Estimation of thoron exhalation rate using LR-115 based passive technique.

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Abstract A simple technique based on solid state nuclear track detector (SSNTD) is proposed which is an off-shoot of the well-known sealed can technique. This technique delves on the significantly different diffusion lengths of radon and thoron in air. This can be made use to selectively record the radon or thoron contributions to the track density of the SSNTD. When the focus is on thoron, films are mounted at two different distances. The farther film records only radon and its progenies and the counts can be subtracted from the nearer film which is meant for thoron. The work deals with optimizing the distance of the near LR-115 films from the sample surface so as to record maximum counts. This depends on the thoron diffusion length and the alpha energy sensitivity of LR-115 film. The thoron exhalation rate measured by this technique is compared with the flow mode measurements using RAD7 radon monitor. The RAD7 measurement is found to underestimate the exhalation rate by 14-20% because of the decay losses

KEYWORDS:

1`. INTRODUCTION:

The soil and the rocks make a two way contribution towards the radiation exposure due to the natural background radiation- firstly, the external exposure due to the presence of primordial radionuclides namely ^{238}U, ^{232}Th and ^{40}K and secondly, the internal exposure due to radon isotopes and their daughter products which are being constantly exhaled from the earth's crust. Hence, the analysis of the radioactivity content of soil at any geographical location plays a major role in the background radiation dose estimation at that location. For India, in particular, the ^{232} Th and hence, ^{220}Rn estimations are very important since the average ^{232}Th activity concentration in soil is significantly greater than the world average. [1]
SSNTD-based sealed can technique is a well-known technique to estimate the radon exhalation rates from soil and construction materials.[2,3] When the bare SSNTD film is exposed to the sample, the tracks formed is due to the collective contribution from radon, thoron and their progenies. A simple way to selectively record the radon or thoron contributions is to make use of their significantly different diffusion lengths in air. While measuring radon, the thoron is treated as interference and this can be avoided by increasing the distance of the film from the sample surface. [4,5] On the other hand, when thoron is measured, films are mounted at two different distances.[6] The farther film records only radon and its progenies and the counts can be subtracted from the nearer film.
In this work, the thoron exhalation from a soil sample is estimated by the above technique of mounting LR-115 films at two different distances. The work deals with optimizing the distance of the LR-115 films from the sample surface so as to record maximum counts for thoron. This depends on the thoron diffusion length and the alpha energy sensitivity of LR-115 film. This method, based on diffusion, is more appropriate for thoron than the technique of forced air circulation with online monitor connected in loop especially when the pump flow rate is low. In such cases, owing to its short half life (55 sec) a sizeable fraction of thoron decays in the air loop before it reaches the monitor. Correction factors have to be applied which depends on the flow rate of the pump.

2. METHODS AND MATERIALS:

The methodology used to estimate the thoron exhalation rate from soil is the well –known SSNTD-based sealed can technique. The SSNTD film used in the present study is the LR-115 film (composition – Cellulose Nitrate).The soil is taken in a container and the films are fixed at a certain distance from the soil

surface. Another set of films are fixed at a greater distance to measure the radon background. After the container is sealed, ^{220}Rn attains steady state concentration within a few minutes and 216Po is constantly in equilibrium with ^{220}Rn. These two are alpha emitters. ^{212}Pb, which follows, is a beta emitter and has a half life of 10.6 h. It decays to ^{212}Bi which emits alpha with a branching ratio of 0.36.
Considering a situation in which an exposure time of 6 hours or less will be sufficient to get significant track density on the film, the contribution to the tracks is mainly from ^{220}Rn and ^{216}Po.This can be shown from calculations using the growth and decay equations given below.The integrated activitiy concentrations of ^{220}Rn and its alpha emitting daughters for the time periods 0-t is given by

$$A_s = \int_0^t A_1\,(t)dt + \int_0^t A_2\,(t)\,dt\; + 0.36\int_0^t A_4\,(t)\,dt \quad (1)$$

Where A_1, A_2, A_4 are activity concentrations of ^{220}Rn, ^{212}Po and ^{212}Bi respectively. After thoron attains equilibrium,

$$A_1 = A^{eq}$$

$$A_2(t) = \lambda_2\, A^{eq}\left\{1 + \frac{e^{-\lambda_2 t}}{(\lambda_1 - \lambda_2)}\right\}$$

$$A_4(t) = \lambda_2\,\lambda_3\lambda_4\, A^{eq}\left\{\frac{1}{\beta_1} + \frac{e^{-\lambda_2 t}}{\beta_2} + \frac{e^{-\lambda_3 t}}{\beta_3} + \frac{e^{-\lambda_4 t}}{\beta_4}\right\}$$

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

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

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

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

Where λ_1, λ_2, λ_3, λ_4 are the decay constants of ^{220}Rn, ^{216}Po, ^{212}Pb and ^{212}Bi respectively and A_{eq} is the equilibrium thoron concentration. The respective values are $\lambda_1 = 1.25 \times 10^{-2}\ s^{-1}$,$\lambda_2 = 4.78\ s^{-1}$,$\lambda_3 = 1.82 \times 10^{-5}\ s^{-1}$ and $\lambda_4 = 1.91 \times 10^{-4}\ s^{-1}$
Substituting the λ_1, λ_2, λ_3, λ_4 values and for t=5h (18000 s) , Eq.(1) works out to be

$$A_s = 18000\ A^{eq} + 18000\ A^{eq} + 6.43\ A^{eq} \quad (1)$$

Soil sample with high ^{232}Th content (Soil1- 33 KBq Kg^{-1}) was chosen for the study. The sample was taken in a PVC container of volume about 2l . Four LR-115 films were fixed on the lid of the container (which is at a distance of 14cm from the soil surface) to measure the collective contribution due to radon and its alpha emitting daughters. Another set of four LR-115 films were fixed near the soil surface at a certain height to measure both the radon, thoron and daughter contributions .The objective of the experiment was to find the optimum height at which the second set of films should be fixed to get the maximum track density. This height is based on the thoron diffusion length in air and the dependence of sensitivity of the LR-115 to alpha energy. LR-115 film registers countable tracks in the energy range 2-4 MeV[7,8].For a bare LR-115 film , the region of influence is bound by 1 to 3 cm [9].

After fixing the two sets of LR-115 films, the lid was closed and the container was sealed to prevent the escape of radon and thoron .The films were exposed for 5 hours. A series of measurements were done by fixing the LR-115 films at different heights from the soil surface– 1.5, 2.5, 3.0, 3.5, 4.5 and 5.5cm.After each exposure, the two sets of LR-115 films (8 Nos. -one set, at a constant height of 14cm from soil surface and another at varying heights mentioned above.) were etched with 2.5 N NaOH solution for 90 min at 60°C . The track density was obtained by the spark counting technique and the height at which the track density is maximum is found. Once this optimum height is found, it is fixed for the rest of the measurements with different soil samples. The total time integrated activity concentration (A_s) is calculated from the track density by using the species independent calibration factor for the bare mode exposure. [9] From equation (1), the equilibrium thoron concentration A_{eq} is found. The thoron surface exhalation rate was then estimated by the relation given below[].

$$E= A_{eq}\lambda\ V/ S \qquad (2)$$

λ is the decay constant (s^{-1}),V is the effective air volume(m^3) and S is the sample's surface area .
The same analysis is carried out for Soil -2 and Soil-3. Since, the thorium activity was relatively lower, the exposure period was increased to 23 hours. The average track densities are given in the table.

$$A_s= 82800\ A^{eq} +82800\ A^{eq} + 22.34\ A^{eq} \qquad (3)$$

The thoron surface exhalation rates of all the soil samples were also estimated by the closed loop technique using RAD7 online monitor to investigate the effect of thoron decay in the loop.

3. RESULTS AND DISCUSSION:

The average track density obtained for Soil-1 for LR 115 films fixed at the above mentioned heights from the soil surface and for the LR-115 films at height 14 cm are given in Table-1. It can be observed that the track density is maximum at 3 cm height above the soil surface. After subtracting the radon background, the tracks can be attributed to alpha counts due to the time integrated ^{220}Rn activity (A_1), ^{216}Po activity (A_2) and to a small extent due to ^{214}Bi activity (A_4) which is already discussed in the previous section.

Height (cm)	Average track density (tracks/cm^2)	Radon background track density (tracks/cm^2)	Net Track density(tracks/cm^2)
1.5	189	70	119
2.5	312	84	228
3.0	436	119	317
3.5	302	127	175
4.5	269	122	147
5.5	149	96	53

The average track density of LR-115 films placed at 3cm height above the soil samples Soil-2 and Soil-3 are given in Table -2 .

Soil	Average track density (tracks/cm^2)	Radon background track density (tracks/cm^2)	Background subtracted Track density(tracks/cm^2)
Soil-2	455	147	308
Soil-3	453	130	323

The equilibrium thoron concentration and hence the thoron exhalations are calculated from Equations (1) , (2) and (3).The values are given in Table-3 . The exhalation rates estimated by flow mode technique using RAD7 monitor are also given in Table-3. It can be seen that, without applying the necessary correction, RAD7 underestimates the exhalation rate. This can be attributed to the decay losses of thoron in the loop.

Soil	A^{eq} (Bq m^{-3})	Thoron surface exhalation rate (KBq m^{-2} h^{-1})	A^{eq} (RAD7) (Bq m^{-3})	Thoron surface exhalation rate (RAD7) KBq m^{-2} h^{-1})
Soil-1	38113 ± 2164	86 ± 5	11200 ± 611	74 ± 4
Soil-2	8050 ± 444	18 ± 1	2342 ± 86	14 ± 1
Soil-3	8442 ± 470	19 ± 1	2532 ± 83	15 ± 1

4. CONCLUSION:

SSNTD based technique has been developed to measure thoron concentration in the presence of radon circumventing the thoron decay loss problem of flow mode technique. The optimum height at which the LR-115 film is to be mounted is found to be 3cm. The RAD7 flow mode measurement is found to underestimate the exhalation rate by 14-20% .

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