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Indoor ^{222}Rn and ^{220}Rn variations: Evidence for Boyle's law

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Background: Radon and its daughter product are a major source of natural radiation exposure, the measurement of radon concentration in dwellings is assuming ever increasing importance. It is known from recent surveys in many countries that radon and its progeny contributes significantly to total inhalation dose and is well established that radon when inhaled in large quantity causes lung disorder. In view of this the authors have measured ^{222}Rn and ^{220}Rn levels in dwellings of different volumes at Bangalore Metropolitan, India. ***Materials and Methods:*** Integrated and long duration measurements of radon were carried out using twin cup dosimeters with Solid State Nuclear Track Detector technique. ***Results:*** Results showed that concentrations of ^{222}Rn, ^{220}Rn, their progenies and dose rates in dwellings decreased with increase in room volume. ***Conclusions:*** The annual effective inhalation dose due to ^{222}Rn, ^{220}Rn and their progenies from the study ranged between 0.2–4.4 with an arithmetic mean of 1.7 ± 1.1 mSv y^{-1}. The dwellers of lower volumes are posed to high dose rates. **Iran. J. Radiat. Res., 2012; 9(4): 231-236**

Keywords: *^{222}Rn, ^{220}Rn, room volume, dose rate.*

INTRODUCTION

Inhalation pathway is reported to be the major contributor to radiation dose received by man from natural sources [(1)]. This path way is mainly contributed by radon and progeny nuclides. Measurement of radon in living atmosphere thus becomes pertinent while assessing dose from natural environment [(2)]. Since radon gas concentration in the environment varies diurnally and seasonally, it is essential that long term integrated measurements are carried out for a meaningful estimate of the gas concentration. Solid state nuclear track detector (SSNTD) based dosimeters are used for long term measurement of radon in atmosphere [(3, 4)]. SSNTD dosimeters can be used in large numbers covering more numbers of sampling locations for integrated long term measurements of radon and progeny nuclides [(5)]. The major sources of indoor radon levels are emanation from soil and building materials, radon release from tap water and natural cooking gas and infiltration from external environment. The topography, type of house construction, soil characteristics, ventilation rate, wind direction, atmospheric parameters and to a certain extend life style of the people influence the concentration of radon inside a house [(6-8)].

Boyle's law states that *at constant temperature* for a fixed mass, the absolute pressure and the volume of a gas are inversely proportional. Robert Boyle derived the law solely on experimental grounds. The law can also be derived theoretically based on the presumed existence of atoms and molecules and assumptions about motion and perfectly elastic collisions. In mathematical terms, this law is:

$$PV = \mathrm{k}$$

where:
p denotes the pressure of the system.
V denotes the volume of the gas.
k is a constant value representative of the pressure and volume of the system.

So long as temperature remains constant the same amount of energy given to the system persists throughout its operation and therefore, theoretically, the value of k will remain constant. Forcing the volume V of the fixed quantity of gas to increase, keeping the gas at the initially measured

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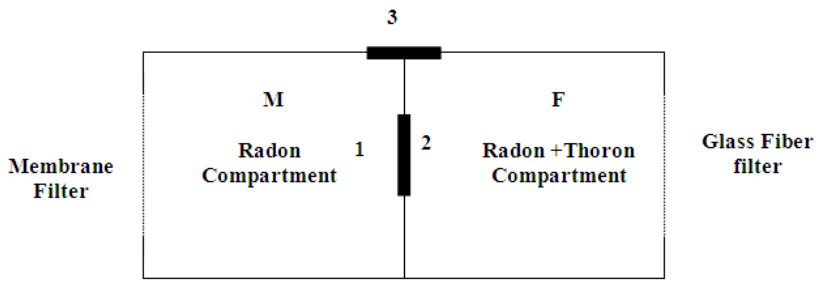
temperature, the pressure p must decrease proportionally. Conversely, reducing the volume of the gas increases the pressure. A common use of this law is to predict how a change in pressure will alter the volume of the gas or vice-versa. Such a problem can be regarded as a two state problem: the initial state (represented by subscript i) and the final state (represented by subscript f). If a sample of gas initially at pressure P_i and volume V_i is subjected to a change that does not change the amount of gas or the temperature, the final pressure P_f and volume V_f are related to the initial values by the expression: $P_i V_i = P_f V_f$. In view of this an attempt is made to observe the volumetric variation of indoor ^{222}Rn and ^{220}Rn. In the present method, the mode of sampling applied was by passive technique using SSNTD based dosimeters for time integrated long duration sampling.

MATERIALS AND METHODS

Twin cup dosimeters developed in Bhabha Atomic Research Centre (BARC), Mumbai, India were used in this study. The dosimeter has two cylindrical cups of equal volumes having radius 3.1 cm and height 4.1 cm as shown in figure 1. The cups are having provision to hold SSNTD films inside the cups and a third SSNTD film outside the cup for progeny measurements. The dosimeter is designed to discriminate ^{222}Rn and ^{220}Rn in mixed field situations, where both the gases are present like in monazite rich deposit areas. Track detector used in the dosimeter is cellulose nitrate films, commercially called LR-115 films, made by Kodak, Pathe. Films of size 3 cm × 3 cm were affixed at the bottom of each cup as well as on the outer surface of the dosimeter. The exposure of the detector inside the cup is termed as *cup mode* and other one exposed openly is termed as *bare mode*. One of the cups has its entry covered with a glass fiber filter paper that permeates both ^{222}Rn and ^{220}Rn gases into the cup and is called *filter cup*. The other cup is covered with a semi permeable membrane sandwiched between two glass fiber filter papers called membrane cup [9]. These types of semi permeable membranes have diffusion coefficient for radon gas in the range of 10^{-8}–10^{-7} cm^2 s^{-1} that permeates more than 95% of the ^{222}Rn gas while it suppress the entry of ^{220}Rn gas to [10] more than 99%. Thus, the SSNTD films inside the membrane cup register tracks that attributes to ^{222}Rn gas alone, while the *filter film* records tracks due to both ^{222}Rn and ^{220}Rn gases. The third film exposed in the bare mode registers alpha tracks produced by both the gases and their alpha emitting progeny. Eappen and Mayya [11] have reported that LR-115 (12 μm) film does not register tracks from deposited activity since E_{max} for LR-115 (12 μm) is 4 MeV and all the progeny isotopes of ^{222}Rn /^{220}Rn emit alphas with energies more than 5 MeV. Thus, uncertainty due to deposited activity on film surface is removed for the bare detector estimate; a reason to choose LR-115 (12 μm) film for bare card estimate.



1. Radon Cup mode SSNTD Film
2. Radon + Thoron Cup mode SSNTD Film
3. Bare mode SSNTD Film

Figure 1. Schematic diagram of twin cup ^{222}Rn/^{220}Rn dosimeter used in the study.

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The dosimeters were kept at a height of about 1.5 m from the ground, considering least disturbance to the occupants. Care is taken while placing the dosimeter such that the active surface of the SSNTD film used in bare mode exposure is kept at a minimum distance of 10 cm away from any surface to avoid tracks due to attenuated alphas reaching from these surfaces. Measurements were completed in each dwelling for a calendar year covering the four seasons prevailing in the area. After exposure, the dosimeters were retrieved and SSNTD films were removed from the dosimeter for etching. The films were then etched in 10% NaOH solution at 60 ºC for 90 minutes [11]. The tracks recorded on LR-115 films were counted using a spark counter [12, 13]. Tracks are converted to gas concentrations using equations (1) and (2):

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

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

where T_m is the track density of the film in membrane compartment (Tr cm^{-2}), d is the period of exposure in days (d), S_m refers to the sensitivity factor of membrane compartment (Tr cm^{-2})/(Bq d m^{-3}), T_f is the track density of the film in filter compartment (Tr cm^{-2}), S_{rf} is the Sensitivity of ^{222}Rn in filter compartment (Tr cm^{-2})/(Bq d m^{-3}) and, C_R and C_T are the concentrations (Bq m^{-3}) of ^{222}Rn and ^{220}Rn, respectively. We followed the protocols given by Eappen and Mayya [11] for processing the exposed films; hence sensitivity factors S_m and S_{rf} are taken from their work for computing the gas concentrations. The progeny concentrations in terms of Working Level (WL) can be written as:

$$R_n(mWL) = \frac{C_R \times F_R}{3.7} \quad (3)$$

$$R_T(mWL) = \frac{C_T \times F_T}{0.275} \quad (4)$$

where F_R and F_T are equilibrium factors for radon and thoron respectively and can be equated with progeny fractions of respective gases as shown in equations (5) and (6).

$$F_R = 0.104F_{RA} + 0.514F_{RB} + 0.37F_{RC} \quad (5)$$

$$F_T = 0.91F_{TB} + 0.09F_{TC} \quad (6)$$

where F_{RA}, F_{RB}, F_{RC}, F_{TB} and F_{TC} are activity fractions of ^{218}Po, ^{214}Pb, ^{214}Bi, ^{212}Pb and ^{212}Bi, respectively. Mayya *et al.* [14] have obtained these activity fractions through ventilation parameters applying a root finding method using the deposition velocities for attached and unattached fractions of the progeny nuclides. Since the data in this study is not sufficient for deriving ventilation dependent F_{xx} factors, bare card results are not used in deriving F values. Such an exercise will be tried later after large number of measurements data are collected from future study. For the present study, inhalation dose is computed using UNSCEAR [1] F values. Indoor occupancy factor for the population is taken as 0.8 and the annual inhalation dose (mSv y^{-1}) is calculated using equation (7).

$$D(mSv\ y^{-1}) = 7000 \times [(0.17 + 9F_R)C_R + (0.11 + 40F_T)C_T] \times 10^{-6}$$

RESULTS AND DISCUSSION

The natural radioactivity contents of soil samples of Bangalore region reported by earlier studies are 15.2, 16.90 and 486.7 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K respectively [15] and the concentrations of ^{238}U, ^{232}Th and ^{40}K in the building rocks of Karnataka region are 33.0, 30.5 and 412.3 Bq kg^{-1} respectively [16]. However, major quantity of bricks used for the construction of the buildings in Bangalore city are brought from places in the city out skirts called Nelamangala, and Magadi and a small quantity from Hoskote, Ramanagara and Channapattana. The average activity concentrations of ^{226}Ra, ^{232}Th and ^{40} K in the soils of Nelamangala and

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Magadi are 31.3 ± 0.6, 52.6 ± 0.9 and 303.1 ± 6.1 Bq kg^{-1} and 16.9 ± 0.6, 57.5 ± 1.1 and 1073 ± 15.6 Bq kg^{-1} respectively [17]. The radon gas is occurring in the groundwater of the study area ranging from 55.96 Bq L^{-1} to 1189.30 Bq L^{-1} [18]. The authors have also reported that the radon concentration is above the permissible limit of 11.83 Bq L^{-1} and at places the concentration is as high as hundred times.

A total of 46 houses were selected for the study and the houses were categorized into six groups with average room volume ranging from 35 to 300 m^3. Each group had minimum of 7 houses. Geometric means of indoor ^{222}Rn and ^{220}Rn levels in the study area are 23.0 and 20.0 Bqm^{-3} with GSDs 2.1 and 2.0 respectively. Cumulative frequencies against the $^{222}Rn/^{220}Rn$ values showed linear regression with correlation coefficient 1 for both the cases. A linear correlation with correlation coefficient nearing one indicates a common factor predominant in the various categories of rooms governing the gas concentrations in these houses. Inhalation dose is calculated using UNSCEAR [1] dose conversion factors and it varied from 0.27 - 4.45 mSv y^{-1} with a geometric mean of 1.34 mSv y^{-1} (GSD 2.1). Variation of ^{222}Rn and ^{220}Rn concentrations with volume of dwellings is shown in figure 2.

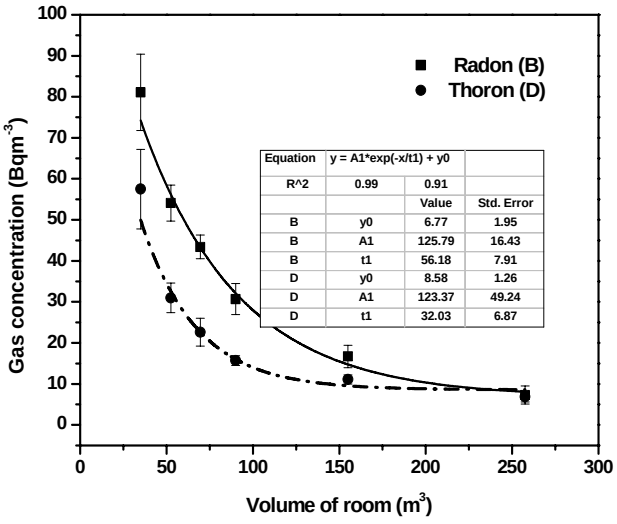

Figure 2. Variation of ^{222}Rn and ^{220}Rn concentrations with volume of dwellings.

It is seen from the figure that the concentrations of ^{222}Rn and ^{220}Rn decrease with increase in volume of the rooms. However, in the case of ^{220}Rn the effect is almost nullified beyond room volumes greater than 100 m^3. If we consider that the exhalation rate for ^{222}Rn and ^{220}Rn from the room surfaces is almost same, assuming that the materials used for construction in these houses are similar, it is expected that the gas concentrations will decrease with increase in volume of the room since the surface to volume ratio decreases with increase in room volume.

Scatter plot of area to volume ratio (A/V) to the volume of room of dwellings for the environment of Bangalore, India is plotted in figure 3. It shows that there is a perfect exponential decay fit with a correlation of 99%.

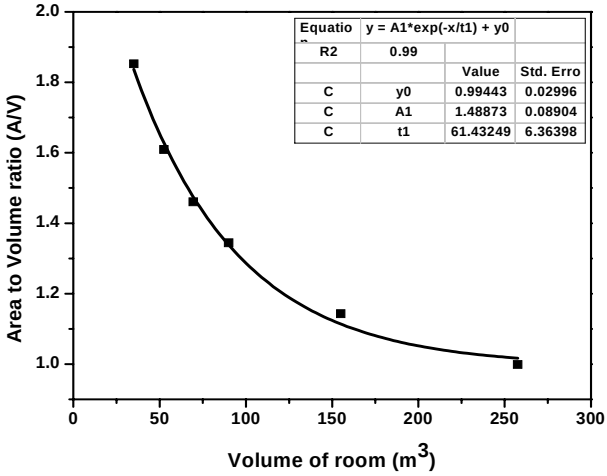

Figure 3. Scatter plot of area to volume ratio (A/V) to the volume of room of dwellings for the environment of Bangalore, India.

It is interesting to note that the fitting parameter 't' in figure 3 is 61.4 which closely match with effective decay value for ^{222}Rn (56.2) in figure 2 from six groups of experimental results. This clearly indicates that the ^{222}Rn values inside dwellings covered under the study is predominantly depended on A/V ratio inside the houses. Effect of ventilation seems negligible when the measurement was carried out for long durations. However, the results of thoron were different compared to ^{222}Rn. The 't'

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value is almost half (32) to that of A/V ratio. One can speculate certain other phenomenon governing the ^{220}Rn values. It is only logical to say that predominance of ^{220}Rn profile inside the room exists to some extend and in rooms having larger volumes concentration of ^{220}Rn is profound from surfaces closer to dosimeter placement.

Figure 4 shows the variation of concentration with inverse volume. It is interesting to note that the variation of concentration against change in the volume of the room with its least square fit leads to a straight line. The correlation between concentration and volume is 95% in all cases and envisages that there is a direct dependence of concentration with volume. The annual effective inhalation dose due to ^{222}Rn, ^{220}Rn and their progenies from the study ranged between 0.2 – 4.4 with an arithmetic mean of 1.7 ± 1.1 mSv y^{-1}.

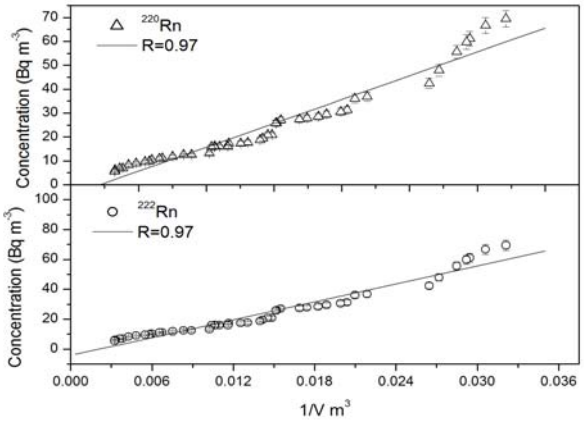

Figure 4. Correlation between ^{222}Rn and ^{220}Rn with inverse of the volume of rooms.

The concentration [19] C of ^{222}Rn/^{220}Rn may be written as:

$$C = \frac{JA}{V(\lambda_0 + \lambda_v)}$$

where, J is the area averaged emission flux, A and V are the surface area and the volume of the room respectively, l_0 is the radioactive decay constant and l_v is the air-exchange rate. For ^{222}Rn, air exchange rates are of the order of 0.1-1 h^{-1}, whereas the decay constant is 0.0076 h^{-1} then $l_v >> l_0$ by neglecting the decay of ^{222}Rn. Therefore, for all the houses in a particular location, J is approximately same and different combination of l_v, A and V will result in different concentrations. However, for ^{220}Rn, the decay constant is 45 h^{-1} whereas air exchange rates are 0.1-1 h^{-1}, $l_0 >> l_v$ by neglecting the ventilation. Then the concentration will depend upon A/V. As a first effort, an attempt is made to interpret the data with respect the volume of the room and its fitting parameters and is shown in figure 3. This result obeys the Boyle's law.

This reveals that the concentration is inversely proportional to the volume of the room provided the activity concentration of radon source is constant. This can also be stated that the product of concentrations and volume of the room is always constant. The variations follow exactly a hyperbolic function and become an experimental proof of the Boyle's law, keeping other parameters constant. The model studies with all these parameters and concentrations need further attention to have a clear picture of the variations of ^{222}Rn and ^{220}Rn in dwellings.

CONCLUSION

The main elucidation of the study is the estimation of the concentration in rooms of different volume due to the indoor radon exposure. The result concludes that the concentrations are more in smaller volume rooms and lower in larger volume rooms. Hence, it is suggested that the lower volume rooms should be well ventilated to reduce the radiation exposure due to radon and its progeny. The study also reveals that 'at constant activity concentrations of radon source for a fixed mass, the concentration of radon and the volume of the room are inversely proportional'.

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