

Simultaneous measurement of radon and thoron using Lucas scintillation cell

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Abstract

A method has been developed for the simultaneous measurement of radon (^{222}Rn) and thoron (^{220}Rn) in a mixed environment using the Lucas scintillation cell (LSC). The method uses counts from two arbitrary counting intervals from zero time with respect to sampling hence, called two count method (TCM). The basis of the method involves formulation of simultaneous linear equations with two unknown variables. The unknown variables represent the radon and thoron activities in the cell. The coefficients of the equations are the integrated activities of radon, thoron and their alpha decay products per unit activities of the parent radionuclides. The method can be used to estimate the radon and thoron concentrations in a mixed environment accurately and quickly, as there is no need to delay the counting to achieve complete decay of thoron. The mathematical basis of the method along with few experimental results is presented in this paper. Results yielded, on comparison, good agreement with the delay count method (DCM) and double filter measurement (DFM) technique. Multiple regression analysis is also carried out to get the concentrations of radon and thoron from the counts obtained for various counting intervals. The corresponding growth factors of radon and thoron also showed good comparison with the average concentrations obtained from the two count approach for different counting intervals. Use of LSC for routine environmental measurements has its limitation due to high minimum detection level (MDL) values. However, LSC is commonly used in uranium mines, high background radiation areas and in calibration experiments. The present methodology is a very convenient approach to measure radon and thoron simultaneously especially in calibration experiments.

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1. Introduction

The Lucas scintillation cell (LSC) is commonly used all over the world for the estimation of radon. The cell was originally devised by Vandilla and Taysum (1955). The cell has been modified since by others (Lucas, 1957; Kristan and Kobal, 1973; Raghavayya, 1977; Quindos-Ponceta et al., 2003; Abbady et al., 2004). The air sample is admitted inside the cell through a filter and the concentration is evaluated using the efficiency factor for the counting system and from the theoretical factors due to buildup of radon decay products with respect to sampling and counting delays. The principle of detection is counting of photons resulting from the interaction of alpha particles produced by radon and its progeny with the ZnS (Ag) scintillator. A photomultiplier (PM) tube assembly counts the photon events. Fig. 1 gives the schematic diagram of a Lucas cell.

Several techniques have been developed to estimate thoron concentration with the Lucas cell (Dyck, 1969; Morse, 1976; Hutter, 1995). Hutter (1995) used a 1 min counting immediately after sampling followed by another counting after a delay of 5 min for a duration of 10 min. The counts in the first counting period due to radon and its progeny are calculated from the known radon concentration and then subtracted from the total counts obtained in the first counting period. The remaining counts are due to thoron and its progeny and are used to calculate the thoron concentration. The overall uncertainty of this method lies between 10% and 20% (90% confidence level).

The method developed in this study is based on the decay of radon, thoron and the build up of their progeny inside the cell. Counts obtained for any interval of time from the start of sampling will correspond to the alpha decay of radon, thoron and progeny concentrations present in the cell during that counting interval. This could be computed theoretically for the counting interval for unit activities of radon and thoron. Hence, the accuracy of either radon or thoron estimation will not be affected

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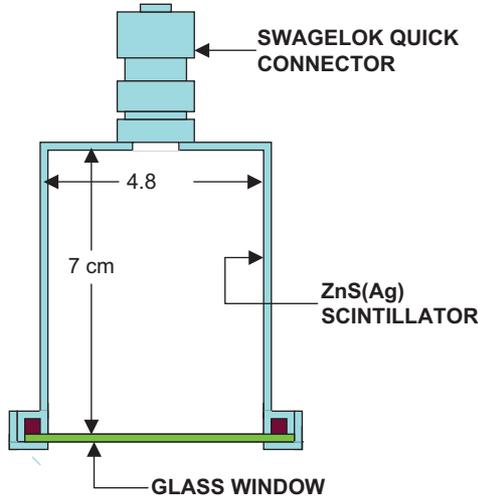


Fig. 1. Schematic diagram of a Lucas cell.

by the relative concentrations of its counterparts. The method is tested successfully against the delay count method (DCM) for radon and the double filter measurement (DFM) technique for thoron.

2. Mathematical formulation

Let c_1 and c_2 be the total disintegrations (efficiency corrected) obtained for the counting intervals $0-t_1$ and $0-t_2$, respectively. These disintegrations are contributed by radon and two of its alpha decay products (^{218}Po and ^{214}Po) and thoron and its three alpha decay products (^{216}Po , ^{212}Bi and ^{212}Po). This information forms the basis for the following simultaneous equations, which can be easily solved to obtain the radon and thoron activities in the cell.

$$c_1 = a_{\text{RN}1}x + a_{\text{TN}1}y, \quad (1)$$

$$c_2 = a_{\text{RN}2}x + a_{\text{TN}2}y. \quad (2)$$

Here x is the radon activity and y is the thoron activity in the cell (unknown variables in Bq). The coefficients $a_{\text{RN}1}$ and $a_{\text{RN}2}$ are the integrated activities (disintegrations) of radon and its alpha emitting daughters for the time periods $0-t_1$ and $0-t_2$, respectively, per unit activity of radon. The integrated activities of ^{222}Rn and its alpha emitting daughters for these time periods can be mathematically expressed as

$$a_{\text{RN}1} = \int_0^{t_1} A_{R1}(t) dt + \int_0^{t_1} A_{R2}(t) dt + \int_0^{t_1} A_{R5}(t) dt, \quad (3)$$

where A_{R1} , A_{R2} and A_{R5} are the activities of ^{222}Rn , ^{218}Po and ^{214}Po , respectively, at time t per unit activity of radon. Similarly $a_{\text{RN}2}$ can be expressed as

$$a_{\text{RN}2} = \int_0^{t_2} A_{R1}(t) dt + \int_0^{t_2} A_{R2}(t) dt + \int_0^{t_2} A_{R5}(t) dt, \quad (4)$$

where

$$A_{R1}(t) = e^{-\lambda_1 t}, \quad (5)$$

$$A_{R2}(t) = \lambda_2 \left\{ \frac{e^{-\lambda_1 t}}{(\lambda_2 - \lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1 - \lambda_2)} \right\}, \quad (6)$$

$$A_{R5}(t) = \lambda_2 \lambda_3 \lambda_4 \lambda_5 \left\{ \frac{e^{-\lambda_1 t}}{\alpha_1} + \frac{e^{-\lambda_2 t}}{\alpha_2} + \frac{e^{-\lambda_3 t}}{\alpha_3} + \frac{e^{-\lambda_4 t}}{\alpha_4} + \frac{e^{-\lambda_5 t}}{\alpha_5} \right\}, \quad (7)$$

where

$$\alpha_1 = (\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)(\lambda_4 - \lambda_1)(\lambda_5 - \lambda_1),$$

$$\alpha_2 = (\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)(\lambda_4 - \lambda_2)(\lambda_5 - \lambda_2),$$

$$\alpha_3 = (\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)(\lambda_4 - \lambda_3)(\lambda_5 - \lambda_3),$$

$$\alpha_4 = (\lambda_1 - \lambda_4)(\lambda_2 - \lambda_4)(\lambda_3 - \lambda_4)(\lambda_5 - \lambda_4),$$

$$\alpha_5 = (\lambda_1 - \lambda_5)(\lambda_2 - \lambda_5)(\lambda_3 - \lambda_5)(\lambda_4 - \lambda_5).$$

In these equations, the symbol λ represents the radioactive decay constants of ^{222}Rn and its daughters such that the subscript 1 stands for the parent and the other subscripts stand for its daughters, respectively.

The coefficients $a_{\text{TN}1}$ and $a_{\text{TN}2}$ represent the integrated activities (disintegrations) of ^{220}Rn and its alpha emitting daughters for the time periods $0-t_1$ and $0-t_2$, respectively, per unit activity of thoron as given below.

$$a_{\text{TN}1} = \int_0^{t_1} A_{T1}(t) dt + \int_0^{t_1} A_{T2}(t) dt + \left[0.36 \int_0^{t_1} A_{T4}(t) dt + 0.64 \int_0^{t_1} A_{T5a}(t) dt \right], \quad (8)$$

$$a_{\text{TN}2} = \int_0^{t_2} A_{T1}(t) dt + \int_0^{t_2} A_{T2}(t) dt + \left[0.36 \int_0^{t_2} A_{T4}(t) dt + 0.64 \int_0^{t_2} A_{T5a}(t) dt \right], \quad (9)$$

where A_{T1} , A_{T2} , A_{T4} and A_{T5a} are the activities of ^{220}Rn , ^{216}Po , ^{212}Bi and ^{212}Po , respectively, at time t per unit activity of thoron. The expressions for these activities are given below.

$$A_{T1}(t) = e^{-\lambda_1 t}, \quad (10)$$

$$A_{T2}(t) = \lambda_2 \left\{ \frac{e^{-\lambda_1 t}}{(\lambda_2 - \lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1 - \lambda_2)} \right\}, \quad (11)$$

$$A_{T4}(t) = \lambda_2 \lambda_3 \lambda_4 \left\{ \frac{e^{-\lambda_1 t}}{\beta_1} + \frac{e^{-\lambda_2 t}}{\beta_2} + \frac{e^{-\lambda_3 t}}{\beta_3} + \frac{e^{-\lambda_4 t}}{\beta_4} \right\}, \quad (12)$$

$$A_{T5a}(t) = \lambda_2 \lambda_3 \lambda_4 \lambda_{5a} \left\{ \frac{e^{-\lambda_1 t}}{\alpha_1} + \frac{e^{-\lambda_2 t}}{\alpha_2} + \frac{e^{-\lambda_3 t}}{\alpha_3} + \frac{e^{-\lambda_4 t}}{\alpha_4} + \frac{e^{-\lambda_{5a} t}}{\alpha_{5a}} \right\}, \quad (13)$$

Table 1
Decay series of ^{222}Rn , ^{220}Rn

Nuclide	Notation	Decay mode	Half-life	Product of decay
^{222}Rn				
^{222}Rn	A_{R1}	α	3.82 days	^{218}Po
^{218}Po	A_{R2}	α	3.11 m	^{214}Pb
^{214}Pb	A_{R3}	β	26.8 m	^{214}Bi
^{214}Bi	A_{R4}	β	19.8 m	^{214}Po
^{214}Po	A_{R5}	α	164 μs	^{210}Pb
^{220}Rn				
^{220}Rn	A_{T1}	α	55.6 s	^{216}Po
^{216}Po	A_{T2}	α	0.15 s	^{212}Pb
^{212}Pb	A_{T3}	β	10.64 h	^{212}Bi
^{212}Bi	A_{T4}	β (64%)	60.55 m	^{212}Po
		α (36%)	60.6 m	^{208}Tl
^{212}Po	A_{T5a}	α	0.305 μs	^{208}Pb

where

$$\beta_1 = (\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)(\lambda_4 - \lambda_1),$$

$$\beta_2 = (\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)(\lambda_4 - \lambda_2),$$

$$\beta_3 = (\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)(\lambda_4 - \lambda_3),$$

$$\beta_4 = (\lambda_1 - \lambda_4)(\lambda_2 - \lambda_4)(\lambda_3 - \lambda_4).$$

The terms α_1 , α_2 , α_3 and α_4 are defined earlier. The term α_{5a} is defined as

$$\alpha_{5a} = (\lambda_1 - \lambda_{5a})(\lambda_2 - \lambda_{5a})(\lambda_3 - \lambda_{5a})(\lambda_4 - \lambda_{5a}),$$

where λ_{5a} is the radioactive decay constant of ^{212}Po , the alpha emitting daughter product of the beta emitting branching of ^{212}Bi in the thoron decay series.

Here, the symbol λ represents the radioactive decay constants of ^{220}Rn and its daughters such that the subscript 1 stands for the parent and the other subscripts stand for its daughters respectively. Table 1 gives the decay series of ^{222}Rn and ^{220}Rn with branching ratios identifying the nuclides with notation used in the above equations.

Eqs. (3), (4), (8) and (9) can be integrated analytically or numerically. The branching fractions (Eqs. (8) and (9)) are taken into account for computations giving weightage to the half life of ^{212}Po atoms in the thoron decay series. Numerical integration based on the trapezoidal rule as well as analytical integration is used in the present study. As an example, the time integrated alpha activity (disintegrations) calculated for radon and thoron from 0 to 1000 s are shown in Fig. 2.

In Eqs. (1) and (2), the unknowns are x and y , and the equations can be solved to obtain the radon and thoron activities in the cell. These activities in the cell can be converted into respective concentrations using the volume of the cell. A computer program has been developed to estimate the radon and thoron concentrations in the cell based on the measured total disintegrations for any two arbitrary counting periods.

For a working formula, the mathematical formulation can be simplified for two specified counting periods. For example, let 0–100 and 0–500 s be the two counting periods chosen for the

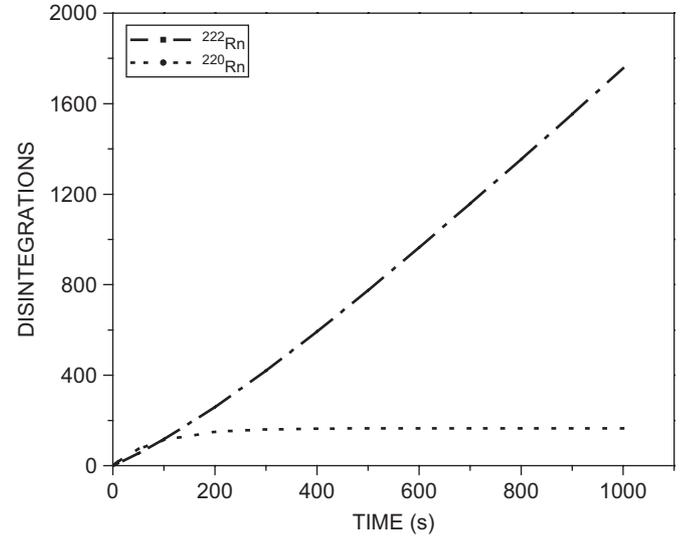


Fig. 2. Integrated disintegrations (theoretical) of ^{222}Rn , ^{220}Rn and their progeny for unit activity of parents.

measurements of radon and thoron. Radon and its alpha emitting daughters will theoretically register 116.75 disintegrations in total (a_{RN1}) for 100 s per unit activity of radon. The same for 500 s (a_{RN2}) is computed as 776.94 disintegrations. Similarly for thoron, these quantities are estimated as $a_{TN1} = 113.11$ and $a_{TN2} = 157.00$ disintegrations. The coefficients in Eqs. (1) and (2) are substituted with the numeric values obtained for the specific counting time intervals and the concentrations of radon and thoron can be calculated using the following simple equations.

$$R_n = (1626.31c_2 - 2257.37c_1)/V, \quad (14)$$

$$T_n = (11170.97c_1 - 1678.65c_2)/V, \quad (15)$$

where R_n is the concentration of radon in the cell (Bq m^{-3}), T_n is the concentration of thoron in the cell (Bq m^{-3}) and V is the volume of the cell (ml). The terms c_1 and c_2 represent the measured total disintegrations for time periods 0–100 and 0–500 s, respectively.

3. Experimental set up

A few laboratory scale experiments were carried out for the estimation of radon and thoron using TCM, DCM and DFM. A leak proof cubical chamber of 0.5 m^3 capacity was used to contain the gases for the experiments. ^{222}Rn was introduced into the chamber from a ^{226}Ra source. Equilibrated ^{222}Rn from the ^{226}Ra source was pumped at different durations to alter the concentration inside the chamber. Thorium powder (^{232}Th) separated from monazite, packed in a porous container was used to generate ^{220}Rn inside the chamber. By varying the thorium powder quantity, ^{220}Rn concentration was varied inside the chamber. A small fan was used inside the chamber for uniform mixing of the gases inside the chamber. A sampling port at one side of the chamber was used for sample collections.

4. Two count method (TCM)

The LSC is coupled to a photomultiplier tube assembly which in turn is connected to an alpha counting system. The volume of the LSC used is 140 ml. Sampling was done by vacuum collection through quick connector system. The air sample collected was filtered through a glass fibre filter, before entry into the Lucas cell, to prevent progeny nuclides entering the cell. Counting of the LSC was started instantly after sampling. The total cumulative counts for every 100 s interval were recorded and counting continued up to 2000 s. A computer program developed based on the TCM was used to estimate the radon and thoron concentrations in the cell. Data obtained was also subjected to a multiple regression analysis making use of the counts obtained for different counting intervals.

5. Delay count method (DCM)

^{222}Rn in the Lucas cell was estimated by the DCM. The sample collected in the LSC was recounted after a delay of more than 3 h. By this time all the ^{220}Rn atoms are decayed and radon inside the cell attains equilibrium with its progeny nuclides. Counts were taken for 10 min. The ^{222}Rn concentration in the cell was estimated using Eq. (16) (Raghavayya et al., 1980).

$$R_n = \frac{6.967 \times 10^{-5} C}{V E e^{-\lambda_R t} (1 - e^{-\lambda_R T})}, \quad (16)$$

where R_n is the concentration of radon in the cell (Bq m^{-3}); V is the volume of the cell (m^3); E is the efficiency of the counter (%); C is the net counts obtained for the counting duration; t is the delay time from sampling to start of the counting time (min); T is the duration of counting (min); and λ_R is the decay constant of radon (min^{-1}). The numerical constant, 6.967×10^{-5} , is the product of the decay constants computed after a delay of 180 min post sampling.

6. Double filter method (DFM)

Immediately after the LSC sample was collected, the port is connected to a double filter system and sampling was done for a few minutes with 10–15 lpm flow rate. As the volume sucked out being small compared to the chamber volume, the concentrations of ^{222}Rn and ^{220}Rn in the air sample was assumed to be same from the start to end of the sampling. The exit filter was then counted for alpha activity at two counting intervals and the ^{222}Rn and ^{220}Rn activities were estimated using the method given by Mayya and Sahni (1994).

7. Results and discussion

7.1. Two count method (TCM)

The TCM was used for estimating the concentrations of ^{222}Rn and ^{220}Rn from the counts obtained for the two sets of counting periods. Since cumulative counts were taken for every 100 s from start of the counting to 2000 s, ^{222}Rn and

Table 2
Statistical estimates for the two count method

Expt. no.	^{222}Rn concentration (Bq m^{-3})		^{220}Rn concentration (Bq m^{-3})	
	Mean	SD	Mean	SD
1	4715	137	21 291	552
2	2262	156	22 125	706
3	1354	101	9361	295
4	946	39	7207	171
5	6929	62	21 787	303
6	5524	505	26 461	2482
7	32 146	632	NDA	NDA
8	54 126	280	24 186	939

NDA, no data available.

^{220}Rn concentration were computed for the different sets of counts available. Table 2 gives the mean concentrations calculated from concentrations obtained for the sets of counts at different counting combinations and their standard deviations. It can be seen that the variation in concentration for different counting intervals is within 10% of the mean concentration for both ^{222}Rn and ^{220}Rn .

For a better estimate, the counts available for the different counting periods are subjected to a multi-regression analysis with counts as the variable parameter and related growth factors of radon and thoron as independent variable. Fig. 3 gives the comparison of radon and thoron concentrations obtained by the two approaches. It could be seen that the radon concentrations are same for both the methods whereas thoron shows lower concentrations for the multiple regression analysis compared to the two count method (slope 0.94). The lower concentration of thoron for the multiple regression analysis could be because of the background build up inside the Lucas cell. It is also seen that when the initial counting intervals were larger (> 500 s), the thoron computations were showing incorrect values.

7.2. Comparison of TCM and DCM

The delay count method was used to estimate the radon concentrations in 10 experiments. Thoron in the cell is decayed with in few minutes and radon will form equilibrium with its progeny nuclides with in about 200 min. Counts obtained after this delay time corresponds to radon only and the concentration is calculated using Eq. (16). The results of these experiments are compared with those obtained from the two count method (Fig. 4). Results show very good agreement between the two methods. The resultant regression line has a slope of 0.96 and a correlation coefficient of 0.999.

7.3. Comparison of TCM and DFM

The radon and thoron concentrations were also estimated using the DFM and the results of five experiments are presented in Table 3. This table also contains the concentrations of radon and thoron estimated by the TCM. The total disintegrations measured, for the two counting intervals (0–100 and 0–500 s),

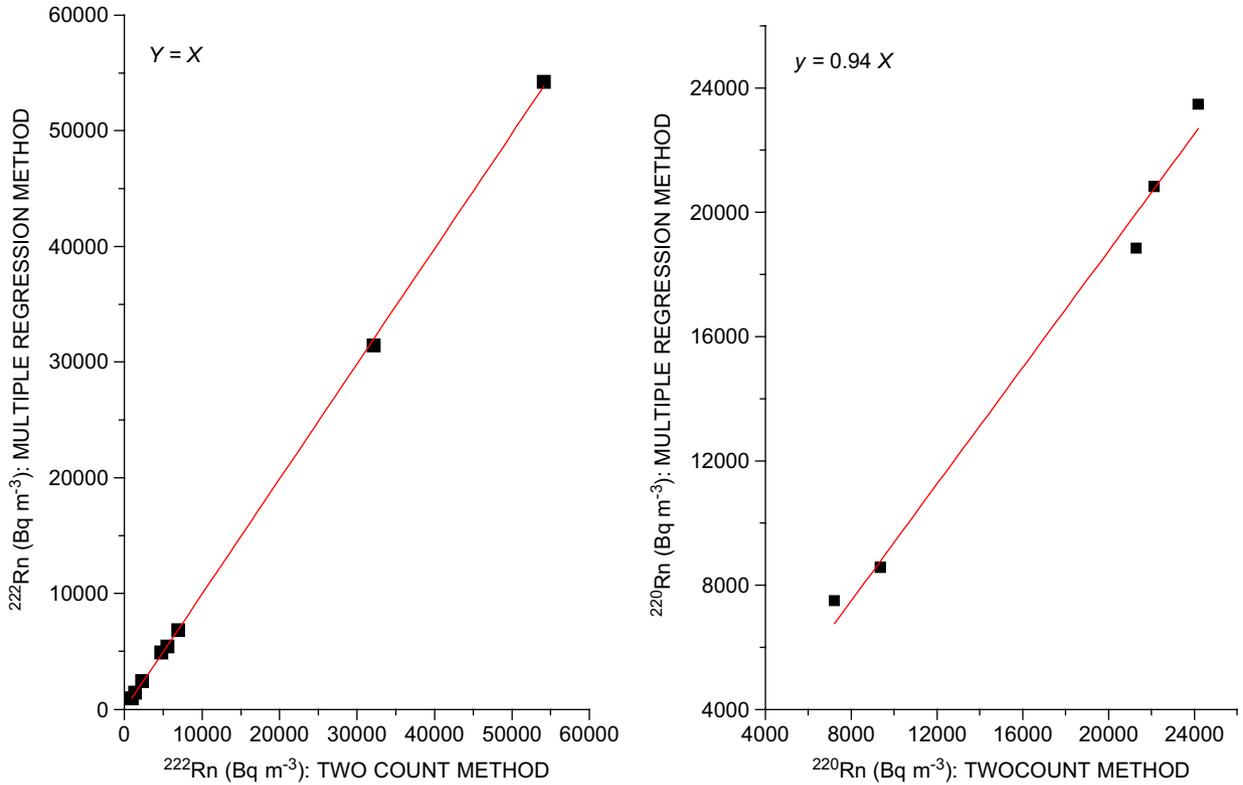


Fig. 3. Comparison of two count and multiple regression methods.

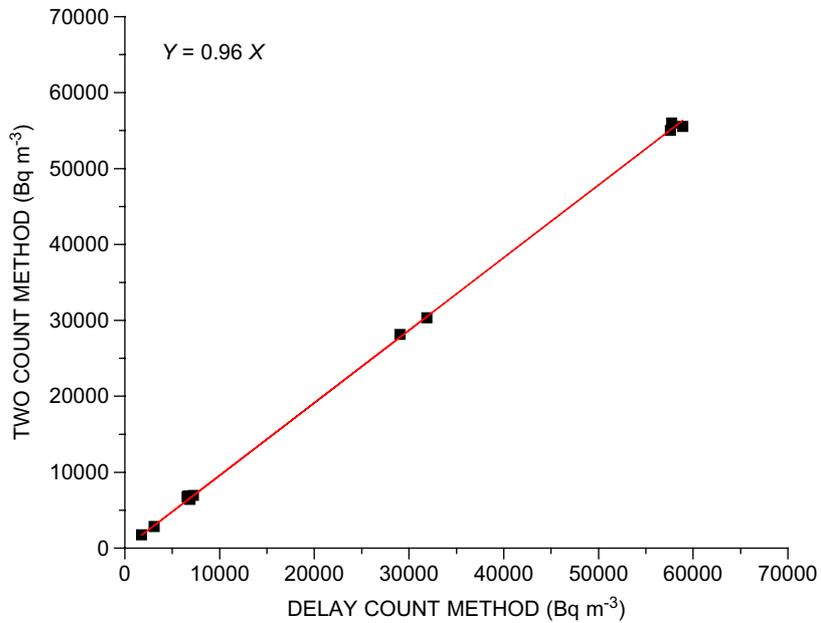


Fig. 4. Comparison of delay count and two count methods for ^{222}Rn .

due to radon, thoron and their alpha decay products are also given in the table.

In the first four experiments, the thoron concentrations are considerably higher than the radon concentrations. The ratio of counts in these cases is almost 2. In the fifth experiment, the radon concentration is more than double than that of thoron

concentration. In this case the ratio of the counts works out to be around 5. The results show very good agreement between the TCM and DFM in all the five experiments, except for the radon concentration in experiment 2. The variation in this case is about 25% whereas all other experiments showed variation within 5% if DFM values are taken as the correct estimation.

Table 3
²²²Rn and ²²⁰Rn concentrations estimated by the two count and double filter methods

Expt. no.	Two count method			Double filter method		
	Measured disintegrations		²²² Rn (Bq m ⁻³)	²²⁰ Rn (Bq m ⁻³)	²²² Rn (Bq m ⁻³)	²²⁰ Rn (Bq m ⁻³)
	0–100 s	0–500 s				
1	315	740	4715	21 291	4689	21 683
2	290	550	2262	22 125	3084	22 061
3	130	268	1354	9361	1356	9546
4	98	197	946	7207	944	7277
5	950	4812	54 126	24 186	54 107	24 142

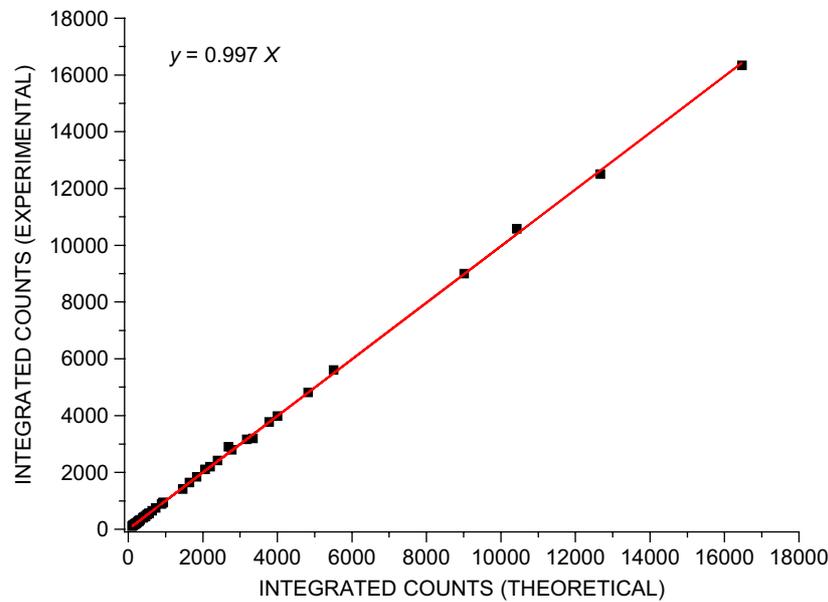


Fig. 5. Comparison of experimental and theoretical counts for different counting intervals.

Assuming that the radon and thoron concentrations estimated by the DFM are exact, the theoretical counts of these concentrations are estimated using Eqs. (1) and (2) for different counting periods. These theoretical counts are compared with the corresponding experimental counts for the same time periods. The correlation between the theoretical and experimental counts is depicted in Fig. 5. The two counts method showed very good agreement. The regression line in Fig. 5 has a slope of 0.997 and a correlation coefficient of 0.999. The one to one correspondence in experimental and theoretical counts indicates that the TCM may be used without significant error in the estimation of radon and thoron concentrations.

8. Minimum detection level

The Lucas cell measurement has limitations with respect to minimum detection level (MDL). It could be seen from Fig. 6 that the MDL depends on the counting duration. For radon alone case, the counting could be done for longer duration there by the MDL could be brought down. The figure shows that for a Lucas cell of volume 140 cm³ with 6 cph background at 75%

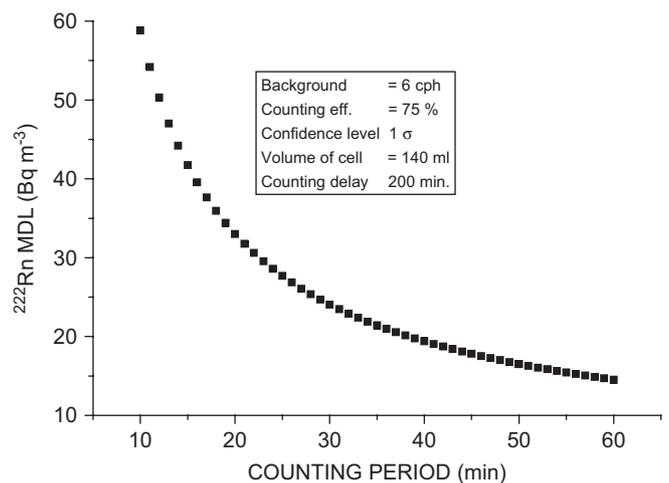


Fig. 6. Minimum detection levels for ²²²Rn using Lucas cell.

counting efficiency, the MDL could be brought down to only 8 Bq m⁻³ at maximum count rate with confidence level at 1σ when the counting period is increased to 60 min. This limitation

hinders the use of Lucas cell for environmental sampling of radon in dwellings. The MDL value for ^{220}Rn by this method is much above a value for normal environmental levels. For the same background and counting efficiency the MDL value of ^{220}Rn is $> 600 \text{ Bq m}^{-3}$. The MDL for ^{220}Rn could be brought down with online sampling. However, in the on-line method the Lucas cell background increases due to deposition of ^{212}Pb atoms (10.6 h half-life). For ^{220}Rn alone measurement, a background correction can be done by a method suggested by Eappen et al. (2007).

9. Conclusions

A method has been developed to estimate the radon and thoron concentrations in a mixed environment using the integrated counts obtained during two arbitrary counting periods. The method is simple and found to be as accurate as the conventional methods like the DFM and the DCM. The main advantage of the TCM is that it saves significant time of measurements. Within 10 min, a good measurement can be obtained using this method, whereas the other methods require a few hours for the same measurements. Unlike the other methods, the two count method can be used to obtain statistical estimates for the same sample based on different counting periods. The multiple regression analysis of the counting data indicates that larger counting intervals in the initial stages of counting give rise incorrect values for thoron concentrations. Hence, it is necessary that counting intervals are maintained at lower durations to obtain accurate results. Results also show that the method is not only applicable in mixed field environments containing radon and thoron, but in radon and thoron alone environments also. The present Lucas cells have limitations in using them for environmental samples because of the comparable MDL value with the atmospheric concentrations. However, the method could be adapted when large volume Lucas cells (Sakamoto and Takakura, 1998) will be used by which the MDL values could be brought down and such cells could be used for environmental radon measurements.

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