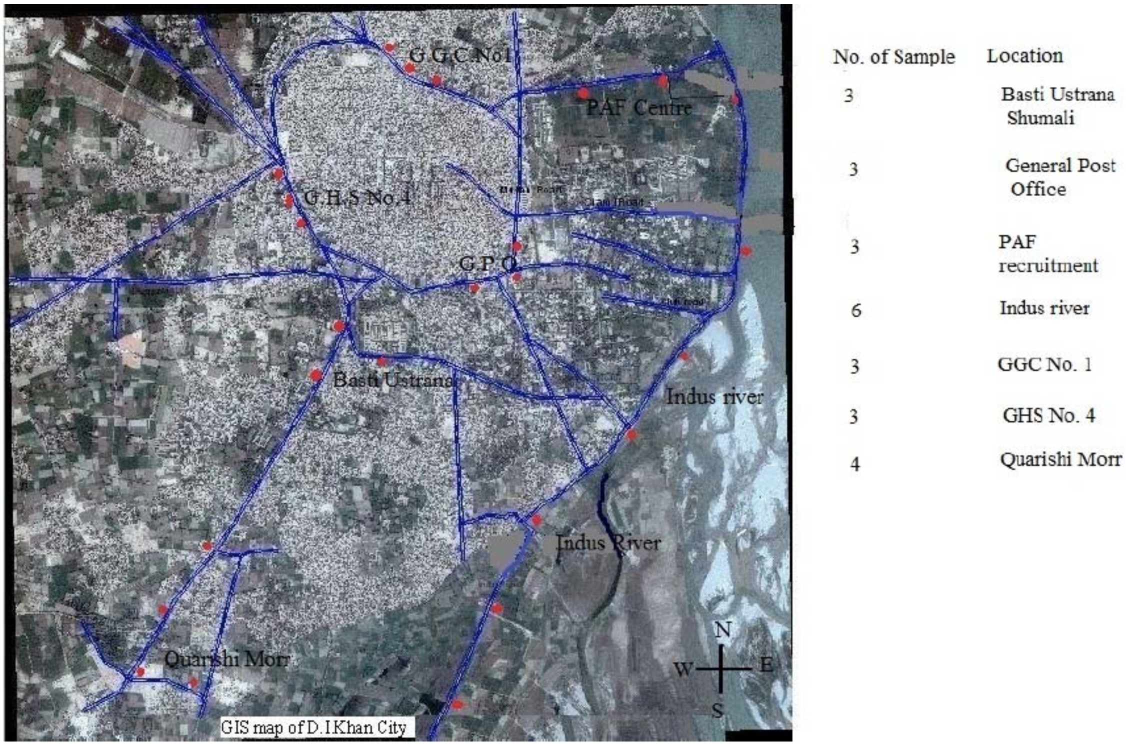


Figure 2: Map indicating the locations of the soil samples collected from Dera Ismail Khan city.

soil samples are indicated in Figure **2**. From each location the soil sample was taken in such a way that 6 inches upper layer of soil was removed because from the soil surface the radium nuclides might have been flushed away by wind or water.

The collected samples were dried in oven at 110 ^{0}C for 24 hours to evaporate all the moisture content. After drying these samples were crushed and passed through different size of sieves to get three different grain sizes ($\leq$0.595 mm, 0.595-2.00 mm and 2.00-4.00 mm) of soil. Then sample of 500 grams each was placed in plastic vessel of height 27.5 cm (volume 10.2×10^3 cm^3) with sample surface area 369.6 cm^2. The thicknesses of three different grain sizes of soil in container were 1.5 cm, 1.1cm and1.3cm. Small pieces of CR-39 track detectors of size 1.5× 1.5 cm^2 were fixed at the bottom of NRPB (National Radiological Protection Board) dosimeters. In order to account the contribution of radon only and to avoid the thoron contribution from the soil surface, CR-39 based NRPB (National Radiological Protection Board) radon dosimeters were installed in these vessels at a height greater than 25 cm from the surface of sample (Figure **3**) [4, 17]. All radon isotopes have diffusion co-efficient D of ~ 0.1 cm^2 sec^{-1} in air. The diffusion length ($L= \sqrt{D\tau}$ where τ is the mean life in seconds) of thoron in air is ~ 2.8 cm. The intensity of the gas falls after each distance L. So thoron due to very short half life (55.6 sec) has negligible amount at distance of 25 cm [4].

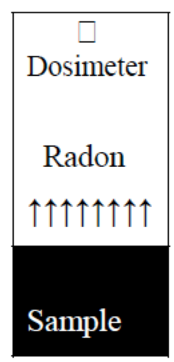


Figure 3: Experimental setup for the measurement of radon exhalation rate of soil.

The containers were hermetically sealed for 30 days. During this period equilibrium level of about 98% was reached between radium and radon. This resulted in exposure of the dosimeters to variable levels of radon concentration (i.e. starting from zero concentration level to equilibrium level). Therefore the effective exposure time of the dosimeters to radon was calculated by using the following relation.

$$T_{effective} = t - \tau\left(1 - e^{-\lambda t}\right) \quad (1)$$

Where τ = Mean life of radon (5.5 days) t = total exposure length (days)

λ = radon decay constant (7.55×10^{-3} h^{-1})

This type of correction is needed for closed system [4]. The effective time calculated from equation (1) is 588.575 hours.

After exposure the CR-39 track detectors were etched at 80^0C in 6N NaOH for sixteen hours.

Track densities were measured by counting the number of tracks in each sample under an optical microscope using the following relation.

$$\rho = \frac{\sum_i N_i}{nA} \tag{2}$$

Where A is the area of field of view, N_i is the total number of tracks and n is the total number of field of view.

The statistical error was estimated by the following relation.

$$\sigma_\rho = \frac{\sqrt{\sum N_i}}{nA} \tag{3}$$

The track density was divided by effective time (588.575 hours) calculated by equation (1) to obtained track production rate.

The measured track densities were related to the radon concentration (K Bq m^{-3}) using calibration factor [17].

$$2.7 \text{ track cm}^{-2} \text{ hr}^{-1} = 1 \text{ K Bq m}^{-3} \tag{4}$$

Radon exhalation rate was measured for each sample by using the following equation [13].

$$F_o = \frac{C(t)\left[\omega A + \lambda V\right]}{A\left[1 - e^{-(\frac{\omega A}{V}+\lambda)t}\right]} \tag{5}$$

Where V is the volume of void space in close container (m^3), A is the surface area of sample (m^2), 't' is radon accumulation time in closed container.

$\omega = \varepsilon \lambda z_o$ is called back diffusion.

λ = Radon decay constant (sec^{-1})

z_o is the thickness of the sample in sealed container and ε is the total porosity.

In a closed container that contains a sample, radon concentration increases with the passage of time from zero to maximum value. Therefore back diffusion takes place after reaching its maximum value, which reduces the concentration of radon by a factor ω in a container.

By putting values of all quantities in equation (5) we can determine radon exhalation rate.

3. RESULTS AND DISCUSSION

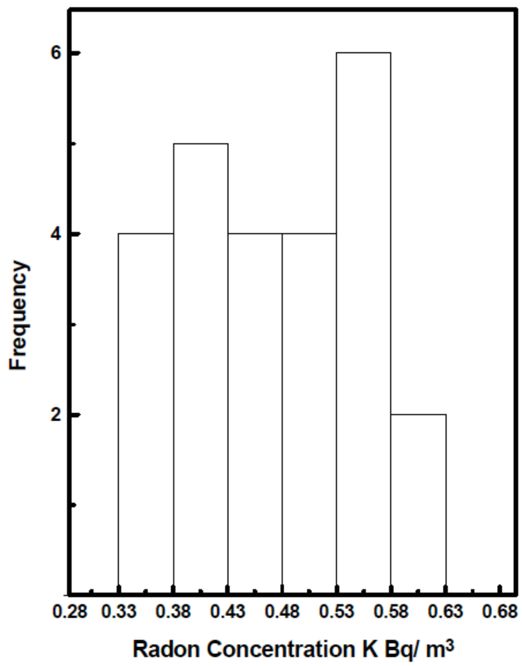
Arithmetic mean of radon concentration in soil samples collected from each location for different grain size is given in Table **1**.

Frequency distributions of radon concentration in all soil samples of Dera Ismail Khan city having grain size ≤0.595 mm, 0.595- 2.00 mm and 2.00- 4.00 mm are given in Graph **1**, **2** and **3** respectively. In Graph **1** two soil samples having grain size ≤0.595 mm have highest radon concentration which is between 0.58 and 0.63 K

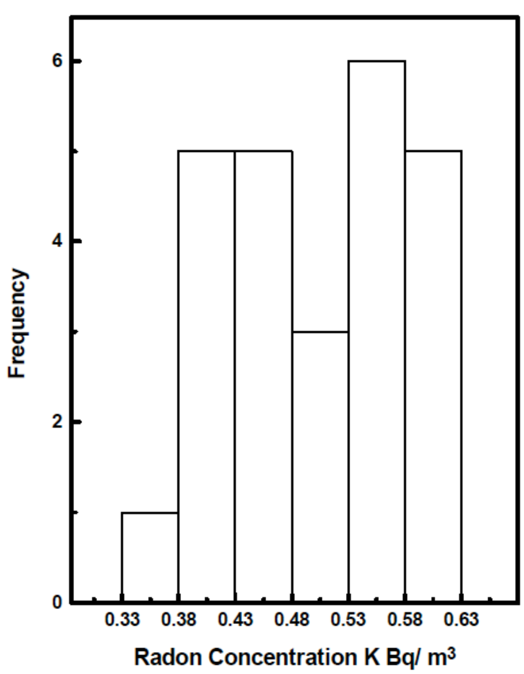
Table 1: Arithmetic Mean of Radon Concentration (K Bq m^{-3}) in Soil Samples having Different Grain Size

Location	No. of samples	Arithmetic Mean of Radon concentration (K Bq m^{-3})		
		grain size 2.00-4.00 mm	grain size 0.595-2.00 mm	grain size ≤ 0.595 mm
Basti Ustrana Sumali	3	0.48±0.05	0.47±0.05	0.45±0.05
General Post Office	3	0.50±0.05	0.50±0.05	0.46±0.05
PAF (Pakistan Air Force) recruitment centre	3	0.47±0.05	0.45±0.05	0.42±0.05
Indus river	6	0.57±0.06	0.54±0.05	0.50±0.05
GGC (Government Girls College) No. 1	3	0.54±0.05	0.51±0.05	0.48±0.05
GHS (Girls High School) No 4	3	0.52±0.05	0.49±0.05	0.47±0.05
Quarishi Morh	4	0.49±0.05	0.47±0.05	0.45±0.05

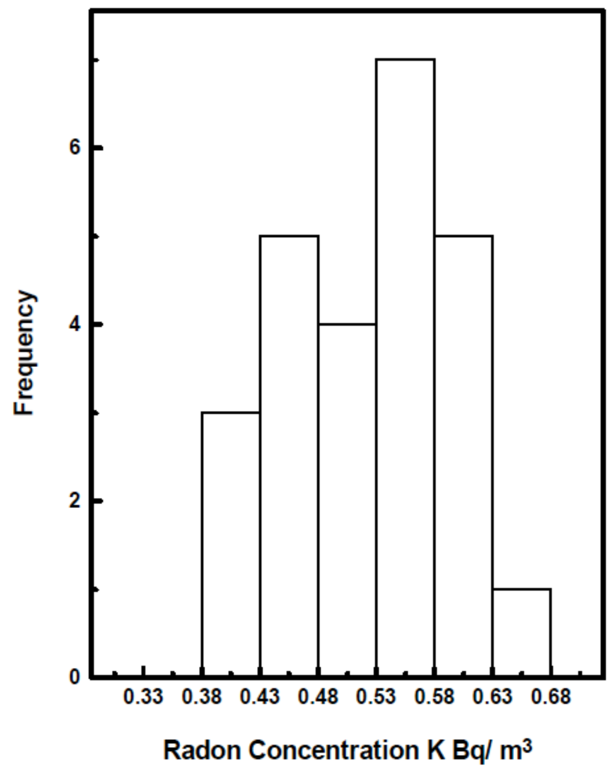
Bq m^{-3}. In Graph **2** five soil samples having grain size 0.595- 2.00 mm have highest radon concentration which is between 0.58 and 0.63 K Bq m^{-3}. In Graph **3** five soil samples having grain size 2.00- 4.00 mm have radon concentration between 0.58 and 0.63 K Bq m^{-3} while one sample has radon concentration between 0.63 and 0.68 K Bq m^{-3}.



Graph 1: Frequency distributions of radon concentration in soil samples having grain size ≤ 0.595 mm, collected from Dera Ismail Khan city.



Graph 2: Frequency distributions of radon concentration in soil samples having grain size 0.595 – 2.00 mm, collected from Dera Ismail Khan city.



Graph 3: Frequency distributions of radon concentration in soil samples having grain size 2.00 – 4.00 mm, collected from Dera Ismail Khan city.

In Table **2** arithmetic mean of radon exhalation rate for soil samples collected from each location, varies from (2.21±0.26) to (2.63±0.26) Bq m^{-2} hr^{-1} for grain size ≤ 0.595 mm, (2.81±0.31) to (3.37±0.31) Bq m^{-2} hr^{-1} for grain size 0.595-2.00 mm and (2.95±0.31) to (3.57±0.38) Bq m^{-2} hr^{-1} for grain size 2.00-4.00 mm. The maximum average value of radon exhalation rate (3.57±0.38) Bq m^{-2} hr^{-1} has been found in samples collected from river side where soil texture is silty clay loam. This is perhaps due to the fact that radioactivity of soil increases with clay content [8].

3.1. Variation in Radon Concentration and Exhalation Rate with Grain Size

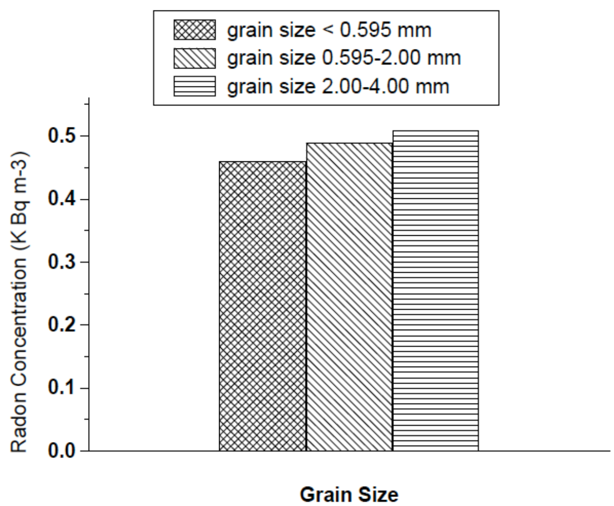
In Table **3** the average measured values of track density, radon concentration and exhalation rate are presented for all samples of different grain size obtained by sieving the samples through different sieves. It is clear from the Table **3** that radon concentration and exhalation rate increases with the grain size. Graph **4** also shows an increase in radon concentration with the grain size. It may be due to the increase in pore space between grains due to which more wide space becomes available for radon to diffuse.

Table 2: Arithmetic Mean of Radon Exhalation Rate (Bq m^{-2} hr^{-1}) in Soil Samples having Different Grain Size

Location	No. of samples	Arithmetic Mean of Radon Exhalation rate (Bq m^{-2} hr^{-1})		
		grain size 2.00-4.00 mm	grain size 0.595-2.00 mm	grain size ≤ 0.595 mm
Basti Ustrana Sumali	3	3.00±0.31	2.94±0.31	2.37±0.26
General Post Office	3	3.13±0.31	3.12±0.31	2.42±0.26
PAF (Pakistan Air Force) recruitment centre	3	2.95±0.31	2.81±0.31	2.21±0.26
Indus river	6	3.57±0.38	3.37±0.31	2.63±0.26
GGC (Government Girls College) No. 1	3	3.39±0.31	3.19±0.31	2.53±0.26
GHS (Girls High School) No. 4	3	3.26±0.31	3.06±0.31	2.48±0.26
Quarishi Morh	4	3.07±0.31	2.94±0.31	2.37±0.26

Table 3: Variation in Track Density, Radon Concentration and Exhalation Rates for Soil Samples with Grain Size

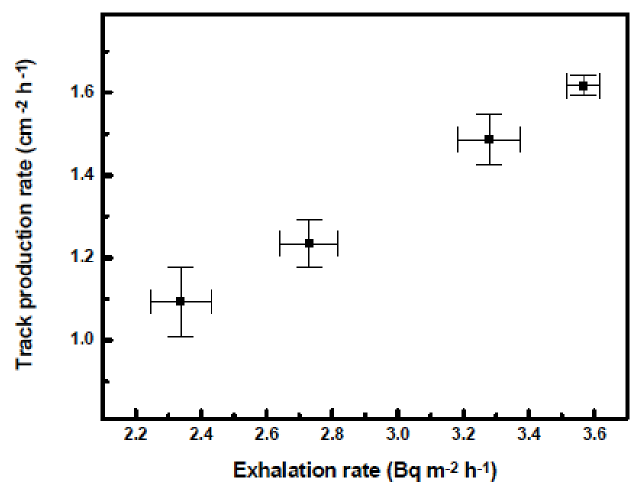
S No.	Grain size mm	Average track density track cm^{-2} hr^{-1}	Average radon concentration K Bq m^{-3}	Average exhalation rate (Bq m^{-2} hr^{-1})
1	≤0.595	1.24±0.14	0.46±0.05	2.43±0.26
2	0.595-2.00	1.32±0.14	0.49±0.05	3.06±0.31
3	2.00-4.00	1.4±0.14	0.51±0.05	3.19±0.32



Graph 4: Average radon concentration (K Bq m^{-3}) versus grain size.

3.2. Correlation of Radon Exhalation Rate with Track Production Rate

Variation of Exhalation rate with respect to track production rate is given in Graph **5**. The experimental points are the average values in the respective Exhalation rate-Track Production rate bins. Bars at points indicate statistical as well as experimental errors. The graph shows a good linear correlation between radon exhalation rate and track production rate for soil samples. This correlation of radon exhalation rate with track production rate indicates the track recording efficiency of CR-39. So it is the most suitable track detector for radon measurements.



Graph 5: Correlation of radon exhalation rate with track production rate for soil samples.

4. CONCLUSIONS

Measurements on soil radon concentration and radon exhalation rate were carried out on totally 7 points in Dera Ismail Khan.

Closed chamber technique was used to determine the radon concentration and exhalation rate for 25 samples collected from different locations of Dera Ismail Khan. There was no vast variation in radon concentration of samples collected along the bank of river and those collected from the rest of the city.

It was concluded that the radon exhalation rate increases with the grain size and it also depends on the soil texture. A good linear correlation between radon exhalation rate and track production rate indicates the track recording efficiency of CR-39. So it can be concluded that CR-39 is the most suitable track detector for radon measurements.

The maximum exhalation rate for samples having grains sizes ≤ 0.595 mm, 0.595-2.00 mm and 2.00-4.00 mm was (2.63±0.26) (Bq m^{-2} hr^{-1}), (3.37±0.31) (Bq m^{-2} hr^{-1}) and (3.57±0.38) (Bq m^{-2} hr^{-1}) respectively. These values of radon exhalation rate were found well below the world average value of 57.60 Bq m^{-2} hr^{-1} [18]. Hence it is suggested that for construction purpose this soil may be used, as it does not pose any health hazards due to low radon exhalation rate.

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