Nuclear Engineering and Technology 52 (2020) 1289–1296

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Original Article

Estimation of natural radionuclide and exhalation rates of environmental radioactive pollutants from the soil of northern India

Vandana Devi, Rishi Pal Chauhan*

Department of Physics, National Institute of Technology, Kurukshetra, Haryana, 136119, India

ARTICLE INFO

Article history:
Received 3 August 2019
Received in revised form
17 November 2019
Accepted 18 November 2019
Available online 19 November 2019

Keywords:
Radon
Exhalation and emanation
Radionuclide concentration
Dose
Health risk

ABSTRACT

The estimation of radioactivity level is vital for population health risk assessment and geological point of view and can be evaluated as rate of exhalation and source concentration (^{226}Ra, ^{232}Th and ^{40}K). The present study deals with the soil samples for investigation of radionuclides content and exhalation rates of radon –thoron gas from different sites in northern Haryana, India. Absorbed dose and associated index estimated in the present study are the measures of environmental radioactivity to inhalation dose. Effective doses received by different tissues and organs by considering different occupancy and conditions are also measured. Exhalation rates of radon and thoron are measured with active scintillation monitors based on alpha spectroscopy namely scintillation radon (SRM) and thoron (STM) monitors respectively. Sample height was optimized before measurement of thoron exhalation rate using STM. Average values of radon and thoron exhalation are found 16.6 ± 0.7 mBqkg^{-1}h^{-1} and 132.1 ± 2.6 mBqm^{-2}s^{-1} respectively. Also, a simple approach was also adopted, to evaluate the thoron exhalation which accomplished a lot of challenges, the results are compared with the data obtained experimentally. The study is useful in the nationwide mapping of radon and thoron exhalation rates for understanding the environmental radioactivity status.

1. Introduction

Radioactivity is widespread in environment due to primordial radionuclides in surrounding and has adverse effects on human health. One of the main concerns for the environmental radioactivity is radon (^{222}Rn), which is the decay product of radium present in earth crust and is recognized as the important source of natural radioactivity exposure. Primary source of radon is soil which contains about a thousand times soil gas concentration than atmosphere concentration. Radon indoor level is influenced by underneath soil gas concentration and is enhanced with the use of materials having high source content for construction purpose. The contribution of environmental radioactivity from various samples can be defined as activity concentration (Bq/kg) of radionuclides (^{226}Ra, ^{232}Th and ^{40}K) present in them and the exhalation rate. Concentration of radon and its sources i.e. radionuclides (radium and thorium) vary with geological and environmental conditions. Therefore, measurement of radionuclides content is an important issue not only for health effect estimation but also from biochemical and geochemical point of view [1–4]. Several studies have been performed to study the level of radon (^{222}Rn) and thoron (^{220}Rn) in dwellings in connection with radon soil gas concentration [5,6]. Knowledge of the amount of natural radionuclide and ^{222}Rn - ^{220}Rn exhalation rates in soil is vital to assess the attainable radiological risk and associated hazards index to human health and additionally to develop standards with their use [7–9].

Radon exhalation rates are reported by several researchers using different techniques but only few studies are done with thoron. Thoron is the short-lived isotope of radon present in the thorium radioactive series. Thoron exhalation rate is likewise a significant issue not just on accounts of difficulties in its estimation due to its short half-life yet in addition because of its high dose conversion factors [1,2,6,7]. In recent past Canister technique was used for radon exhalation rate measurement but the difficulty with the technique arises because of thoron interference and leakage. Also, thoron exhalation cannot be measured with this technique [10–12]. This study deals with the radon as well as thoron exhalation rate measurement using scintillation cells equipped with different monitors and sampling arrangement i.e. flow mode or diffusion mode for thoron and radon respectively. Present study

---

* Corresponding author.
E-mail addresses: vandanac821@gmail.com (V. Devi), chauhanrpc@gmail.com (R.P. Chauhan).

https://doi.org/10.1016/j.net.2019.11.016

also deals with the difficulties associated with thoron measurement.

The present study evaluates both exhalation rates (^{222}Rn and ^{220}Rn) and their radionuclide contents from the soil samples in the proximity of Karnal district, lies on the western bank of the river Yamuna in northern Haryana, India. Study is performed for this inhabited area by keeping in mind that this area has not been studied for environmental radon and thoron so far. Researchers have contemplated the radioactivity level and ^{222}Rn - ^{220}Rn exhalation rate in soil [6,7,13–15] in the northern India. But a combined study of radionuclides and exhalation rates with active techniques has not been performed for the study area so far. The estimations of radiological parameters which are screening tool for the risk appraisal with the utilization of these materials for construction purpose and obtained results are compared with universal suggested criteria as well as with other studies [6–8]. This study is also helpful in mapping of exhalation rates in country by providing radon exhalation data with geographical location of each sampling site.

2. Experimental procedure

2.1. Study area and sample collection details

Area under examination belongs to the vast Indo-Gangetic plain to the southwest of Shivalik hills. This area represents almost an alluvial plain without any conspicuous topographical features. Map of the study area along with sample collecting sites is represented in Fig. 1. The district is bounded by latitudes 29.25° and 29.59° and longitudes 76.27° and 77.13°. The district is well populated and rich in educational and research institutes. Clayey loam and sandy loam are the major soil types and Yamuna River is one of the sources of drainage. Study is performed with soil samples from thirty locations by collecting 3–4 samples from diverse sites of study area with their exact geographical location. The samples were first made free of moisture by drying them at 100 °C in oven till the constant weight was achieved and homogenized by sieving using mesh of sieve size 150 μm. The samples were then processed to carry out further studies of radionuclides and exhalation rates.

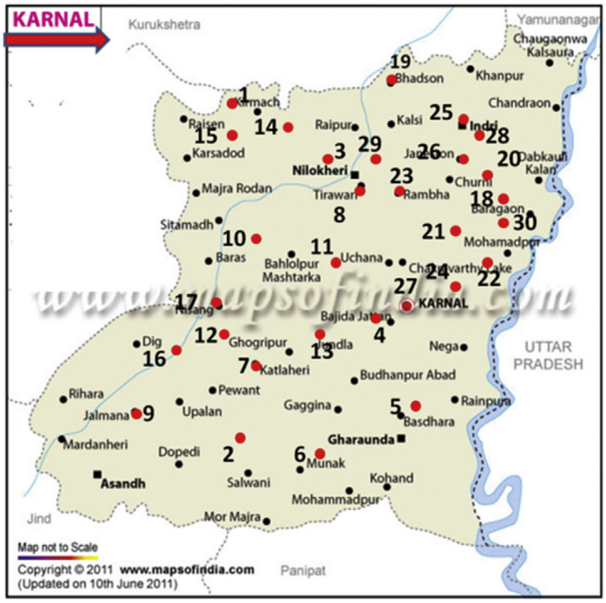

Fig. 1. Guide of the region under study marked with sample collecting sites (by red dots). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

2.2. Measurement of radionuclides (^{226}Ra, ^{232}Th and ^{40}K) content in samples under study

For gamma dose estimation, measurement of present radionuclides in the samples are important and carried out using NaI(Tl) scintillation detector. The samples were packed in measuring beaker of same dimension that was utilized for standardization of detector of 8 cm in height and 7 cm in diameter. Packing was ensured to be air tight by sealing the box. To ensure secular equilibrium between radionuclide and decay products they were then placed for at least one month. NaI(Tl) used for the measurement is 5.08 × 5.08 cm^2 in size and coupled to1K MCA card. Before starting the measurement detector was calibrated using Co and Cs source. Counts were obtained after a counting period of 4–5 h. Activity concentrations of radionuclides were calculated in Bq/kg by using the counts per second, efficiency and transition probability for the corresponding peak under study [10,16].

2.3. Estimation of radon exhalation and emanation rates

The rate of radon emission per unit area i.e. ^{222}Rn exhalation rate is measured in the collected soil samples utilizing Scintillation radon monitor (SRM) developed and calibrated by Bhabha Atomic Research Centre, Mumbai, India. SRM is an active device dependent on the alpha particles detection resulting from the decay of radon and its produced progenies into the cell. Measuring arrangement consists of cylindrical accumulator of height 5 cm and radius 15 cm, with provision to attach cell from upper side. ZnS(Ag)is utilized as scintillation material in the cell have volume of 150 cc. The sample to be tested, of known weight (1 kg) was placed into the radon-tight accumulator and the accumulator attached to SRM in sensitive mode. Radon starts to build up inside and enters into the cell after crossing the pin hole arrangement which used to avoid thoron interference present into the accumulator. Time cycle of one hour was used for measurement and an inbuilt algorithm displayed the radon concentration at regular interval of one hour using a conversion factor of 1.2 cph/Bqm^{-3}. Radon mass exhalation rate (E) in (Bq kg^{-1}h^{-1}) is calculated by analysis of growing radon concentration (C) with time (t) using the following relation (1) [5,7,15].

$$C = \frac{EM}{V\lambda_e}\left(1 - e^{-\lambda_e t}\right) + C_i e^{-\lambda_e t} \tag{1}$$

Emanation factor (ε), measures the radon that finally enters the permeable arrangement of the sample after its creation in grains of sample was calculated by the following equation (2) [4,17].

$$\varepsilon = E/A_{Ra}\ \lambda \tag{2}$$

Where V account for the effective volume including the volume of the scintillation cell and accumulator, λ_e is the effective decay of radon account for natural decay, leakage and back diffusion rate, C_i is initial concentration.

2.4. Estimation of thoron exhalation and emanation rates

The precise estimation of ^{220}Rn exhalation rate is crucial from the perspective of radiation assurance particularly for the remarkable areas having high thoron source. Thoron exhalation rate in the soil samples under investigation was estimated utilizing scintillation thoron monitor (STM). In light of small half life of thoron (55 s), it can't be estimated utilizing a similar plan as utilized for radon exhalation rate estimation. So as to recognize thoron all the more adequately, thoron sampling was done in flow mode in a closed loop arrangement including accumulator, monitor and pump represented schematically in Fig. 2. In the flow mode operation, pump was kept ON continuously and cycle of 15 min was used for complete mixing. In the small air volume and with the use of pump to accomplish the proper air mixing, thoron concentration attains saturation very quickly.

After achieving stable thoron concentration in the accumulator, the ^{220}Rn exhalation rate (J) rising up out the material would thus be able to be determined from the experimentally registered data of thoron concentration (C) by taking mean of concentrations in the close chamber and surface (A) to volume (V) ratio of chamber using the following relation (3) [4,10,17,18].

$$J = \frac{CV\lambda}{A} \tag{3}$$

Mass exhalation rates J_m for ^{220}Rn can be estimated knowing the density of soil samples (ρ) and its diffusion length (l) with the following relation [7–10].

$$J_m = \frac{J}{\rho l} \tag{4}$$

Since ^{220}Rn diffusion length is small, therefore, only the top surface of samples enclosed in the chamber would contribute to thoron in the chamber. Therefore, the ^{220}Rn mass can be perceived as thoron emanation rather its exhalation and utilized in present investigation [7–10].

3. Results and discussion

3.1. Radionuclides and radium equivalent concentration in soil samples

For the estimation of potential hazard, radionuclides content are measured using gamma spectroscopy with scintillation detector in Bq/kg for the study area. Obtained results of activity concentrations of radium, thorium and potassium together with the statistical uncertainty are depicted in Table 1. The obtained values of radium content ranged from 41.8 ± 2.9 Bq/kg to 71.9 ± 5.6 Bq/kg with average of 52.0 ± 1.3 Bq/kg while thorium in soil samples ranged from 136.9 ± 30.2 to 257.1 ± 10.3 Bq/kg with average of 187.0 ± 6.8 Bq/kg. Presence of potassium content (^{40}K) in materials is also responsible for external radiation exposure by emitting gamma radiations. Therefore, potassium contents are also measured in all samples under study and its concentration ranged from 737.4 ± 38.5 Bq/kg to 2172.9 ± 52.2 Bq/kg with average of 1332.6 ± 92.3 Bq/kg. Some samples show concentration of potassium on higher side which is due to soil contamination with the use of fertilizers. Study area is known as rice bowl of India and have good rice production region which is allowed to dispose in the agriculture land. Hence the green paddy contains varying amounts of potassium content in the soil samples. The used fertilizers and TENORM materials are responsible for slightly higher values of potassium content [19].

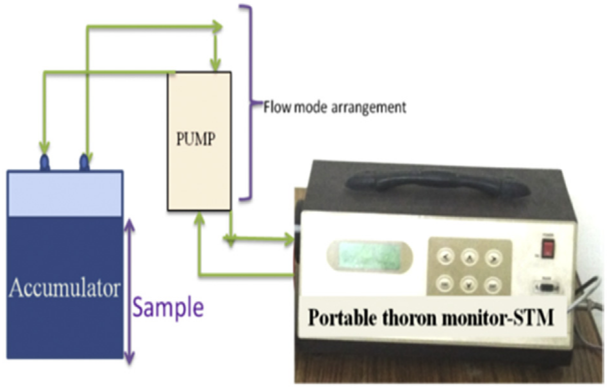

Fig. 2. A schematic representation of experimental setup for thoron exhalation measurement.

For the comparison of different samples having non-uniform radionuclide concentration, a common index in terms of radium equivalent is used. For the assessment of the radiological hazard and also to overcome non-uniformity in concentrations of individual radionuclides, radium equivalent is calculated using the radionuclides content [1]. Radium equivalent vary from 309.8 to 562.7 Bq/kg with average of 422.0 ± 10.9 Bq/kg. The estimated values of Ra_{eq} are found on somewhat higher side than referenced worldwide value, which can be explained on the basis that it is a common index and is the weighted sum of all three radionuclides concentration [1–3,20]. As explained earlier that some samples have high values of potassium content which are responsible for slightly higher radium equivalent for the samples. Study area lies on the south of Shivalik hills and higher values of radioactivity contents present in soil and sand of hilly area have been found [7,14]. This region is famous for agriculture especially for rice production and Yamuna river water is the main source of irrigation. Yamuna is originating from hilly region which brought down the soil from hill sand which accounts for radioactivity in this area. Flowing water extracts the material containing high radium and thorium content which can account for the obtained values of radium equivalent activity higher for some samples. Hence being the southwestern bank of the river the region may have some higher level of radioactivity in the sand mixed soil [5–7,13]. The information of radioactivity levels is valuable so as to set the standards and guidelines in the light of global recommendations. Due to the increasing social concern, a large number of research groups are engaged in the estimation of natural radioactivity on national as well as worldwide levels. The concentration of the radionuclides found here are under the recommended level of UNSCEAR and OCED [1,20].

3.2. Radiological hazard and risk index assessment

Materials having radionuclides content are health hazardous and their hazard index depends on the concentrations of radionuclides present in them with their use in construction and daily life. Different hazardous terms and index were calculated from the measured radionuclides concentration [1,21,22]. Assessed radiological risk emerging from the utilization of soil in development of abodes in terms of absorbed dose and different index are presented in Table 2. Estimated values of alpha index lies from 0.21 to 0.36 with average of 0.26 ± 0.01. Values of alpha dose for all the samples under study are less than one which is recommended for safety [1,21]. Also, calculated average value of gamma dose is 1.55 ± 0.44, is within the safe limit because value of that index less than 2 i.e. <2 corresponds to 0.3 mSv annual dose rate. Additionally, inferred estimations of annual effective dose rates do not surpass the average worldwide exposure of 2.4 mSvy^{-1} arises from natural sources [1–3,21]. Hence potential radiation hazard posed by these materials for construction purpose is not hazardous as to restrict their use. Therefore, they can be exempted from the restrictions concerning level of environmental radioactivity in the investigation area. In this manner, these soils can be exempted from the restrictions concerning radioactivity. The human beings are exposed to different level of environmental radioactivity which varies according to geographical variation, environmental condition and

Table 1
Radionuclides content and radon-thoron exhalation rates in soil samples.

Sample Code (Location)	Radium ^{226}Ra A_{Ra} (Bq/kg)	Thorium ^{232}Th A_{Th}(Bq/kg)	Potassium ^{40}K A_K(Bq/kg)	Radium equivalent $A_{Ra} + 1.43 \times A_{Th} + 0.077 \times A_K$ Ra_{eq} (Bq/kg)	Radon exhalation (mBq$\mathbf{Kg}^{-1}\mathbf{h}^{-1}$)	Thoron exhalation (mBq$\mathbf{m}^{-2}\mathbf{s}^{-1}$)
K-1 (76°54′38″,29°53′48″)	46.4 ± 3.2	158.6 ± 12.1	737.4 ± 38.5	329.9	12.9 ± 0.9	137.9 ± 8.7
K-2 (76°54′50″, 29°45′31″)	48.0 ± 1.7	214.1 ± 11.0	870.0 ± 4.7	421.1	15.3 ± 1.2	122.2 ± 10.2
K-3 (76°55′37″, 29°50′17″)	41.8 ± 2.9	157.3 ± 10.1	1860.4 ± 1.5	409.9	21.1 ± 1.1	152.3 ± 11.5
K-4 (76°54′55″, 29°40′48″)	65.9 ± 6.8	166.2 ± 1.4	808.8 ± 24.3	365.8	19.5 ± 1.5	142.7 ± 14.6
K-5 (76°46′22″, 29°47′16″)	48.2 ± 5.8	168.5 ± 5.4	1764.1 ± 64.4	425.1	15.2 ± 1.3	138.0 ± 9.8
K-6 (76°45′26″, 29°49′30″)	54.3 ± 4.2	163.5 ± 16.4	780.1 ± 5.9	348.2	19.4 ± 0.9	145.9 ± 11.5
K-7(76°49′49″, 29°38′18″)	54.3 ± 2.5	249.9 ± 1.4	1961.0 ± 31.7	562.7	14.5 ± 0.7	118.6 ± 8.6
K-8(76°55′49″, 29°48′43″)	58.7 ± 2.5	250.2 ± 31.6	790.4 ± 34.4	477.4	18.2 ± 1.1	142.0 ± 9.5
K-9(76°42′21″, 29°35′17″)	43.4 ± 3.1	145.7 ± 8.9	753.6 ± 13.1	309.8	9.9 ± 0.7	109.9 ± 13.5
K-10(76°47′33″, 29°46′14″)	51.8 ± 2.9	155.6 ± 6.5	1793.6 ± 69.9	412.5	17.9 ± 0.9	134.0 ± 16.5
K-11(76°53′23″, 29°43′52″)	55.1 ± 1.9	223.1 ± 27.4	1759.7 ± 45.3	509.6	18.1 ± 0.6	129.4 ± 18.4
K-12(76°45′50″, 29°36′35″)	49.9 ± 2.5	153.5 ± 0.7	1798.7 ± 58.7	407.9	12.7 ± 1.0	122.6 ± 15.3
K-13(76°52′30″, 29°39′09″)	51.3 ± 2.7	222.6 ± 18.3	837.7 ± 34.4	434.1	18.0 ± 0.9	131.0 ± 18.6
K-14(76°52′36″, 29°52′45″)	45.8 ± 2.3	147.9 ± 15.8	1838.5 ± 109.5	398.9	17.3 ± 1.1	147.5 ± 14.2
K-15(76°47′58″, 29°51′15″)	49.5 ± 6.4	139.3 ± 17.9	1719.5 ± 32.6	381.0	18.8 ± 1.7	138.3 ± 17.6
K-16(76°43′34″, 29°39′54″)	49.1 ± 1.8	233.7 ± 10.6	783.9 ± 42.2	443.7	20.3 ± 1.3	138.7 ± 9.7
K-17(76°45′10″, 29°41′13″)	47.6 ± 4.6	172.0 ± 2.7	1815.6 ± 4.3	433.4	16.5 ± 1.3	144.5 ± 10.3
K-18(76°07′03″, 29°48′35″)	51.8 ± 6.1	165.4 ± 12.4	1068.2 ± 109.9	370.6	16.7 ± 1.5	136.1 ± 15.6
K-19(76°58′45″, 29°54′35″)	48.5 ± 2.0	235.0 ± 17.6	1158.4 ± 40.8	473.7	22.8 ± 1.9	143.2 ± 9.6
K-20(77°06′28″, 29°50′20″)	43.3 ± 1.3	166.4 ± 8.2	866.0 ± 16.2	347.9	13. ± 0.9	126.5 ± 8.8
K-21(77°02′35″, 29°49′02″)	54.7 ± 4.1	231.9 ± 34.7	864.1 ± 46.9	452.8	18.4 ± 1.2	119.1 ± 12.3
K-22(77°06′46″, 29°47′28″)	43.3 ± 3.2	257.1 ± 10.3	1728.7 ± 86.3	544.1	13.0 ± 0.8	110.2 ± 18.5
K-23(77°00′05″, 29°47′34″)	55.0 ± 3.8	171.2 ± 26.4	1937.7 ± 17.7	449.0	19.7 ± 0.5	148.3 ± 17.9
K-24(77°03′54″, 29°44′14″)	44.5 ± 4.4	228.9 ± 15.9	779.3 ± 8.9	431.9	10.3 ± 0.6	112.2 ± 10.6
K-25(77°02′28″, 29°52′05″)	53.4 ± 2.4	136.9 ± 30.2	1925.1 ± 71.5	397.4	25.0 ± 1.5	157.9 ± 12.2
K-26(77°08′48″, 29°49′52″)	64.4 ± 9.3	185.8 ± 5.1	2172.9 ± 52.2	497.4	13.0 ± 1.1	115.9 ± 11.1
K-27(77°59′29″, 29°42′52″)	56.1 ± 6.4	164.2 ± 3.5	1795.7 ± 17.7	429.2	17.3 ± 0.8	110.1 ± 10.5
K-28(77°04′36″, 29°51′36″)	56.7 ± 8.6	168.4 ± 17.2	950.0 ± 44.5	370.6	11.9 ± 0.9	123.3 ± 9.9
K-29(76°56′41″, 29°48′57″)	71.9 ± 5.6	209.3 ± 19.5	1085.7 ± 16.2	454.8	17.9 ± 1.4	148.7 ± 15.6
K-30(77°05′18″, 29°45′24″)	55.8 ± 4.4	166.6 ± 40.7	974.4 ± 159.2	369.0	13.5 ± 1.5	115.0 ± 13.4
AM	52.0 ± 1.3	187.0 ± 6.8	1332.6 ± 92.3	422 ± 10.9	16.6 ± 0.7	132.1 ± 2.6

* AM = Mean and SE = standard error = SD/√ N, Where SD is standard deviation and N is the no of observations.

Table 2
Estimated various radiological dose and index in soil samples.

	Representative level index	Absorbed gamma dose (D_R) (nGy/h)	Annual effective dose (H_R) (mSv/y)	Alpha Index I_α	Gamma index I_γ
Formula used [1,20–22]	$\frac{A_{Ra}}{150}+\frac{A_{Th}}{100}+\frac{A_K}{1500}$	$A_{Ra} + 1.1A_{Th} + 0.080A_K$	$D_R \times 8766 \times 0.2 \times 0.7 \times 10^{-6}$	$\frac{A_{Ra}}{200Bq/kg}$	$\frac{A_{Ra}}{300}+\frac{A_{Th}}{200}+\frac{A_K}{3000}$
Min.	2.25	139.5	0.17	0.21	1.12
Max.	4.17	257.8	0.32	0.36	2.08
Average	3.10	192.5	0.24	0.26	1.55
SE	0.08	5.1	0.01	0.01	0.44

individual's living standard. The activity concentrations of radionuclides in the soil give essential and principal information of radioactivity with an impact on general wellbeing, plants and creatures [7,8].

3.2.1. *Effective dose rate to different tissues and organs*

Dose to different body organs and tissues H_{organs} is a function of fraction of time spend i.e. occupational factor (*OF*) and can be calculated by

$$H_{organs} = OF \text{ x } A_{edr} \text{ x } F \tag{5}$$

Where, A_{edr} and F refer to annual effective dose rate and dose conversion factor respectively. Values of conversion factors are different for different organs and are given by O'Brien and Sanna [23]. In literature, occupancy factor is 0.8 which is commonly used all over the world, but according to living standards of peoples in India, Sharma et al. [6] gave satisfactory reasons for the variability of occupational factors. Different occupancy factors were analyzed according to the populace indoor occupancy. The fraction of time spend depends on climate, season and life style etc. can be taken as 0.1 (<0.1 assumed for peoples live in towns/villages amid the summer season.), 0.5 (for residential communities and urban areas, the part of the time spent inside in the midst of the summer season is 0.5.), 0.3 (the weighted normal of all the populace range) and 0.8(commonly used occupancy factor). Hence, here dose to different body organs (Lungs, Ovary, Red bone marrow RBM, Testes and whole-body WB) is calculated using the average value of annual effective dose, occupancy factors and corresponding conversion factor for different organs. Comparison of annual effective dose to different body organs for different occupancy factor is given in Fig. 3 and shows the straight forward dependence of dose to the occupancy factor. The estimated dose due to radionuclide content to different body tissues and organs was found to be well within the recommendation of ICRP [3].

3.3. *Radon exhalation and emanation rates in samples under study*

Radon exhalation rate was measured using closed accumulator technique in which the sample enclosed hermetically in

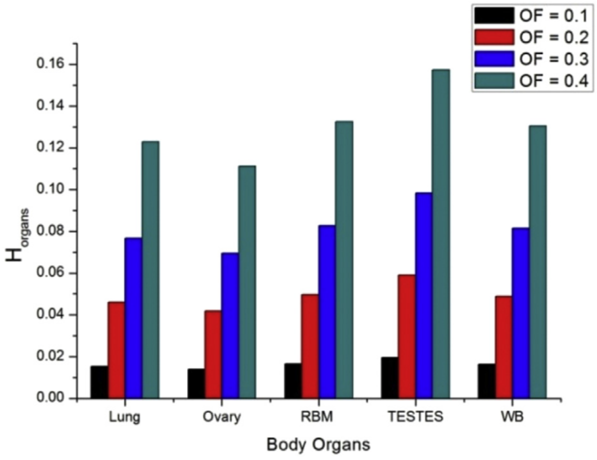

Fig. 3. Annual effective doses to different body organs for different occupancy.

accumulator [4]. Radon concentration builds up to attain a saturation concentration in the closed chamber. The build-up of ^{222}Rn concentration (C) in accumulator was measured for one hour cycle represented in Fig. 4 and continued until it gets saturated. This equilibrium concentration estimated with the following relation.

$$C = C_{\mathrm{eq}}\left(1 - e^{-\lambda_e t}\right) \tag{6}$$

Where, C_{eq} is the asymptotic value of ^{222}Rn concentration and λ_e refers to effective decay constant. Equilibrium concentration for the samples under study ranged from 96.1 ± 4.8 to 219.9 ± 3.2 Bq/m^3 with average concentration of 146.7 ± 5.8 Bq/m^3. This varying equilibrium radon concentration is because of different radioactivity content in samples (Table 1) and different soil parameters which results into different values of exhalation rates.

Estimated values of radon exhalation rates are depicted in Table 1. Radon mass exhalation varies from 9.9 ± 0.7 mBqkg^{-1}h^{-1} to 25.0 ± 1.5 mBqkg^{-1}h^{-1}, with average exhalation value of 16.6 ± 0.7 mBqkg^{-1}h^{-1}. The obtained ^{222}Rn exhalation rates are below the world wide average value and also less than that exhaled by the samples of coal and fly ash [19]. Radon emanation which is measures the amount of ^{222}Rn comes into pore space after its production is calculated from radium content present in samples and final radon exhalation rate (Table 1) from the samples using equation (2). Emanation factor in the collected soil samples are found to be 2.69 to 6.73 with average emanation factor of 4.30 ± 0.2. Values of observed emanation factors are of same order as observed by Sahoo et al. [17] for different construction materials. A positive correlation of 0.51 is presented in Fig. 5 between ^{222}Rn emanation and its accumulated concentration in the chamber. In the accumulator containing a known weight of sample, accumulated concentration depends on the amount of radon coming from grain to pore space. This concentration increase with an increase in the emanation factor is represented by positive correlation.

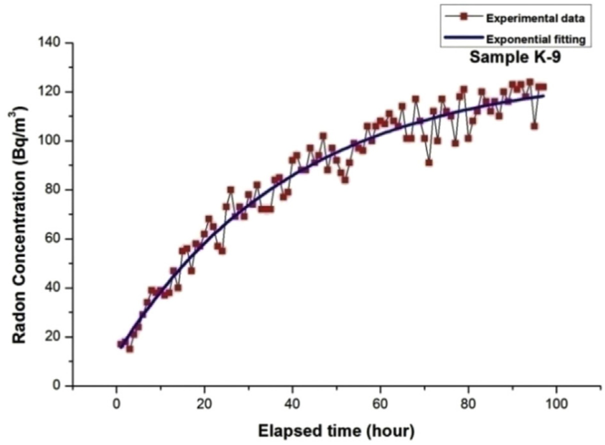

Fig. 4. Typical graph showing radon builds up concentration with time cycle of one hour.

3.4. Thoron exhalation and emanation rates in samples under study

In recent past thoron was underestimated because of difficulties in measurement and calibration which makes its case very different from that of radon due to its shorter half life (55s) as compared to long lived ^{222}Rn. Different half life of these two results into their distinct behavior such as different diffusion length, time required in attainment of stable concentration (Figs. 4 and 7), height and amount of sample taken, dispersion behavior (uniformity) and hence requirement of chamber size for exhalation measurement [11,18,25]. Due to these difficulties associated with thoron the same measurement apparatus and procedure used for the radon case cannot be applicable to thoron hence, the different monitors. Scintillation monitors are employed for radon and thoron exhalation measurements in different modes i.e. diffusion mode (SRM) and flow mode (STM) respectively. Because of difficulty in accomplishing uniformity for thoron in the chamber used for radon of volume 3.5 × 10^{-3} m^3, a different chamber of small volume 1.3 × 10^{-3} m^3 is used for thoron exhalation measurement. Thoron uniformly cannot be achieved without forces air mixing in the large chamber because of short diffusion length. A pump was used to achieve a certain level of homogeneity between the scintillation cell and accumulator of STM. Establishment of uniformity in case of thoron requires the optimal minimum chamber size and forced air mixing within closed circuit [18,25]. Fig. 6 represents the radon concentration growth with time measured using SRM which shows that radon attains a asymptotic value after several cycles of one hour while thoron concentration measured using STM with small air volume attains a stable concentration very quickly for the time cycle of 15 min (Fig. 7). During measurement of thoron concentration in the accumulator present air volume is only 33% of total chamber volume (STM) and is very less as compare to accumulator used in SRM. Air volume during thoron measurement is only 0.4 × 10^{-3} m^3 which is only 12% of the volume of accumulator used in SRM.

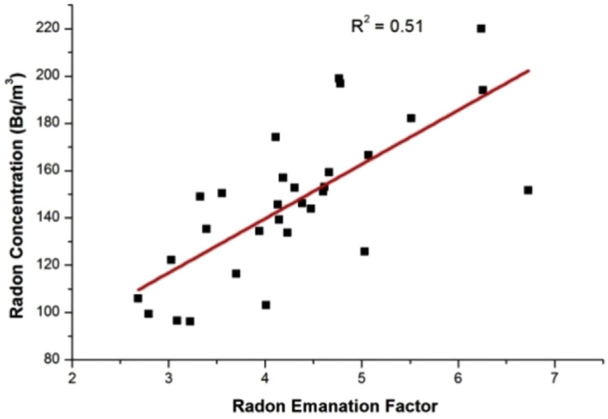

Fig. 5. Radon concentrations in correlation with radon emanation factor.

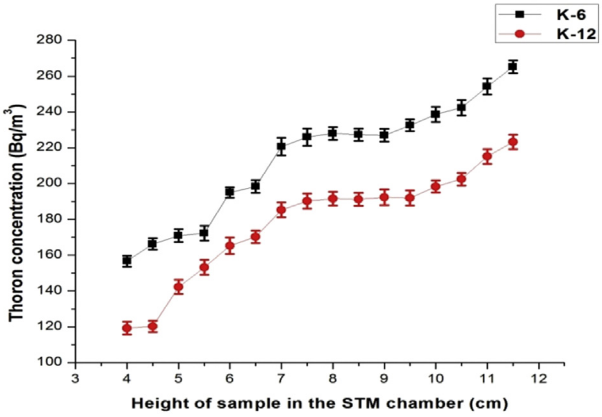

Fig. 6. Optimization of sample height for thoron exhalation measurement using STM.

The diffusion length of ^{220}Rn in the soil is less as compared to that of radon which makes its origin mainly from upper surface. The consequences of the thoron exhalation measurements rely upon the thickness of sample placed which must be taken into account. The ^{220}Rn concentration (C) present in the chamber includes its generation and decay rate. In order to optimize the sample height for thoron, concentration was measured experimentally corresponding to varying height presented in Fig. 6. Initially thoron concentration increases with the height of sample in the chamber because more the amount of sample taken in the chamber more the thoron will be emerging out in chamber. Then, thoron concentration attains a constant value within a range of sample height because only few upper layers contribute to thoron concentration because of small diffusion length of thoron in soil (1 cm) and air (2.3 cm) in the certain air volume present. After this thoron concentration again rise due to the emission of fixed amount of thoron concentration in the small air volume than earlier [11,18,25]. For the exhalation rate measurement optimized height range should be correspond to constant thoron concentration which is 7.5 cm–9.5 cm in this case. Therefore, to measure the thoron exhalation rates of the soil samples under study, the height of all samples was kept at 8 cm in order to keep up a generally small air volume in the accumulator to ensure uniform mixing at the operating flow rate.

Thus, with the forced air mixing in the accumulation chamber and this optimized sample height, one can have the thoron uniform distribution inside the chamber and thoron exhalation rates can be calculated using thoron concentration in equation (3). Attainment of steady state thoron concentration is illustrated graphically in Fig. 7 [14,24]. Calculated values of exhalation rates for 220Rn for the soil samples under study are depicted in Table 1 with arithmetic mean and standard deviation. Thoron surface exhalation varies from 109.9 ± 13.5 to 157.9 ± 12.2 mBqm-2s-1 with an average of 132.1 ± 2.6 mBqm-2s-1. Also the emanation rate of 220Rn can be calculated using its diffusion length and sample density for all the samples under study and is varying from 5.50 to 7.89 with an average of 6.60 ± 0.13.^{222}Rn and ^{220}Rn exhalation rates are below the recommendation level [1,2], therefore the region is safer as the exhalation rates of 222Rn and 220Rn are concerned. The variation in 222Rn and 220Rn exhalation rate can also be attributed because of different radionuclide content and soil substructure of samples as shown in Table 1.

The highest and lowest value of exhalation for both ^{222}Rn and ^{220}Rn are obtained from the same soil samples. Minimum value of radon mass exhalation rate is 9.9 ± 0.7 mBqkg^{-1}h^{-1} and thoron surface exhalation rate is 109.9 ± 13.5 mBqm^{-2}s^{-1} from the sample K-9 while the maximum value of radon exhalation (25.0 ± 1.5 mBqkg^{-1}h^{-1}) and thoron exhalation (157.9 ± 12.2 mBqm^{-2}s^{-1}) are obtained from the sample K-25. Also, Fig. 8 represents an observed positive correlation between ^{222}Rn and ^{220}Rn exhalation rates.

3.5. Estimation of thoron exhalation using radon exhalation rates

Detection of thoron and measurement of its exhalation rate is important by taking its radiological risk into consideration especially for some regions having its high level; on the other hand its measurement is not a simple task but consists of a lot of challenges described above. Any alternate method rather than the direct measurement if available can be easily adopted by one for the ^{220}Rn exhalation rates calculation in the samples. One such theoretical approach is provided by Magnoni et al. [12], to estimate the exhalation rates of thoron using the radon exhalation rates and the source term concentration both for radon and thoron. Present study has measured all the parameters required for such approach to predict thoron exhalation using that of radon and can estimate the correlation between predicted and experimentally measured values using STM. Magnoni proposed the relation given below to estimate thoron exhalation (J_{Th}), in equation (7) using the radon exhalation rate (J_{Rn}), radium concentration (A_{Radium}) and thorium concentration($A_{Thorium}$) [12].

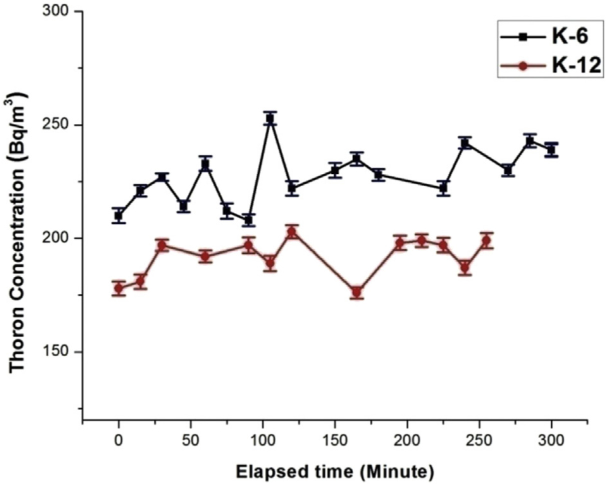

Fig. 7. Typical graph showing thoron stable concentration with 15 min time cycle.

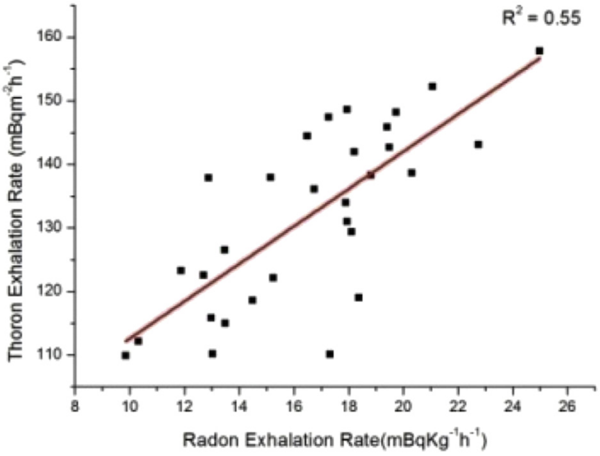

Fig. 8. Correlation of ^{220}Rn exhalation rate with ^{222}Rn mass exhalation rate in soil samples.

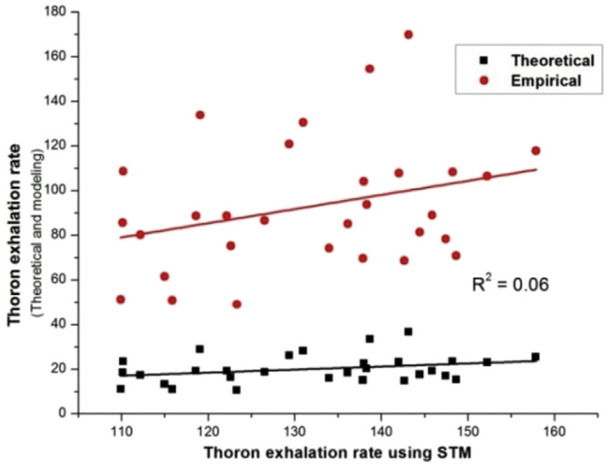

Fig. 9. Represents the correlation between experimental and theoretical thoron exhalation.

$$J_{Th} = J_{Rn} \cdot \frac{A_{Thorium}}{A_{Radium}} \cdot \sqrt{\frac{\lambda_{Thoron}}{\lambda_{Radon}}} \tag{7}$$

Where λ_{Thoron} and λ_{Radon} represents the decay constant for thoron and radon respectively. Fig. 9 presented the estimated thoron exhalation from radon exhalation based on the above relation with black dots. A test of this method had also been performed by experimentally measuring ^{222}Rn and ^{220}Rn exhalation rate along with theoretical calculations for the sienites sample utilizing alpha spectrometry technique. Result of test performed by Magnoni et al. also shows significantly greater than those obtained theoretically, which is also observed in this study. However, based on the test results, Magnoni et al. proposed an empirical relation to model the thoron exhalation using the same parameters as in equation (7). Based on the assumption that the exhalation rate is not only depends on the diffusion but is the result of direct recoil of gas atoms from the upper surface with alpha decay, he deduced an general empirical relationship between the ^{222}Rn and ^{220}Rn exhalation rates as following [12].

$$\frac{J_{Th}}{J_{Rn}} = \frac{A_{Thorium}}{A_{Radium}} .357 \tag{8}$$

This relation can also be used as screening tools for other stony materials such as granites and similar rocks for practical purposes. The values for thoron exhalation rates obtained using the parameters estimated in the present study in the empirical relation (equation (8)) are used to find he correlation with the experimentally obtained values by red dots in Fig. 9. Correlation found between the experimentally observed values and the values obtained using theoretical and empirical relation is $R^2 = 0.06$, but the values of exhalation are more comparable for empirical relation. Also, study conducted by Tokonami (2010) [25] founds no correlation between radon and thoron, proposed formula by Magnoni et al. [12] needs some more validation to use this as a screening tool for ^{220}Rn exhalation rate estimation.

In order to study the environmental radioactivity level, a comparative study was performed with the nearby regions of the study area. On comparison with the other studies of northern India conducted in recent times, it is observed that the values of exhalation rates for ^{222}Rn and ^{220}Rn are below than in the hilly areas and comparable to nearby plain area of indo Gangetic plane. A comparison of the study area with other nearby hilly and plain regions in terms of radon exhalation rates represented in Table 3.

Table 3
Comparison of present study results with recent studies of ^{222}Rn exhalation rates conducted in north Indian regions.

S.N.	Location	Radon exhalation rate (mBqKg^{-1}h^{-1})	Technique Used (Reference)
1.	Morni Hills	122 ± 1	Canister technique [14]
2.	Yamuna nagar	73 ± 3	-do-
3.	Naraingarh	50 ± 2	-do-
4.	Shivalik foot hills	50 ± 1–143 ± 6	Canister technique [13]
5.	Udhampur (J&K)	11.57–65.62	SRM [6]
6.	Kurukshetra	6–31	SRM [4]
7.	Yamunanagar	29.2–73.1	SRM [7]
8.	Ambala	28.2–60.7	-do-
9.	Panchkula	34.3–76.8	-do-
10.	Karnal	9.9–25.0	SRM (Present study)

4. Conclusions

The measurements of radionuclides (^{226}Ra, ^{232}Th and ^{40}K) along with exhalation rates of radon-thoron in the samples from northern part of India are presented. The following conclusions are drawn from the study:

- Estimated values of radioactivity content using gamma spectroscopy and different radiation hazard index are within the recommendation levels by UNSCEAR and ICRP.
- Uniformity and stability during thoron measurement are achieved with air volume of 0.4×10^{-3} m^3 and forced thoron mixing. Optimized height for measurement lies in the range corresponding to constant thoron concentration and found at top of the chamber.
- Average value of radon mass exhalation found 16.6 ± 0.7 mBqkg^{-1}h^{-1}, while that of thoron surface exhalation was found 132.1 ± 2.6 mBqm^{-2}s^{-1}which are less than the world average values. Positive correlations are observed between ^{222}Rn and ^{220}Rn exhalation rates from the soil samples.
- A different approach for ^{220}Rn exhalation rate measurement, by measuring only the corresponding rate for radon (SRM) was also tested and found a poor correlation hence, needs some more verification.
- A comparison with other studies indicated that the place and use of soil present in study area for construction is considered to be safe for human habitation.

This study will be helpful in bridging the source radionuclides concentration for the area under study for future research expeditions and is also helpful in mapping of country by providing radiological data with exact geographical location of each sampling site.

Declaration of competing interest

There is no conflict of interest of any kind with person or organization.

Acknowledgments

The authors are thankful to the Director, National Institute of Technology Kurukshetra and the Head, Department of Physics, NIT Kurukshetra for providing the facilities for the completion of the work and financial help in the form of fellowship.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.net.2019.11.016.

References

[1] UNSCEAR, Sources and Effects of Ionizing Radiation (Exposures from Natural Radiation Sources), 2000. New York.

[2] ICRP Publication 115, Lung Cancer Risk from Radon and Progeny and Statement on Radon, 2010, https://doi.org/10.1016/j.icrp.2011.08.011.

[3] S. Abdullahi, A.F. Ismail, S. Samat, Determination of indoor doses and excess lifetime cancer risks caused by building materials containing natural radionuclides in Malaysia, Nucl. Eng. Technol. 51 (2019) 325–336, https://doi.org/10.1016/j.net.2018.09.017.

[4] V. Devi, A. Kumar, R.P. Chauhan, A study on radionuclides content and radon exhalation from soil of Northern India, Environ. Earth Sci. 78 (2019) 506, https://doi.org/10.1007/s12665-019-8512-9.

[5] C. Papachristodoulou, K. Stamoulis, K. Ioannides, Temporal variation of soil gas radon associated with seismic activity: a case study in NW Greece, Pure Appl. Geophys. (2019), https://doi.org/10.1007/s00024-019-02339-5.

[6] S. Sharma, A. Kumar, R. Mehra, R. Mishra, Radiation hazards associated with radionuclides and theoretical evaluation of indoor radon concentration from soil exhalation of Udhampur District, Jammu and Kashmir State, India, J. Soils Sediments 19 (2019) 1441–1455, https://doi.org/10.1007/s11368-018-2125-x.

[7] R.P. Chauhan, A. Kumar, N. Chauhan, M. Joshi, P. Aggarwal, B.K. Sahoo, Ventilation effect on indoor radon–thoron levels in dwellings and correlation with soil exhalation rates, Indoor Built Environ. 25 (2016) 203–212, https://doi.org/10.1177/1420326X14542887.

[8] M.A.E. Abdel-Rahman, S.A. El-Mongy, Analysis of radioactivity levels and hazard assessment of black sand samples from Rashid area, Egypt, Nucl. Eng. Technol. 49 (2017) 1752–1757, https://doi.org/10.1016/j.net.2017.07.020.

[9] A.K. Mahur, R. Kumar, D. Sengupta, R. Prasad, Estimation of radon exhalation rate, natural radioactivity and radiation doses in fly ash samples from Durgapur thermal power plant, West Bengal, India, J. Environ. Radioact. 99 (2008) 1289–1293, https://doi.org/10.1016/j.jenvrad.2008.03.010.

[10] P. Ujić, I. Čeliković, A. Kandić, Z. Žunić, Standardization and difficulties of the thoron exhalation rate measurements using an accumulation chamber, Radiat. Meas. 43 (2008) 1396–1401, https://doi.org/10.1016/j.radmeas.2008.03.003.

[11] A. Kumar, R.P. Chauhan, Back diffusion correction for radon exhalation rates of common building materials using active measurements, Mater. Struct. 48 (2015) 919–928, https://doi.org/10.1617/s11527-013-0203-5.

[12] M. Magnoni, E. Chiaberto, A. Prandstatter, E. Serena, R. Tripodi, Thoron exhalation rate in stony materials: a simplified approach, Constr. Build. Mater. 173 (2018) 520–524, https://doi.org/10.1016/j.conbuildmat.2018.04.053.

[13] R.P. Chauhan, P. Chauhan, A. Pundir, S. Kamboj, V. Bansal, R.S. Saini, Estimation of dose contribution from 226Ra, 232Th and 40K radon exhalation rates in soil samples from Shivalik foot hills in India, Radiat. Prot. Dosim. 158 (2014) 79–86, https://doi.org/10.1093/rpd/nct190.

[14] A. Pundir, R. Singh, S. Kamboj, Measurement of radon concentration and exhalation rates in soil samples of some districts of Haryana and Himachal in India, Researcher 6 (2014) 71–76.

[15] L.M. Singh, M. Kumar, B.K. Sahoo, B.K. Sapra, R. Kumar, Study of radon, thoron exhalation and natural radioactivity in coal and fly ash samples of kota super thermal power plant, Rajasthan, India, Radiat. Prot. Dosim. 171 (2016) 196–199, https://doi.org/10.1093/rpd/ncw057.

[16] V. Devi, A. Kumar, R.P. Chauhan, Radiation doses due to background radioactivity in soil from inhabited area of Northern Haryana, in: Advances in Basic Science (Icabs 2019), 2019, p. 120010, https://doi.org/10.1063/1.5122506.

[17] B.K. Sahoo, D. Nathwani, K.P. Eappen, T.V. Ramachandran, J.J. Gaware, Y.S. Mayya, Estimation of radon emanation factor in Indian building materials, Radiat. Meas. 42 (2007) 1422–1425, https://doi.org/10.1016/j.radmeas.2007.04.002.

[18] S.D. Kanse, B.K. Sahoo, B.K. Sapra, J.J. Gaware, Y.S. Mayya, Powder sandwich technique: a novel method for determining the thoron emanation potential of powders bearing high 224Ra content, Radiat. Meas. 48 (2013) 82–87, https://doi.org/10.1016/j.radmeas.2012.10.014.

[19] H. Azeez, H. Mansour, S. Ahmad, Effect of using chemical fertilizers on natural radioactivity levels in agricultural soil in the Iraqi Kurdistan region, Pol. J. Environ. Stud. 29 (2019) 1–10, https://doi.org/10.15244/pjoes/106032.

[20] NEA-OECD, Exposure to Radiation from Natural Radioactivity in Building Materials, 1979, pp. 1–34. https://www.oecd-nea.org/rp/reports/1979/exposure-to-radiation-1979.pdf.

[21] EC, Radiation Protection 112, Radiological Protection Principles Concerning the Natural Radioactivity of Building Materials, 1999. Finland, https://eceuropa.eu/energy/sites/ener/files/documents/112.pdf.

[22] S. Righi, L. Bruzzi, Natural radioactivity and radon exhalation in building materials used in Italian dwellings, J. Environ. Radioact. 88 (2006) 158–170, https://doi.org/10.1016/j.jenvrad.2006.01.009.

[23] K. O'Brien, R. Sanna, The distribution of absorbed dose-rates in humans from exposure to environmental gamma rays, Health Phys. 30 (1976) 71–78.

[24] A.K. Mahur, R. Kumar, M. Mishra, D. Sengupta, R. Prasad, An investigation of radon exhalation rate and estimation of radiation doses in coal and fly ash samples, Appl. Radiat. Isot. 66 (2008) 401–406, https://doi.org/10.1016/j.apradiso.2007.10.006.

[25] S. Tokonami, Why is 220Rn (thoron) measurement important? Radiat. Prot. Dosim. 141 (2010) 335–339, https://doi.org/10.1093/rpd/ncq246.