Aerosol and Air Quality Research, 18: 811–819, 2018

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2017.10.0405

Technical Note

Measurement of Variation of Radon-Thoron and their Progeny Concentrations in Dwellings using Pin Hole Based Dosimeters

Deep Shikha[1], Vimal Mehta[1*], Rishi Pal Chauhan[2], Gurmel Singh Mudahar[3]

[1] *PG Department of Physics, SGTB Khalsa College, Anandpur Sahib-140 118, India*
[2] *Department of Physics, National Institute of Technology, Kurukshetra-136 119, India*
[3] *Department of Physics, Punjabi University, Patiala-147 001, India*

ABSTRACT

In the present investigation, a newly designed pinhole-based radon–thoron dosimeter with LR-115 track detectors was used for the integrated determination of radon, thoron and their progeny levels in the indoor air of the dwellings of the Union Territory (U.T.) Chandigarh for checking the indoor air quality. The soil and the building materials are the major sources of the indoor radon, and the contribution of these towards indoor radon levels depends upon the radium content and exhalation rates and therefore can be used as a primary index for radon levels in the dwellings. As a result, the radon exhalation rate of some soil samples from the study area was measured using the active technique. The exhalation rate for samples of sand available from the study area, utilized for construction purposes, was also measured.

The concentration of indoor radon varied from 24.2 ± 1.1 Bq m^{-3} to 62.1 ± 3.1 Bq m^{-3}. The thoron concentration was found to vary from 3.0 ± 0.1 Bq m^{-3} to 99.2 ± 4.9 Bq m^{-3}. The annual inhalation dose received by the inhabitants of these dwellings is within safe limits. A good positive correlation was found between the indoor radon concentration and the radon mass exhalation rate of the soil samples from the study area.

Keywords: Indoor radon; Indoor thoron; Annual effective dose; Exhalation rate.

INTRODUCTION

Radon is a noble but a natural radioactive gas that evolves from the radioactive decay of radium. The radium content of the soil is considered to be constant due to its long half-life. Atmospheric radon originates exclusively from the earth's surface (Chambers *et al.*, 2016a). Like radon, thoron is also a radioactive gas that originates from the radioactive decay of thorium present throughout the earth's crust and in many building materials. But radon is more dangerous than thoron because it has long half-life of 3.8 days (Chambers *et al.*, 2016b), in comparison to thoron's half-life of 55.6 sec. The decay of radon, ^{222}Rn, and thoron, ^{220}Rn, in the environment acts as a source of low vapor pressure decay products that attach to other nuclei or aerosol particles. Its decay products, ^{218}Po (3.0 min), ^{214}Pb (26.8 min) and ^{212}Pb (10.64 h) (Papastefanou, 2009), may attach to aerosol particles and be inhaled, which is known to cause lung cancer (Brookins, 1998). Many other studies have also produced similar results (Cohen, 1987; National Research Council, 1988; Nazaroff and Nero, 1988; Momcilovic and Lykken, 2007). Besides radon, thoron and its progeny, $PM_{2.5}$ is also one of the factors, interaction with which causes various diseases, such as bronchitis, allergies, lung cancer, and heart disease, that become the cause of premature deaths (Chelani, 2017). $PM_{2.5}$ may act destructively, as it can be inhaled easily (due to its small length which is equal to 3.6% of the diameter of a human hair) deep inside the lungs and can penetrate the pulmonary alveolar cells, thus mobbing into the blood circulatory system (Xing *et al.*, 2017). It's long been recognized that the air quality of indoor environments is a critical factor for determining human health, but complete data of different areas about this is still missing (Saraga *et al.*, 2017). To determine the health effects of inhaled pollutants, it is necessary to measure the dose deposited in the respiratory tract during exposure (Rissler *et al.*, 2017). But the most prominent cause of lung cancer is radon, so the focus of study in this paper is the measurement of radon, thoron and its progeny concentration and the annual effective dose received by the general public due to these radionuclides.

After generation, radon and thoron migrate through soil into dwellings, but their concentration in dwellings depends on many factors, such as their rate of exhalation in the soil,

* Corresponding author.
Tel.: +919855460699
E-mail address: mehta_vimal78@yahoo.co.in

ventilation conditions and the construction material of the dwelling. Radon poses grave health hazards beyond certain indoor levels, as declared by many worldwide agencies, including USEPA (United States Environmental Protection Agency), UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation), ICRP (International Commission on Radiological Protection) and WHO (World Health Organization). In many developed countries, radon awareness, the measurement of radon, and mitigation programmes exist. Many studies were conducted worldwide to prepare a national geographic information system based on a radon-thoron map (Stoulos *et al.*, 2003; Stojanovska *et al.*, 2013). People in developed countries are concerned about this problem and get their home surveys done (whether for existing older homes or newly built ones). But in countries such as India, Bangladesh and Pakistan, awareness of this problem is low due to low literacy rates along with the nonexistence of specialized awareness programmes. So the people of these countries are more prone to exposure to radon and its health hazards. For these countries, the best remedy is to measure indoor radon levels at as many places as possible and report them if the levels measured are found to be more than the safe limit.

This study is aimed at measuring the concentration of indoor radon and thoron in the dwellings of different places in the study area having different air exchange conditions (depending upon the number of doors and windows) in U.T. Chandigarh, for which very little data is available in the literature. The previous studies of the different areas near the study area were conducted by bare-mode or twin cup dosimeter techniques, which have had their own limitations reported elsewhere (Sahoo *et al.*, 2013). The newly developed pinhole-based radon-thoron dosimeters were used for the study of this area (Sahoo *et al.*, 2013). The calibration of the dosimeters and the systematic errors has been dealt elsewhere (Sahoo *et al.*, 2013). Another important task of this study was to study the correlation of soil exhalation rates with the measured concentration levels of the radon and thoron. The radon mass exhalation rates were measured by the active technique by measuring the growth of radon in a closed sealed accumulator (Sahoo *et al.*, 2007). Scintillation Radon Monitor (SRM) was used to measure the radon growth in the accumulator.

METHODS

Geology of the Area

The Union Territory of Chandigarh is in the foothills of Shivalik Hills, which is a part of the fragile Himalayan ecosystem. The city is the joint capital of both the states of Haryana and Punjab. It is approximately 114 km² in area and is surrounded by Haryana, Himachal Pradesh, Punjab and Uttaranchal. As per the census of 2011, the total population of the city was 1,054,686 persons, with a population density of 9252 persons/sq. km (Gupta, 2007). The altitude of the city ranges from 304 to 365 meters above mean sea level. It is located between 30°40'N and 30°46'N latitude and between 76°42'E and 76°51'E longitude (Gupta, 2007).

Chandigarh observes four seasons, viz. summer, autumn, winter and the rainy season. Summer starts in mid-March and lasts till mid-June, the rainy season starts in late June and lasts till mid-September, autumn starts in mid-September and lasts till mid-November and winter starts in mid-November and lasts till mid-March. The two hottest months are May and June, in which the temperature varies from 40°C and 25°C, respectively (Gupta, 2007). It is surrounded by the Mohali and Ropar districts in Punjab and the Panchkula and Ambala districts in Haryana.

Fig. 1 shows the areas in the different locations of Chandigarh where the measurements of indoor radon and radon exhalation were conducted. The dosimeters were installed in different dwellings at different places of Chandigarh, keeping uniformity in mind. The data of the annual inhalation dose received by the dwellers was analyzed according to the guidelines given by the International Commission on Radiological Protection (ICRP, 2009). The soil samples for the measurement of radon exhalation rates from Chandigarh were taken from the dwellings themselves or an area close to the dwellings. However, some of the soil samples were collected from different parts of Chandigarh, as shown in Fig. 1 (shown by different symbols in the figure). A few sand samples from a local source (Khuda Lahora) and other sources available in hardware stores in Chandigarh were taken.

Indoor Radon-Thoron, their Progeny Concentration and Annual Effective Dose Study

In this study, we report the concentration of radon-thoron measured in single-family dwellings. The study of indoor radon concentrations, which may be representative of population exposure, was carried out in a random sample of dwellings for feasibility reasons. Solid-state nuclear track detectors (SSNTDs) were used in single entry pinhole-based radon thoron dosimeters (PRTM) for the measurement of indoor radon levels. The advantages of this dosimeter over the twin cup dosimeters were discussed by Sahoo *et al.* (2013).

This dosimeter consists of two chambers that are cylindrical in shape and has a length of 4.1 cm and a radius of 3.1 cm. An internal coating of the chambers with metallic powders was done to have zero electric field inside so as to have a uniform deposition of the progenies formed from the gases throughout the volume (Sahoo *et al.*, 2013). The schematics of the pinhole dosimeter are shown in Fig. 2.

The two chambers are separated centrally by a pinhole disc having four pinholes. The pinholes are 2 mm in length and 1 mm in diameter. The disc acts as a thoron discriminator. There is a common entry for both the gases in the dosimeter cups. The first chamber, called the "radon + thoron" chamber, contains a glass fiber filter paper (with a pore size of 0.7 µm) through which the gas enters; then, only radon gas diffuses to the second chamber, called the "radon" chamber, through pinholes that prevent the entry of thoron into this chamber. LR-115 (Type II) solid-state nuclear track detector strippable films, 2 cm × 2 cm in size, were fixed at the opposite ends of the entry face in each chamber. The detector of the first chamber measures the alpha particle tracks due to both the radon and thoron, while the detector

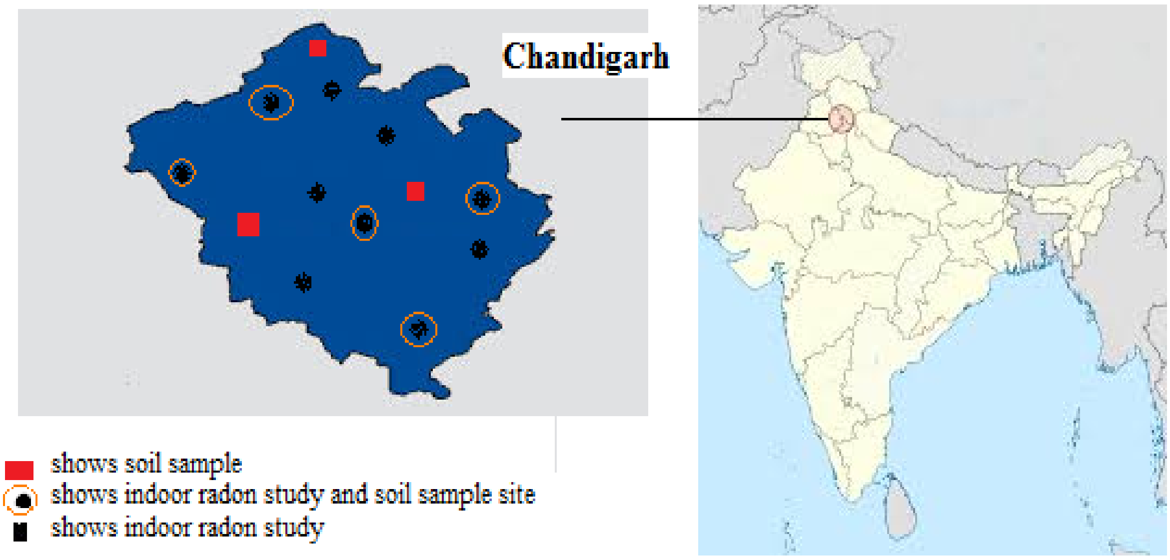

Fig. 1. Location map of the study area.

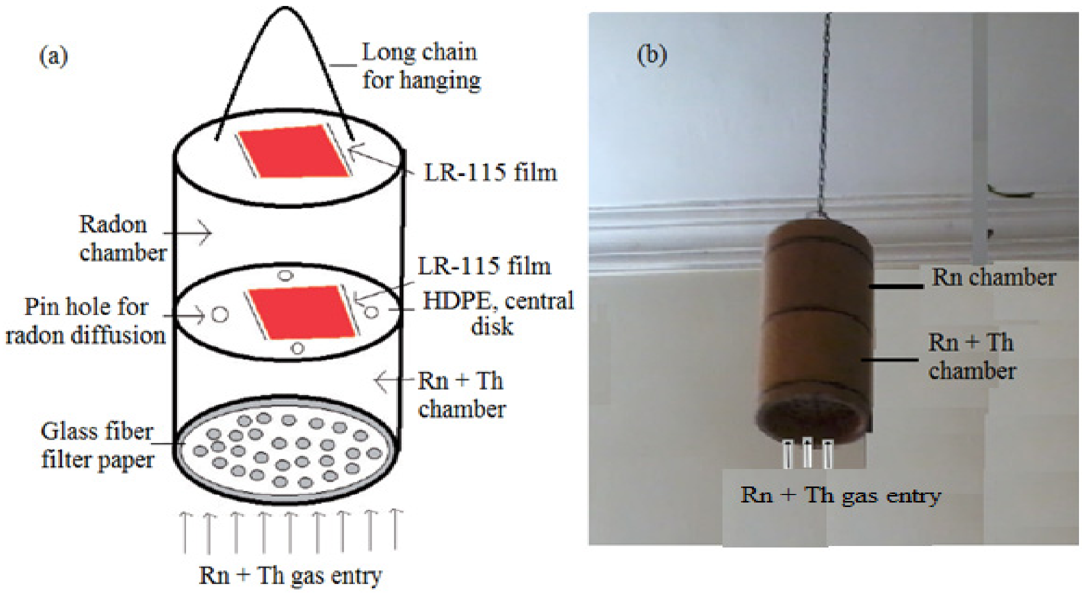

Fig. 2. (a) Schematic diagram (b) Actual single entry radon, thoron dosimeter cup.

of the second chamber measures the alpha tracks due to radon only.

The pinhole dosimeters were hung in the rooms about 1 meter below the ceiling and more than 2 m above ground level. This was done to avoid any direct exposure of the LR-115 films to the alpha particles emitted from the construction material of the ceiling. The dwellers were instructed regarding the detectors and told to clean and ventilate their homes as they normally would. Information regarding the dimensions of dwellings, and the building and roof materials as well as ventilation was gathered during the deployment of dosimeters in the dwellings. The detectors were placed for a period of three months, after which they were removed from the dwellings. During the whole span of study, proper procedures were followed to minimize any extra exposure of the films before and after their exposure in the dwellings. Each dosimeter was prepared shortly before deployment in the dwelling. After retrieval from the dwelling, the detector films were stored in highly ventilated rooms and sealed in radon-proof steel almirah before being processed. The etching and counting of tracks from exposed detectors were carried out by the method described elsewhere (Mehta *et al.*, 2015a, b). The counted track density is converted into the radon and thoron concentration according to the following relations:

$$C_T = \frac{T_2 - d.C_R.K_R^{'}}{d.K_T} \quad (1)$$

$$C_R = \frac{T_1}{d.K_R} \quad (2)$$

where C_R and C_T are the radon and thoron concentrations and T_1 is the track density observed in the "radon" chamber. K_R is the calibration factor of radon in the "radon" chamber (0.0170 ± 0.002 tr. cm^{-2} per Bq.d.m^{-3}); d is the number of days of exposure. T_2 is the track density observed in the "radon + thoron" chamber; K_R' (0.0172 ± 0.002 tr. cm^{-2} per Bq.d.m^{-3}) and K_T (0.010 ± 0.001 tr. cm^{-2} per Bq.d.m^{-3}) are the calibration factors of radon and thoron in the "radon + thoron" chamber (Sahoo *et al.*, 2013). The inhalation dose was calculated using conversion coefficients of 9 and 40 nSv/h/(Bq m^{-3}) with equilibrium factors of $F_R = 0.4$ and $F_T = 0.1$ for radon and thoron, respectively (UNSCEAR, 2006). The dose coefficients for radon and thoron were calculated using a conversion coefficient of 0.17 nSv for radon and 0.11 nSv for thoron. The annual inhalation dose (mSv yr^{-1}) may be provided using the formula (UNSCEAR, 1993):

$$D\ (\text{mSv}) = \{(0.17 + 9F_R)\, C_R + (0.11 + 40F_T)\, C_T\} \times 7000 \times 10^{-6} \tag{3}$$

Radon Exhalation Rate Study (Soil Samples)

Active measurement of radon growth in a closed sealed accumulator was done to measure the radon exhalation rate from soil samples (Petropoulos *et al.*, 2001; Sahoo *et al.*, 2007). The soil samples were taken from the dwellings (if available) or from areas close to the dwellings. Some more soil samples were taken from different parts of the city for uniformity of results. The soils were dried before being sealed in the leak proof exhalation chamber. The chamber was connected to Scintillation Radon Monitor (SRM) to determine the growth of the radon in the accumulator. The measurement was carried out till the radon growth reached a saturated concentration. The growth data was then fitted to Eq. (4) (Kumar *et al.*, 2014) for estimating the radon mass exhalation rate (J_m):

$$C = \frac{J_m M}{V\lambda_e}\left(1 - e^{-\lambda_e t}\right) + C_0 e^{-\lambda_e t} \tag{4}$$

where J_m represents the radon mass exhalation rates in Bq kg^{-1} h^{-1}, and M and V represent the mass of the soil sample and the effective volume of the chamber, including the volume of the scintillation cell, respectively. λ_e represents the effective decay constant, which is the sum of the radon decay constant, radon back diffusion constant and chamber leakage rates, if any, and C_0 is the initial radon concentration in chamber.

Radon Exhalation Rate Study (Sand Samples)

The closed canister technique was used to find the radon exhalation rates of the sand samples (Abu-Jarad *et al.*, 1980; Mehta *et al.*, 2015b). The sand samples collected from the hardware stores in the area under study were dried in an oven, and 100 g of them were placed in plastic cans similar to those used in the calibration experiment of Singh *et al.* (1997) and were sealed. The sensitive side of the SSNTD film was fixed on the lid of the canister to freely expose it to radon. After the exposure period of 100 days, the detectors were removed and processed, and the tracks were counted as per the procedure given elsewhere (Mehta *et al.*, 2015a). The track density was converted into radon activity using the calibration factor of 0.056 tr. cm^{-2} d^{-1}/Bq m^{-3} obtained from an earlier calibration experiment (Singh *et al.*, 1997). The radon mass and surface exhalation rates from the soil sample can be calculated by following relation:

$$E_A = \frac{CV\lambda}{A\left(T + \frac{1}{\lambda}\left(e^{-\lambda T} - 1\right)\right)} \tag{5}$$

$$E_M = \frac{CV\lambda}{M\left(T + \frac{1}{\lambda}\left(e^{-\lambda T} - 1\right)\right)} \tag{6}$$

where C is the equilibrium radon activity inside the canister, V and A are the volume and area of cross-section of the canister, M is the mass of the sample, λ is the radon decay constant and T is the time of exposure.

RESULTS AND DISCUSSION

Concentration of Indoor Radon, Thoron and Annual Dose

The pinhole-based radon-thoron dosimeters were used in autumn to measure the indoor radon and thoron levels in some dwellings of Chandigarh, and the results obtained are listed in Table 1. The necessary information regarding the latitude and longitude of the dwellings chosen is given in this table. The levels of indoor radon, indoor thoron and their progeny concentration are also given along with the amount of the annual dose received by the dwellers. It also has the number of effective doors and windows (small + large), which remained open for at least 8 hrs a day during the study period (the dwellings actually had more windows than reported, but a few remained closed most of the time due to inaccessibility or due to the habit of the occupants). The indoor radon levels in Chandigarh were found to vary from 24.2 ± 1.1 Bq m^{-3} to 62.1 ± 3.1 Bq m^{-3}. The indoor thoron levels varied from 3.0 ± 0.1 Bq m^{-3} to 99.2 ± 4.9 Bq m^{-3}. The average values of indoor radon and thoron were found to be 39.9 Bq m^{-3} and 31.1 Bq m^{-3}, respectively. The progeny levels of radon and thoron were found to be 2.61 to 6.7 mWL and 0.08 to 2.7 mWL, respectively. The averages of the progeny levels for radon and thoron were 4.3 mWL and 0.8 mWL, respectively. The annual inhalation dose for the dwellers was found to be 0.83–3.89 mSv with an average value of 1.9 mSv.

Fig. 3 shows the variation in average indoor radon-thoron levels with the effective number of outlets available in the dwellings for air circulation. This shows the decrease in indoor radon-thoron levels with the increase in the number of outlets. Similar trends are shown in Table 2, which provides the value of the average annual inhalation dose received by the dwellers according to the air circulation conditions, which depend upon the effective number of outlets/vents (doors and windows) operating in the specific room where the dosimeter has been placed. It indicates that the value of the annual dose received by the dwellers was on the lower side when more doors and rooms were open for more than 8 hrs a day. This result supports one of the methods of mitigating radon exposure, which is the evacuation of internal air.

Table 1. Radon, thoron and their progeny levels in some dwellings of Chandigarh in autumn season.

Location	Door (D) + Windows (W)	Latitude & Longitude	Radon Concentration C_R (Bq m^{-3}) AM + SE	Thoron Concentration C_T (Bq m^{-3}) AM + SE	Progeny levels of Radon C_R (mWL)	Progeny levels of Thoron C_T (mWL)	Annual Dose (mSV)
DW-1	1D + 1W	30°44'29"N 76°44'12"E	62.10 ± 3.10	9.87 ± 0.49	6.71	0.27	1.92
DW-2	1D + 2W	30°45'22"N 76°46'32"E	40.52 ± 2.03	10.30 ± 0.52	4.38	0.28	1.37
DW-3	1D	30°44'22"N 76°44'35"E	39.22 ± 1.96	99.22 ± 4.96	4.24	2.68	3.89
DW-4	1D	30°44'17"N 76°44'36"E	56.21 ± 2.81	36.65 ± 1.83	6.08	0.99	2.54
DW-5	1D + 1W	30°44'17"N 76°44'36"E	43.14 ± 2.16	71.35 ± 3.57	4.66	1.93	3.19
DW-6	1D + 1W	30°45'20"N 76°46'22"E	32.06 ± 1.6	19.36 ± 0.9	3.46	0.52	1.40
DW-7	2D + 2W	30°42'31"N 76°44'40"E	27.90 ± 1.1	3.0 ± 0.1	3.00	0.08	0.83
DW-8	1D + 2W	30°41'42"N 76°45'20"E	42.94 ± 2.20	19.35 ± 1.01	4.58	0.51	1.58
DW-9	2D + 1W	30°44'06"N 76°45'55"E	31.40 ± 1.52	6.12 ± 0.29	3.40	0.16	1.01
DW-10	1D + 2W	30°42'22"N 76°44'23"E	24.18 ± 1.3	36.18 ± 1.5	2.61	0.98	1.68

AM = Arithmetic Mean, SE = Standard Error = σ/√N, where σ is SD (Standard Deviation) and N is the no. of observations, DW = Dwelling.

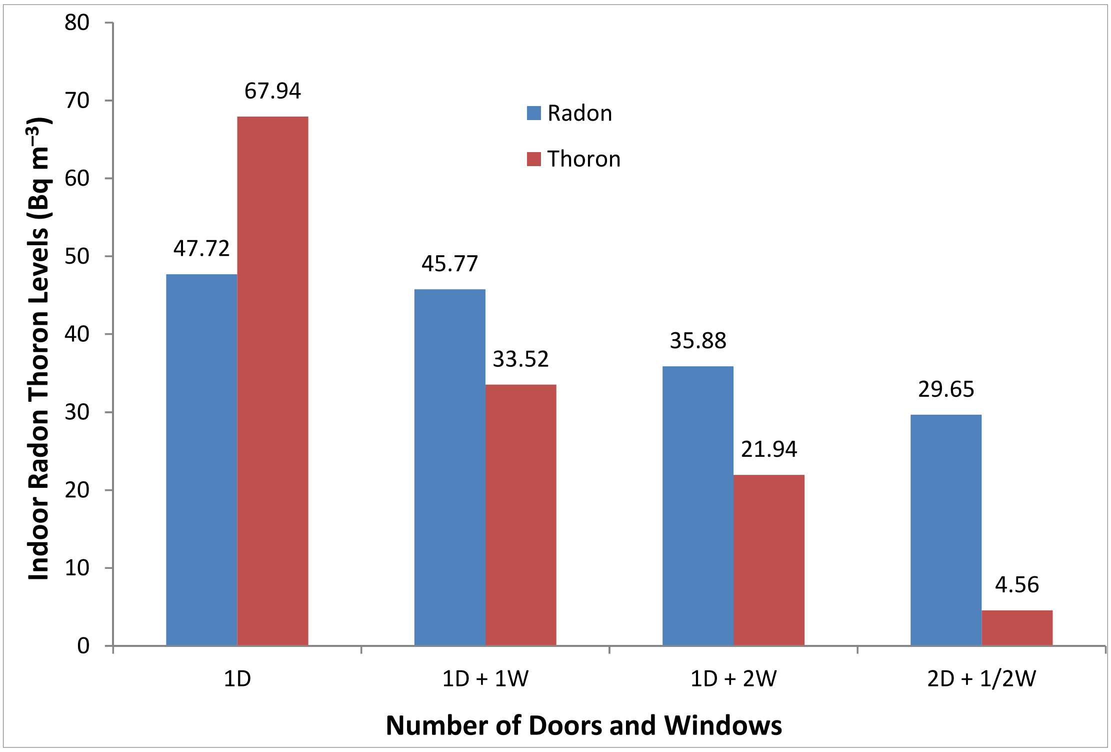

Fig. 3. Variation of indoor radon-thoron levels effective number of outlets for air circulation (D = Doors, W = Window).

Table 2. Dependence of average annual dose upon the air circulation conditions of the dwellings.

S. No	Air Circulation (depends on number of doors and windows)	No. of dwellings	Average annual Dose (mSv)
1	Low (1D)	2	3.22
2	Moderate (1D + 1W)	3	2.17
3	High (1D + 2W)	3	1.54
4	Very high (2D + 1/2W)	2	0.92

D = Door, W = Window.

Soil and Sand Samples Radon Exhalation Rates

The results of the radon exhalation study on different soils collected from Chandigarh are listed in Table 3. The minimum radon mass exhalation rate measured was 1.96 mBq kg^{-1} h^{-1}, and the maximum was 12.52 mBq kg^{-1} h^{-1}. The average value was found to be 5.26 ± 1.18 mBq kg^{-1} h^{-1}. The results of this study in this area show lower values than those of the worldwide average.

The results of the study on both the mass and the surface exhalation rates of the sand samples from different sources available in hardware stores in Chandigarh are shown in Table 4. The values of the mass exhalation rates and surface exhalation rates vary from 0.9 to 1.9 mBq kg^{-1} h^{-1} and 20.1 to 42.7 mBq m^{-2} h^{-1}, respectively. The average values of the mass and surface exhalation rates were found to be 1.4 ± 0.2 mBq kg^{-1} h^{-1} and 31.0 ± 4.9 mBq m^{-2} h^{-1}, respectively. The table also contains the values of the equilibrium radon concentration, which varies from 24.6 to 52.4 Bq m^{-3} with an average of 38.0 ± 5.7 Bq m^{-3}.

A very good positive correlation (R^2 = 0.97) was found between indoor radon concentration and radon mass exhalation rate of the area of Chandigarh as shown in Fig. 4. This correlation is an indication of the direct relationship between the radium content of the soil and the construction materials with the values of radon levels to be present in the dwellings.

CONCLUSIONS

The indoor radon and thoron levels were found to range from 24.2 ± 1.1 Bq m^{-3} to 62.1 ± 3.1 Bq m^{-3} and from 3.0 ± 0.1 Bq m^{-3} to 99.2 ± 4.9 Bq m^{-3}, respectively. The average values of the indoor radon and thoron were 39.9 Bq m^{-3} and 31.1 Bq m^{-3}, respectively. Since the variations of the values for indoor radon activity for all the dwellings of the area under study are well below the recommended level of 300 Bq m^{-3}, radon poses no harmful effects to the people in this area. The average annual inhalation dose received by the dwellers was found to be 0.83 to 3.89 mSv with an average value of 1.9 mSv. However, the amount of this dose is somewhat dependent on the effective number of vents operating in the dwellings. The measured values of the radon mass exhalation rates for the soil in Chandigarh varied from 1.96 to 12.52 mBq kg^{-1} h^{-1} with an average of 5.26 ± 1.18 mBq kg^{-1} h^{-1}. The values of the mass exhalation rates and surface exhalation rates for the sand in the study

Table 3. Radon exhalation rates from soil samples of Chandigarh using scintillation radon monitor.

Sr. No	Location	Radon mass exhalation rates (mBq kg^{-1} h^{-1})
1	DW-2	4.21
2	DW-3	4.22
3	DW-4	5.78
4	DW-7	2.89
5	DW-10	1.96
6	S-1(Near PGI)	7.14
7	S-2(Mullanpur barrier)	12.52
8	S-3 (Khuda Lahoran	3.38
	AM ± SE	5.26 ± 1.18

AM = Arithmetic Mean, SE = Standard Error = σ/√N, where σ is SD (Standard Deviation) and N is the no. of observations, S = Site, DW = Dwelling.

Table 4. Equilibrium radon concentration, mass exhalation and surface exhalation rate in the sand samples.

Sr. No.	Location	Codes	Equilibrium Radon concentration (Bq m^{-3})	Mass exhalation rate (mBq kg^{-1} h^{-1})	Surface exhalation rate (mBq m^{-2} h^{-1})
1	Chandigarh (Khuda	Sand-1	39.2	1.4	31.9
2	Lahora)	Sand-2	35.8	1.3	29.2
3	Other Sources (from	Sand-3	24.6	0.9	20.1
4	hardware store)	Sand-4	52.4	1.9	42.7
	AM ± SE		38.0 ± 5.7	1.4 ± 0.2	31.0 ± 4.9

AM = Arithmetic Mean, SE = Standard Error = σ/√N, where σ is SD (Standard Deviation) and N is the no. of observations.

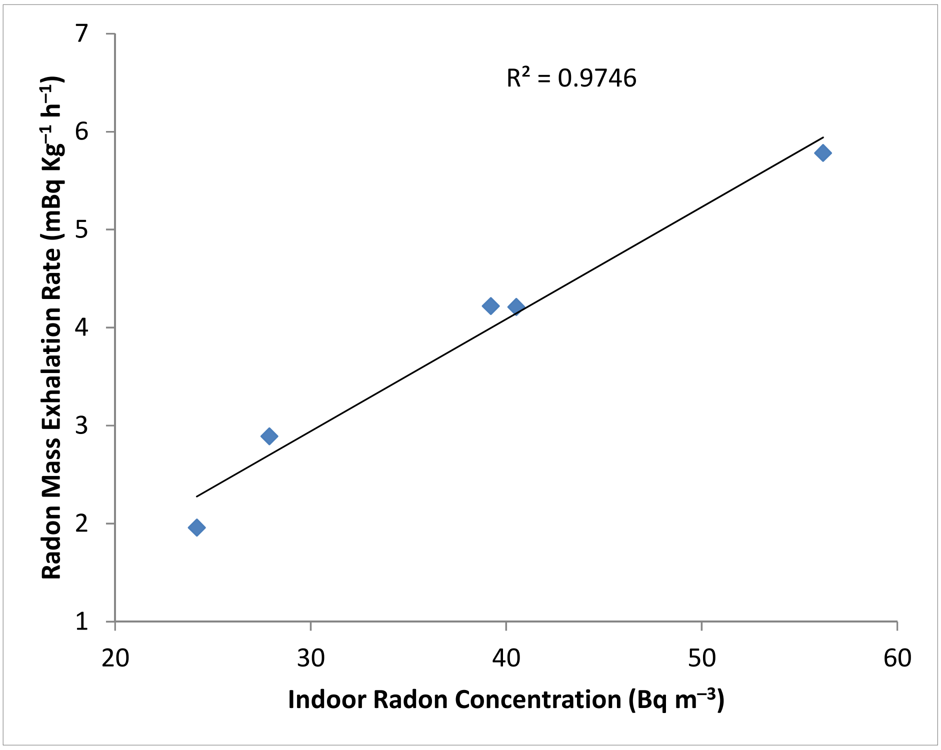

Fig. 4. Correlation between indoor radon concentrations and radon mass exhalation rate.

Table 5. Comparison of indoor radon and annual average dose of the present study with other studies of the nearby areas measured by pin hole dosimeters.

S. No.	Location	Radon Concentration (Bq m^{-3})	Annual Dose (mSV)	Reference
1	Jalandhar, Punjab	36.0	1.27	Mehra *et al.*, 2016a
2	Ludhiana, Punjab	28.0	--	Mehra *et al.*, 2016b
3	Kapurthala, Punjab	42.0	1.69	Mehra *et al.*, 2016a
4	Mansa, Punjab	54.0	--	Bangotra *et al.*, 2015
5	Mukatsar, Punjab	56.0	--	Bangotra *et al.*, 2015
6	Rupnagar, Punjab	20.3	1.3	Mehta *et al.*,, 2015b
7	Hamirpur, Himachal Pradesh	85.0	2.13	Bajwa *et al.*, 2016
8	Kurukshetra, Haryana	21.0	--	Chauhan *et al.*, 2016
9	Panchkula, Haryana	24.0	--	Chauhan *et al.*, 2016
10	Yamunanagar, Haryana	36.0	--	Chauhan *et al.*, 2016
11	Karnal, Haryana	23.0	--	Chauhan *et al.*, 2016
12	**Chandigarh**	39.9	1.9	**Present Study**

area varied from 0.9 to 1.9 mBq kg^{-1} h^{-1} and 20.1 to 42.7 mBq m^{-2} h^{-1}, respectively. The average values of the mass and surface exhalation rates for the sand samples were found to be 1.4 ± 0.2 mBq kg^{-1} h^{-1} and 31.0 ± 4.9 mBq m^{-2} h^{-1}, respectively. The exhalation rate was found to be lower than the average value found in worldwide studies. A strong positive correlation (R^2 = 0.97) was found between indoor radon and the mass exhalation rate of the soil in Chandigarh. The exhalation study of the soil and sand in this place shows values within the safe limits, which indicate safe levels of indoor radon. This indication was confirmed by the results of the indoor radon study. The observed radon values in the present study are comparable with other studies reported for the dwellings of nearby areas, as shown in Table 5.

ACKNOWLEDGMENTS

The author acknowledges the Director of NIT Kurukshetra and the Head of the Department of Physics, NIT Kurukshetra, for the experimental facility provided for the completion of the work. The help received from dwellers of the study area is also thankfully acknowledged.

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Received for review, October 20, 2017
Revised, January 14, 2018
Accepted, January 15, 2018