Chapter 25
Effect of Different Building Materials on Indoor Radon/Thoron and Associated Health Hazards

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Abstract Diverse building materials are utilized for construction all over the world. These emit radioactive pollutants and are liable for continuous exposure of ionizing radiations (radon/thoron) to the indoor environment. As per World Health Organization, radon is recognized as the second significant cause of lung disease subsequent to smoking. Thus, it is of much importance to measure the level of these radioactive gases in dwellings which are constructed using different building materials. The purpose of this chapter is to analyze the impact of different building materials on indoor radon and thoron levels and to find out radon prone regions as it has associated health hazards. In this chapter, indoor radon (^{222}Rn), thoron (^{220}Rn) activity and radon/thoron progeny level were simultaneously measured in 150 houses of twenty locals of district Palwal, Haryana, India. Passive detectors (pinhole twin-cup dosimeter and direct radon/thoron progeny sensors) were utilized for time-integrated monitoring of the exposure period of four months. The variation of radon and thoron gases was observed such as H2 (mud house) > H1 (cemented house) > H3 (traditional house) > H4 (modern house) and H2 (mud house) > H1 (cemented house) > H4 (modern house) > H3 (traditional house) respectively. Similar trends were observed in case of progeny.

Keywords Building materials · DRPS/DTPS · Effective dose · Indoor environment · Pinhole dosimeter · Progeny · Radon/thoron

25.1 Introduction

The primordial radionuclides like ^{238}U and ^{232}Th in soil, rocks, and water are accountable for the presence of ^{222}Rn and ^{220}Rn in indoor and outdoor environments. A radionuclide decays into its daughter products and releases energy as alpha, beta or gamma particles. Then the newly formed nuclei become the head of the decay chain

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and further decay. Radon and Thoron are the daughter nuclides of ^{238}U and ^{232}Th decay chain. Uranium-238 decay series indicates that the primordial radionuclide uranium-238 is unstable radionuclide and therefore decays into thorium-234 with the release of an alpha particle of energy 4.2 meV to attain stability. But thorium-234 is also unstable and further decay into uranium-234 by emitting beta particles. Uranium-234 further decays into thorium-230 by emitting an alpha particle. Thorium-230 decays into radium-226 by emitting an alpha particle. This radium isotope (^{226}Ra) is a generator of Radon-222 after releasing an alpha particle. Similarly, the thorium-232 decay series indicates that thorium-232 primordial radionuclide is also unstable and goes to radioactive decay. After three alpha decays it reaches Thoron (Radon-220). In both, the decay series only Radon and thoron radionuclides are found in gaseous form. Thus, radon and thoron further decay to attain stability. Decay products of radon are listed as short-lived and long-lived progeny. The short-lived decay products of Radon are ^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po, and long-lived decay products are ^{210}Pb, ^{210}Bi, ^{210}Po. Thoron short-lived decay products are ^{216}Po, ^{212}Pb, ^{212}Bi, ^{212}Po. Uranium-238 decay series stops at lead-206 which is a stable nuclide. In this decay series 8 alpha, 6 beta, and associated gamma energy are released. Thorium-232 decay series stops at lead-208 which is a stable nuclide. In this decay series 6 alpha, 4 beta, and associated gamma energy are released. In the periodic table Radon comes in between metals and nonmetals. Thus it is metalloid and has properties of both. It is present diagonally in the periodic table. It is an inert gas and chemically non-reactive. The chemical symbol of Radon is Rn with atomic weight 222 and atomic number 86. It has protons and electrons, 86 protons and 86 electrons in its atomic structure. As the electro-negativity decreases with increase in the atomic number in the column of the periodic table, Radon has low electro-negativity compared to other noble gases. The solubility increases with an increase in atomic number therefore Radon is more soluble than other inert gases. Its solubility is more in organic liquids as compared to water. Also, it was observed that heat vaporization increases and ionization energy decreases with increase in atomic number.

Radon and thoron are inert, carcinogenic and alpha emitter gases. Radon and thoron reach the air viz., indoor environment through many paths. The soil under or nearer the dwellings and construction material are the major sources of the radon gas to the indoor environment whereas only building materials are major sources for thoron gas to the indoor environment. Radon and thoron gas reached the environment from rocks and soil grains through two fundamental processes. The first and the second process is emanation and exhalation from the material grain and the matrix using many transport mechanisms. The process through which radon atoms liberate from the solid mineral grains to the air filled pores is known as emanation. Consequently, transportation of radon gas from the pores of the air to the atmosphere is called exhalation. The transportation of radon in soil pores is significantly due to advection brought out by pressure driven flow of soil gas and diffusion brought out by concentration gradient. The molecular weight of radon is almost 8 times that of air so travels closer to the ground and the progeny can be accumulated as solid radioactive offshoots on water, vegetation and surface of soil. The diffusion of the radon in air is due to Brownian motion. It is well understood that molecular diffusion and advection are responsible for Radon/Thoron transportation through emanation and exhalation processes.

Radon gas reaching the dwellings can accumulate for a longer time (half-life is 3.8 days) and can travel up to 3 m in the air. Thoron gas cannot migrate up to longer distances because of its shorter half-life (55.6s). Firstly, Radon/Thoron gases reach the indoor environment from building materials, soil nearer, or under the ground surface. They diffuse to the indoor environment and decay by emission of alpha particles. The decay products of Radon/Thoron are the isotopes of polonium, bismuth, and lead. These decay products are the combination of coarse and fine fractions of polonium, bismuth, and lead. Most of these progenies are positively charged particles and less are neutral particles. These charged particles can attach with the air vapors and trace gases present in the atmosphere and form clusters and further deposits to surfaces (Fares et al. 2011). Otherwise, these charged particles can interact with aerosol and sulfur dioxide and then become neutral and deposit to the surface. This deposition is mainly governed by the gravity force and Brownian diffusion. Thus, it can occur either by transport towards the surface or by precipitation of aerosol particles on the surface.

The pressure difference, moisture, porosity of the medium, permeability, and temperature has a significant influence on radon concentration in soil. Radon and thoron are transported to the indoor environment through cracks in surface soil, joints of walls and floors of dwellings, pores in buildings materials, etc. The source of radon into the indoor environment is through water. Household activities such as showering, laundering, dishwashing and other activities are the ways to transport radon to an indoor environment. Parameters like temperature, atmospheric pressure, ventilation condition, moisture content, permeability of soil, emanation coefficient, etc. affect the concentration of radon in the environment (Janik et al. 2015).

The World Health Organization reported that over exposure of radon may be considered as a key factor of lung diseases for non-smokers (WHO 2009). During breathing, they may enter the human body and can damage the bronchial epithelial living cells. Secretion and Bronchial stem cells in airways are the primary target cells for lung cancer induction due to the exposure of radon. The deposited daughters of the radionuclide decay and emit ionizing particles such as alpha, beta, and gamma. The densely ionizing alpha radiation causes the potential damage to the deoxyribonucleic acid (DNA) of lung cells. This will cause the initiation of the chain of events leading to lung cancer. However beta particles and gamma rays have lower biological effectiveness and longer range thus have negligible effects on lung tissues. Health hazards from radon are relatively smaller than what is expected from its progeny–^{218}Po, ^{214}Po, ^{214}Pb, and ^{214}Bi due to their longer half-life. The major contribution (>50%) of total dose of ionizing radiation received by the general public is due to radon, thoron, and their progeny. The results of occupational investigations as well as residential studies indicate the human carcinogenicity of radon (IARC 2011). Thirteen case controls studies in Europe and seven case control studies in North America indicated that increase in indoor radon gas is associated with lung cancer risk (Darby et al. 2005). It is predicted by Elío et al. (2018) that household radon can considerably increase the risk of lung diseases. Thus, due to its radiological impact on humans it is very important to simultaneously measure radon, thoron and their progeny levels in indoor regions.

Various investigations were carried out earlier for simultaneous estimation of radon and thoron gases by passive methods in India and other countries (Singh et al. 2019a). Solid state nuclear track detectors based passive equipment such as pinhole dosimeters, Raduet dosimeters, direct progeny sensors were used for these measurements worldwide. In the present investigation, LR-115 type II film based pinhole dosimeters and direct progeny sensors were used to register the alpha tracks formed due to decay of radon/thoron and their progeny. In the past many studies have focused on radon measurements using passive methods. However, thoron had been often neglected and considered as an interrupting factor in the measurement of radon. This may not be entirely correct in the context of India which has regions consisting of thorium rich soil and high background radiation areas (HBRAs). In addition, contrary to the gas measurement, limited investigations were done for direct measurement of decay products. The estimation of decay products concentration is usually calculated from gas concentrations using the equilibrium factor approach. But, for estimating thoron decay products concentration this may not be an appropriate method. Moreover, since the inhalation doses are dominantly because of the decay products of radon and thoron, and not due to the gases, it is important to measure the decay products directly. Thoron along with its progeny is also dominant contributor to annual effective dose due to inhalation as reported in many investigations. Moreover in Indian scenario, the thoron contribution for inhalation dosimetry has also been acknowledged. The exponential decay in thoron concentration from the surface of the wall has been observed. However, a quantification of dose contribution either from thoron or its progeny alone suggests that the majority of dose will come from thoron progeny (~98%) with a very little contribution (~2%) from thoron gas. Therefore in this study, thoron gas and its progeny have been monitored separately using a radon-thoron discriminating dosimeter (Sahoo et al. 2013) and Direct Thoron Progeny Sensor (Mishra and Mayya 2008) respectively and both quantity have been used for dose calculations. Hence the estimated dose will be dominated by the measured value of thoron progeny which is more or less uniform in dwellings and gives reliable results. However, for the completeness of dose, we have added the marginal contribution by thoron gas too.

The aim of this chapter is to compare the impact of different building materials on radon/thoron levels. Secondly, the data will be compared with the safety limits recommended by various agencies like WHO, UNSCEAR, ICRP, etc. For the present work, the area chosen for investigation is under reported and from a geographical point of view it is very important to understand the effect of radiation in this region. This study is conducted under a major project provided by the Board of Research in Nuclear Sciences (BRNS), Bhabha Atomic Research Center (BARC), Mumbai, Government of India. The project will cover the radiation measurement in soil, water, and air of this region. The data provided by this paper will be helpful for the researchers to understand the effect of building materials on radiations. This study is a part of seasonal monitoring (summer, rainy and winter seasons) of indoor radon/thoron and their progeny level in villages of district Palwal, Haryana, India.

25.2 Geology of the Study Area

The study area of the present investigation is located in the southern part of the state of Haryana of Northern India. The latitude and longitude extend from 27°50′29″N to 28°12′30″N and from 77°17′47″E to 77°22′47″E. The area includes 282 villages and the city region of Palwal. It is bounded by districts Gurugram and Faridabad in north-west, by district Mewat in west, and by state Uttar Pradesh in east. About 270 km^2 are irrigated by surface water sources and 770 km^2 by ground water sources. The entire study region has almost flat plains. The soil of this region is tropical and brown. Organic contents in soil are in the range of 0.2 to 0.4% and pH of soil lies in between 6.5 and 8.5. The underground water sources such as borewells, hand-pumps, tap water, etc. and surface water sources such as river Yamuna, Gurugram and Agra canals are present in this region. Tropical and brown soil is present in the major part of district Palwal. Geo-morphological information of the district indicates that organic content in soil varies from 0.2 to 0.4% and in Hathin block it varies from 0.41 to 0.75%. The pH of soil varies from 6.5 to 8.7. The entire study region has almost flat plains. In Palwal, 770 km^2 are irrigated by borewell and 270 km^2 by canals. The sand and gravel are major water-bearing formations. Hydro-geological information of the district describes the study region engrossed by the Indo-Gangetic alluvial plain of the Quaternary age. The main underground water horizon made up of alluvium comprises gravel, kankar, and sands silt. This hydro-geological, geo-morphological, and geological information of this region is based on a report of the Central Ground Water Board (CGWB 2013), government of India.

25.3 Materials and Methods

25.3.1 Preliminary Survey of the Study Area

For impactful study and systematic investigation, a survey has been done before the deployment of detectors. Outdoor gamma level was measured during the preliminary survey to categorize the region into zones to get the zone-wise distribution of radionuclides. During this survey gamma level is measured at a height of 1 m from the ground to avoid any lead or interference by decay products generated in air. The progeny of radon gas (^{214}Pb, ^{214}Bi and ^{210}Pb) and thoron (^{212}Pb, ^{212}Bi and ^{208}Tl) present in the soil are the main sources of gamma radiations. Therefore, quantification of outdoor gamma level is also performed to explore any correlation with parent nuclides such as radon and thoron gases. The Geiger Muller counter based Survey meter (Polimaster PM/1405, Garmin Instrument, Republic of Belarus) was used to measure outdoor gamma level at one meter height from the earth surface. Survey meter incorporates a large energy compensated Geiger Muller tube for precise measurement of the ambient equivalent dose rate of the gamma radiation in the range from background level to 100 mSv/h (10 R/h). It has a gamma energy response from

0.05 to 3 meV and can be used for dose rate measurement varying from 0.01 to 130 mSv h^{-1} suggesting suitability for environmental gamma surveys. It has a calibration accuracy of $\pm(20 + 1/H)$ % where H is the dose rate in μSv h^{-1}. Also, the different types of residential houses in the study region were observed which further categorized based on construction building materials.

25.3.2 Categorization of Investigated Houses

The building material of houses, underground and surrounding surface soil, water used in houses are major contributors to indoor radon level while only building materials are a major source of indoor thoron level as it has a very short half-life. Several building materials such as gypsum, black cement, white cement, stone dust, bricks, marble, tiles, granite, and POP were tested by researchers to find out the level of radon concentration, radon mass exhalation rate, and thoron surface exhalation rates. A wide variation was observed in the level of these radioactive elements in these building materials. It indicated that the different building materials have different impacts on these radioactive elements therefore a systematic investigation was required for it. Therefore the dwellings of the study region were categorized into four types viz H1, H2, H3 and H4 on the basis of building materials used for construction as shown in Fig. 25.1a, b, c, and d respectively.

The first category (H1) included the dwellings having roofs made up of girder and stone slab, walls of houses made up of fired bricks covered with plaster layer and floor covered with cement-plaster layer. Generally, these types of dwellings are present in most of the study region area and nearby areas of similar geological conditions.

The second category (H2) included the dwellings having thatched roofs, walls of houses made up of fired bricks covered with a layer of mixture of clay soil and cow/buffalo dung and open ground floor with coats of the same mixture used on walls. However, the quantity of such dwellings is less compared to the H1 category but peoples all over study regions used these dwellings in present time also. Therefore, this category was also a point of interest for monitoring the radionuclide elements.

The third category (H3) included the dwellings having roof made up of concrete and beam and the roof structure standing on the columns of beam, walls of houses made up of fired bricks and columns at corner and middle of walls which covered with plaster layer and floor covered with cement-plaster layer. These types of dwellings are replacing the category of H1 in the present scenario. Dwellings which were made up of H1 category building materials when damaged over a long period of time are replaced by H3 category. Thus, this category was also a point of interest for measurements of radionuclide pollutants level.

The fourth category (H4) is named as modern houses. It includes the dwellings having roof made up of concrete and beam and the roof structure stand on the columns of beam same as of third category, walls of houses made up of fired bricks and columns at corner and middle of walls which covered with plaster layer and floor covered with

Fig. 25.1 Interior view of investigate houses **a** houses of type H1, **b** houses of type H2, **c** houses of type H3, and **d** houses of type H4 of district Palwal, Southern Haryana, India

cement-plaster layer, also the walls (up to 3 to 4 feet height) and floor covered with tiles, marble, stones etc.

However, there were two more categories viz mud houses and Haveli (tradition in some regions). Mud houses were made up of thatched roofs, walls of houses made up of clay pieces with a layer of mixture of clay soil and cow/buffalo dung and open ground floor with coats of the same mixture used on walls. However, the quantity of such dwellings is very less compared to other residential dwellings. Haveli were made up of small fired brick walls with an open or covered floor. Stone pillars were part of the attraction in this category and had a roof made up of wooden pieces and stone slab. But these were very few in quantity and not to be used for residential or work purposes by the public in current time therefore neglected for investigation.

25.3.3 Pin-Hole Based Dosimeter and Deposition Based Direct Progeny Sensors (DRPS/DTPS)

Pin-hole dosimeter relies upon radon/thoron isolation technique. The length and the radius of the two compartments of the pin-hole dosimeter are 4.1 cm and 3.1 cm respectively. The segregation of the two compartments is done with the help of the disc having thickness 2 mm and 4 pin-holes of diameter 1 mm. The front compartment is a radon + thoron chamber and the rear one is of radon. The complete dimension of the dosimeter is chosen in such a way that the thoron entry into the rear compartment is prohibited. The paper of thickness 0.56 μm is placed at the entry face. The air containing both radon and thoron enters into the front chamber and subsequently the air containing only radon diffuses to the rear chamber through pin-holes. Inner surface of the dosimeter compartments and central disc is coated by metallic substances such as nickel to form a neutral electric field inside the compartment volume. It helps to uniform deposition of charged progeny throughout the inner surface of the dosimeter. Solid-State Nuclear Track Detectors (SSNTDs) are insulating solids widely used for passive measurements. These detectors include plastics, inorganic crystals, glasses, etc. Cellulose nitrate (CN 85, LR-115), allyl diglycol carbonate (CR-39), bisphenol-A polycarbonate (Makrofol, Lexan), etc. are used as SSNTDs. To measure the activity of radionuclides in the air or in powder samples the SSNTDs are widely used. The alpha track etch technique is one of the most widely used techniques to register the tracks created from ionizing radiations (alpha particles). When the radionuclides such as radon and thoron decay they emit alpha particles which can be detected by using SSNTDs films. The alpha particles when passed through the passive detectors release their energies and leave the tracks in detector films. The registration of tracks (latent tracks) in a given SSNTD depends on the orientation, energy, etc. of ionizing particles. These tracks cannot be visualized through a scanning electron microscope and optical transmission microscope because the size of latent tracks is very small (diameter in the range of 1–10 nm). Therefore, by using suitable chemical etchant or reagent the size of these tracks can be enlarged. Thus, fully developed tracks can be visualized or counted by a transmission optical microscope also or by using a spark counter. SSNTDs are easy to handle, are unaffected by humidity, store data up to many years, have low cost, used for time integrating measurements, etc. The type-II LR-115 film (Kodak Path, France) having 12 μm thick cellulose nitrate on a 100 μm thick polyester base has been used to record the tracks generated by alpha particles due to radon and thoron gases inside the chamber. The track recording efficiency of these detectors ranges from 1.7 to 4.8 meV.

DRPS (deposition based direct radon progeny sensors) and DTPS (deposition based direct thoron progeny sensors) were used for measurement of radon/thoron progeny levels in the indoor dwellings. A combination of LR-115 film and a suitable absorber has been used in DRPS and DTPS. DRPS comprises LR-115 film and an absorber of 37 μm thickness (25 μm Mylar sheet and 12 μm cellulose nitrate). The detecting efficiency of DRPS is upto alpha particles of energy 7.67 meV emitted from Polonium-214. DTPS comprises LR-115 film and an absorber of 50 μm thick Mylar sheet. The detecting efficiency of DTPS is upto alpha particles of energy 8.78 meV emitted from Polonium-212. The number of tracks estimated by DRPS and DTPS are used for the calculation of EEC (Equilibrium Equivalent Concentration) using the suitable sensitivity factor. The minimum detection limit for DTPS is 0.1 Bq m^{-3}, whereas that for DRPS is 1.0 Bq m^{-3} which arises due to intrinsic background track density. Intrinsic background tracks are those tracks which are registered on the detector films during transit, manufactured, or packaging period. Detectors having a size of 2.5 cm × 2.5 cm were loaded in DRPS and DTPS were used for the present investigation. The pin holes twin cup dosimeter has been calibrated against standard radon and thoron sources (Model RN 1025 and TH 1025, Pylon, Canada) in a 0.5 m^3 calibration chamber available at Bhabha Atomic Research Centre (BARC), Mumbai, India. Relative humidity controls from 10 to 99% and temperature from 20° to 50 °C in the calibration chamber. DRPS/DTPS were calibrated with active Working level monitors from Tracer lab, Grab-filter-paper sampling and alpha-counting at BARC, Mumbai India.

Dosimeters along with DTPS/DRPS were deployed based on the weight factor assigned to each category of houses after regression analysis. Detectors were deployed for a period of four months (July–October) during the rainy season in district Palwal, Haryana, India according to the standard protocol of Bhabha Atomic Research Center, Mumbai. Detectors were collected back from the dosimeters and progeny sensors on completion of the monitoring time. During this exposure period, tracks are registered on the detectors. These tracks cannot be visualized through a transmission optical microscope because the size of latent tracks is very small (diameter in the range of 1–10 nm). Therefore, by using suitable chemical etchant or reagent the size of these tracks can be enlarged. Thus, fully developed tracks can be visualized or counted by an optical transmission microscope or spark counter. In this study, a constant etching bathtub (model PSI-CTB1) is used for the etching of detectors. It has three compartments (tub), a temperature controller, a timer, a heater coil, and a pump for the circulation of water. The first calibration of the equipment has been carried out by the manufacturer. However, we also calibrated it for bulk etch removal rate of unexposed detector films. A solution of 2.5 N NaOH is prepared and filled in all three compartments of the tub. The temperature is set at 60 °C temperature and after 25 min the solution in all compartments are checked by the thermometer to ensure the temperature of the solution. The detectors are marked by punching at the corner and loaded in a cartridge and put into the compartments. The timer is set for 90 min at this temperature. After the completion of etching time, the cartridges are removed from the compartments and washed with flowing tap water. The detectors are then washed in distilled water and dried for one hour and now the detectors are ready for spark counting.

After the etching process, the next step is to find out the number of tracks on detector films registered due to the alpha particles. Spark counter (we used model PSI-SC1) is used for this purpose. In the spark counter, the thin etched track detector (about 8–10 μm thick) is placed between two electrodes forming a capacitor. The bottom electrode is a thick conductive electrode, commonly made of brass. The thin LR-115 detector is placed on this electrode. The aluminized Mylar is placed on the detector such that the aluminized surface faces the detector as well as the thin electrode. A heavy weight is placed on the top to ensure good contact between the electrodes, the detector, and the aluminized film. These track densities were used to estimate the radon/thoron activity and progeny concentration.

The radon (C_R) and thoron (C_T) gas level were calculated from the number of tracks per unit area observed in exposed LR-115 detector and given by Eqs. (25.1) and (25.2) respectively;

$$C_R\left(\mathrm{Bq\ m^{-3}}\right) = \frac{(T_1 - B)}{d \cdot K_R} C_R \tag{25.1}$$

$$C_T\left(\mathrm{Bq\ m^{-3}}\right) = \frac{\left(T_2 - d \cdot C_R \cdot K_{R'} - B\right)}{d \cdot K_T} C_R \tag{25.2}$$

where T_1 track density (rear chamber), B is the number of tracks per unit area raised from background, d is monitoring days, K_R has the value 0.017 ± 0.002 tr cm^{-2} d^{-1} Bq m^{-3} (rear chamber calibration factor), T_2 track density (front chamber), $K_{R'}$ has the value 0.0172 ± 0.0 02 tr cm^{-2} d^{-1} Bq m^{-3} (front chamber calibration factor for radon) and K_T has the value 0.010 ± 0.001 tr cm^{-2} d^{-1} Bq m^{-3} (front chamber calibration factor for thoron).

The EETC is calculated from Eq. (25.3) and the EERC from Eq. (25.4);

$$EETC\left(\mathrm{Bq\ m^{-3}}\right) = \frac{(T_T - B)}{S_T} \tag{25.3}$$

$$EERC\left(\mathrm{Bq\ m^{-3}}\right) = \frac{(T_{Rn} - B)}{S_R} \tag{25.4}$$

where T_T is the track density in DTPS, S_T and S_R are sensitivity factors for thoron and radon progeny respectively, T_{Rn} track density from radon progeny in DRPS;

$$T_{Rn}\left(Bqm^{-3}\right) = T_{DTPS} - (\eta_{RT}/\eta_{TT})T_{DRPS} \tag{25.5}$$

where T_{DRPD} and T_{DTPS} are total number of tracks in DRPS and DTPS respectively, η_{RT} (0.01 ± 0.0004) and η_{TT} (0.083 ± 0.0004) track registration efficiency for thoron progeny in DRPS and DTPS respectively.

Total annual effective dose (AED_{Rn+Th}) due to inhalation was estimated. It is the sum of annual effective dose calculated from measured concentration of radon (C_{Rn}) along its progeny that is EERC (equilibrium equivalent radon concentration) AED_{Rn} and calculated from thoron (C_{Th}) along its progeny that is EETC (equilibrium equivalent thoron concentration) AED_{Th}.

Annual effective dose due to inhalation of radon and its progeny is calculated from equation

$$\mathrm{AED_{Rn}(mSv\ y^{-1})} = (\mathrm{C_{Rn}} \times \mathrm{FC_{Rn}} + \mathrm{EERC} \times \mathrm{FC_{EERC}}) \times 8750 \times \mathrm{O_F} \times 10^{-6} \tag{25.6}$$

Annual effective dose due to inhalation of thoron and its progeny is calculated from equation

$$\mathrm{AED_{Th}(mSv\,y^{-1})} = (\mathrm{C_{Th}} \times \mathrm{FC_{Th}} + \mathrm{EETC} \times \mathrm{FC}_{EETC}) \times 8750 \times \mathrm{O_F} \times 10^{-6} \tag{25.7}$$

where FC_{Rn} (0.17 nSv Bq^{-1} h^{-1} m^3) and FC_{Th} (0.11 nSv Bq^{-1} h^{-1} m^3) are dose conversion factors for radon and thoron concentration respectively, FC_{EERC} (9 nSv Bq^{-1} h^{-1} m^3) and FC_{EETC} (40 nSv Bq^{-1} h^{-1} m^3) are dose conversion factors for radon progeny concentrations and standard occupancy factor (O_F) is 0.8 for 1 year exposure period.

25.4 Results and Discussion

25.4.1 Distribution of Radionuclides

A heterogeneous distribution of radon, thoron, and their progeny concentrations was observed in this study. Literature review revealed that the distribution of radon and thoron are affected by local geology of testing sites, environmental parameters such as moisture contents, atmospheric pressure, temperature difference inside and outside of the houses, radon emanation factor of soil under the surface of floors, etc. The measured radon and thoron activities vary from 4 to 175.1 Bq m^{-3} with an average of 28.2 ± 2.1 Bq m^{-3} and from 2.1 to 195.2 Bq m^{-3} with an average of 29.5 ± 2.8 Bq m^{-3} respectively. The overall thoron concentration was found higher as compared to radon level in district Palwal, Haryana, India. It is due to thorium rich soil in the earth's crust of India. The average radon level observed here is less than the world average value reported for indoor dwellings of 40 Bq m^{-3} (Singh et al. 2019a) and also less than the indoor radon reference level of 100 Bq m^{-3} (WHO 2009) and 200 Bq m^{-3} of ICRP (2014). Radon was found higher than 100 Bq m^{-3} in three dwellings and it can be attributed to natural geology of location, more exhalation of radon from joints of walls or cracks, etc. Therefore, further measurements are required at these locations to ensure the reasons for high values of radon. The average thoron level was observed at 29.5 ± 2.8 Bq m^{-3} which is nearly 3 times higher than the worldwide average value reported for dwellings of 10 Bq m^{-3}. However this will not significantly affect the total dose contribution as thoron has negligible effect on the total dose.

The average radon and thoron concentrations are found highest in mud houses (type H2) which can be attributed to excessive emission of these radioactive gases from the open floor (mud surface). The walls of these houses were made up of fired bricks covered with a layer of clay soil and cow/buffalo dung therefore continuous emission of radon and thoron gases from cracks and opening of walls are responsible for elevated levels. Also, the dimensions of these types of dwellings in comparison with other categories of houses is generally small resulting in improper ventilation and raised the level of radon and thoron. It is also in agreement with results of other investigations (Sannappa and Ningappa 2014; Suman et al. 2020). Radon concentration found second highest in cemented houses (type H1). In cemented houses walls are of fired bricks covered with cement plaster and floor is also cemented; this reduces the emission of radon and thoron gases from earth's crust due to low permeability of cement plaster. The average radon concentration is found lowest in modern houses (H4). However use of enhanced materials like marble, granite, tiles, stones etc. on walls and floors of the dwellings is expected for higher radon emission inside the houses. But this trend was not observed in our study. Thus the results in the present investigation contradict the results of other investigations in the case of elevated radon levels in modern houses (Singh et al. 2019a). Most probable reason for low levels of radon in modern houses is the proper ventilation conditions of rooms having two doors and a window which leads to air exchange between rooms and the outdoor atmosphere. The variation of radon and thoron gases was observed such as H2 (mud house) > H1 (cemented house) > H3 (traditional house) > H4 (modern house) and H2 (mud house) > H1 (cemented house) > H4 (modern house) > H3 (traditional house) respectively.

The variation of EERC is from 1.1 to 41.4 Bq m^{-3} and has an average of 9.1 $\pm$ 0.02 Bq m^{-3} and EETC is from 0.2 to 6.8 Bq m^{-3} and has an average of 1.2 $\pm$ 0.01 Bq m^{-3}. As per the ICRP, limits of average EERC and EETC are 2–50 Bq m^{-3} and 0.04–2 Bq m^{-3} respectively and the present data confirms that the progeny concentration is within the limits in this study area. The average values of EERC (9.1 $\pm$ 0.02) and EETC (1.2 $\pm$ 0.01) in the present study were found below the world average values of 15 Bq m^{-3} for EERC and 0.5 Bq m^{-3} for EETC. In the present investigations, thoron concentration was found higher than radon concentration but reversed in case of progeny. This can be attributed to the different effects of environmental parameters such as temperature and pressure gradient, moisture content, and ventilation conditions, etc. on the gases and their solid decay products. The overall concentration of radon progeny was found higher than the thoron progeny. It is presumably due to higher deposition velocities of radon progeny than the thoron progeny on detector surfaces. Mean values of decay products of radon and thoron gases are found highest in mud houses (type H2) similar to the results observed for radon and thoron gases. The variation of decay products of radon and thoron was observed such as H2 (mud house) > H1 (cemented house) > H3 (traditional house) > H4 (modern house) and H2 (mud house) > H1 (cemented house) > H4 (modern house) > H3 (traditional house) respectively same as observed for radon and thoron gases.

The box whisker plots of measured indoor radon, thoron, radon progeny (EERC), and thoron progeny (EETC) are shown in Fig. 25.2. The upper and lower whisker represents maximum and minimum concentration value respectively. The top line

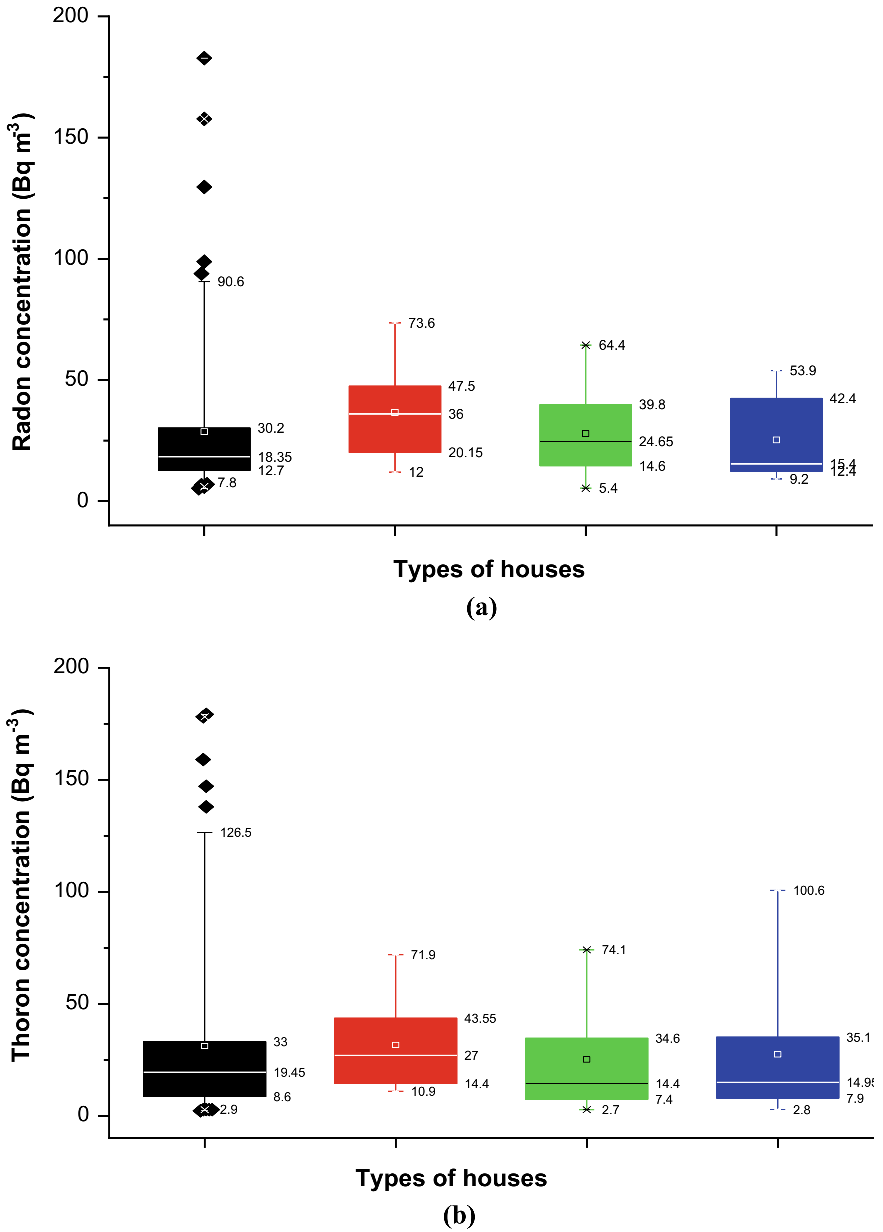
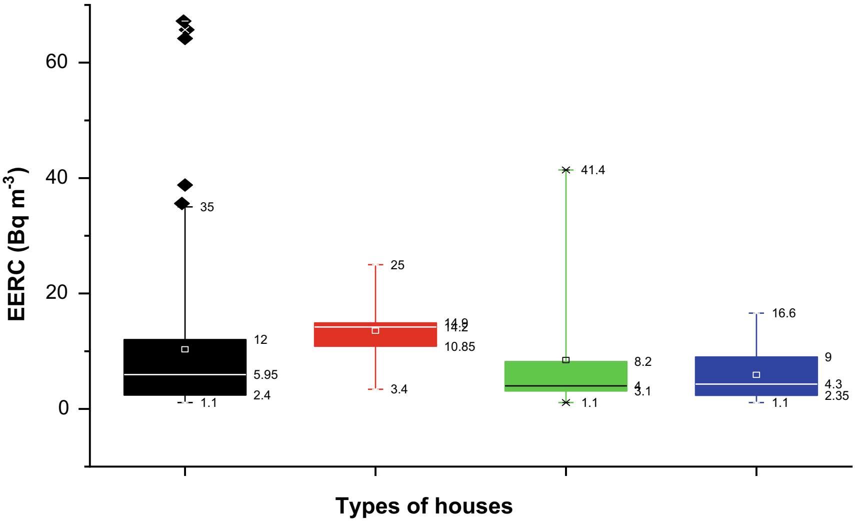
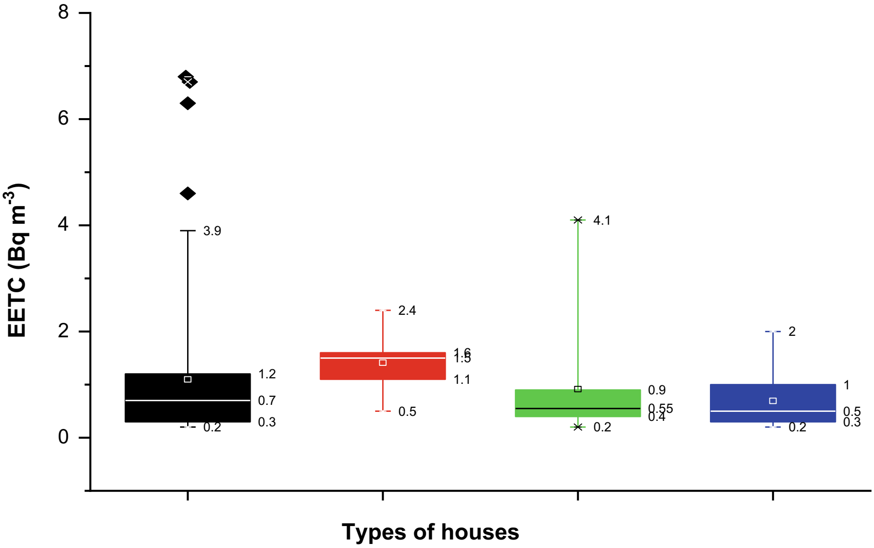


Fig. 25.2 Box-whisker plots of measured **a** radon, **b** thoron, **c** EERC, and **d** EETC in different type of houses of district Palwal, Southern Haryana, India (for H1 type represents by black colour plot, for H2 type represents by red colour plot, for H3 type represents by green colour plot, and for H4 type represents by blue colour plot)



(c)



(d)

Fig. 25.2 (continued)

of the box indicates the third quartile that is 75 percentile, the middle line indicates the second quartile or median that is 50 percentile, and the lower line represents the first quartile that is 25 percentile. Outliers (represented by dots above or below the maximum or minimum values in box-whisker plot) are values which lie 1.5 times greater or lower than the top or below lines of the box. The minimum, maximum, mean, first quartile, median, and third quartile values for radon and thoron and their progeny concentration for different types of houses are shown in Table 25.1.

Table 25.1 Variation of radon, thoron and their decay products (EERC and EETC) concentration in different type of investigated houses for rainy season of district Palwal, Haryana, India

Parameters	Statistical parameter	House category			
		Type H1	Type H2	Type H3	Type H4
Radon (Bq m^{-3})	Min	5.2	11.9	5.4	9.2
	Max	90.6	73.5	64.4	53.9
	Mean	47.4	36.6	27.9	25.2
	1st quartile	12.7	20.1	14.6	12.4
	2nd quartile (median)	18.3	36.0	24.6	15.4
	3rd quartile	30.2	47.5	39.8	42.3
Thoron (Bq m^{-3})	Min	2.9	10.9	2.7	2.8
	Max	126.5	71.9	74.1	100.6
	Mean	31.1	31.6	25.1	27.3
	1st quartile	8.6	14.4	7.4	7.9
	2nd quartile (median)	19.4	27.0	14.4	14.9
	3rd quartile	33.0	43.5	34.6	35.0
EERC (Bq m^{-3})	Min	1.1	3.4	1.1	1.1
	Max	35	25	41.4	16.6
	Mean	10.3	13.0	8.2	5.9
	1st quartile	2.4	10.8	3.1	2.3
	2nd quartile (median)	5.9	12.0	4.0	4.3
	3rd quartile	12.0	13.0	8.4	9.0
EETC (Bq m^{-3})	Min	0.2	0.5	0.2	0.2
	Max	3.9	2.4	4.1	2.0
	Mean	1.1	1.5	0.7	0.7
	1st quartile	0.3	1.1	0.4	0.3
	2nd quartile (median)	0.7	1.4	0.6	0.5
	3rd quartile	1.2	1.6	0.7	1.0

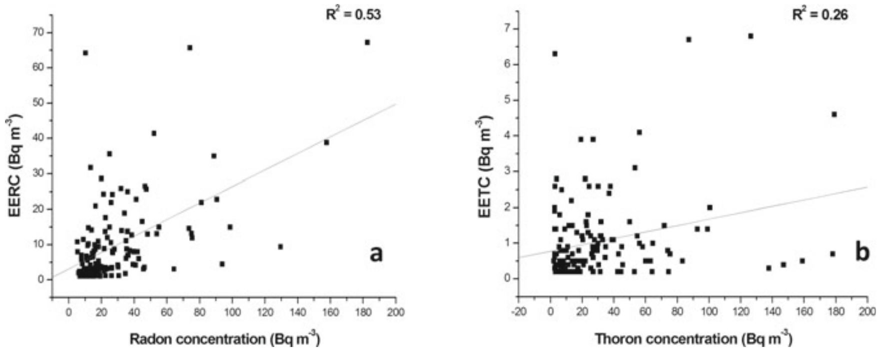


Fig. 25.3 Scatter plot along with Pearson's correlation (R^2) coefficient between **a** radon with EERC and **b** thoron with EETC

25.4.2 Correlation Among Gases and Their Progeny

The scatter plot along with Pearson's correlation coefficient (R^2) between radon and EERC and between thoron and EETC are shown in Fig. 25.3. Radon and EERC have moderate positive correlation ($R^2 = 0.53$) whereas thoron and EETC found to have a weak positive correlation with ($R^2 = 0.26$). Radon and thoron are the gases whereas their decay products are heavy metals and isotopes of lead, bismuth, and lead. Also, the half-life of parent nuclei that is radon and thoron is less compared to their daughter products. Therefore, solid decay products stay for a longer period in the environment compared to the parent nuclides. This weak correlation between thoron and EETC is attributed to the strong influence of moisture contents and ventilation conditions in dwellings of the study region. Also, the environmental parameters affect differently both the parent nuclides (gases) and their daughter nuclides (solid).

25.4.3 Frequency Distribution of Radon, Thoron, and Their Progeny

Frequency distribution of radon, thoron, EERC, and ETC in dwellings of study region is shown in Fig. 25.4. It indicates that 87% of dwellings have radon concentration below the value 50 Bq m^{-3}, 80% of dwellings have thoron concentration below the value 50 Bq m^{-3}, 97% of dwellings have radon progeny concentration below the value 50 Bq m^{-3}, and 97% of dwellings have thoron progeny concentration below the value 4.5 Bq m^{-3}. Three locations in case of radon and 7 locations in case of thoron exceed the value of 100 Bq m^{-3}. Since, the contribution of thoron itself is negligible towards the total annual effective dose due to inhalation of thoron along its progeny therefore it indicates that the thoron is not hazardous in dwellings of the study region. This heterogeneous distribution is most likely due to topography,

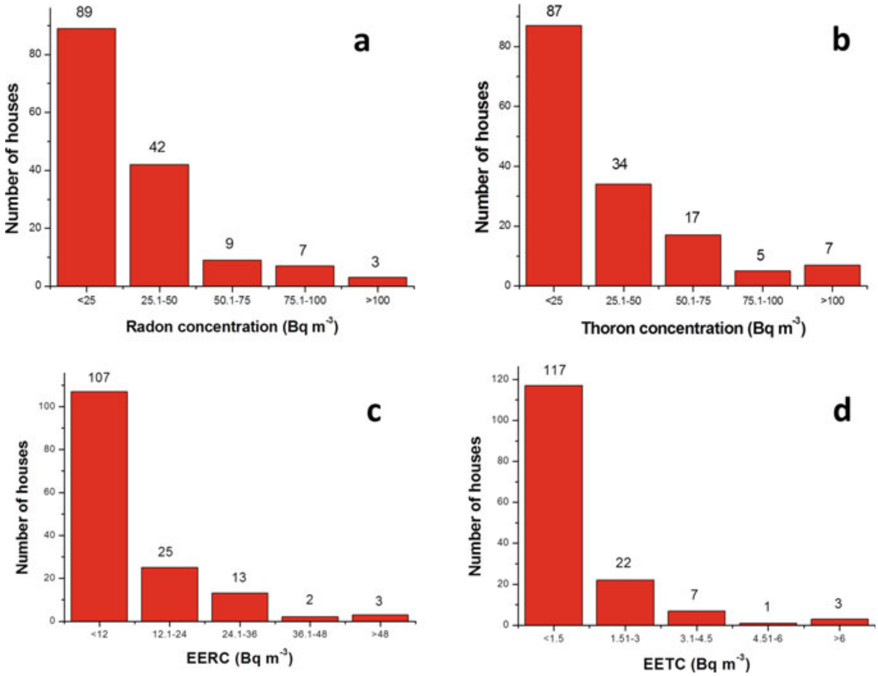


Fig. 25.4 Frequency distribution of measured **a** radon, **b** thoron, **c** EERC, and **d** EETC concentration in houses of district Palwal, Southern Haryana, India

diverse geological location of investigated houses, different building materials used in construction of investigated dwellings, influence of environmental factors such as moisture contents, temperature gradient in dwellings, etc.

25.4.4 Annual Effective Dose Due to Inhalation

Annual effective dose due to inhalation of radon and its progeny is found in the range of 0.07 to 1.11 mSv y^{-1} with an average of 0.29 ± 0.01 mSv y^{-1}. Annual effective dose due to inhalation of thoron and its progeny is found in the range of 0.03 to 0.38 mSv y^{-1} with an average of 0.15 ± 0.01 mSv y^{-1}. The estimated total annual effective inhalation dose due to radon, thoron, and their decay products varies from 0.1 to 1.1 mSv y^{-1} with an average of 0.4 ± 0.01 mSv y^{-1}. Singh et al. (2015) reported that the total annual effective dose rate due to inhalation in Tosham region of Haryana, India varies from 1.33 to 2.44 mSv y^{-1}. Singh et al. (2019a) reported that total AED_{Rn+Th} in the region of district Faridabad of Haryana, India varied from 0.15 to 0.45 mSv y^{-1}. Kumar et al. (2020) reported that total AED_{Rn+Th} in the region of district Dadri of Uttar Pradesh, India varies from 0.29 to 2.06 mSv y^{-1}. Thus, results of the present study are similar to the results of nearby regions. Estimated total annual effective dose was observed within the prescribed safe limits of 3–10 mSv y^{-1} and 10 mSv y^{-1}. It concludes that no radiological hazards are associated with distribution of these radionuclides.

25.4.5 Comparison of Results with Other Investigations of Nearby Regions

The results of the present investigation are compared with outcomes of the previous investigations carried out in nearby regions of India. Sannappa and Ningappa (2014) reported that the indoor radon and thoron in the nearby granite region of Karnataka, India varies from 16–170 Bq m^{-3} and 18–300 Bq m^{-3} respectively. Singh et al. (2015) conducted an indoor investigation of radon, thoron, EERC, and ETC in Tosham region of Haryana, India in rainy season and reported that their values varies from 37 to 80 Bq m^{-3} for radon, from 53 to 80 Bq m^{-3} for thoron, from 12 to 23 Bq m^{-3} for EERC, and from 2 to 7 Bq m^{-3} for EETC. Bangotra et al. (2019) conducted an indoor study in the houses of Muktsar and Mansa districts of Punjab, India and reported that the radon, thoron, EETC, and EERC varies from 19–88 Bq m^{-3}, 22–77 Bq m^{-3}, 11–50 Bq m^{-3}, and 0.7–7 Bq m^{-3} respectively. Singh et al. (2019a) reported that radon, thoron, EERC, and ETC varies from 5.3–128.8 Bq m^{-3}, 9–183.6 Bq m^{-3}, 1.1–18.9 Bq m^{-3}, and 0.1–1.9 Bq m^{-3} respectively in the dwellings of district Faridabad of southern Haryana, India. Kumar et al. (2020) reported that radon, thoron, EERC, and EETC in the nearby region of national capital power station, district Dadri of state Uttar Pradesh of India, varies from 9.7–64.9 Bq m^{-3}, 34–90 Bq m^{-3}, 3.3–27.2 Bq m^{-3}, and 0.3–1 Bq m^{-3} respectively. Thus, it concluded that the results of the present investigation are comparable with results of nearby regions of India.

25.4.6 Seasonal Comparison of Results of Present Investigation

The outcomes of summer season (exposure period of 4 months) for the same study region are published (Singh et al. 2019b) and in the present chapter, results of measurements have been performed for the 2nd season that is the rainy season (exposure period of 4 months) are reported. Thus, the inter-comparison will provide the seasonal variation of radioactive gases and their solid decay products. The average values of measured indoor radon and thoron were found 28.2 ± 2.1 and 29.5 ± 2.8 Bq m^{-3} in the present investigation i.e. rainy season and 28.6 ± 0.03 and 30 ± 0.04 Bq m^{-3} in summer season respectively. No significant change in the average values of indoor radon and thoron is observed. Moreover, the lifestyle of peoples of the present study region is comparatively different in both seasons. In the summer season people mostly use fans, coolers, air-conditioners, etc. in their homes than during the rainy season. Thus, ventilation conditions are different in both seasons which lead to comparatively high levels of radon and thoron in the summer season. But it contradicts our study. However, significant change observed in distribution of radon and thoron dwelling wise. In the rainy season radon and thoron levels are found maximum in mud houses and minimum in modern houses but it is completely opposite for summer season. Due to temperature gradients inside and outside the dwellings of mud in the rainy season, these gases accumulate up to longer time inside the dwellings.

25.5 Conclusions

The measured average indoor thoron gas concentration (29.5 ± 2.8 Bq m^{-3}) is about 3 times higher than the world average value of 10 Bq m^{-3}. The average values for radon concentration, EERC, and EETC found to be 28.2 ± 2.1, 9.1 ± 0.02, and 1.2 ± 0.01 respectively were found below the world average values of 40, for radon, 15 Bq m^{-3} for EERC, and 0.5 Bq m^{-3} for EETC. The overall level of thoron gas was found to be higher than radon but results were reversed in case of EERC and EETC.

Overall no significant change in average values of radon and thoron concentration is observed compared with summer season outcomes. However, dwelling wise comparison showed that results are completely opposite for mud and modern houses. The variation of radon and thoron gases was observed such as H2 (mud house) > H1 (cemented house) > H3 (traditional house) > H4 (modern house) and H2 (mud house) > H1 (cemented house) > H4 (modern house) > H3 (traditional house) respectively. Similar trends were observed in case of progeny. The higher concentration of gases and their solid decay products were found in dwellings of zone 2 than zone 1.

Radon and EERC have moderate positive correlation with Pearson's correlation coefficient ($R^2 = 0.53$) whereas thoron and EETC found to be weak positive correlation with Pearson's correlation coefficient ($R^2 = 0.26$).

Frequency distribution of radon, thoron, EERC, and EETC shows that the radionuclides in the indoor region are widely distributed. This heterogeneous distribution is presumably due to topography, different geological location of investigated houses, different building materials used in construction of investigated dwellings, influence of environmental factors such as moisture contents, temperature gradient in dwellings, etc.

The estimated total AED_{Rn+Th} varies from 0.1 to 1.1 mSv y^{-1} with an average of 0.4 ± 0.01 mSv y^{-1}. The measured concentration of radon, thoron, EERC, and EETC are found within prescribed limits of WHO, ICRP, and UNSCEAR. It indicates that no radiological hazards are associated with radon, thoron, and their progeny in the present study region.

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