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# Comparative study of indoor radon, thoron with radon exhalation rate in soil samples in some historical places at Jaipur, Rajasthan, India

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# ABSTRACT

As the radon progeny contribute a major part of natural radiation dose to general population, attention has been given to the large scale and long term measurement of radon and its progeny. Recent epidemiological evidence suggests that inhalation of low level radon and its progeny in dwellings may contribute towards the cause of lung cancer. Thoron and its progeny contribute little for the radiation dose in normal back ground region due to its small half life. In this comparative study Solid State Nuclear Track Detectors (SSNTD's) based twin chamber dosimeters were used for estimating radon (<sup>222</sup>Rn), Thoron (<sup>220</sup>Rn) gases and Inhalation dose in some historical places at Jaipur, Rajasthan, India using twin chamber dosimeter cups. The dosimeters employ two LR-115 type II plastic track detector peliculable films, cellulose nitrate detector films inside each of the two chambers fitted with filter and polymeric membrane for the discrimination of radon and thoron. Soil samples were also collected simultaneously from different geological formations of the same area for laboratory measurement of radon exhalations rate. Radon concentrations are found to vary from (18.4 ± 3.1) Bq m<sup>-3</sup> to (62.1 ± 5.7) Bq m<sup>-3</sup>, whereas thoron concentrations vary from (5.9 ± 0.6) Bq m<sup>-3</sup> to (22.0 ± 2.6) Bq m<sup>-3</sup>. Radon activity and radon exhalation rates in the soil samples were also measured by using "Sealed can technique" using LR 115-type II nuclear track detectors. Radon activities are found to vary from (294.2) to (868.4) Bqm<sup>-3</sup> with an average value of (566.0) Bqm<sup>-3</sup>. Radon exhalation rates in the soil samples vary from (146.8) to (312.2) mBq m<sup>-2</sup> h<sup>-1</sup> with an average value of (203.4) mBq m<sup>-2</sup> h<sup>-1</sup>.

Key words: Radon, Thoron, Dwellings, Inhalation dose, Soil; LR-115 SSNTD; Radon exhalation rate.

### **INTRODUCTION**

The two significant isotopes of radon are <sup>222</sup>Rn, the immediate decay product of <sup>226</sup>Ra, derived from the uranium decay series and <sup>220</sup>Rn, the immediate decay product <sup>224</sup>Ra. Exposure to Radon (<sup>222</sup>Rn) and its progeny in indoor atmosphere can result significant inhalation risk to population particularly to those living in homes with much higher levels of radon. Natural radiation which originates from the Earth crust, cosmic radiations etc. are the major contributors to the total background exposures to human population. All radiations gives a world average value of

2.4 mSv for the annual effective dose equivalent from natural back ground radiation of which 1.4 mSv comes from the radon, thoron and their daughter products[1-3]. <sup>222</sup>Rn is an inert radioactive gas with a half-life of 3.8 days and belongs to the radioactive uranium series. In recent years, <sup>222</sup>Rn has been used as tracer for the origin and trajectory of air masses [4,5]. Thoron, (<sup>220</sup>Rn) is a natural decay product of thorium series. It has a half-life of 55.6 seconds and also emits alpha rays. Radon is a radiological poison and a carcinogen. Some of the daughter products from radioactive decay of radon (such as polonium) are also toxic. Since radon is a gas, its decay products form a very fine dust that is both toxic and radioactive. This can potentially stick in the lungs after inhalation and do far more damage than the radon itself [6]. Although these elements occur in virtually all types of rocks and soils, their concentrations vary with specific sites and geological materials. As an inert gas, radon can move freely through the soil from its source; the distances are determined by factors such as rate of diffusion, effective permeability of the soil and by its own half-life. The inhalation of short-lived daughter products of naturally occurring radon is a major contributor to the total radiation dose to exposed subjects. Radon progenies might be inhaled and deposited more or less deeply onto the bronchio-pulmonary tree, depending upon the granulometry of the particles on which they become attached. Under specific conditions, such as those prevailing in the uranium mining environment, lung dose due to radon progenies may be sufficiently high to cause an increase in the occurrence of lung cancer. Measurements of indoor radon are of importance because the radiation dose to human population due to inhalation of radon and its daughters contributes more than 50% of the total dose from natural sources [3]. In the present work an effort has been made to make indoor radon/thoron estimation in some historical places at Jaipur, Rajasthan, India by using SSNTD,s in Plastic Twin Chamber Dosimeter cups. The important factors that have got influence up on the indoor radon / thoron concentration are (1) Properties of the building construction materials and the ground. Here the radon exhalation rate from the building construction materials or the ground is dependent on the uranium/thorium content, density and the porosity of the material (2) Indoor radon / thoron concentrations are also influenced by the ventilation rate and metrological parameter.

#### MATERIALS AND METHODS

#### 2.1 Estimation of radon, thoron and inhalation dose

The radon - thoron dosimeter employed for the measurements is made up of a twin cup cylindrical system, developed at the Bhabha Atomic Research Centre (BARC) and is reported else where [7,8]. Figure 1 shows the schematic diagram of the twin cup dosimeter. Each chamber has a length of 4.1cm and a radius of 3.1cm. The SSNTD-1 placed in compartment M, measures radon alone which diffuses into it from the ambient air through a semi-permeable membrane (Latex) of 25 $\mu$ m thickness having diffusion coefficient in the range of 10<sup>-8</sup>-10<sup>-7</sup> cm<sup>2</sup>s<sup>-1</sup> [9,10]. It allows the build up of about 90% of the radon gas in the compartment and suppresses thoron gas concentration by more than 99% (The mean time for radon to reach the steady state concentration inside the cup is about 4.5 h). On the other hand, the glass fiber filter paper of 0.56mm thickness in the compartment F allows both radon and thoron gases to diffuse in and hence the tracks on SSNTD-2 placed in this compartment F, are related to the concentrations of both the gases. By subtracting the result of SSNTD-2 to SSNTD-1, thoron concentration has been determined. The choice of the detector LR-115 is made in view of the fact that detector does not develop tracks originating from the progeny alphas deposited on them [8] and therefore are ideally suited for air concentration measurements. These dosimeters with membrane and the LR-115type II plastic track detector film have been suspended from the mid-point of the ceiling of the houses at a height of about 2.5m from the ground level. At the end of the 100 days the dosimeters are retrieved to lab. The exposed detectors have been etched in 10%NaOH at  $60^{\circ}$ C for a period of 1 hour in a constant temperature bath. After etching, the detectors have been pealed off from the plastic base and counted using a spark counter. From the counts the track density of the films has been calculated. The calibration factors have been obtained by using the setup described by Eappen and Mayya (2004) [8].

Calibration factors have been calculated as ( $S_m = 0.019 \text{ T.cm}^{-2}d^{-1}\text{per Bq m}^{-3}$ ) for SSNTD-1 (compartment M) and ( $S_{tf} = 0.016 \text{ T.cm}^{-2}d^{-1}\text{per Bq m}^{-3}$ ) for SSNTD-2 (compartment F) and used in the present study.

From track density concentration of radon ( $C_R$ ) and thoron ( $C_T$ ) were calculated using the sensitivity factor determined from the controlled experiments [7,11,12].

$$C_R(Bqm^{-3}) = \frac{T_m}{d \times S_m} \tag{1}$$

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$$C_T(Bqm^{-3}) = \frac{(T_f - d \times C_R \times S_{rf})}{d \times S_{rf}} \qquad (2)$$

Where,  $C_R$  -Radon concentration;  $C_T$  -Thoron concentration;  $T_m$ = Track density in membrane compartment;  $T_f$  - track density in filter compartment and d -Exposure time

The inhalation dose (D) in mSv y<sup>-1</sup> was estimated using the relation:

$$D = \{0.17 + 9F_R)C_R + (0.11 + 32F_T)C_T \times 7000 \times 10^{-6} \qquad \dots \qquad (3)$$

Where  $F_R$  and  $F_T$  are equilibrium factor for radon and thoron respectively. The values are taken as 0.4 and 0.1 for radon and thoron given by UNSCEAR, (2000) [13]. The values of radon and thoron concentrations and inhalation dose obtained by using relations 1, 2 and 3.

#### 2.2 Radon exhalation rate

For the measurement of radon exhalation rate in soil samples collected simultaneously from different geological formations of the same area "Sealed can technique" was used. Equal amount of samples (100 gm) were placed in the "Cans" (diameter 7.0 cm and height 7.5 cm as shown in Fig.-2) similar to those used in the calibration experiment Singh et al., (1997) [14]. LR-115 Type II solid state nuclear track detector ( $2 \text{ cm} \times 2 \text{ cm}$ ) was fixed on the top inside the cylindrical "Can". The cans are sealed for 100 days and thus the sensitive lower surface of the detector is freely exposed to the emergent radon so that it could record the tracks of alpha particles resulting from the decay of radon in the remaining volume of "Can". Radon and its daughters reach an equilibrium concentration after 4 hours and hence the equilibrium activity of emergent radon can be obtained from the geometry of can and the time of exposure. After the exposure for 95 days the detectors were taken out and etched in 2.5 N NaOH at  $60^{\circ}$  C for a period of 90 min in a constant temperature water bath. The resultant alpha-particle tracks were counted using an optical microscope at a magnification of  $400 \times$ . From the track density the radon activity was calculated using a calibration factor of 0.056 track cm<sup>-2</sup> d<sup>-1</sup> (Bqm<sup>-3</sup>)<sup>-1</sup>, obtained from an earlier calibration experiment Singh et al., (1997) [14]. Radon exhalation rate is obtained from the following expression [15-17].

$$E = \frac{CV\lambda}{A\left[T + \frac{1}{\lambda}\left\{e^{-\lambda T} - 1\right\}\right]}$$
(4)

where, E is radon surface exhalation rate ( Bq  $m^{-2}h^{-1}$  ); C is a integrated radon exposure as measured by LR -115 solid state nuclear track detector ( Bq  $m^{-3}h$ ); V is the effective volume of can ( $m^{3}$ );  $\lambda$  is the decay constant for radon ( $hr^{-1}$ ); T is the exposure time (hr); A is the area of the can ( $m^{2}$ ).

#### 2.3 Indoor internal exposure due to radon inhalation:

The risk of lung cancer from domestic exposure of radon and its daughters can be estimated directly from the indoor inhalation exposure (radon) effective dose. The contribution of indoor radon concentration from soil samples can be calculated from the following expression [16, 17].

$$C_{Rn} = \frac{E_x \times S}{V \times \lambda_v} \tag{5}$$

where  $C_{Rn}$  is the radon concentration (  $Bqm^{-3}$ );  $E_x$  is radon exhalation rate (  $Bqm^{-2}h^{-1}$ ); S is radon exhalation area (  $m^2$ ); V is room volume ( $m^3$ ), and  $\lambda_v$  is air exchange rate ( $h^{-1}$ ). The maximum radon concentration from the building material was assessed by assuming the room as a cavity with S/V = 2.0 m<sup>-1</sup> and air exchange rate of 0.5 h<sup>-1</sup>. The annual exposure to potential alpha energy  $E_p$  (effective dose equivalent) is then related to the average radon concentration  $C_{Rn}$  by following expression:

$$E_{P}[WLM.y^{-1}] = \frac{8760 \times n \times F \times C_{Rn}}{170 \times 3700}$$
(5)

where,  $C_{Rn}$  is in Bqm<sup>-3</sup>; n is the fraction of time spent indoors; 8760, the number of hours per year; 170, the number of hours per working month and F is the equilibrium factor for radon and is taken as 0.4 as suggested by UNSCEAR, (2000) [13]. Radon progeny equilibrium is most important quantity, where dose calculations are to be made on the basis of the measurement of radon concentration, it may have value 0<F<1. Thus, the values of n=0.8 and F=0.4 were used. From radon exposure the indoor inhalation exposure (radon) effective dose were estimated by using a conversion factor of 3.88 mSv (WLM)<sup>-1</sup> by ICRP, (1993) [18].

# **RESULTS AND DISCUSSION**

The measured concentration of (<sup>222</sup>Rn), Thoron (<sup>220</sup>Rn) gases and inhalation dose in some historical places at Jaipur, Rajasthan, India using twin chamber dosimeter cups are shown in Table-1. Radon concentrations are found to vary from  $18.4 \pm 3.1$  Bq m<sup>-3</sup> to  $62.1 \pm 5.7$  Bq m<sup>-3</sup>, whereas thoron concentrations vary from  $5.9 \pm 0.6$  Bq m<sup>-3</sup> to  $22.0 \pm 2.6$  Bq m<sup>-3</sup>.

Inhalation dose due to radon and thoron concentrations is estimated to vary from 0.99 mSv  $y^{-1}$  to 1.67 mSv  $y^{-1}$ . The International Commission on Radiation Protection ICRP-65, (1993) [18] has recommended that remedial action against radon and its progeny is justified above a continued effective dose of 10 m Sv, while an action level within the range of 3-10 mSv  $y^{-1}$  has been proposed. The action level for radon concentration should be in the range between 200 and 600 Bq m<sup>-3</sup>. The measured values are below the recommended action levels.

It is worth mentioning that it is difficult to predict the radon exhalation rate from the concentration of uranium or its decay series products in the sample, since the radon exhalation rate depends also on the texture and grain size composition [19]. As soil is frequently used as building material in different forms, it is important to assess the radiation risk to public. Although radon levels tend to be very high in the con- fined spaces of underground drifts, elevated radon levels are also found in open pit mines and around U-mill tailings. From open pit mines the radioactive emissions are radioactive fugitive dust and radon gas.

Soil samples collected from the same area, values of radon activity, radon exhalation rate and indoor inhalation exposure (radon)-effective dose are given the Table-2.

The measured values of radon activity and radon exhalation rates in these soil samples were measured by using "Sealed can technique" using LR 115-type II nuclear track detectors. Radon activities are found to vary from 294.2 to 868.4 Bqm<sup>-3</sup> with an average value of 566.0 Bqm<sup>-3</sup>. Radon exhalation rates in these samples vary from 146.8 to 312.2 mBqm<sup>-2</sup>h<sup>-1</sup> with an average value of 203.4 mBqm<sup>-2</sup>h<sup>-1</sup> and the indoor inhalation exposure (radon)-effective dose in these soil samples vary from  $17.3 \times 10^{-3}$  to  $36.8 \times 10^{-3}$  mSv y<sup>-1</sup> with an average value of  $23.9 \times 10^{-3}$  mSv y<sup>-1</sup>.

Name of Place	C <sub>R</sub> (Bq m <sup>-3</sup> )	C <sub>T</sub> (Bq m <sup>-3</sup> )	D (Inhalation dose) (mSv y <sup>-1</sup> )
Hawa Mahal	$18.4 \pm 3.1$	$22.0 \pm 2.6$	0.99
Albert Park	$39.5\pm4.6$	$13.2 \pm 1.3$	1.36
Amer Fort -I	53.7 ±5.3	$7.9 \pm 0.7$	1.59
Amer Fort-II	$50.3 \pm 5.2$	$6.8 \pm 0.6$	1.49
Pratap Mandir	$56.8 \pm 5.5$	$7.7 \pm 0.7$	1.67
Nahargarh-I	$43.4 \pm 4.6$	$15.8 \pm 1.5$	1.51
Nahargarh-II	$46.3 \pm 5.0$	$5.9 \pm 0.6$	1.36
Jantar Mantar	$62.1 \pm 5.7$	$11.1 \pm 0.9$	1.66
Mahamandir Temple	$32.6 \pm 4.1$	$15.5 \pm 1.6$	1.22
Garland fort	43.1 ±4.6	$6.1 \pm 0.6$	1.28
Minimum value	$18.4 \pm 3.1$	$5.9 \pm 0.6$	0.99
Maximum value	$62.1 \pm 5.7$	$22.0 \pm 2.6$	1.67

Table-1 Indoor radon, thoron concentration and inhalation dose in some historical places at Jaipur Rajasthan, India

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Table-2 Radon activity, radon exhalation rate and Indoor inhalation exposure (radon)-effective dose in some soil samples collected from historical places at Jaipur Rajasthan, India

Name of Place	Radon Activity (Bqm <sup>-3</sup> )	Radon Exhalation (mBq m <sup>-2</sup> h <sup>-1</sup> )	Indoor inhalation exposure (radon)-effective dose ( mSv y <sup>-1</sup> )
Hawa Mahal	409.2	147.0	$17.3 \times 10^{-3}$
Albert Park	794.8	285.6	$33.6 \times 10^{-3}$
Amer Fort -I	420.0	150.8	$17.8 \times 10^{-3}$
Amer Fort-II	813.6	292.4	34.5×10 <sup>-3</sup>
Pratap Mandir	817.2	293.6	$34.6 \times 10^{-3}$
Nahargarh-I	294.2	105.6	12.5×10 <sup>-3</sup>
Nahargarh-II	411.4	147.8	17.5×10 <sup>-3</sup>
Jantar Mantar	868.4	312.2	$36.8 \times 10^{-3}$
Mahamandir Temple	422.8	152.0	17.9×10 <sup>-3</sup>
Garland fort	408.4	146.8	17.3×10 <sup>-3</sup>
Minimum value	294.2	146.8	17.3×10 <sup>-3</sup>
Maximum value	868.4	312.2	$36.8 \times 10^{-3}$
Average Value	566.0	203.4	$23.9 \times 10^{-3}$
S. D.	213.8	76.9	9.04× 10 <sup>-3</sup>



1.

Radon cup mode SSNTD Film Radon+ Thoron Cup mode SSNTD Film 2.



Fig.-1 Photograph and Schematic diagram of radon- thoron twin chamber dosimeter cup



Fig.-2 Experimental setup for the measurement of radon exhalation rate using "Sealed Can Technique"

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