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Analysis of Atmospheric Concentrations of Radon and Thoron Using Beta Counting Technique

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Abstract

This paper presents a detailed theory and experimental procedure for measurement and analysis of mixed radon and thoron in the environment. The technique has been successfully applied to the study of seasonal variations of radon and thoron in the atmosphere around Rajshahi, Banladesh, during the years 1989-1991. The maximum radon concentration in outdoor air was observed in the winter from December to January while the indoor radon concentration was found to be maximum during the monsoon months of July and August. The implication of the results is briefly discussed in the paper.

Key Words: radon; thoron; radon progeny; secular equilibrium; radon concentration; seasonal variation.

1. Introduction

The earth's crust and most common building materials contain trace amounts of ^{238}U and ^{232}Th wihch decay to radon ($^{222}_{R}n$ and thoron (^{220}Rn), respectively. The radon gas molecules diffuse out of the ground through pore spaces in rocks and soils and mix with the atmosphere. Inhalation of radon and its daughters can cause a significant health hazard when they are present in enhanced levels in enclosed indoor environments such as human dwellings if they are poorly ventilated and if the radon input from the soil or the building materials in high. It has been suggested that the indoor radon concentration in the U. S. is responsible for about 10% of the total risk of lung cancer [1].

Diring the past decade, numerous devices for assaying the radon content of specific air samples have been developed, but there is not a single technique that can meet all

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the reqirements of the different types of radon measurements. the choice of the most appropirate technique depends on the particular information needed, the type of radon survey, the cost of the apparatus, etc.

However, when considering the use of a specific technique for radon monitoring, one of the most important parameters to be considered is its accuracy. Still, there is some confusion between accuracy and precision; the repoducibility of replicate measurements (i.e. precision) is erroneously taken as an indication of the accuracy of a result.

Although radon measurements via electronic counting of alpha or beta radiations emitted by radon daughters by way of active sampling of air is a standard technique, we have made an attempt to analyze radon and thoron in air employing a beta counting method in measuring the filter activity with an adequate theoretical framework.

2. Theory

The build up of radon and thoron progeny atoms on the filter during the sampling period, t_s, is decribed by the following differential equations [2,3]:

$$\frac{dN_1(t_s)}{dt_s} = \nu n_1 - \lambda_1 N_1(t_s) \quad (1)$$

$$\frac{dN_2(t_s)}{dt_s} = \nu n_2 - \lambda_1 N_1(t_s) - \lambda_2 N_2(t_s) \quad (2)$$

$$\frac{dN_3(t_s)}{dt_s} = \nu n_3 - \lambda_2 N_1(t_s) - \lambda_3 N_3(t_s) \quad (3)$$

and

$$\frac{dN_4(t_s)}{dt_s} = \nu n_4 - \lambda_4 N_4(t_s) \quad (4)$$

$$\frac{dN_5(t_s)}{dt_s} = \nu n_5 + \lambda_5 N_5(t_s), \quad (5)$$

where $N_1(t_s), N_2(t_s), N_3(t_s), N_4(t_s), N_5(t_s)$ are the numbers of atom of RaA(^{218}Po), RaB(^{214}Pb), RaC(^{214}Bi), ThB(^{212}Pb) and ThC(^{212}Bi on the filter, n_1, n_2, n_3, n_4 and n_5 are the numbers of atoms of RaA, RaB, RaC, ThB and ThC per unit volume of air passing through a filter per unit time ($m^3 \cdot \min^{-1}$) and $\lambda_1, \lambda_2, \lambda_3, \lambda_4$,and λ_5 are the decay constants (min^{-1}) of the radon and thoron daughters, respectively.

These equations are solved to get the number of radon and thoron progeny atoms which were collected on the filter paper during the sampling period, t_s:

$$N_1(t_s) = \frac{\nu n_1}{\lambda_1}\left[1 - e^{-\lambda_1 t_s}\right] \quad (6)$$

$$N_1(t_s) = \frac{\nu(n_1 + n_2)}{\lambda_2}\left[1 - e^{-\lambda_2 t_s}\right] + \frac{\nu n_1}{\lambda_2 - \lambda_1}\left[e^{-\lambda_2 t_s} - e^{-\lambda_1 t_s}\right] \quad (7)$$

$$N_3(t_s) = \frac{\nu(n_1+n_2+n_3)}{\lambda_3}\left[1-e^{-\lambda_3 t_s}\right] + \frac{\nu(\lambda_2 n_2-\lambda_1 n_1-\lambda_1 n_2)}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_2)}\left[1-e^{-\lambda_3 t_s}-e^{-\lambda_2 t_s}\right] + \frac{\nu\lambda_2 n_2}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_2)}\left[e^{-\lambda_3 t_s}-e^{-\lambda_1 t_s}\right] \quad (8)$$

$$N_4(t_s) = \frac{\nu n_4}{\lambda_4}\left[1-e^{-\lambda_4 t_s}\right] \quad (9)$$

$$N_5(t_s) = \frac{\nu(n_4+n_5)}{\lambda_5}\left[1-e^{-\lambda_5 t_s}\right] + \frac{\nu n_4}{\lambda_5-\lambda_4}\left[1-e^{-\lambda_5 t_s}-e^{-\lambda_4 t_s}\right] \quad (10)$$

In an atmosphere, secular equilibrium exists when the only input is that of radon and the only losses are by radioactive decay. Departures from this equilibrium occur when additional sources or removal processes alter the relative abundances of radon and its daughters in the air. For indoor radon, the most important of the removal processes are ventilation and the plateout of its daughters on surface within the house. Hence radioactive equilibrium is seldom found in ventilated air spaces.

In the present analysis, it has been assumed that the activities in the atmosphere are in secular equilibrium with radon and its daughters and thoron and its daughters. On the basis of this assumption, Eqns.(6) and (7) yield the ratio of the activity of RaB to the activity of RaA, $R_1(t_s) = \lambda_2 N_2/\lambda_1 N_1$, after the filtering process proceded has for a time t_s:

$$R_1(t_s) = \frac{\frac{\lambda_2+\lambda_1}{\lambda_2}(1-e^{-\lambda_2 t_s}) + \frac{\lambda_2}{\lambda_2-\lambda_1}(e^{-\lambda_2 t_s}-e^{\lambda_1 t_s})}{(1-e^{-\lambda_1 t_s})} \quad (11)$$

Similarly, Eqns.(7) and (8) yield the ratio of the activity of RaC to the activity of RaB:

$$R_2(t_s) = \frac{\frac{(\lambda_1\lambda_2+\lambda_1\lambda_3+\lambda_2\lambda_3)1-e^{-\lambda_3 t_s})}{\lambda_2\lambda_3} - \frac{\lambda_3\lambda_1^2(e^{-\lambda_3 t_s}-e^{-\lambda_2 t_s})}{\lambda_2(\lambda_2-\lambda_1)(\lambda_3-\lambda_2)} + \frac{\lambda_2\lambda_3(e^{-\lambda_3 t_s}-e^{-\lambda_1 t_s})}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)}}{\frac{\lambda_1+\lambda_2}{\lambda_2}(1-e^{-\lambda_2 t_s}) + \frac{\lambda_2}{\lambda_2-\lambda_1}(e^{-\lambda_2 t_s}-e^{-\lambda_1 t_s})} \quad (12)$$

The variations of $R_1(t_s)$ and $R_2(t_s)$ with sampling time, t_s, are shown in Figures 1 and 2.

Once the flow of air through the filter stops, the equations governing the decay of radon and thoron progeny atoms on the filter are

$$\frac{dN_1(t)}{dt} = -\lambda_1 N_1(t) \quad (13)$$

$$\frac{dN_2(t)}{dt} = \lambda_1 N_1(t) - \lambda_2 N_2(t) \quad (14)$$

$$\frac{dN_3(t)}{dt} = \lambda_2 N_2(t) - \lambda_3 N_3(t) \quad (15)$$

$$\frac{dN_4(t)}{dt} = -\lambda_4 N_4(t) \quad (16)$$

$$\frac{dN_5(t)}{dt} = \lambda_4 N_4(t) - \lambda_5 N_5(t) \quad (17)$$

The solutions of these equations are as follows,

$$N_1(t) = N_1^0 \left[e^{-\lambda_1 t}\right] \quad (18)$$

$$N_1(t) = N_1^0 \left(\frac{\lambda_1}{\lambda_2 - \lambda_1}\right) \left[e^{-\lambda_1 t} - e^{-\lambda_2 t}\right] + N_2^0 \left[e^{-\lambda_2 t}\right] \quad (19)$$

$$N_3(t) = N_1^0 \left[\frac{\lambda_1\lambda_2 \cdot e^{-\lambda_1 t}}{(\lambda_3 - \lambda_1)(\lambda_2 - \lambda_1)} - \frac{\lambda_1\lambda_2 \cdot e^{-\lambda_2 t}}{(\lambda_3 - \lambda_2)(\lambda_2 - \lambda_1)} + \frac{\lambda_1\lambda_2 \cdot e^{-\lambda_3 t}}{(\lambda_3 - \lambda_2)(\lambda_3 - \lambda_1)}\right] + N_2^0 \left(\frac{\lambda_2}{\lambda_3 - \lambda_2}\right) \left[e^{-\lambda_2 t} - e^{-\lambda_3 t}\right] + N_3^0 \left[e^{-\lambda_3 t}\right] \quad (20)$$

$$N_4(t) = N_4^0 \left[e^{-\lambda_4 t}\right] \quad (21)$$

$$N_5(t) = N_4^0 \left(\frac{\lambda_4}{\lambda_5 - \lambda_4}\right) \left[e^{-\lambda_4 t} - e^{-\lambda_5 t}\right] + N_5^0 \left[e^{-\lambda_5 t}\right], \quad (22)$$

where $N_1^0 = N_1(t_s)$, $N_2^0 = N_2(t_s)$, $N_3^0 = N_3(t_s)$, $N_4^0 = N_4(t_s)$, and $N_5^0 = N_5(t_s)$, are the number of atoms of RaA, RaB, RaC, ThB and ThC, respectively on the filter at the end of the sampling period, and $N_1(t), N_2(t), N_3(t), N_4(t)$ and $N_5(t)$ are the corresponding numbers on the filter at any time, t, after the end of sampling.

Taking the activities of RaA, RaB and RaC at the end of the sampling period to be A_1^0, A_2^0 and A_3^0, respectively, and using the relationships $A_2^0/A_1^0 = R_1(t_s)$ and $A_3^0/A_2^0 = R_2(t_s)$, one obtains for the beta activites $A_2 = \lambda_2 N_2(t)$ and $A_3 = \lambda_3 N_3(t)$ the expressions

$$A_2(t) = A_2^0 \left[\frac{1}{R_1(t_s)} \cdot \frac{\lambda_2}{\lambda_2 - \lambda_1} \left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) + e^{-\lambda_2 t}\right] \quad (23)$$

$$A_3(t) = A_2^0 \left[\frac{1}{R_1(t_s)} \cdot \frac{\lambda_2\lambda_3 \cdot e^{-\lambda_1 t}}{(\lambda_3 - \lambda_1)(\lambda_2 - \lambda_1)} - \frac{1}{R_1(t_s)} \cdot \frac{\lambda_2\lambda_3 \cdot e^{-\lambda_2 t}}{(\lambda_3 - \lambda_2)(\lambda_2 - \lambda_1)} + \frac{1}{R_1(t_s)} \cdot \frac{\lambda_2\lambda_3 \cdot e^{-\lambda_3 t}}{(\lambda_3 - \lambda_2)(\lambda_3 - \lambda_1)} + \frac{\lambda_3}{\lambda_3 - \lambda_2} \left(e^{-\lambda_2 t} - e^{-\lambda_3 t}\right) + R_2(t_s) e^{-\lambda_3 t}\right] \quad (24)$$

where t is measured from the end of the sampling period.

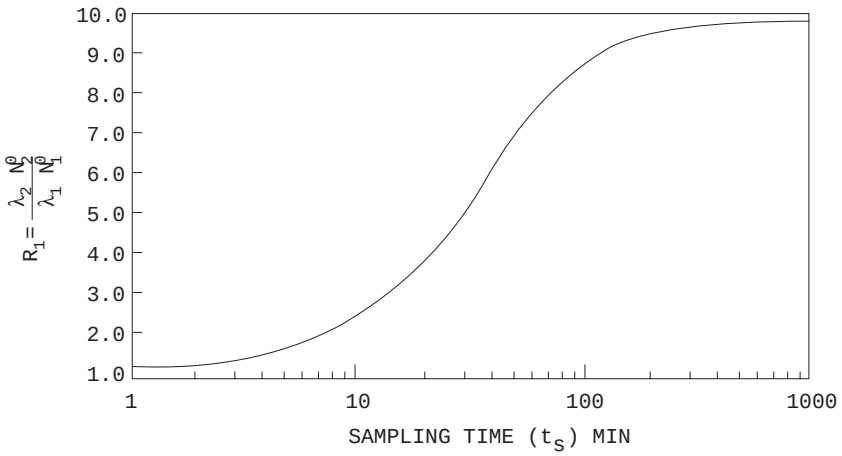

Figure 1. Dependence of the initial RaB-RaA ratio on sampling time

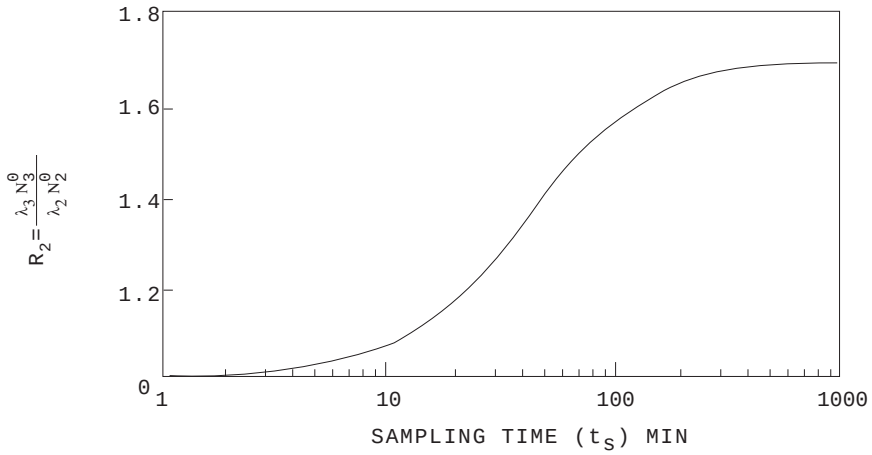

Figure 2. Dependence of the initial RaC-RaB ratio on sampling time

The observed variation of counting rate with time, or the total beta activity on the filter paper at a time t, after the end of filtration should be given primarily by the sum of the activities of RaB and RaC:

$$B_t = [A_2(t) + A_3(t)]\varepsilon f \tag{25}$$

where ε is the detection efficiency and f is the filtering efficiency.

Substituting the values of $A_2(t)$ and $A_3(t)$ from Eqns. (23) and (24) into Eqn.(25), we have

$$B_t = A_2^0 \varepsilon f \left[\frac{1}{R_1(t_s)} \cdot \frac{\lambda_2}{\lambda_2 - \lambda_1} \left(e^{-\lambda_1 t} - e^{-\lambda_2 t} \right) + e^{-\lambda_2 t} + \frac{\lambda_2 \lambda_3 \cdot e^{-\lambda_1 t}}{R_1(t_s)(\lambda_3 - \lambda_1)(\lambda_2 - \lambda_1)} - \frac{\lambda_2 \lambda_3}{R_1(t_s)} \cdot \frac{e^{-\lambda_2 t}}{(\lambda_3 - \lambda_2)(\lambda_2 - \lambda_1)} \right.$$
$$\left. \frac{\lambda_2 \lambda_3}{R_1(t_s)} \cdot \frac{e^{-\lambda_3 t}}{(\lambda_3 - \lambda_2)(\lambda_3 - \lambda_1)} + \frac{\lambda_3}{\lambda_3 - \lambda_2} \left(e^{-\lambda_2 t} - e^{-\lambda_3 t} \right) + R_2(t_s) \cdot e^{-\lambda_3 t} \right] \quad (26)$$

Knowing B_t experimentally, A_2^0 is calculated using Eqn.(26).
Since $A_2^0 = \lambda_2 N_2^0$ therefore, from Eqn.(7),

$$A_2^0 = \nu(n_1 + n_2)\left(1 - e^{-\lambda_2 t_s}\right) + \nu n_1 \frac{\lambda_2}{\lambda_2 - \lambda_1} \left(e^{-\lambda_2 t_s} - e^{-\lambda_1 t_s} \right) \quad (27)$$

Using the condition of secular equilibrium as before, Eqn. (27) reduces to:

$$A_0^0 = \nu n_2 \left[\left(1 + \frac{\lambda_2}{\lambda_1} \right) \left(1 - e^{-\lambda_2 t_s} \right) + \frac{\lambda_2^2}{\lambda_1(\lambda_2 - \lambda_1)} \left(e^{-\lambda_2 t_s} - e^{-\lambda_1 t_s} \right) \right]. \quad (28)$$

From Eqn.(28), n_2 is calculated, and the radon (^{222}Rn) concentration is finally obtained from the equation

$$C_{Rn} = \lambda_{Rn} n_{Rn} = \lambda_2 n_2 \quad (29)$$

For the decay products of thoron (^{220}Rn), the sum ThB+ThC was measured. However, because of the relatively small counting rate due to these activities and the limited statistical accuracy of he actual measurements, it is sufficient to approximate this contribution by a simple exponential of half-life 10.64h, corresponding to the decay of ThB. Eqn.(9) gives the activity of ThB on the filter at the instant of terminating the filtration:

$$A_4^0 = \nu n_4 \left[1 - e^{-\lambda_4 t_s} \right]$$

The beta activities of ThB+ThC at a time t after the end of filtration can be written as

$$B_t' = A_4^0 e^{-\lambda_4 t} = \nu n_4 \left[1 - e^{-\lambda_4 t_s} e^{-\lambda_4 t} \right]$$

Hence, the concentration of ^{212}Pb in atmospheric air is obtained from

$$C_{\mathrm{Th}} = \lambda_4 n_4 = \frac{B_t' \lambda_4 e^{\lambda_4 t}}{\nu \left[1 - e^{-\lambda_4 t_s} \right]}. \quad (30)$$

The relative magnitude of this contribution depends on a number of external circumstances; it can be experimentally estimated as described in the analysis of results.

4. Experimental

The beta counting technique was employed in measuring the filter activity. The apparatus used here consists of an air sampler capable of drawing air at $0.5\text{m}^3\cdot\text{min}^{-1}$ through a Millipore (type AA) filter paper and a shielded beta-sensitive end-window Gm tube (window diameter 3.5 cm) connected to a scalar. The air sampling equipment was calibrated using appropriate instrumentation. The collection efficiency of the filter paper was found to be almost 80%. This was determined by fixing a second filter at the outlet of the vacuum pump and measuring its activity immediately after the end of sampling. A metal grid was fixed at the inlet of the pump which served as a mechanical support for the filter. The active filter area was $\sim 8.5\text{cm}^2$.

While performing the experiment, air was drawn through a sheet of filter paper for 15-90 min. After the sampler was turned off, the filter paper was removed and was placed under the GM tube for the measurement of beta activity. The activity of the sample was then followed for more than eight hours, through a series of 5-min counts. In order to obtain the overall beta counting efficiency of the GM tube, a thin, disc-shaped standard ^{90} Sr+^{90}Y beta source (0.1μCi), whose beta-radiation is similar to the beta-radiation of RaB and RaC was used for calibration. The observed count rate was corrected for background and for the dead time of the GM tube.

The outdoor and indoor radon daughter levels were measured in different seasons at the University Campus of Rajshahi. The periods of measurement included the following months: winter= December-Jauary; spring=February- March, summer=April-June; monsoon=July-August; autumn=September-November. At least three measurements were made every month, and seasonal concentrations of radon and thoron were obtained from the average of monthly data. The nuclear physics laboratory was chosen as the reference room for indoor radon measurements. The ventialion condition of the room was kept identical throughout the period of investigation. In addition, indoor radon measurements were also carried out in several offices and laboratories in the physics building under different ventilation conditions.

In order to verify the accuracy and reliability of the measurement technique, reproducibility measurements were conducted in a special room of the physics building without any windows. The room was kept closed for at least 10d before each measurement was made. The room was contaminated with a weak radium source to further increase the radon activity. The time interval was sufficient to build-up of radon, thoron and their progeny in the room approximately to their initial levels. Seven measurements for beta-activity were performed successively using a 30-min sampling time.

5. Analysis of Results

Figures 3 and 4 show the results of 15-and 30-min sampling times, respectively. During the first three hours or so the radon daughter products would have decayed almost completely and the subsequent counts are due to the decay of thoron daughter products only. The curves clearly reveal the effects of the thoron decay products. The straight line fitted to the tail of these decay curves corresponds to the half-life 10.64 of ThB. When

the ordinates of this line are subtracted from the observed counting rates, the resulting rates should be due solely to RaB and RaC. The function $[A_2(t) + A_3(t)]\varepsilon f$ is shown normalized to these points. The calculated counting rates due to RaB and RaC alone are shown in Figure 4 only (broken lines). A good fit is observed between the data and calculated count rates.

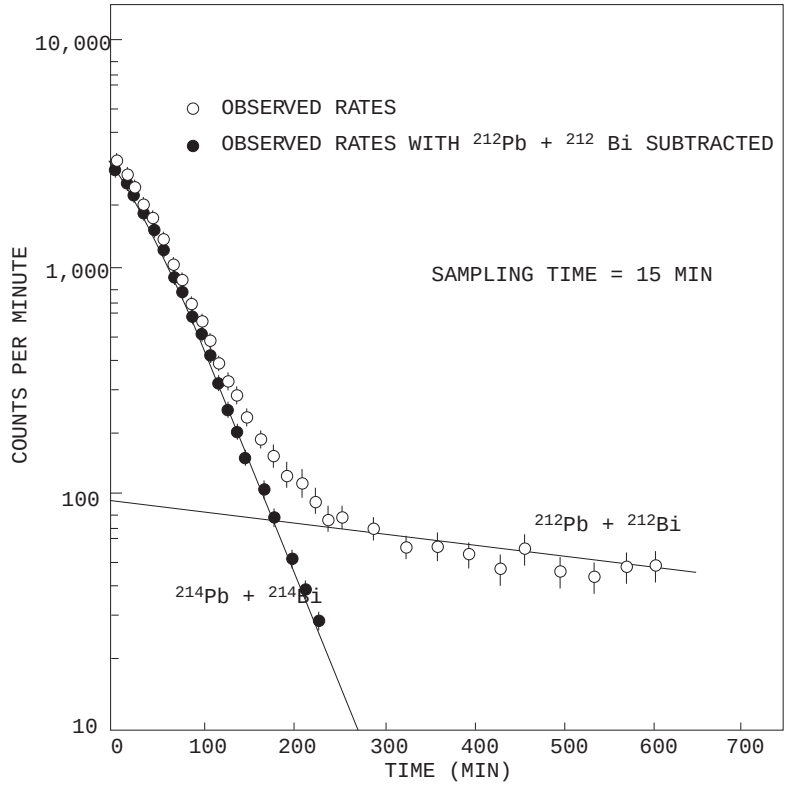

Figure 3. The variation of counting rate with time following a 15-min sampling period

The results of reproducibility measurements are provided in Table 1 shows reasonably good reproducibility of the estimation of the radon and thoron concentrations. Table 2 gives the seasoal mean concentrations of radon and thoron in the surface air (both outdoor and indoor) which are displayed in Figure 5 and 6. The outdoor radon concentration was found to be maximum during winter season while the maximum indoor radon concentration was observed during the monsoon months of July and August. The increased indoor radon activity during monsoon season is probably due to the filling of pores in the top soil layers with water, thus increasing the resistance for radon escape to the outdoor atmosphere and increasing the migration indoors.

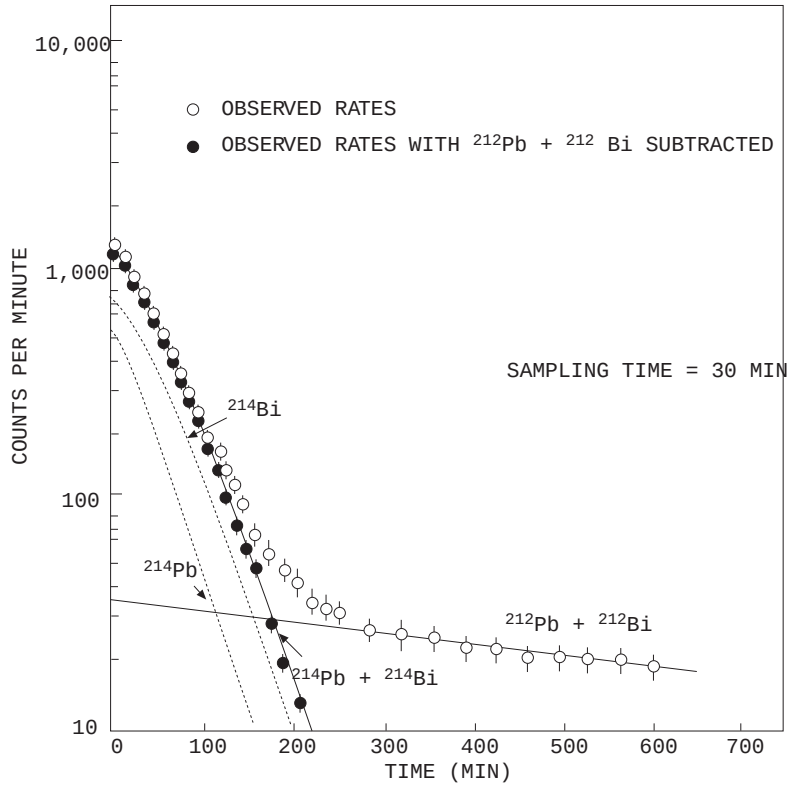

Figure 4. The variation of counting rate with time following a 30-min sampling period

During spring and summer at Rajshahi, the weather turns windy and both indoor and outdoor radon concentrations are found to be lower than those of the winter season. Particularly, the lowest indoor radon concentration during these seasons must be due to a large scale mixing of the indoor radon with outside air depending on the natural ventilation rate.

The results of radon and thoron concentration measurements in several offices and laboratories in the physics building yielded average radon and thoron concentrations of 31.78 Bq.m^{-3} and 2.27 Bq.m^{-3}, respectively. These values are far below the current upper limit of 150 Bq. m^{-3} recommended by the U.S. Environmental Protection Agency [4] for indoor radon-in-air concentration. The mean indoor concentration was normally found to be about three to six times more than that of outdoor radon activity.

Table 1. Reproducibility of radon and thoron measurements *

Assembled unit	Radon concentration ($Bq \cdot m^{-3}$)	Thoron concentration ($Bq \cdot m^{-3}$)
1	95.20	5.14
2	96.03	5.61
3	89.35	5.82
4	97.81	6.21
5	97.01	4.12
6	99.12	5.90
7	98.41	7.24
Mean	96.13	5.72

* Statistical error is ∼5%

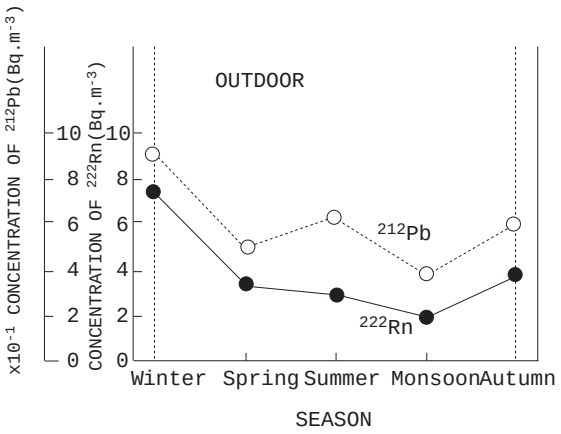

Figure 5. Seasonal variations of ^{222}Rn and ^{212}Pb concentrations in open atmosphere

6. Conclusions

Radon and thoron concentrations in air can be analyzed with reasonably good statistical accuracy by utilizing simple and inexpensive instrumentation. However, the actual values of radon and thoron concentrations are likely to be slightly larger than those of the presently estimated values, when equilibrium factor is properly taken into account. For a proper interpretation of the results obtained from the measurements of radon and thoron in air, it appears that one should study more extensively the seasonal variations of radon levels in this region and relate them with meteorological parameters.

The measurements also reveal that the proportion of ^{220}Rn: ^{222}Rn in environmental air of this geographic region of Bangladesh is quite low, and the net effect is that the thoron daughters may be neglected in most radon exposure studies.

Based on the above technique further nation-wide measurements are planned.

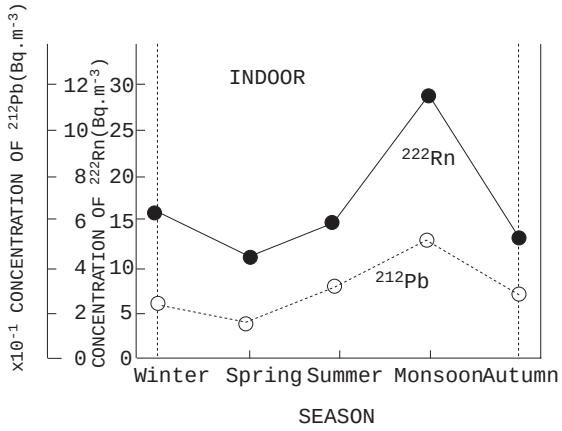

Figure 6. Seasonal variations of ^{222}Rn and ^{212}Pb concentrations inside the laboratory

Table 2. Seasonal variation of ^{222}Rn and ^{212}Pb concentrations* in the surface air

	Outdoor activity		Indoor activity	
Season	Concentration of ^{222}Rn $(Bq \cdot m^{-3})$	Concentration of ^{212}Pb $(Bq \cdot m^{-3})$	Concentration of ^{222}Rn $(Bq \cdot m^{-3})$	Concentration of ^{212}Pb $(Bq \cdot m^{-3})$
Winter	7.83	0.94	60.60	2.19
Spring	3.36	0.48	86.86	1.66
Summer	2.86	0.61	15.01	3.25
Monsoon	1.94	0.34	29.90	5.38
Autumn	3.37	0.55	13.77	3.20

* Statistical error is $\sim$5%

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