

MEASUREMENT OF RADON EXHALATION RATE, RADIUM ACTIVITY AND ANNUAL EFFECTIVE DOSE FROM BRICKS AND CEMENT SAMPLES COLLECTED FROM DERA ISMAIL KHAN

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Received 2013-11-14; Revised 2013-11-18; Accepted 2013-12-21

ABSTRACT

Radon concentration, exhalation rate, radium activity and annual effective dose have been measured from baked and unbaked bricks and cement samples commonly used as construction material in the dwellings of Dera Ismail Khan City, Pakistan. CR-39 based NRPB radon dosimeters and RAD7 have been used as passive and active devices. The values of radon concentration for baked, unbaked bricks and cements obtained from passive and active techniques were found in good agreement. Average values of radon exhalation rates in baked, unbaked bricks and cement were found $(1.202 \pm 0.212) \text{ Bq m}^{-2} \text{ h}^{-1}$, $(1.419 \pm 0.230) \text{ Bq m}^{-2} \text{ h}^{-1}$ and $(0.386 \pm 0.117) \text{ Bq m}^{-2} \text{ h}^{-1}$ and their corresponding average radium activity and annual effective dose were found $(0.956 \pm 0.169) \text{ Bq/kg}$, $(1.13 \pm 0.184) \text{ Bq/kg}$, $(0.323 \pm 0.098) \text{ Bq/kg}$ and $(33.96 \pm 5.99) \mu\text{Sv y}^{-1}$, $(40.3 \pm 6.51) \mu\text{Sv y}^{-1}$ and $(10.94 \pm 3.28) \mu\text{Sv y}^{-1}$, respectively. Radon concentration, exhalation rate and their corresponding radium activity and annual effective dose were found higher in unbaked bricks as compared to baked bricks and cement but overall values of radon exhalation rate, annual effective dose and radium activity were found well below the world average values of $57.600 \text{ Bq m}^{-2} \text{ h}^{-1}$, $1100 \mu\text{Sv y}^{-1}$ and 370 Bq/kg , respectively.

Keywords: Radon, CR-39, NRPB Dosimeter, RAD7

1. INTRODUCTION

High concentration of radon and its decay product is widely known to be dangerous to human health. It is possibly associated with different types of cancer and especially with lung cancer. Emission percentage of radon into atmosphere coming from a few meters above the ground is at least 80% (Shweikani *et al.*, 1995). Radon is a natural radioactive gas which is found everywhere and cannot be avoided. It has very serious effects on human health due to large scale abundance of its parents and its longer half life. Radon has been considered as second leading cause of lung cancer after smoking (IARC, 2004). A 2.4 mSv

average annual dose has been estimated from natural radiation sources to the world population. Two-third of average annual dose is the internal exposure and one-third is external exposure (Wang, 2002). Inhalation of radon gas and its daughters will expose the lung tissue to short-lived alpha emitting radio nuclides, which will increase the risk of lung cancer. One of the major factors of increasing skin cancer is radon gas, where it is due to the deposition of radon on the skin, the alpha particles are suspected to include damage to the epithelial cell. Kidney related diseases have also been observed in some people exposed to radon. The reason is that kidney receives the highest dose compared to other body organs, after

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radon is transferred from the lung to the kidney by blood (Henshaw *et al.*, 1990).

Different sources are responsible for the presence of radon and its daughters in houses. One of the major sources of concentration of radon in houses is the building materials used for construction. Cracks in walls, floors, joints or pipe holes and lines provide a channel for radon which flows to the surface and ultimately enters houses. In addition, the radon production rate in houses depends on the concentrations in the building materials, subsoil, porosity, density of the uranium and radium activity in materials used for construction (Cartwright *et al.*, 1978).

The presence of radon is expected everywhere, but found in high concentration in soil and rocks enriched in radium and uranium elements. Most of these soils and rocks are compacted and processed to produce building materials that are used for workplaces and houses in which we work and live. There are many concerns about the escape of radon from building materials into the workplaces and houses. All nations, including Pakistan, use building materials in workplaces and houses such as cements, granite, tiles, clay, sand, bricks and ceramic. If radon emanated from these materials exceeds the action level of 200 Bq.m^{-3} , then remedial action should be taken to reduce health hazards due to exposure to indoor radon.

Numerous studies in Pakistan have been conducted to measure radon concentration in air, soil and building materials (Faheem *et al.*, 2010; Nasir and Ahmad, 2012; Nasir and Shah, 2012; Rafique *et al.*, 2011a; 2011b; Rahman *et al.*, 2012). Therefore, the study of radon exhalation from building materials commonly used in Dera Ismail Khan Dwellings is an important step in diminishing indoor radon levels into significantly below the action level.

2. DETAIL OF SAMPLES

The main objectives of this study were to measure the radon concentration, radon exhalation rate, radium contents and annual effective dose from bricks and cements used as building materials in Dera Ismail Khan City, Pakistan. Twenty samples of bricks (both unbaked and baked) were collected from different bricks kilns and five representative samples of cements were collected from different building materials suppliers in Dera Ismail Khan City. The details on the samples collection of cement are given in **Table 1** and that of bricks in **Table 2**. In **Table 1** cement samples are given the names of SS-1 to SS-5, while in **Table 2** SS-6 to SS-24 are baked bricks and SS-7 to SS-25 are unbaked bricks.

Table 1. Cement samples from various factories

Sample name	Factory name
SS-1	Maple leaf cement
SS-2	Kohat cement
SS-3	Lucky cement
SS-4	Askari cement
SS-5	Cherrat cement

Table 2. Locations of the collected bricks samples from various kilns

Sample name	No. of samples	Location
SS-6,7	2	New sarhad bricks
SS-8,9	do	Ahmad khan bricks
SS-10,11	do	Azhar bricks
SS-12,13	do	Baho bricks
SS-14,15	do	Shabarat bricks
SS-16,17	do	Malik sohail bricks
SS-18,19	do	New baho bricks
SS-20,21	do	Malik bricks
SS-22,23	do	Kerri hamad khan bricks
SS-24,25	do	Kerri nawab bricks

3. EXPERIMENTAL TECHNIQUE

The collected samples were brought to Laboratory of Radiation Physics, Gomal University Dera Ismail Khan and were dried in oven at 110°C for 24 h to evaporate all the moisture contents. Bricks samples were crushed and homogenized by passing through sieve. After processing samples of 500 g each of bricks and cement, they were placed in plastic containers of 27.5 cm in height (volume $9.66 \times 10^{-3} \text{ m}^3$) with sample surface area 0.03696 m^2 (**Fig. 1b**). The thickness of bricks in the containers was 1.6 cm and that of cement was 1.5 cm. CR-39 based NRPB radon dosimeters were fitted at a distance of 25 cm from the surface of the samples in these containers, so as to account only the radon contribution and to avoid the role of thoron from the surface of sample (Howarth *et al.*, 2002). The containers were hermetically sealed for 30 days. About 98% of equilibrium level is reached between radium and radon during this time period. This resulted in the exposure of the radon to variable levels of radon concentration (i.e., starting from zero concentration level to equilibrium level). Therefore, effective exposure time was calculated by using the following relation (Durrani and Ilic, 1997):

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

Where:

τ = Mean life of radon (5.5 days)

t = Total exposure length (30 days)

λ = Decay constant of radon ($7.55 \times 10^{-3} \text{ h}^{-1}$)

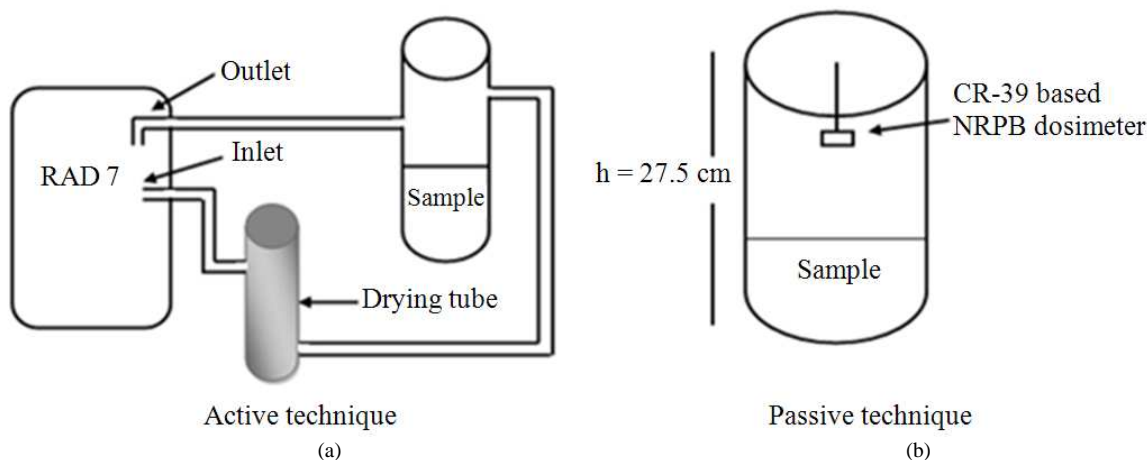


Fig. 1. Building materials samples under exposure of (a) RAD7 and (b) CR-39 NTDs

This type of correlation is needed only for closed system. The effective time calculated from Equation (1) was 588.575 h.

After the exposure, CR-39 detectors were etched in 6N NaOH at 70°C for 8 h and studied under optical microscope at 400× magnification.

The track densities were measured using the following Equation (2):

$$\rho = \frac{\sum N_i}{nA} \quad (2)$$

Where:

A = The area of field of view, N_i is the total number of tracks and

n = The total number of field of views

The statistical error was calculated using Equation (3):

$$\sigma_\rho = \frac{\sqrt{\sum N_i}}{nA} \quad (3)$$

To obtain the track production rate, the measured track density was divided by effective time (588.575 h).

The track density was converted into radon concentration using calibration factor (Howarth *et al.*, 2002):

$$2.7 \text{ track/cm}^2 \text{ h} = 1 \text{ kBq/ m}^3$$

Radon exhalation rate was measured for each sample by using Equation (4) (Rafique *et al.*, 2011a):

$$F_0 = \frac{C(t)[\omega A + \lambda V]}{\left[1 - e^{-\left(\frac{\omega A}{V} + \lambda\right)t}\right]} \quad (4)$$

Where:

V = The volume of void space in close container (m^3)

A = The surface area of sample (m^2) and t is radon accumulation time in closed container

ω = Back diffusion and

C(t) = Measurement of radon concentration.

The radium activity of the samples was calculated by using Equation (5) (Somogyi, 1990; Abd-Elzaher, 2012):

$$C_{\text{Radium}} = \rho h A_c / K T_c M \quad (5)$$

where, ρ , h and A_c are the track density (track/cm^2), distance between the detector and sample (m) and area of cross section of container (m^2), respectively. K is the sensitivity factor ($0.2315 \text{ tracks cm}^{-2} \text{ d}^{-1}(\text{Bq/m}^3)^{-1}$) (Ismail and Jaafar, 2009).

Annul effective dose from the collected samples was calculated by using following relation (Ackers *et al.*, 1984; Khan, 1994):

$$D_{\text{Rn}} = t_f T (\text{DCF})(S_R / V_R)(1/\lambda_v)(0.45 - 0.15 \lambda_v) \sum F_0 \text{si} \quad (6)$$

where, t_f is the fraction of time a person spent indoor (0.8), T is total hours in year (8760 h/y), DFC is dose conversion factor, ($9 \text{ nSv.h}^{-1}/\text{Bq.m}^{-3}$) or (6 mSv/ WLM) (UNSCEAR, 2000). S_R/V_R is the ratio of surface area to volume ($1.5 \text{ to } 3.0 \text{ m}^{-1}$), λ_v is the constant of ventilation ($0.3 \text{ to } 1.0 \text{ h}^{-1}$), ($0.45-0.15 \lambda_v$) is the dimension less

equilibrium factor for radon for the ventilation rate in the interval of 0.1-2 exchanges per hour, F_0 is radon exhalation rate ($\text{Bq m}^{-2} \text{h}^{-1}$). To estimate the annual effective dose from the building materials, the following constant factors were used: Dimension of room was assumed as a chamber with ($6 \times 4 \times 3 \text{ m}^3$ i.e., $V = 72 \text{ m}^3$), using S_R/V_R ratio of 1.5 m^{-1} and typical value of the ventilation rate λ_v of 1 air changes per hour.

In order to support the results of Passive technique, RAD7 manufactured by DurrIDGE Company USA was used as an active technique at the School of Physics, Universiti Sains Malaysia by making small modification of the design (**Fig. 1a**).

RAD7 has an air pump and a solid state alpha detector which consists of semiconductor material. Silicon is usually used as semiconductor material which directly converts alpha radiation to an electrical signal. Solid state devices have the ability to distinguish radon from thoron by the emitted alpha particle energy. Bq m^{-3} was used as a standard unit in this study.

For the measurement of radon concentration in building materials by RAD7, samples of 300 g each were placed in a container of 11 cm in height and 6 cm in diameter then connected to RAD7 which enables to degas the radon from the sample into closed loop. Before each measurement, RAD7 was purged for ten minutes to reduce the humidity by using laboratory drying unit. Thirty minutes measuring time at Grab protocol was taken for all samples.

4. RESULTS

Table 3 shows the results of radon concentrations of bricks (both unbaked and baked) obtained by passive and active techniques. The range of radon concentration obtained by passive and active techniques in unbaked bricks varies from (0.1588 ± 0.0300) kBq m^{-3} to (0.3121 ± 0.0420) kBq m^{-3} and (0.128 ± 0.031) kBq m^{-3} to (0.291 ± 0.042) kBq m^{-3} respectively, while in case of

baked bricks its values varies from (0.1362 ± 0.0278) kBq m^{-3} to (0.2837 ± 0.0401) kBq m^{-3} and (0.110 ± 0.021) kBq m^{-3} to (0.241 ± 0.098) kBq m^{-3} , respectively. **Table 4** represents the radon exhalation rate from unbaked and baked bricks calculated by Equation (4). As may be seen from the table, radon exhalation rate was found to vary from (1.068 ± 0.202) $\text{Bq m}^{-2} \text{h}^{-1}$ to (2.100 ± 0.282) $\text{Bq m}^{-2} \text{h}^{-1}$ in the case of unbaked bricks, while for baked bricks, its values varies from (0.915 ± 0.187) $\text{Bq m}^{-2} \text{h}^{-1}$ to (1.909 ± 0.269) $\text{Bq m}^{-2} \text{h}^{-1}$.

Table 5 represents the results of radon concentrations of cement measured by both techniques and radon exhalation rate. Radon concentration measured by passive and active techniques were (0.0794 ± 0.0212) kBq m^{-3} and (0.065 ± 0.021) kBq m^{-3} for white cement, while in the case of gray cement, its values ranged from (0.0454 ± 0.0160) kBq m^{-3} to (0.0681 ± 0.0196) kBq m^{-3} and (0.040 ± 0.013) kBq m^{-3} to (0.060 ± 0.020) kBq m^{-3} , respectively. For white cement, the value of radon exhalation rate was (0.511 ± 0.136) $\text{Bq m}^{-2} \text{h}^{-1}$, while in the case of gray cement its values were varied from (0.292 ± 0.103) $\text{Bq m}^{-2} \text{h}^{-1}$ to (0.438 ± 0.126) $\text{Bq m}^{-2} \text{h}^{-1}$.

Radium activity and annual effective dose from bricks, calculated by Equation (5 and 6), respectively are presented in **Table 6**. The maximum value of radium activity in unbaked bricks was (1.676 ± 0.225) Bq/kg and the minimum value was (0.853 ± 0.161) Bq/kg , while in the case of baked bricks, the maximum value was (1.524 ± 0.215) Bq/kg and the minimum value was (0.731 ± 0.149) Bq/kg . Annual effective dose from unbaked and baked bricks vary from (30.3 ± 5.7) $\mu\text{Sv y}^{-1}$ to (59.6 ± 8.0) $\mu\text{Sv y}^{-1}$ and (25.9 ± 5.3) $\mu\text{Sv y}^{-1}$ to (54.2 ± 7.6) $\mu\text{Sv y}^{-1}$, respectively. Radium activity and annual effective dose in white cement were (0.427 ± 0.114) Bq/kg and (14.5 ± 3.8) $\mu\text{Sv y}^{-1}$ respectively, while for gray cement its values ranged from (0.244 ± 0.086) Bq/kg to (0.366 ± 0.105) Bq/kg and (8.2 ± 2.9) $\mu\text{Sv y}^{-1}$ to (12.4 ± 3.5) $\mu\text{Sv y}^{-1}$, respectively as given in **Table 7**.

Table 3. Radon concentration in baked and unbaked bricks

Sample name	Radon concentration (kBq m^{-3}) (baked) RAD7	Radon concentration (kBq m^{-3}) (baked) CR-39 NTDs	Radon concentration (kBq m^{-3}) (unbaked) RAD7	Radon concentration (kBq m^{-3}) (unbaked) CR-39 NTDs
SS-6,7	0.165 ± 0.036	0.1759 ± 0.0315	0.204 ± 0.039	0.1985 ± 0.0335
SS-8,9	0.147 ± 0.031	0.1418 ± 0.0283	0.131 ± 0.025	0.1700 ± 0.0318
SS-10,11	0.191 ± 0.037	0.1872 ± 0.0326	0.221 ± 0.037	0.2155 ± 0.0348
SS-12,13	0.114 ± 0.028	0.1532 ± 0.0294	0.153 ± 0.033	0.2043 ± 0.0340
SS-14,15	0.189 ± 0.036	0.2213 ± 0.0358	0.216 ± 0.034	0.2497 ± 0.0376
SS-16,17	0.151 ± 0.035	0.1475 ± 0.0289	0.175 ± 0.034	0.1644 ± 0.0305
SS-18,19	0.241 ± 0.098	0.2837 ± 0.0401	0.291 ± 0.042	0.3121 ± 0.0420
SS-20,21	0.110 ± 0.021	0.1362 ± 0.0278	0.128 ± 0.031	0.1588 ± 0.0300
SS-22,23	0.144 ± 0.029	0.1418 ± 0.0283	0.184 ± 0.040	0.1816 ± 0.0321
SS-24,25	0.145 ± 0.030	0.1929 ± 0.0330	0.265 ± 0.045	0.2553 ± 0.0380

Table 4. Radon exhalation rate in backed and unbaked samples

Sample name	Radon Exhalation rate (Bq m ⁻² h ⁻¹) (baked) CR-39 NTDs	Radon Exhalation rate (Bq m ⁻² h ⁻¹) (unbaked) CR-39 NTDs
SS-6,7	1.183±0.212	1.335±0.225
SS-8,9	0.954±0.190	1.144±0.209
SS-10,11	1.259±0.219	1.450±0.234
SS-12,13	1.031±0.197	1.374±0.228
SS-14,15	1.489±0.240	1.680±0.253
SS-16,17	0.992±0.194	1.106±0.205
SS-18,19	1.909±0.269	2.100±0.282
SS-20,21	0.915±0.187	1.068±0.202
SS-22,23	0.993±0.190	1.222±0.216
SS-24,25	1.298±0.222	1.718±0.255

Table 5. Radon concentration and exhalation rate in cement samples

Sample name	Radon concentration (kBq m ⁻³) CR-39 NTDs	Radon concentration (kBq m ⁻³) RAD7	Exhalation rate (Bq m ⁻² h ⁻¹) CR-39 NTDs
SS-1	0.0794±0.0212	0.065±0.021	0.511±0.136
SS-2	0.0454±0.0160	0.040±0.013	0.292±0.103
SS-3	0.0567±0.0179	0.058±0.019	0.365±0.115
SS-4	0.0510±0.0170	0.052±0.018	0.328±0.109
SS-5	0.0681±0.0196	0.060±0.020	0.438±0.126

Table 6. Radium activity and Annual effective dose from baked and unbaked bricks samples

Sample name	CR-39 NTDs			
	Radium activities (Bq/kg) Baked	Radium activities (Bq/kg) unbaked	Annual Effective dose (μSv y ⁻¹) Baked	Annual Effective dose (μSv y ⁻¹) unbaked
SS-6,7	0.944±0.169	1.066±0.180	33.5±6.0	37.9±6.3
SS-8,9	0.761±0.152	0.914±0.166	27.0±5.3	32.4±5.9
SS-10,11	1.005±0.175	1.158±0.187	35.7±6.28	41.8± 6.6
SS-12,13	0.823±0.158	1.097±0.182	29.2±5.59	39.0±6.4
SS-14,15	1.188±0.190	1.341±0.202	42.2±6.8	47.7±7.1
SS-16,17	0.792±0.155	0.883±0.163	28.1±5.5	31.4±5.8
SS-18,19	1.524±0.215	1.676±0.225	54.2±7.6	59.6±8.0
SS-20,21	0.731±0.149	0.853±0.161	25.9±5.3	30.3±5.7
SS-22,23	0.761±0.152	0.975±0.172	27.0±5.3	34.7±6.1
SS-24,25	1.036±0.177	1.371±0.204	36.8±6.3	48.7±7.2

Table 7. Radium activity and annual effective dose from cements

Sample name	CR-39 NTDs	
	Radium activities (Bq/kg)	Annual effective dose (μSv y ⁻¹)
SS-1	0.427±0.114	14.5±3.8
SS-2	0.244±0.086	8.2±2.9
SS-3	0.305±0.096	10.3±3.2
SS-4	0.274±0.091	9.3±3.0
SS-5	0.366±0.105	12.4±3.5

5. DISCUSSION

Positive and strong correlation has been observed between the results of passive and active techniques. The linear correlation value for both techniques was

0.917, as shown in **Fig. 2**. The value of the correlation was statistically significant and 99% confidence interval. Data shown in **Table 4** indicate that radon exhalation rate is comparatively higher in unbaked bricks while it is lower in baked bricks. It is due to the

fact that radon emanation in the case of unbaked bricks can escape from a few centimetres below the surface and the quantity of radon released from materials depends on the internal surface area, while in the case of baked bricks it emanates from the surface layer (Rawat *et al.*, 1991). It can also be seen that radon exhalation rate in these samples varies from sample to sample, which is due to porosity of building materials (Folkerts *et al.*, 1984) and the difference of uranium and radium contents (Ramachandran and Ramu, 1989). The values of white cement were found higher as compared to gray cement. It has been observed from the results that like radon exhalation rate, the values of radium activity and annual effective dose in unbaked bricks are higher than that of baked bricks and cement.

With regards to radon exhalation rate, a large amount of data has been published in literature. Some of these data reported by different researchers in different countries are presented in **Table 8**. High value of radon exhalation rate ($0.11 \text{ Bq m}^{-2} \text{ h}^{-1}$) was reported in unbaked bricks and lower value of radon exhalation rate was reported ($0.09 \text{ Bq m}^{-2} \text{ h}^{-1}$) in baked bricks in India (Khan, 1994). In the present work, high value of radon exhalation rates were found in unbaked bricks and lower values were found in baked bricks. In Pakistan, (Faheem and Matiullah, 2008) reported radon exhalation rates in Punjab ($0.212 \pm 0.034 \text{ Bq m}^{-2} \text{ h}^{-1}$) and ($0.23 \pm 10.030 \text{ Bq m}^{-2} \text{ h}^{-1}$) in bricks and cements, respectively. These values of radon exhalation rate are close to the results of the present study.

Table 8. Comparison of radon exhalation rate of building materials with previous work

Material	Present work ($\text{Bq m}^{-2} \text{ h}^{-1}$)	Previous work ($\text{Bq m}^{-2} \text{ h}^{-1}$)	Country	References
Baked Bricks	0.915±0.187 to 1.909±0.269	0.09	India	(Khan, 1994)
		0.215	India	(Rawat <i>et al.</i> , 1991)
		0.245-0.365	Pakistan	(Rahman <i>et al.</i> , 2007)
		0.212±0.034	Pakistan	(Faheem and Matiullah, 2008)
		0.078 - 0.117	Algeria	(Amrani and Cherouati, 1999)
Unbaked Bricks	1.068±0.202 to 2.100±0.282	0.11	India	(Khan, 1994)
		0.417	India	(Rawat <i>et al.</i> , 1991)
Gray Cement	0.292±0.103 to 0.438±0.126	0.083 - 0.140	Algeria	(Amrani and Cherouati, 1999)
		0.231±0.030	Pakistan	(Faheem and Matiullah, 2008)
White Cement	0.0794±0.0212	0.04	India	(Khan, 1994)
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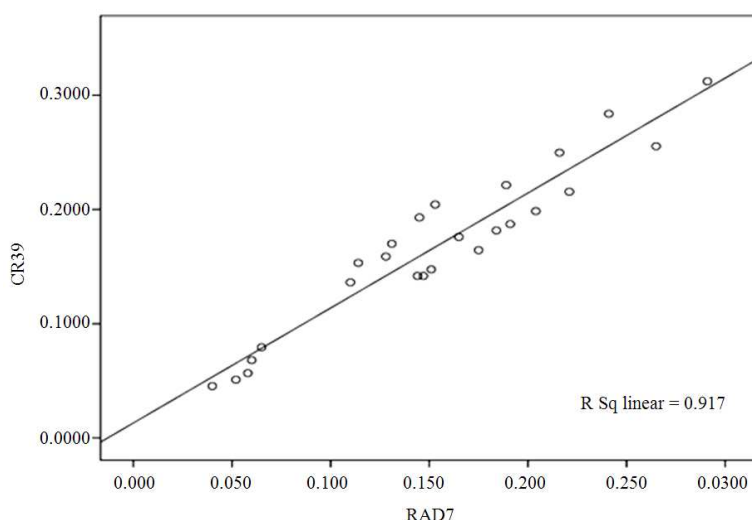


Fig. 2. Linearity between passive and active techniques for measurement of radon concentration

6. CONCLUSION

Radiological parameters in building materials (bricks and cement) locally used in Dera Ismail Khan have been determined. Radon concentrations were measured by passive and active techniques and the results are in good agreement. Results obtained from the current study shows that the average radon exhalation rate (1.419 ± 0.230) Bq $m^{-2} h^{-1}$, radium activity (1.13 ± 0.184) Bq/kg and annual effective dose (40.35 ± 6.51) $\mu Sv y^{-1}$ from unbaked bricks have comparatively higher values as compared to baked bricks (1.202 ± 0.212) Bq $m^{-2} h^{-1}$, (0.956 ± 0.169) Bq/kg, (33.96 ± 5.99) $\mu Sv y^{-1}$ and cement (0.386 ± 0.117) Bq $m^{-2} h^{-1}$, (0.323 ± 0.098) Bq/kg, (10.94 ± 3.28) $\mu Sv y^{-1}$ respectively, but overall values of radon exhalation rate, radium activity and annual effective dose are below the world average value of 57.600 Bq $m^{-2} h^{-1}$ (UNSCEAR, 2000), 370 Bq/kg (OECD, 1979) and 1100 $\mu Sv y^{-1}$, respectively and thus are safe to be used as building materials according to the international recommendations. The values were found minimum for cement samples, therefore, from the health point of view it is suggested that cement houses are safer than mud houses. The present work was designed to study the radon level, radium activity and annual effective dose from building materials. Further studies are needed to determine radon level and natural radioactivity in water, soil, food and vegetation samples and their risk factors should be investigated.

7. ACKNOWLEDGEMENT

One of the researchers (Nisar Ahmad) is thankful to Universiti Sains Malaysia and TWAS for financial support in the form of TWAS-USM Fellowship.

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