Determination of Radon Exhalation Rates and Emanation Factor of Some Soil Samples Collected From Southern Seashore of Kerala, India

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Abstract

Weathering and erosion of rocks are the main cause for the formation of mineral deposits in beach sand and soil. There are quite a few identified monazite sand-bearing placer deposits causing environmental high radiation area along its long coastline of Kerala and Tamil Nadu in India. The extent and effects of the radioactivity in the regions like Chavara and Neendakara in Kollam, Kerala is well known. Following a pilot study along the southern west coast of Kerala state, India we found certain regions with higher levels of natural radioactivity. Later made a detailed investigation of activity concentration of $^{226}$Ra, radon exhalation rates and radon emanating factor of those locations namely Kovalam and Varkala. The results indicates that most of the soil sample shows higher specific activity of $^{226}$Ra and there is a linear relation to $^{222}$Rn mass exhalation rate. But radon emanation factor is not so linear to the specific activity of Radium in sample analysed.

Key words: Natural radioactivity, specific activity, mass exhalation rates, emanation factor

Introduction

The ionizing radiation exposure of human beings from natural sources is a continuing and inescapable feature of life on earth¹. There are two main contributors to natural radiation exposures: high energy cosmic ray particles incident on the earth’s atmosphere and radioactive nuclides that originated in the earth’s crust. Radon is a radioactive noble gas that does not chemically react with other element. But it can change the physical
properties of surrounding medium. Radon is produced due to the radioactive decay of radium in earth crust. It’s concentration in the ground depends on the radium content of the soil and the emanation power of soils and rocks\(^2,3\). Radium is a decay product of uranium in the naturally occurring uranium series. When radium decays in soil, the resulting atoms of radon isotopes first escape from the mineral to air-filled pores. The rate at which radon escapes from soil into the surrounding air is known as radon exhalation rate of the soil. This may be measured by either per unit area or per unit mass of the soil.

About 20% of the natural radiation dose is due to external radiation from terrestrial radioactivity\(^4,5\). When radon gas is inhaled, densely ionizing alpha particles emitted by deposited short-lived decay products of radon \(^{218}\)Po and \(^{214}\)Po can interact with biological tissue in the lungs leading to damage and causes lung cancer\(^6\). Due to the hazardous effect of radon exhalation on human health, it has great significance to conduct measurements of radium content in the soil. Higher values of \(^{226}\)Ra in soil have greater contribution in the enhancement of environmental radon\(^7\).

In the present study investigations have been carried out to measure the radium content and radon mass exhalation rates in soil samples collected from some areas of Varkala and Kovalam in Kerala, India using a smart radon monitor. The importance of this work, is that, it represents the first study on measuring radon mass exhalation rates and radium content in Varkala and Kovalam in Kerala.

Incidentally these two locations are famous tourist destinations with thick population. These shallow beaches attract tourists from India and abroad. Kovalam has three beaches separated by rocky outcroppings in its 17 km coastline and the three together form the crescent of the Kovalam beach. Varkala is a coastal town located 50 km north-west of Thiruvananthapuram (Trivandrum). In addition to the floating population (tourists) average population in these regions are about 60,000 each.

**Materials and Methods**

*Collection and preparation of sample:*

Each sample was taken maintaining a distance of about 200m from each other. About 0.75 - 1.00 kg of sample was collected from each location. These samples were first weighed, and then dried at 110\(^\circ\)C in a dry air oven for 24 hours. The homogenized and sieved samples were transferred to cylindrical plastic container of 7 cm height and 5.5 cm in diameter. All the sample containers were sealed hermetically and were stored for eight weeks to ensure the secular equilibrium between \(^{226}\)Ra (of the \(^{238}\)U) with their radioactive progenies.

*Gamma spectroscopy:*

The gamma spectrometer is a 5\(\times\)4" NaI(Tl) detector. Samples were analysed for \(^{238}\)U (\(^{226}\)Ra) by gamma spectroscopy. The counting period for samples and background was set for 10,000 s. The activity of \(^{238}\)U was evaluated from 1764 keV gamma of \(^{214}\)Bi.

Preliminary observations of the parameters like background efficiency and MDL for the Gamma ray spectrometer were done periodically. The sample kept for attaining secular equilibrium were analysed using the Gamma ray spectrometer. The samples in the standard bottle were placed into the detector for counting and left for recording spectrum for 10,000s. By selecting the respective peaks for the isotopes, the region of interest (ROI) are noted and the corresponding gross counts are calculated. The net activity was determined by deducting background radiation from the gross count.

*Radon monitor-Smart RnDuo:*

The SMART RnDuo is a portable continuous activity monitor for radon (\(^{222}\)Rn), thoron (\(^{220}\)Rn) and gross alpha in the sampled air. The detection principle is based on detection of alpha particles, emitted from
sampled radon/thoron and its decay products formed inside the detector volume, by scintillation with ZnS:Ag. The counts obtained for each interval are converted to radon/thoron/alpha activity concentration using a smart algorithm implemented in the micro-controller. The advanced algorithm has been developed by Bhabha Atomic Research Centre, Mumbai based on radioactive decay and growth laws. This algorithm accounts for the counts obtained from the decay products formed during the previous intervals.

The radon emission potential from soil sample is governed by radon mass exhalation rate (Jm). Jm can be estimated by performing measurements using smart radon monitor- SMART Rn Duo of the sample in a closed accumulation chamber (also called mass exhalation chamber) and monitoring the build-up of radon concentration in the chamber at regular time intervals. Typically about 350 – 500 g of soil or any powd sample may be enclosed in a leak tight metallic chamber coupled to the SMART Rn Duo. Measurement cycle should be 1 hour. Sampling should be done preferably by diffusion mode. Hence Detector probe should be connected to the mass exhalation chamber directly. The radon concentration C(t) at time t since closing the chamber builds up according to the formula.

\[ C(t) = \frac{JmM}{V \lambda_e} \left[ 1 - e^{-\lambda t} \right] + C_0 e^{-\lambda t} \]

Where

- \( C_0 \) is the \(^{222}\text{Rn} \) concentration (Bq m\(^{-3}\)) present in the chamber volume at t = 0
- M is the total mass of the dry sample (kg)
- V is the effective volume (residual air volume of exhalation chamber + Porous Volume of Sample + internal volume of SMART Rn Duo (m\(^3\))
- \( \lambda_e \) is the effective decay constant for \(^{222}\text{Rn} \), which is sum of the leak rate (if existing) and the radioactive decay constant of \(^{222}\text{Rn} \) (h\(^{-1}\))
- t is the measurement time (h)

In linear approximation (i.e. by restricting measurement time within 12 hrs), Eq(1) can be converted to

\[ C(t) = \frac{JmM}{V} t + C \]

Upon least square fitting of the data to the above equation one may obtain Jm from the fitted slope value with the information of the mass M of the sample and residual air volume, V of the set up.

**Emanation factor**:

Emanation factor is defined as the fraction of radon atoms generated that escape the solid phase in which they are formed and become free to migrate through the bulk medium.

\[ \text{Emanation factor } f = \frac{Jm}{C_{Ra} \lambda} \]

C\(_{Ra}\) Radium concentration in Bq/kg

**Result and Discussion**

Activity concentrations of and \(^{226}\text{Ra}\), Mass exhalation rate of \(^{222}\text{Rn} \) and emanation factor in the soil samples from Kovalam are shown in Table 1. The values of activity concentration of radium are given in Bq/kg of dry weight. Below Detectable Value (BDL) of \(^{226}\text{Ra} \) is 4.7 Bq/kg.
Table 1: Values of radium concentration, Radon mass exhalation rates and emanation factor of soil samples from Kovalam

| Location | C_{Ra} (Bq/kg) | Mass exhalation rate (mBq/kg/h) | Emanation factor |
|----------|----------------|---------------------------------|------------------|
| Kovalam 1 | 96.7±3.6       | 6.80                            | 9.38             |
| 2        | 62.5±5.7       | 4.60                            | 9.81             |
| 3        | 35.2±1.8       | 2.68                            | 10.16            |
| 4        | 41.8±2.3       | 2.95                            | 9.41             |
| 5        | 53.2±2         | 3.70                            | 9.27             |
| 6        | 41.0±1         | 3.20                            | 10.41            |
| 7        | 53.4±8.3       | 3.65                            | 9.11             |
| 8        | 42.8±8.4       | 3.40                            | 10.59            |
| 9        | 65.6±8.9       | 4.50                            | 9.15             |
| 10       | 41.7±4         | 3.20                            | 10.23            |
| 11       | 91.3±8         | 6.43                            | 9.39             |
| 12       | 85.5±2.9       | 6.07                            | 9.47             |
| 13       | 34.3±6.2       | 2.57                            | 10.00            |
| 14       | 98.8±6.8       | 6.98                            | 9.41             |
| 15       | 34.2±4.4       | 2.61                            | 10.16            |
| Mean     | 58.53          | 4.22                            | 9.73             |
| Max      | 98.8           | 6.80                            | 10.59            |
| Min      | 34.2           | 2.57                            | 9.11             |
| STDEV    | 23.67          | 1.59                            | 0.49             |

In Kovalam the range and mean values (in brackets) of the activities for $^{226}$Ra are 34.2 – 98.8 (58.53) Bq/kg, Mass exhalation rate varies 2.57-6.8 (4.22) (mBq/kg/h), Emanation factor ranged between 9.11-10.59 (9.73) respectively.

Figure 1: Mass exhalation rate Vs. Radium concentration of soil samples from Kovalam

![Graph showing linear relationship between radium concentration and mass exhalation rate](image)

Figure 1 represent the radium concentration values are found to be linearly dependent with radon mass exhalation rates.
Table 2: Values of radium concentration, Radon mass exhalation rates and emanation factor of soil samples from Varkala

| Location | C_{Ra} (Bq/kg) | Mass exhalation rate (mBq/kg/h) | Emanation factor |
|----------|----------------|---------------------------------|-----------------|
| 1        | 191.02±9.6     | 5.30                            | 3.70            |
| 2        | 200.25±10      | 5.15                            | 3.43            |
| 3        | 104.31±5.2     | 3.93                            | 5.02            |
| 4        | 142.27±7.1     | 4.41                            | 4.14            |
| 5        | 197.28±9.9     | 5.10                            | 3.45            |
| 6        | 192.02±6       | 5.04                            | 3.50            |
| 7        | 58.46±2.9      | 3.14                            | 7.17            |
| 8        | 311.96±15.6    | 6.81                            | 2.91            |
| 9        | 264±13.2       | 6.45                            | 3.26            |
| 10       | 187.38±9.4     | 5.04                            | 3.59            |
| 11       | 102.82±5.1     | 3.56                            | 4.61            |
| 12       | 112.52±5.6     | 4.07                            | 4.82            |
| 13       | 20.52±1        | 2.91                            | 18.89           |
| 14       | 274.10±13.7    | 6.58                            | 3.20            |
| 15       | 148.01±7.4     | 4.16                            | 3.75            |
| Mean     | 167.13         | 4.78                            | 5.03            |
| Max      | 311.96         | 6.80                            | 18.89           |
| Min      | 20.52          | 2.91                            | 2.91            |
| STDDEV   | 80.52          | 1.20                            | 3.98            |

Table 2 shows in Varkala, the range and mean values (in brackets) of the activities for $^{226}$Ra are 20.52–311.96 (167.13) Bq/kg, Mass exhalation rate varies 2.91-6.8(4.78) (mBq/kg/h), Emanation factor ranged between 2.91-18.89 (5.03) respectively.

Figure 2: Mass exhalation rate Vs Radium concentration of soil samples from Varkala.

Figure 2 reflects the radium concentration values and radon mass exhalation rates of Varkala are found to be linear.
The uneven distribution of radioactivity concentration for different locations has been observed. The variation among radionuclides in beach soil may be due to the continuous wave action, results in the deposition of heavy minerals along the sea shore. The emanation factor of soil grains is variable because soil is generally composed of many kinds of mineral grains of different rock origins.

**Conclusion**

The activity concentration is unevenly distributed across the study area. In Varkala and Kovalam more than 90% of samples showed higher specific activities for $^{226}\text{Ra}$. $^{222}\text{Rn}$ is the immediate daughter element of $^{226}\text{Ra}$, results showed there is a linear relation to $^{222}\text{Rn}$ mass exhalation rates to that of $^{226}\text{Ra}$ concentration values in samples analysed. Results reflects that proper precautions are necessary for masking the $^{222}\text{Rn}$ emission inside a dwelling when soil is used for construction of houses.

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