Journal of Radioanalytical and Nuclear Chemistry (2020) 324:949–961
https://doi.org/10.1007/s10967-020-07133-5

Dynamics of radionuclides activity, radon exhalation rate of soil and assessment of radiological parameters in the coastal regions of Kerala, India

K. Nadira Mahamood[1] · P. V. Divya[1] · V. Vineethkumar[1] · V. Prakash[1]

Received: 30 July 2019 / Published online: 15 April 2020

Abstract
The activity concentration of natural radionucides such as ^{40}K, ^{226}Ra and ^{232}Th and radon surface and mass exhalation rates of the soil were assessed and the related radiometric parameters have been estimated from the activities of the samples from beaches of coastal Kerala including some high background radiation areas (HBRAs). The activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th were estimated using NaI (Tl) gamma ray spectrometry and the radon mass and surface exhalation rate has been estimated using the 'Can technique' with LR-115 solid state nuclear track detectors. The estimated values indicate that the radionuclide concentrations in the study area were within the recommended limit except for Kollam district when compared with world average values. Radon exhalation rate found to vary according to grain size of the soil. Dose parameters have been evaluated in order to verify the radiological protection of the general public. The results of the present systematic investigation are presented and discussed in detail in the manuscript.

Keywords Radionuclide · Soil grain size · Exhalation rate · NaI (Tl) detector · Can technique · Annual effective dose

Introduction

Human being is continuously exposed to ionizing radiations from natural sources and is an inescapable feature of life on earth. The two main contributors to natural radiation sources are high-energy cosmic ray particles incident on the earth's atmosphere and radioactive nuclides that are originated in the earth's crust, which are present everywhere in the environment with half-lives comparable to the age of earth and their decay products. External exposure to human is mainly due to the presence of terrestrial radionuclides such as ^{238}U and ^{232}Th series and singly occurring ^{40}K in rocks and soils [1]. Radon is one of the significant sources of natural radiation which is a naturally occurring radioactive isotope of uranium series with half life of 3.82 days. The decay products of radon include ^{218}Po, ^{214}Po, ^{214}Pb and ^{214}Bi, which can be attached to aerosol, may lead to greater biological effects through inhalation and lead to lung cancer on continuous exposure for a long time. Therefore, studies on activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th radionuclides, radon exhalation rate and estimation of associated dose parameters in soil assumes great significance [2].

Radon is present in indoor and outdoor air since its parent nuclide ^{226}Ra occurs in trace amount throughout the earth. When ^{226}Ra decays, the fraction of the liberated ^{222}Rn in the soil grain escapes in to the air/water filled pore space and migrates through the pore space by the process known as emanation. Emanation fraction or emanation coefficient is the ratio of fraction of radon emanated to the total amount of radon generated in the soil grain [3]. The emanation happened mainly by two processes; diffusion and recoil in which diffusion do not contribute much to emanation process due to extremely short diffusion length (10^{-13} to 10^{-32}) of radon in the soil [4]. So the emanation fraction mainly comes from the recoil process, for which the range of recoil distance of radon in the common minerals is 20–70 nm, in water is 100 nm and in air is 63 nm. The emanation of radon continues until it decays or releases in to the air and this process is known as exhalation [3]. The radon exhalation rate is the rate of radon escapes from the soil surface to the atmospheric air and is measured either per unit area or per unit mass of the soil. The two mechanisms are involved in the exhalation

✉ V. Prakash
prakashamv@gmail.com

[1] Department of Studies & Research in Physics, Payyanur College, Kannur, Kerala 670 327, India

process; emanation and transport. These two factors are affected by uranium concentration, radium concentration, soil texture, permeability and grain size [3, 4]. Earlier studies on the effect of grain size on radon concentration reveal that radon exhalation rate from the soil depend on the grain size of the sample [4].

The contribution of radionuclides to external gamma dose rate at a location can be determined only by monitoring the area since the actual radiation level caused by radionuclide varies from place to place ie. The dose rate depends up on the geological location [5]. The high radiation levels have been observed in many countries due the enrichment of radioactive minerals in soil and sand leading to the formation of what are known as high background radiation areas [1]. India is one of the few countries in the world having high natural background radiation levels. The reported high background radiation areas in India are the southwest coastal region of Kerala, Ullal in Karnataka and Manavalakuruchi in Tamilnadu [6]. It is important to determine the background levels of radionuclides in soils of coastal regions of Kerala, in order to compare and provide more clear idea about the distribution of radionuclides in the normal and high background radiation areas. An attempt has been made to investigate the radionuclide enrichment and exhalation rates using gamma ray spectrometry and Can technique respectively in the soil samples of Kerala beaches including HBRAs. Radium equivalent activity, absorbed dose rate, hazard parameters, annual effective doses etc. were also estimated and feasible conclusions have been drawn.

Materials and methods

Sample collection and preparation

Soil samples have been collected from different sampling locations which were identified based on the background radiation intensity using Micro-R Survey Meter UR-705 (range 0–10,000 μR h^{-1}). Figure 1 shows the location map of the sampling stations along the coastal region of Kerala. The population density in the study area is 859 per square kilometre as per the census 2011. The soil samples have been collected and prepared following the standard procedure [7]. About 27 soil samples were collected from 9 locations of different beaches at 1 km distance from the sea line. ie, three samples weighing about 1 kg have been collected from each location a few meters apart from each other based on the similarity in background radiation intensity and the samples from the same locations were mixed for homogeneity and were crushed and dried at approximately 110 °C for 24 h in order to remove the complete moisture content. The dried samples were sieved using British standard mesh of 250 μm grain size. About 250 g of each sample was filled in plastic containers and were sealed with adhesive taps to make them air tight. The containers were stored for a period of one month in order to attain secular equilibrium between radium, thorium and their daughter products before doing gamma spectrometric analysis.

Activity determination

The activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in the soil samples were estimated using a high efficiency NaI (Tl) detector. The well-type receptacle NaI (Tl) scintillation assembly provides maximum absorption of radiation from a sample by approximating 4pi geometry. The scintillation detector of size 5 cm × 5 cm was used along with Multichannel Analyzer. The spectrometer was calibrated using RG-U, RG-Th and RG-K, as standard sources. These are standard sources for Uranium, Thorium and Potassium, procured from International Atomic Energy Agency, Vienna. The full-width at half maximum (FWHM) was 60.78 keV with resolution 8.46%, for the ^{137}Cs (661 keV) peak. The detector was shielded with lead blocks of size 6" × 3" × 1.75", to reduce background counts due to terrestrial gamma ray radiations. The samples were analyzed with an acquisition time of 40,000 s, using GSPEC software to obtain the gamma-ray spectrum with good statistics. The gamma peaks corresponding to 1460.8 keV (^{40}K), 1764 keV (^{214}Bi) and 2614 keV (^{208}Tl) were considered for evaluating the activity concentration of ^{40}K, ^{226}Ra and ^{232}Th respectively [8]. A sample spectrum obtained using NaI (Tl) gamma-ray spectrometer is shown in Fig. 2.

The simultaneous equation method was used to determine the activity concentration of ^{40}K, ^{226}Ra and ^{232}Th. The simultaneous equations [9] are given below.

$$C_1 = T_{2.61} \tag{1}$$

$$C_2 = T_{1.76} - F_1 C_1 \tag{2}$$

$$C_3 = T_{1.46} - F_2 C_3 - F_3 C_2 \tag{3}$$

where C_1, C_2 and C_3 are compton corrected net counts under the photopeaks of the characteristic gamma rays of ^{232}Th, ^{226}Ra and ^{40}K respectively; and $T_{2.61}$, $T_{1.76}$ and $T_{1.46}$ are the total integral counts under photopeaks of ^{208}Tl, ^{214}Bi and ^{40}K respectively. $F_1 = 0.1$, $F_2 = 0.33$ and $F_3 = 1.34$ are Compton contribution factors. These factors are determined by recording the spectra of standard sources of each radionuclide.

The activity of radionuclides was estimated using the equation [10]

$$A = (C \pm SD) \times \frac{100}{E} \times \frac{100}{a} \times \frac{1000}{W} \text{Bq kg}^{-1} \tag{4}$$

where $_C$ is the Compton corrected net counts under the photopeak, SD is the standard deviation, E is the photopeak

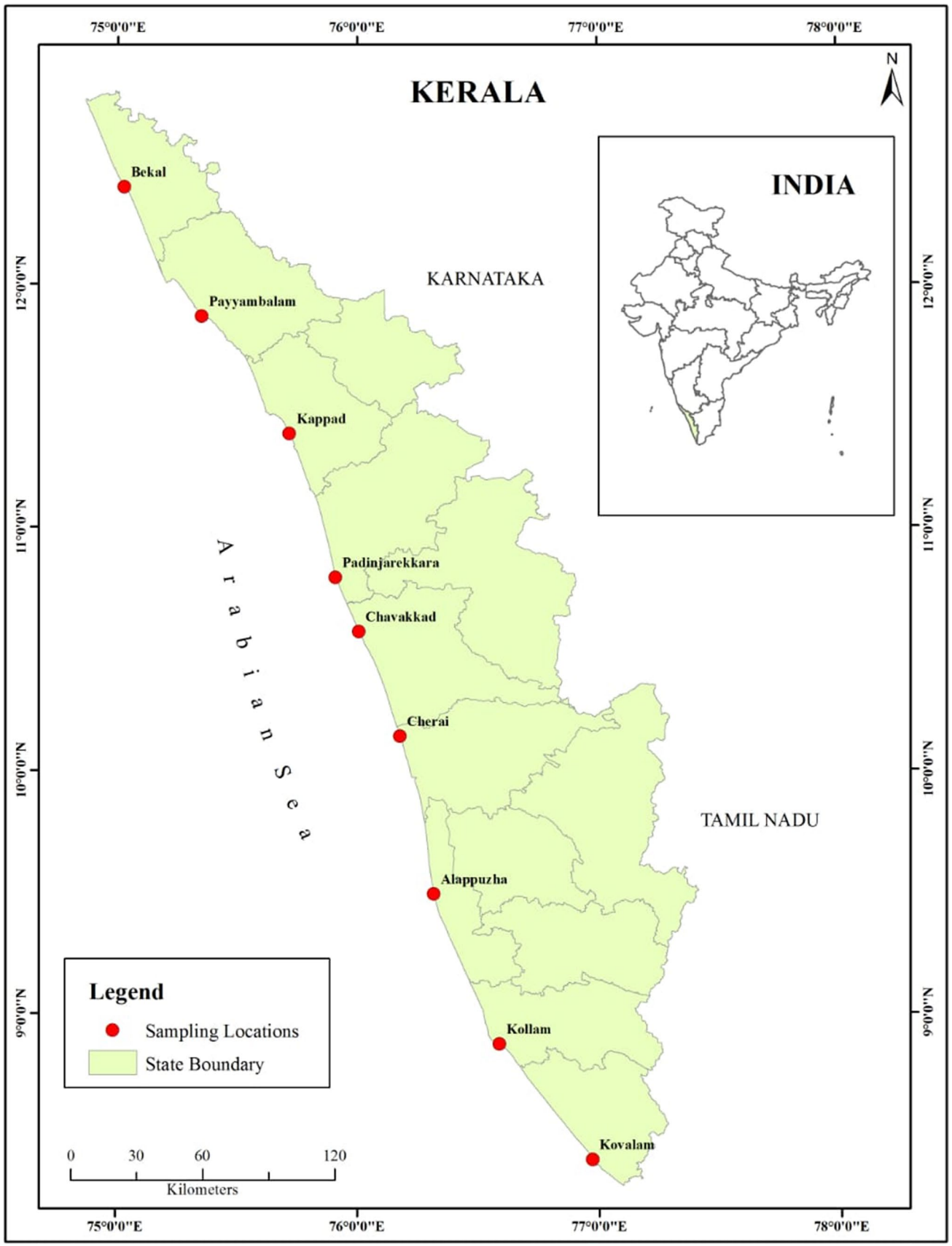

Fig. 1 Location map of the sampling stations

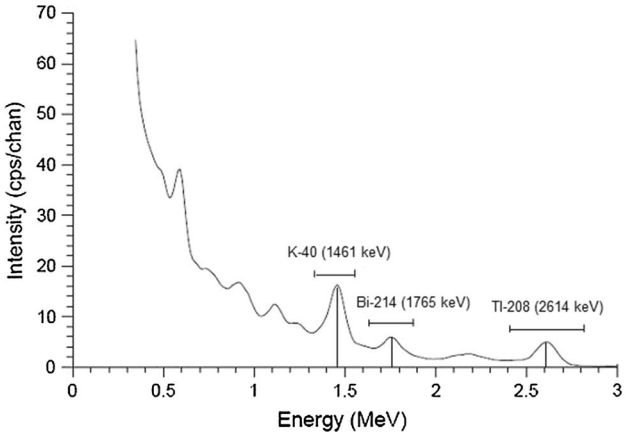

Fig. 2 Gamma-ray spectrum from NaI (Tl) detector

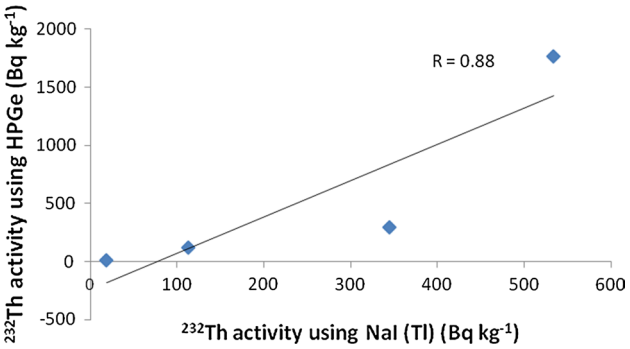

Fig. 3 Correlation between the measurements of NaI (Tl) and HPGe

efficiency (%) of the detector, a is the abundance of the characteristic gamma ray [7, 11] and W is the weight of the sample in grams. The minimum detectable activity at 95% confidence level for 40,000 s counting time and 500 g sample weight was found to be 2.6 Bq kg^{-1} for ^{40}K, 0.9 Bq kg^{-1} for ^{226}Ra and 0.5 Bq kg^{-1} for ^{232}Th.

Inter-comparison measurements have been carried out in order to validate the results. The activity concentration obtained using NaI (Tl) have been correlated with the values obtained by the HPGe detector (Canberra) having efficiency 50%. The results of the activitiy concentrations of ^{40}K, ^{226}Ra and ^{232}Th were found to be in good agreement. A sample result of inter-comparison measurements (correlation coefficient is 0.88) for ^{232}Th in some of the soil samples is shown in Fig. 3. Consistency in correlation between the radioactivity concentration of ^{232}Th measured by NaI (Tl) and HPGe is not expected as the analysis have been made during different times and the efficiency of the detectors were totally different. The samples were from different locations including HBRAs and the inconsistency was shown by some samples having higher ^{232}Th activity concentration, may be attributed to decrease in counting efficiency due to increase in gamma ray energy.

Radiological parameters

Radium equivalent activity

The Ra_{eq} is a single index used to compare the activity of materials containing different amount of ^{40}K, ^{226}Ra and ^{232}Th which can explain uniformity in exposure to radiation. The index in Bq kg^{-1} is calculated following the relation

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (5)$$

where A_{Ra} is activity of radium in Bq kg^{-1}, A_{Th} is activity of thorium in Bq kg^{-1} and A_K is activity of ^{40}K in Bq kg^{-1}. The relation is based on the assumption that 1 Bq kg^{-1} of ^{226}Ra, 0.7 Bq kg^{-1} of ^{232}Th and 13 Bq kg^{-1} of ^{40}K produce the same gamma ray dose [11]

Absorbed dose rate

Absorbed dose is the quantity to measure energy deposited in matter by ionizing radiation per unit mass. Absorbed dose is used to calculate dose uptake in living tissue in both radiation protection and radiology. Absorbed gamma dose rate (nGy h^{-1}) in air above the ground surface due to the activity of ^{226}Ra, ^{232}Th, and ^{40}K is calculated using the following formula [12]

$$D = 0.462\ A_{Ra} + 0.604\ A_{Th} + 0.042\ A_K \quad (6)$$

where D is the absorbed dose rate and A_{Ra}, A_{Th}, and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K expressed in Bq kg^{-1}.

Annual effective dose equivalent

In order to estimate the annual effective dose equivalent, it is necessary to use the conversion coefficient from the absorbed dose in air to the effective dose (0.7 Sv Gy^{-1}) and the outdoor occupancy factor (0.2) proposed by UNSCEAR [12]. Since a typical resident in a location would spend about 80% a day in indoor and 20% in outdoor, the outdoor annual effective dose in mSv y^{-1} is determined by the following relation

Annual effective dose equivalent,

$$AEDE = D \times 8766 \times 0.2 \times 0.7 \times 10^{-6} \quad (7)$$

where D is the absorbed dose rate in indoor air, 8766 h is the average annual time duration for exposure to radiation.

External hazard index (H_{ex})

The external hazard index indicates the radiological suitability of a material. The external hazard index due to the emitted gamma rays, from the soil samples was calculated following the relation [13]

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (8)$$

Internal hazard index (H_{in})

Radon and its progeny, being a radiolabel hazard to humanity, internal exposure to radon and its progeny is quanitaively expressed in terms of internal hazard index. This criterion is applied whether ^{226}Ra plays the dual danger role in terms of gamma radiation and indoor radon [14]. It can be calculated by the relation [12]

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (9)$$

Radon exhalation rate

Assessment of radium activity concentration and radon exhalation rate in soil samples have been carried out using the Closed Can technique [4, 15–19]. This method is considered as the most efficient method for the estimation of time integrated radon exhalation rate [18]. In such measurements, the accuracy depends upon the material used and its amount as well as on the geometry and dimension of the Can [18, 20]. About 27 soil samples were collected from different beaches at 1 km distance from the sea line of coastal Kerala. The samples were crushed and dried at approximately 110 °C for 24 h. The dried samples were sieved using British standard mesh, fractionated in the sizes 1000–500, 500–250, 250–125 and < 125 µm and separately collected. For each grain size, a 100 g sample was placed in a cylindrical can of 10 cm height and 8 cm diameter. Schematic diagram of closed can technique is shown in Fig. 4 [3]. In each can, the α-sensitive LR-115 type-II solid state nuclear track detector of size 3 × 3 cm was fixed on the inner upper surface of the can [21–23]. The sensitive face of the detector is freely exposed to the sample material in the can so that it can record the tracks due to alpha particles resulting from the decay of radon following the decay of radium [4]. The LR-115 films were kept at approximately 7 cm distance from the sample in order to avoid the track contribution due to thoron as per the protocol [24]. The cans were sealed and stored for a period of 90 days. After the exposure, the detectors were removed and subjected to a chemical etching

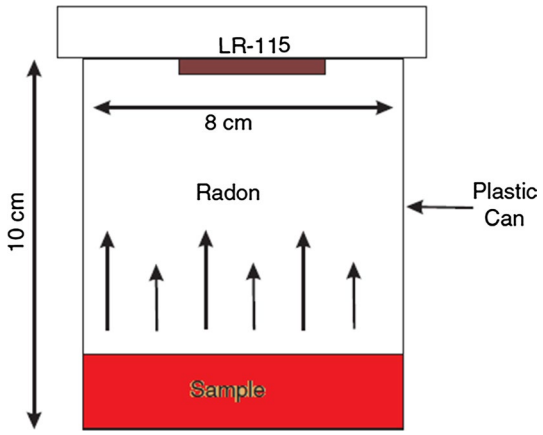

Fig. 4 Experimental set up of closed can technique

process in 2.5 N NaOH solution at 60 °C for 90 min in a constant temperature water bath for the revelation of tracks. The etched detectors were thoroughly washed and the red sensitive layer was stripped for spark counting. The dried detectors were then used for alpha counting using the spark counter (PSI-SC1) procured from Polltech instruments. The detectors were first pre-sparked at 900 V and then tracks were counted at an operating voltage of 500 V two or three times.

Effective radium content

The accumulated radon concentration was obtained from the track density (track cm^{-2}), using the calibration factor of 0.056 tr. cm^{-2} d^{-1} Bq^{-1} m^3 obtained from an earlier calibration experiment [19, 25]. Once the radioactive equilibrium is established in sealed can, the alpha particles released from the radon can be used to determine the radium content (Bq kg^{-1}) of soil samples [3, 20]. The radium concentration in soil samples, C_{Ra} was calculated following the relation (10) [3, 26]

$$C_{Ra} = \frac{\rho hA}{KT_eM} \quad (10)$$

where ρ is the track density, h (m) is the distance between soil sample and the detector, K is the calibration factor, A (m^2) is the area of cross section of the can and M (kg) refers to the mass of soil sample. In Eq. (10), T_e (days) denotes the effective exposure time. The detectors in cans record the ^{222}Rn concentration starting from zero activity to equilibrium activity. Therefore, it is needed to determine the effective exposure time.[3, 27].

The effective exposure time (T_e) which is related to the actual exposure time t and decay constant λ for ^{222}Rn, was calculated using the Eq. (11)

$$T_e = t - \frac{1}{\lambda}(1 - e^{-\lambda t}) \quad (11)$$

The value of λ was calculated using the relation (12)

$$\lambda = \frac{0.693}{t_{1/2}} \quad (12)$$

where $t_{1/2}$ is the radon half life (3.82 days) [4]

Radon exhalation rate

The radon exhalation rate in terms of area and mass is calculated from the following relations (13) and (14)

$$E_A = \frac{CV\lambda}{AT_e} \quad (13)$$

$$E_M = \frac{CV\lambda}{MT_e} \quad (14)$$

where E_A is the radon surface exhalation rate (Bq m^{-2} h^{-1}) and E_M is the radon mass exhalation rate (Bq kg^{-1} h^{-1}), C is the integrated radon exposure (Bq m^{-3} h), V is the effective volume of can (m^3), λ is the decay constant for radon (h^{-1}), and A is the area covered by the can (m^2) and M is the mass of the sample taken in the can (kg) [28]

Indoor annual effective dose equivalent from exhalation rate

The radon concentration in the soil samples contributing to indoor air due to surface exhalation can be calculated following the relation (15) [29, 30]

$$C_{Rn} = \frac{E_A S}{V\lambda_v} \quad (15)$$

where C_{Rn} is radon concentration (Bq m^{-3}), E_A is radon exhalation rate (Bq m^{-2} h^{-1}); S is the internal surface area of the room (m^2), V is the room volume (m^3), and λ_v is the air exchange rate (h^{-1}). The maximum radon concentration from the building material was assessed by assuming the room as a cavity with S/V = 2.0 m^{-1} and air exchange rate of 0.5 h^{-1} [31, 32]. The annual exposure to potential alpha energy, Ep (WLM y^{-1}) is then related to the average radon concentration C_{Rn} following the expression [29–32]

$$E_P = \frac{C_{Rn} \times 8760 \times n \times F}{170 \times 3700} \quad (16)$$

where n is the fraction of time spent indoors taken as 0.8, 8760 is the number of hours per year, F is the equilibrium factor for radon taken as 0.4 as recommended by the UNSCEAR [12], since radon progeny equilibrium is the most important factor while calculating the dose based on the measurement of radon concentration. It may be assumed the value in the range 0 < F < 1 [29]. The equilibrium factor F quantifies the state of equilibrium between radon and its daughters. A working level month can be defined as the amount of radon short lived decay products in equilibrium with a radon concentration of 3700 Bq m^{-3} during a working month of 170 h [30]. From radon exposure, the effective dose equivalents (E_{Rn}) can be calculated using the dose conversion factor of 3.88 mSv $(WLM)^{-1}$ [29, 30, 33]

Alpha index

The excess alpha radiation due to the radon inhalation originating from the soil samples can be assessed using the parameter alpha index, I_α, using the following formula [4, 34]

$$I_\alpha = \frac{C_{Ra}}{200} \quad (17)$$

where C_{Ra} is the effective radium content of the soil samples in Bq kg^{-1}. The recommended limit of activity concentration of ^{226}Ra is 200 Bq kg^{-1} and when it exceed the limiting value may leads the radon exhalation from the soil samples could cause indoor radon concentrations exceeding 200 Bq m^{-3} [34, 35]

Results and discussion

Soil is one of the important sources of natural radioactivity which mainly contributes to indoor radon. It is important to assess its emanation potential to estimate the radiation risk to the population and the radon exhalation rates. Coastal region of Kerala constitutes high and low background radiation areas (HBRAs and NBRAs). So a comparison of exhalation rates of soil samples from HBRAs and NBRAs have also done during the present study. The activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th of soil samples have been estimated and presented in Table 1.

The activity concentration of ^{40}K varies from 13.4 to 152 Bq kg^{-1} with a mean value of 79.8 ± 47.8 Bq kg^{-1}, the activity concentration of ^{226}Ra varies from 5.6 to 23.4 Bq kg^{-1} with a mean value of 14.9 ± 6.6 Bq kg^{-1} and the activity concentration of ^{232}Th varies from 12.7 to 534 Bq kg^{-1} with a mean value of 205.1 ± 182.8 Bq kg^{-1}. The minimum ^{40}K and ^{226}Ra activity concentration was shown by the soil samples of Kappad beach where as the maximum ^{40}K and ^{226}Ra activity concentration was shown by the Kollam beach soil. The minimum ^{232}Th activity concentration was shown by the Payyambalam beach soil samples and maximum activity concentration was shown

Table 1 Activity concentration of ^{40}K, ^{226}Ra and ^{232}Th

Sampling locations	Activity concentration (Bq kg^{-1})		
	^{40}K	^{226}Ra	^{232}Th
Kovalam	112 ± 1	17.7 ± 0.3	412 ± 1.4
Kollam	152 ± 0.72	23.4 ± 0.7	534 ± 1.7
Alapuzha	82.6 ± 0.3	9.4 ± 0.4	345.3 ± 1.4
Cherai	42.5 ± 0.8	23.2 ± 0.5	149.1 ± 0.9
Chavakkad	78.9 ± 0.7	19.1 ± 0.34	96.5 ± 0.6
Padinjarekkara	127 ± 2	15.9 ± 0.79	163.8 ± 1.6
Kappad	13.4 ± 0.6	5.6 ± 0.43	18.9 ± 0.94
Payyambalam	17.5 ± 0.9	11.6 ± 0.91	12.7 ± 0.76
Bekal	92 ± 2	7.9 ± 0.51	113.3 ± 1.2
Min	13.4	5.6	12.7
Max	152	23.4	534
Mean	79.8	14.9	205.1
SD	47.8	6.6	182.8
Variance	2288.7	43.0	33,423.7
Skewness	− 0.10	− 0.02	0.82
Kurtosis	− 1.01	− 1.49	− 0.56

Table 2 Radiological parameters

Sampling locations	Ra_{eq} activity (Bq kg^{-1})	Absorbed dose rate D (nGy h^{-1})	AEDE (mSv y^{-1})	H_{ex}	H_{in}
Kovalam	615.5	261.7	0.32	1.66	1.71
Kollam	798.7	339.7	0.42	2.16	2.22
Alapuzha	509.6	216.4	0.27	1.38	1.40
Cherai	239.7	102.6	0.13	0.65	0.71
Chavakkad	163.2	70.4	0.09	0.44	0.49
Padinjarekkara	259.9	111.6	0.14	0.70	0.74
Kappad	33.7	14.6	0.02	0.09	0.11
Payyambalam	31.1	13.8	0.02	0.08	0.12
Bekal	177.0	75.9	0.09	0.48	0.50
Min	31.1	13.8	0.02	0.08	0.11
Max	798.7	339.7	0.42	2.16	2.22
Mean	314.3	134.1	0.16	0.85	0.89
SD	267.6	113.5	0.14	0.72	0.73

by the Kollam beach soils. The UNSCEAR recommended world average values of ^{40}K, ^{226}Ra and ^{232}Th are 400, 35 and 30 Bq kg^{-1} respectively [12]. Study shows that the average values of ^{40}K and ^{226}Ra activity concentrations are within the recommended limit where as the average thorium activity concentration was higher than the world average value. The higher radionuclides concentration was shown by the soil samples of Kollam district which is a well reported high background radiation area. Comparatively higher thorium activity concentration was shown by Kollam beach samples may be due to heavy mineral deposits and geological & geophysical factors. The high thorium activity concentrations were reported in Varkkala and other areas of Kollam district [36, 37] even though the origin of thorium deposition on the west coast of India is not well established [38]. The other sampling stations except Kollam district are coming under normal background radiation areas show activity concentration within the recommended limit.

The statistical analysis of radionuclide activity concentration is also given in Table 1. It is clear that, skewness and kurtosis values of all radionuclides slightly differ from zero indicates asymmetric distribution. The ^{40}K and ^{226}Ra show negative skewness values indicate, values are extending towards negative direction. Positive skewness value of ^{232}Th means that asymmetric tail extending towards positive direction. The negative kurtosis values of ^{40}K, ^{226}Ra and ^{232}Th indicates flat distribution.

The radiological parameters such as radium equivalent activity (Ra_{eq}), absorbed dose rate (D), annual effective dose equivalent (AEDE), external hazard index (H_{ex}) and internal hazard index (H_{in}) were also calculated in order to understand the health effects contributed by the radionuclide activity concentration in the samples of the study area and are given in Table 2.

Comparatively higher radium equivalent activity was shown by the samples collected from Kollam region (798.7 Bq kg^{-1}) and low values are shown by the samples of Payyambalam region (31.1 Bq kg^{-1}). However, the average radium equivalent activity corresponding to all samples (314.3 Bq kg^{-1}) is lower than the recommended upper limit 370 Bq kg^{-1} by the UNSCEAR [12]. Absorbed dose obtained from the study area ranges from 13.8 nGy h^{-1} (Payyambalam) to 339.7 nGy h^{-1} (Kollam) with a mean value of 134.1 nGy h^{-1}. It can be seen that, the average value of AD was higher than the world average value of 57 nGy h^{-1} [12]. A large variation of absorbed dose among selected locations has been observed due to distinct geological characteristics, unequal distribution of radionuclides in the environment and human interference.

The outdoor annual effective dose was lower in Kappad & Payyambalam region (0.02 mSv y^{-1}) and higher in Kollam region (0.42 mSv y^{-1}). It can be seen that, the average AEDE value (0.16 mSv y^{-1}) was slightly higher than the world average value of 0.07 mSv y^{-1} [12] indicates that radionuclide concentration in the study area may not contribute any radiation hazard to the populace. Average values of H_{ex} and H_{in} were less than unity indicates that exposure to radiation below the estimated level does not cause any kind of adverse health effects to the inhabitants in the study area.

The effective radium content (C_{Ra}), surface exhalation rate (E_A), mass exhalation rate (E_M), radon concentration contributing to indoor air (C_{Rn}), indoor inhalation exposure (radon) effective dose (E_{Rn}) and alpha index (I_α) of the soil samples of different grain sizes analyzed by can

technique, were given in Tables 3 and 4. The effective radium content (Table 3) was found to vary from 0.32 to 2.17 Bq kg^{-1} with a mean value of 1.20 ± 0.64 Bq kg^{-1} for 1000–500 µm, from 0.48 to 2.36 Bq kg^{-1} with a mean value of1.43 ± 0.63 Bq kg^{-1} for 500–250 µm, from 0.78 to 2.40 Bq kg^{-1} with a mean value of 1.74 ± 0.52 Bq kg^{-1} for 250–125 µm, and from 0.89 to 3.10 Bq kg^{-1} with a mean value of 2.25 ± 0.72 Bq kg^{-1} for < 125 µm grain size soil samples. Higher effective radium content was observed for smaller grain size fractions of the soil samples. The higher radium activity concentration was shown by the soil samples of Kovalam beach where the background radiation shown was abnormally high within 1 km distance from seashore. The lower effective radium content was shown by the soil samples of Kappad beach which comes under NBRAs. The average value of effective radium content obtained in the present study is lower than the recommended value of 370 Bq kg^{-1} as recommended by the Organization for Economic Cooperation and Development [21, 39]

The surface exhalation rate (Table 3) found to vary from 48 to 327 mBq m^{-2} h^{-1} with a mean value of 180 ± 97 mBq m^{-2} h^{-1} for 1000–500 µm, from 72 to 355 mBq m^{-2} h^{-1} with a mean value of 215 ± 95 mBq m^{-2} h^{-1} for 500–250 µm, from 118 to 361 mBq m^{-2} h^{-1} with a mean value of 261 ± 79 mBq m^{-2} h^{-1} for 250–125 µm and from 134 to 466 mBq m^{-2} h^{-1} with a mean value of 338 ± 108 mBq m^{-2} h^{-1} for < 125 µm grain size soil samples. It can be seen that surface exhalation rate varies appreciably in different grain size fractions. The higher exhalation rate was shown by smaller fractions of soil samples (< 125 µm) and lower rate was shown by large fractionated grain sizes (1000–500 µm). The similar results were reported by Jagadeesha and Narayana [4]. The obtained mass exhalation rates varied from 2.4 to 16.4 mBq kg^{-1} h^{-1} with a mean value of 9.1 ± 4.9 mBq kg^{-1} h^{-1} for

Table 3 Effective radium content (C_{Ra}), surface exhalation rate (E_A) and mass exhalation rate (E_M) of soil samples

Sampling locations	Effective radium content C_{Ra} (Bq kg^{-1})				Surface exhalation rate E_A (mBq m^{-2} h^{-1})			
	1000–500 µm	500–250 µm	250–125 µm	< 125 µm	1000–500 µm	500–250 µm	250–125 µm	< 125 µm
Kovalam	2.17	2.36	2.40	3.10	327	355	361	466
Kollam	2.06	2.18	2.36	2.79	310	328	355	420
Alapuzha	0.79	0.88	1.99	2.66	120	132	301	400
Cherai	1.74	1.97	2.01	2.69	261	296	303	404
Chavakkad	0.86	1.10	1.33	2.14	129	166	200	322
Padinjarekkara	0.73	1.09	1.39	1.63	110	164	209	246
Kappad	0.32	0.48	0.78	0.89	48	72	118	134
Payyambalam	1.10	1.23	1.65	2.09	165	185	248	316
Bekal	1.03	1.57	1.69	–	155	237	255	–
Min	0.32	0.48	0.78	0.89	48	72	118	134
Max	2.17	2.36	2.40	3.10	327	355	361	466
Mean	1.20	1.43	1.74	2.25	180	215	261	338
SD	0.64	0.63	0.52	0.72	97	95	79	108

Sampling locations	Mass exhalation rate E_M (mBq kg^{-1} h^{-1})			
	1000–500 µm	500–250 µm	250–125 µm	< 125 µm
Kovalam	16.4	17.8	18.2	23.4
Kollam	15.6	16.5	17.9	21.1
Alapuzha	6.0	6.6	15.1	20.1
Cherai	13.1	14.9	15.2	20.3
Chavakkad	6.5	8.3	10.1	16.2
Padinjarekkara	5.5	8.2	10.5	12.4
Kappad	2.4	3.6	5.9	6.7
Payyambalam	8.3	9.3	12.5	15.9
Bekal	7.8	11.9	12.8	–
Min	2.4	3.6	5.9	6.7
Max	16.4	17.8	18.2	23.4
Mean	9.1	10.8	13.1	17.0
SD	4.9	4.8	3.9	5.4

Table 4 Radon concentration in the indoor air (C_{Rn}), annual effective dose equivalent (E_{Rn}) and alpha index (I_α) of soil samples

Sampling locations	Radon conc. in the room C_{Rn} (mBq m^{-3})				Annual effective dose equivalent E_{Rn} (μSv y^{-1})			
	1000–500 μm	500–250 μm	250–125 μm	< 125 μm	1000–500 μm	500–250 μm	250–125 μm	< 125 μm
Kovalam	1306	1420	1446	1863	22.6	24.6	24.9	32.2
Kollam	1241	1313	1421	1678	21.5	22.7	24.6	29.0
Alapuzha	478	529	1202	1600	8.3	9.2	20.8	27.7
Cherai	1046	1184	1210	1617	18.1	20.5	20.9	27.9
Chavakkad	517	664	800	1286	8.9	11.5	13.8	22.2
Padinjarekkara	440	655	836	983	7.6	11.3	14.5	17.0
Kappad	190	286	471	535	3.3	4.9	8.2	9.3
Payyambalam	661	738	992	1263	11.4	12.8	17.2	21.8
Bekal	621	946	1020	–	10.7	16.4	17.6	–
Min	190	286	472	535	3.3	4.9	8.2	9.3
Max	1306	1420	1446	1863	22.6	24.6	24.9	32.2
Mean	722	860	1044	1353	12.5	14.9	18.1	23.4
SD	386	382	314	433	6.7	6.6	5.4	7.5

Sampling locations	Alpha index I_α (mBq kg^{-1})			
	1000–500 μm	500–250 μm	250–125 μm	< 125 μm
Kovalam	10.9	11.8	12.0	15.5
Kollam	10.3	10.9	11.8	13.9
Alapuzha	3.9	4.4	9.9	13.3
Cherai	8.7	9.8	10.1	13.4
Chavakkad	4.3	5.5	6.7	10.7
Padinjarekkara	3.7	5.4	6.9	8.2
Kappad	1.6	2.4	3.9	4.4
Payyambalam	5.5	6.1	8.2	10.5
Bekal	5.2	7.9	8.5	–
Min	1.6	2.4	3.9	4.4
Max	10.9	11.8	12.0	15.5
Mean	5.9	7.1	8.7	11.2
SD	3.2	3.2	2.6	3.6

1000–500 μm, from 3.6 to 17.8 mBq kg^{-1} h^{-1} with a mean value of 10.8 ± 4.8 mBq kg^{-1} h^{-1} for 500–250 μm, from 5.9 to 18.2 mBq kg^{-1} h^{-1} with a mean value of 13.1 ± 3.9 mBq kg^{-1} h^{-1} for 250–125 μm and from 6.7 to 23.4 mBq kg^{-1} h^{-1} with a mean value of 17.0 ± 5.4 mBq kg^{-1} h^{-1} for < 125 μm grain size. Results show that comparatively higher exhalation rates are shown by the soil samples of HBRA beaches such as Kovalam and Kollam than beaches of NBRAs such as Kappad, Payyambalam and Bekal. However the average values of exhalation rates are lower than the world average value of 57.6 Bq m^{-2} h^{-1} [12, 30].

In order to find out the annual effective doses contributed by the exhaling radon in the indoor air, contribution of radon from soil to indoor radon was determined using the estimated surface exhalation rates. It can be seen that radon concentration contributing to indoor air (Table 4) varies from 190 to 1306 mBq m^{-3} with a mean value of 722 ± 386 mBq m^{-3} for 1000–500 μm, from 286 to 1419 mBq m^{-3} with a mean value of 860 ± 382 mBq m^{-3} for 500–250 μm, from 472 to 1446 mBq m^{-3} with a mean value of 1044 ± 314 mBq m^{-3} for 250–125 μm and from 535 to 1863 mBq m^{-3} with a mean value of 1353 ± 433 mBq m^{-3} for < 125 μm grain size. The calculated values of the annual effective dose equivalents obtained from radon concentration contributing to indoor air was also given in Table 4. It was found to vary from 3.3 to 22.6 μSv y^{-1} with a mean value of 12.5 ± 6.7 μSv y^{-1} for 1000–500 μm, from 4.9 to 24.6 μSv y^{-1} with a mean value of 14.9 ± 6.6 μSv y^{-1} for 500–250 μm, from 8.2 to 25.0 μSv y^{-1} with a mean value of 18.1 ± 5.4 μSv y^{-1} for 250–125 μm and from 9.3 to 32.2 μSv y^{-1} with a mean value of 23.4 ± 7.5 μSv y^{-1} for < 125 μm grain size. It can be seen that, the estimated annual effective dose is less than the prescribed limit of 0.3 mSv y^{-1} [29, 40].

Most of the houses from the sample collected coastal area are made up of soil and red stones. Hence, soil and building

materials play an important role in contributing to indoor radon concentration due to exhalation from these materials. From the present study, it can be seen that maximum concentration was shown by the soil samples of Southern coastal Kerala which are well reported HBRAs and the exhalation rate increases with decrease in grain size. The variation may be attributed to varying geological structure of the sampling sites, differences in the concentration of uranium and radium, porosity, permeability, density and grain size. As grain size decreases, surface area increases, which may turn increase in pore space between the grains leads to more radon diffusion.

The I_α value of the soil samples (Table 4) found to be varying from 1.6 to 10.9 mBq kg^{-1} with a mean value of 5.9 ± 3.2 mBq kg^{-1} for 1000–500 µm, from 2.4 to 11.8 mBq kg^{-1} with a mean value of 7.1 ± 3.2 mBq kg^{-1} for 500–250 µm, from 3.9 to 12.0 mBq kg^{-1} with a mean value of 8.7 ± 2.6 mBq kg^{-1} for 250–125 µm and from 4.4 to 15.5 mBq kg^{-1} with a mean value of 11.2 ± 3.6 mBq kg^{-1} for < 125 µm grain size soil samples. The recommended exemption and upper level of radium concentration in building materials such as soil, tiles, granite, cement etc. are 100 and 200 Bq kg^{-1} respectively. Therefore, it can be say that, when the activity concentration of radium exceeding 200 Bq kg^{-1} leads to increase the indoor radon concentration beyond 200 Bq m^{-3} and on contrary, radium activity concentration below 100 Bq kg^{-1} may not lead to increase the indoor radon concentration above 200 Bq m^{-3}. So the recommended limit of radium activity concentration is 200 Bq kg^{-1} for which $I_\alpha = 1$ [34]. The present study shows that the estimated I_α values are less than 1.

Table 5 gives the statistical data of radon concentration based on different grain sizes of the soil samples such as 1000–500 µm, 500–250 µm, 250–125 µm and < 125 µm. Figures 5, 6, 7 and 8 show the frequency distribution curves of radon concentration in the grain sizes 1000–500 µm, 500–250 µm, 250–125 µm and < 125 µm of soil samples respectively. From the table, it is clear that, the skewness and kurtosis values for all the grain sizes slightly differ from zero which indicates the distribution is asymmetric. The positive skewness values of 1000–500 µm and 500–250 µm grain sizes are nearer to zero indicates approximately normal distribution. While the negative skewness values of 250–125 µm and < 125 µm

Table 5 Statistical data of radon concentration (Bq m^{-3}) based on grain size of the soil samples

Parameter	1000–500 µm	500–250 µm	250–125 µm	< 125 µm
Minimum	73.89	111.07	183.04	207.59
Maximum	506.94	551.04	561.06	723.02
Mean	280.25	333.59	405.31	525.17
Median	240.97	286.51	395.83	560.12
SD	149.85	148.12	121.95	168.21
Variance	22,454.51	21,940.84	14,872.97	28,295.9
Skewness	0.49	0.18	−0.44	−0.93
Kurtosis	−1.06	−1.09	−0.16	0.49

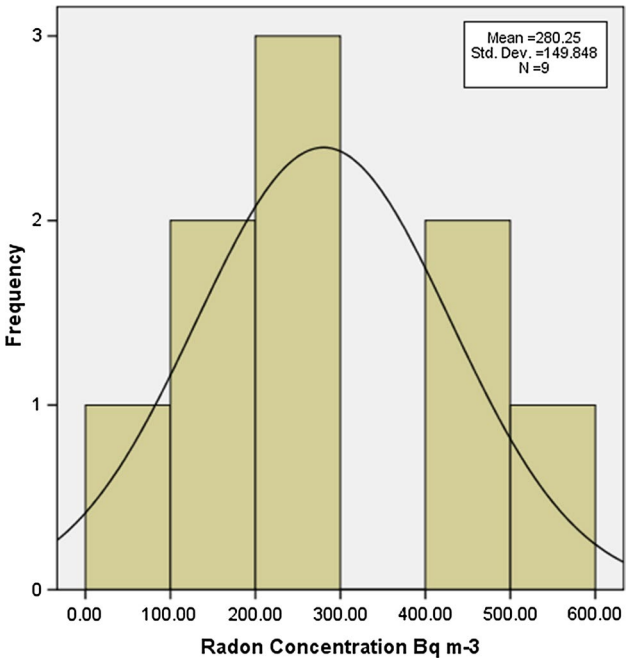

Fig. 5 Frequency distribution curve of 1000–500 µm grain size soil

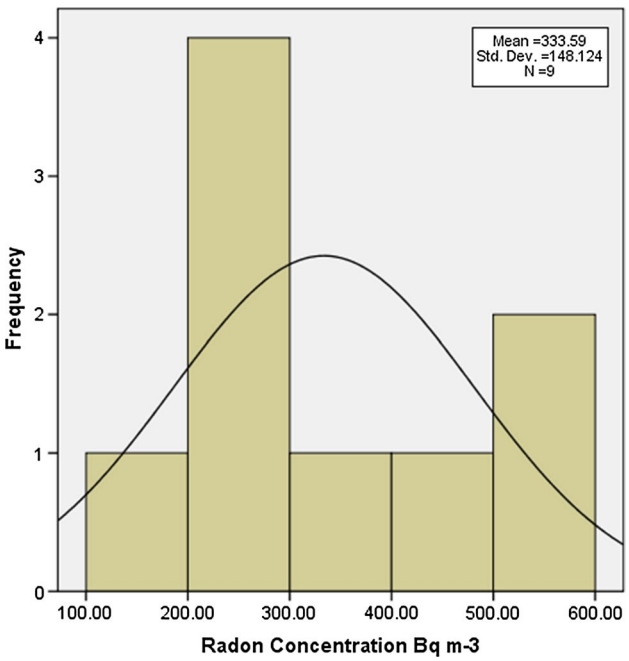

Fig. 6 Frequency distribution curve of 500–250 µm grain size soil

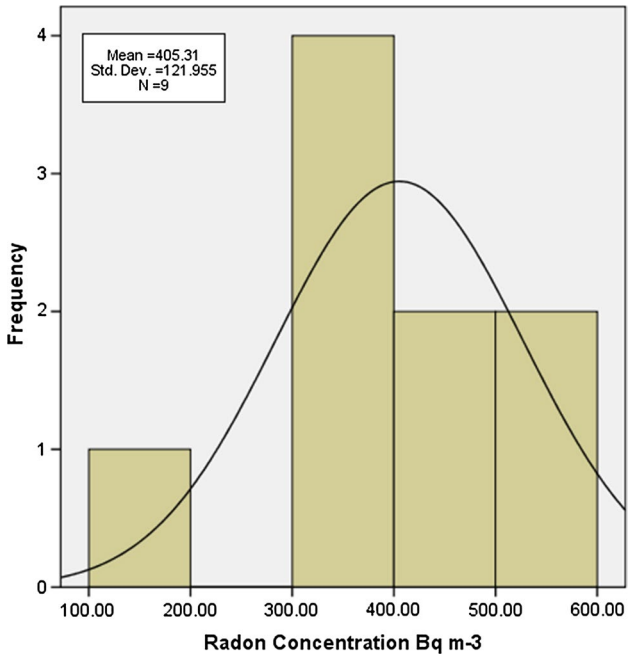

Fig. 7 Frequency distribution curve of 250–125 µm grain size soil

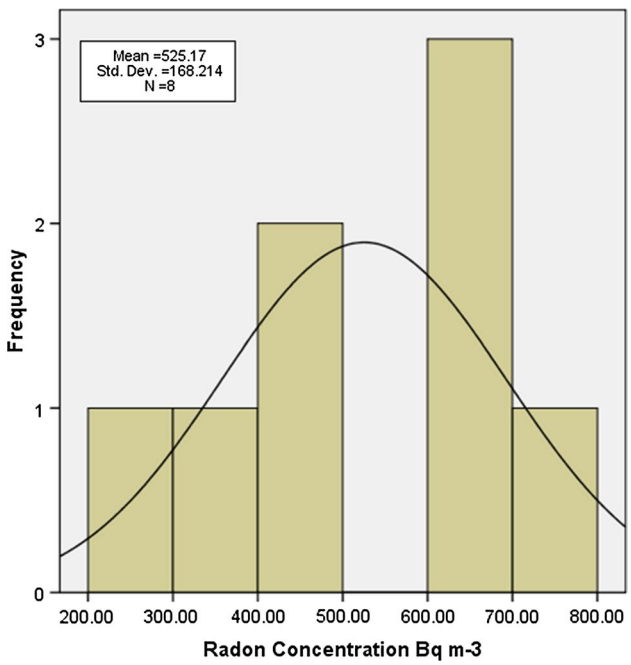

Fig. 8 Frequency distribution curve of < 125 µm grain size soil

grain sizes show that the values are slightly extending towards the negative direction and the left tail is being slightly longer than right tail. The positive kurtosis values indicate peaked distribution while the negative kurtosis values indicate flat distribution.

A comparison of radon surface exhalation rate and mass exhalation rate values obtained in the soil samples of coastal Kerala have been done with the studies reported from various parts of the world and is given in Table 6. It can be seen that there is a large discrepancy between the present study and reported values of exhalation rates. The inconsistency may be due to variation in the radium content of the soil samples, lithology of the selected sampling sites and measurement errors from the tracks contribution due to thoron.

A correlation study has been carried out regarding the activity concentration of ^{226}Ra obtained from the gamma spectrometric analysis and radon surface exhalation rate obtained by Can technique and is plotted in Fig. 9. The obtained regression coefficient of 0.63 indicates that the variation of analyzed values according to change in sampling location using these two methods is almost linear. Hence, the study appreciates the applicability of these two methods in radionuclide analysis.

Conclusion

Systematic investigations of radionuclides activity concentration have been undertaken using gamma spectrometric analysis and the exhalation rates have been estimated using closed can technique in different grain sizes for the soil samples of coastal Kerala. The results show that the estimated parameters vary significantly among each sample according to varying geological conditions and background radiation content of the study area. The higher activity concentration and exhalation rates were shown by soil samples collected from west coastal areas of Kerala, which are well reported high background radiation areas. The exhalation rates also vary according to different grain sizes of the soil in which higher values were obtained for smaller grain size soil samples and vice versa. The correlation analysis indicates that estimated values using these two methods show linear variation according to change in sites and hence implies the applicability of these methods in radionuclide analysis. From the estimated radiological parameters, some values come beyond the recommended limit of UNSCEAR especially of Kollam beach, the place coming under known high background radiation areas. However, the exhalation rates and related parameters were found below the recommended level by the UNSCEAR 2000. The study concludes that the coastal zone of Kerala constitutes unpolluted marine ecosystem except Kollam and therefore, the area can be used for leisure, recreation and tourism & the soil samples of these areas can be utilized for construction purpose safely.

Table 6 Comparison of radon surface exhalation rate and radon mass exhalation rate values in soil samples of coastal Kerala with those reported from different regions of the world

Surface exhalation rate E_A (mBq m^{-2} h^{-1})	Mass exhalation rate E_M (mBq kg^{-1} h^{-1})	Region	References
63.61–973.23	23.45–359.49	Karanataka, India	[2]
10.4–67.2	0.45–2.9	Punjab, India	[19]
41.8–907	8.39–182	Karnataka, India	[4]
–	39.1–91.2	Himachal Pradesh, India	[21]
83.96–419.81	2.11–10.55	Uttar Pradesh, India	[28]
98.18–234.37	2.52–7.07	Punjab, India	[23]
730–5180	35.76–253.15	Sakarya, Turkey	[3]
502.34–2062.53	–	Assam, India	[22]
31–507	1.16–19.13	Libya	[41]
480–15,370	8.31–233.70	Egypt	[27]
47.59–465.74	2.39–23.39	Coastal Kerala, India	Present study

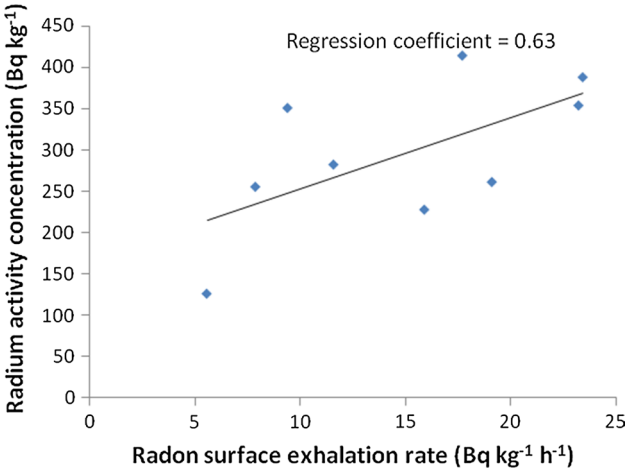

Fig. 9 Correlation of radium activity concentration and radon exhalation rate

Acknowledgements The first author wishes to acknowledge the University Grants Commission for providing Maulana Azad National Fellowship to carry out the research work.

References

1. D'Cunha P, Narayana Y (2012) Elevated natural radioactivity in soil samples of coastal Kerala, India. J Environ Res Dev 7(2):700–704
2. Kaliprasad CS, Vinutha PR, Narayana Y (2017) Natural radionuclides and radon exhalation rate in the soils of Cauvery River Basin. Air Soil Water Res 10:1–7
3. Tabar E, Yakut H, Kus A (2016) Measurement of the radon exhalation rate and effective radium concentration in soil samples of southern Sakarya, Turkey. Indoor Built Environ 27:1–11
4. Jagadeesha BG, Narayna Y (2018) Effect of grain size on radon exhalation rate in the soils of Hassan district of Southern India. Radiochemistry 60(3):328–333
5. Shanthi G, Thampi Thanka Kumaran J, Allen Gnana Raj G, Maniyan CG (2010) Measurement of activity concentration of natural radionuclides for the assessment of radiological indices. Radiat Prot Dosim 141(1):90–96
6. Jose RM, Byju SB, Anilkumar S, Chougaonkar MP (2015) External gamma dose levels in the soil samples of HBRAs of Kerala and Tamil Nadu, India. Int J Fundam Phys Sci 5(2):54–57
7. Volchock LH, de Planque G (eds) (1983) EML procedure manual, 26th edn. Environmental Measurement Laboratory, New York
8. Alnour IA, Wagiran H, Ibrahim N, Laili Z, Omar M, Hamzah S, Idi BY (2012) Natural radioactivity measurements in the granite rock of quarry sites, Johor, Malaysia. Radiat Phys Chem 81:1842–1847
9. Venunathan N, Kaliprasad CS, Narayana Y (2016) Natural radioactivity and sediments and river bank soil of Kallada River of South India and associated radiological risk. Radiat Prot Dosim 171:271–276
10. IAEA/RCA (1989) Regional workshop on environmental sampling and measurement of radioactivity for monitory purpose, Kalpakkam, pp 85–92
11. Prakash MM, Kaliprasad CS, Narayana Y (2017) Studies on natural radioactivity in rocks of Coorg district, Karnataka state, India. J Radiat Res Appl Sci 10:128–134
12. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (2000) Sources, effects and risks of ionizing radiation, report to the General Assembly, with annexes
13. Lu X, Chao S, Yang F (2014) Determination of natural radioactivity and associated radiation hazard in building materials used in Weinan, China. Radiat Phys Chem 99:62–67
14. Kuzmanovic P, Todorovic N, Nikolov J, Hansman J, Vranicar A, Knezevic J, Miljevic B (2019) Assessment of radiation risk and radon exhalation rate for granite used in the construction industry. J Radioanal Nucl Chem 321:565–577
15. Abu-Jarad F, Fremlin JH, Bull R (1980) A study of radon emitted from building materials using plastic alpha track detectors. Phys Med Biol 25(4):683–694
16. Khan AJ, Prasad R, Tyagi RK (1992) Measurement of radon exhalation rate from some building materials. Nucl Tracks Radiat Meas 20:609–610
17. Kumar R, Sengupta D, Prasad R (2003) Natural radioactivity and radon exhalation studies of rock samples from Surda Cu deposits in Singhbhum shear zone. Radiat Meas 36:551–553
18. Rajesh-Kumar AK, Mahur N, Sulekha-Rao D, Prasad SR (2008) Radon exhalation rate from sand samples from the newly discovered high background radiation area at Erasama beach placer deposit of Orissa India. Radiat Meas 43:S508–S511

19. Mehta V, Singh TP, Chauhan RP, Mudahar GS (2015) Radon exhalation rates from some soil samples of Kharar, Punjab. Adv Mater Radiat Phys 1675:030102
20. Somogy G, Abdel-Fateh H, Hunyadi I, Toth SM (1986) Measurement of exhalation and diffusion parameters of radon in solids by plastic track detectors. Nucl Tracks 12:701–704
21. Bala P, Kumar V, Mehra R (2017) Measurement of radon exhalation rate in various building materials and soil samples. J Earth Syst Sci 126(31):1–8
22. Kakati RK, Kakati L, Ramachandran TV (2013) Measurement of uranium, radium and radon exhalation rate of soil samples from Karbi Anglong district of Assam India using EDXRF and Can technique method. APCBEE Procedia 5:186–191
23. Kumar A, Kaur A (2014) A study of radon concentration in water and radon exhalation rate in soil samples belonging to Kapurthala district, Punjab, India. Adv Appl Sci Res 5(1):43–47
24. Sahoo BK, Agarwal TK, Gaware JJ, Sapra BK (2014) Thoron interference in radon exhalation rate measured by solid state nuclear track detector based can technique. J Radioanal Nucl Chem 302:1417–1420
25. Singh AK, Jojo PJ, Khan AJ, Prasad R, Ramchandran TV (1997) Calibration of track detectors and measurement of radon exhalation rate from solid samples. Radiat Prot Environ 3:129–133
26. Sharma DK, Kumar A, Kumar M, Singh S (2003) Study of uranium, radium and radon exhalation rate in soil samples from some areas of Kangra district, Himachal Pradesh, India using solid state nuclear track detectors. Radiat Meas 36:363–366
27. El-Zaher MA (2013) A comparative study of the indoor radon level with the radon exhalation rate from soil in Alexandria city. Radiat Prot Dosim 154(4):490–496
28. Choudhary AK (2014) Measurement of radon activity and exhalation rate in soil samples from Banda district India. Radiat Prot Environ 37:161–164
29. Gupta M, Mahur AK, Varshney R, Sonkawade RG, Verma KD, Prasad R (2013) Measurement of natural radioactivity and radon exhalation rate in fly ash samples from a thermal power plant and estimation of radiation doses. Radiat Meas 50:160–165
30. Feng T, Lu X (2015) Natural radioactivity, radon exhalation rate and radiation dose of fly ash used as building materials in Xiangyang China. Indoor Built Environ 25(4):1–9
31. Mahur AK, Kumar R, Sengupta D, Prasad R (2008) Estimation of radon exhalation rate, natural radioactivity and radiation doses in fly ash samples from Durgapur thermal power, plant West, Bengal, India. J Environ Radioact 99:1289–1293
32. Mahur AK, Kumar R, Mishra M, Sengupta D, Prasad R (2008) An investigation of radon exhalation rate and estimation of radiation doses in coal and fly ash samples. Appl Radiat Isot 66:401–406
33. ICRP (1993) International Commission on Radiological Protection (Oxford: Pergamon Press) ICRP Publication No. 65
34. Amen RM (2015) A study of radon emitted from building materials using solid state nuclear track detectors. J Radiat Res Appl Sci 8:516–522
35. European Commission (EC) (1999) Radiation Protection Unit. Radiological protection principles concerning the natural radioactivity of building materials. Radiation Protection, p 112
36. Ramsiya M, Joseph A, Eappen KP, Visnuprasad AK (2019) Activity concentrations of radionuclides in soil samples along the coastal areas of Kerala India and the assessment of radiation hazard indices. J Radioanal Nucl Chem 320:291–298
37. Divya PV, Kaliprasad CS, Narayana Y, Prakash V (2019) Distribution of natural radionuclides and assessment of excess lifetime cancer risk along coastal areas of Varkala in Kerala. J Radioanal Nucl Chem 322(1):1–7
38. Iyer MR (2018) Origin of thorium deposits in Kerala beach sands. Radiat Prot Environ 38:98–101
39. NEA-OECD, Nuclear Energy Agency. Exposure to radiation from natural radioactivity in building materials (1979) Report by NEA Group of Experts, Organization for Economic Corporation and Development OECD, Paris
40. ICRP (2007) International Commission for Radiological Protection. New York.
41. Saad AF, Abdallah RM, Hussein NA (2013) Radon exhalation from Libyan soil samples measured with the SSNTD technique. Appl Radiat Isot 72:163–168