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Natural Radionuclides and Radon Exhalation Rate in the Soils of Cauvery River Basin

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ABSTRACT: In this study, systematic measurement of activity concentrations of ^{40}K, ^{226}Ra, and ^{232}Th and radon exhalation rate has been done in soil samples of Cauvery River environment. The activity was measured using HPGe gamma-ray spectrometer, and the mean values of ^{40}K, ^{226}Ra, and ^{232}Th in the soil samples were found to be 182 ± 4, 34 ± 2, and 19 ± 1 Bq kg^{-1}, respectively. The radon exhalation rate was measured by "Can technique" using SSNTD (LR-115) films. The mean values of radium concentration, surface exhalation, and mass exhalation rate were found to be 118.95, 293.61, and 108.53 mBq kg^{-1} h^{-1}, respectively. The radiological hazard indices due to natural radioactivity were calculated and compared with international recommended values, which are lower than the recommended level. The radon exhalation rate is lower than the recommended level.

KEYWORDS: Cauvery, radon, gamma dose, radioactivity, soil

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Introduction

Soil is the top surface of the earth's crust and it is formed due to the weathering of rocks. The soil contains radionuclides headed by the radioactive decay series ^{238}U, ^{232}Th, and singly occurring ^{40}K. ^{238}U and ^{232}Th contribute about 30% to 60% of the internal radiation dose, and the worldwide average annual effective dose per person ranges from 1.4 to 2.4 mSv y^{-1}, which depends on the concentration of natural radionuclides present in a particular location.[1] Radon (^{222}Rn) is a naturally occurring radioactive isotope of ^{238}U series, which has half-life of 3.82 days. Radon is one of the significant sources of natural radiation. The radon decay products which are attached in aerosol cause greater biological effect through inhalation and can lead to lung cancer on prolonged exposure. ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po are the short-lived decay products of radon; these decay products give rise to maximum dose through emission of alpha and beta particles. Therefore, the studies on radon exhalation rate, radionuclides distribution, and its associated dose rate in the soil are significant. The riverine environs are of greater interest due to the population, which lives near river basin. The soils found near river basin are also used for construction of materials such as bricks and plastering material to walls in areas of South Karnataka, India. The published data on radionuclides concentration in riverine environs are sparse, and therefore, an attempt was made in the present investigation to study natural radionuclides distribution and radon exhalation rate in the soils of Cauvery River basin.

Materials and Methods

Sample collection and preparation

The sampling stations along Cauvery River are indicated in Figure 1. Sampling station K1 corresponds to the upper reaches of the river and K14 corresponds to the lower reaches. The soil samples from the river basin were collected following standard procedures.[2] The collected soil samples were brought to the laboratory and oven dried at 110°C till constant dry weight was obtained. The dried samples were sieved through 250-μm mesh and then stored in a sealed polyvinyl chloride container for 30 days to attain secular equilibrium between radium and its progeny.[3]

Activity measurement

The activity concentrations of ^{40}K, ^{226}Ra, and ^{232}Th in soil were measured using a high-resolution N-type HPGe (NGC 3019, DSG) detector–based gamma spectrometry system. The detector was shielded using thick lead blocks to avoid interference of external gamma radiations. The output of the detector was analyzed using a 16-K multi channel analyzer (MCA-3 series/P7882; FAST ComTec). The spectrometer was calibrated using International Atomic Energy Agency (IAEA) standard reference materials. The standards used were RG-U, RG-Th, and RG-K for uranium, thorium, and potassium, respectively. The gamma spectrum was obtained with a counting period of 20 000 seconds. The peaks corresponding to 1.46 MeV (^{40}K), 609.31 keV (^{214}Bi), and 911.07 keV (^{223}Ac) were considered for evaluating the activity levels of ^{40}K, ^{226}Ra, and ^{232}Th, respectively.[4]

Radon exhalation rate

The radon exhalation rate in the soil samples of the Cauvery River was determined by "Sealed Can Technique" using SSNT detectors. About 100 g of the dried and sieved (250 μm) soil sample was taken in each "Can" (diameter: 7.0 cm and height: 10.5 cm),

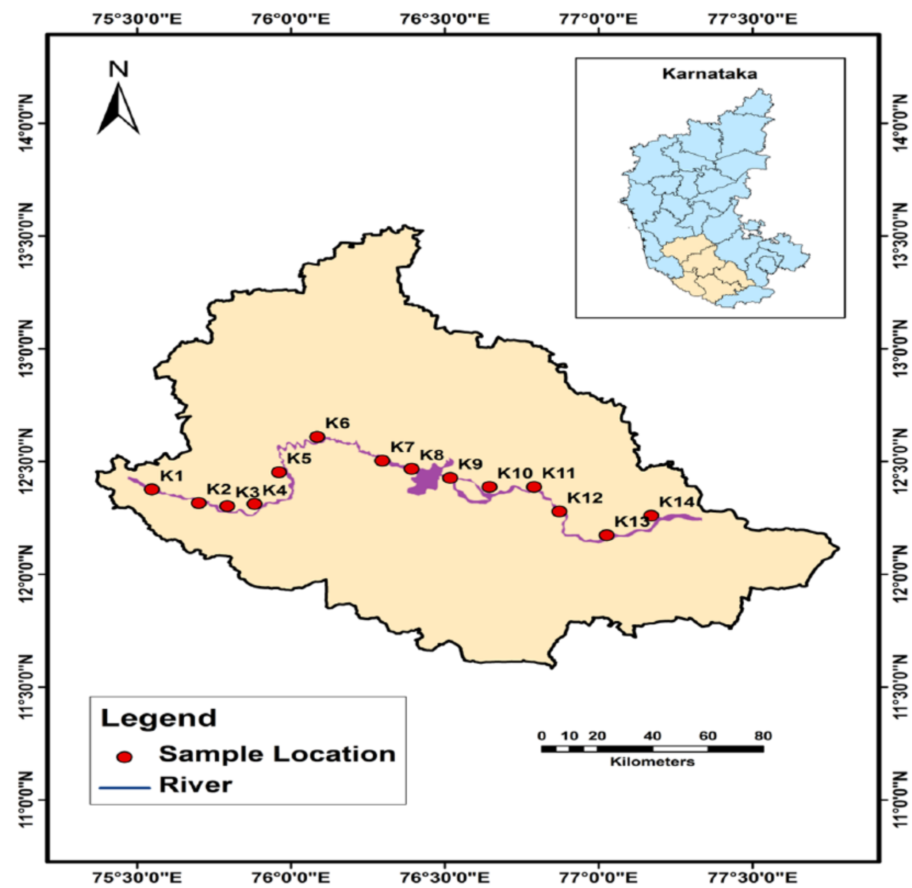

Figure 1. Cauvery River basin map.

and LR-115 Type II SSNT detector (3 cm × 3 cm) was fixed on the top inside of each "Can."[5] The "Can" was kept airtight for 4 hours to allow the radon and its progeny to reach equilibrium. The equilibrium activity of the emergent radon was calculated based on the geometry of the "Can" and the time of exposure. The LR-115 film inside the can was exposed to radon for 90 days. The films (detectors) were removed and then etched in 2.5 N NaOH at 60°C ± 1°C for a period of 60 minutes in an etching bath at constant temperature to enlarge the tracks produced from the alpha particles emitted from the decay of radon.[6] The background track density of the detector was measured using unexposed detectors under the same etching condition. The enlarged alpha tracks were counted using spark counter. Precounting was done at 900 V and finally track density was determined at 450 V, in the plateau region of the counter.

The effective radium concentration C_{Ra} can be calculated using the following formula[7]:

$$C_{Ra} = \frac{\rho h A}{M K T_e} \tag{1}$$

where ρ is the track density (0.056 tracks cm^{-2} d^{-1} [Bq m^{-1}]), T_e is the effective exposure time in an hour, h is the distance between detector films and surface of the specimen sample, M is the mass of the sample, A is the area of cross section of the cylindrical can, and k is the sensitivity factor and its value is k = 0.0312 tracks m^{-2} d^{-1} Bq^{-1} m^{-3}.

Surface exhalation rate "E_A" is obtained from the following expression[8]:

$$E_A = \frac{CV\lambda}{A\left[T + \frac{1}{\lambda}\{e^{-\lambda T} - 1\}\right]} \tag{2}$$

This formula is also modified to calculate the mass exhalation rate "E_M":

$$E_M = \frac{CV\lambda}{M\left[T + \frac{1}{\lambda}\{e^{-\lambda T} - 1\}\right]} \tag{3}$$

where E_A is measured in Bq m^{-2} h^{-1} and E_M in Bq kg^{-1} h^{-1}, V is the effective volume of can (m^3), C is the integrated radon exposure as measured by LR-115 solid-state nuclear track detectors (Bq m^{-3} h), T is the exposure time (h), l is the decay constant for radon (h^{-1}), A is the area of the can (m^2), and M is mass of the sample.

Results and Discussions

Activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K

The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in soil samples of Cauvery River basin were measured using HPGe gamma-ray spectrometer, and the results are presented in Table

Table 1. Activity of ^{226}Ra, ^{232}Th, ^{40}K in Cauvery River soil samples.

SAMPLE LOCATIONS	^{40}K, Bq kg^{-1}	^{226}Ra, Bq kg^{-1}	^{232}Th, Bq kg^{-1}
K1	24.5 ± 1.56	65.0 ± 2.55	12.3 ± 1.11
K2	160.0 ± 4.00	17.9 ± 1.34	21.1 ± 1.45
K3	100.5 ± 3.17	25.8 ± 1.60	13.7 ± 1.17
K4	58.5 ± 2.41	23.6 ± 1.53	4.5 ± 0.67
K5	181.9 ± 4.26	25.2 ± 1.58	12.3 ± 1.11
K6	530.6 ± 7.28	19.9 ± 1.41	22.0 ± 1.48
K7	236.2 ± 4.86	32.3 ± 1.79	39.4 ± 1.98
K8	79.5 ± 2.82	33.3 ± 1.82	5.7 ± 0.75
K9	242.7 ± 4.92	30.0 ± 1.73	22.1 ± 1.48
K10	351.0 ± 5.92	42.0 ± 2.05	31.9 ± 1.78
K11	316.9 ± 5.62	17.7 ± 1.33	2.9 ± 0.53
K12	33.3 ± 1.82	40.1 ± 2.00	5.3 ± 0.72
K13	221.8 ± 4.71	55.6 ± 2.35	46.9 ± 2.16
K14	3.8 ± 1.17	45.5 ± 2.13	26.9 ± 1.64

1. The measured activity ranged from 18 ± 1 to 65 ± 3 Bq kg^{-1} for ^{226}Ra, 3 ± 0.5 to 47 ± 2 Bq kg^{-1} for ^{232}Th, and 14 ± 1 to 531 ± 7 Bq kg^{-1} for ^{40}K. The measured activity of the radionuclides varied from location to location. These variations are mostly due to the changes in local geology. The physical and chemical sorting processes also contributed to the variation of radionuclides in the soil from different locations.[9] The mean activity of ^{40}K, ^{226}Ra, and ^{232}Th is 182 ± 4, 34 ± 2, and 19 ± 1 Bq kg^{-1}, respectively. In all the locations, activity concentration is in the order of $^{40}K > {}^{226}Ra > {}^{232}Th$; the concentration of ^{232}Th is much lower than the ^{226}Ra and ^{40}K. The ^{40}K activity concentration dominates over ^{226}Ra and ^{232}Th activity. The activity concentrations of ^{40}K were found to be lower in upper reaches of the river and higher values were observed in lower reaches. The ^{40}K in the upper reaches decreases by leaching process and may also be diluted with increase in organic matter and soil water contents.[10,11] In all the locations, the average concentration of ^{226}Ra, ^{232}Th, and ^{40}K is lower than the world average values (world average of ^{226}Ra, ^{232}Th, and ^{40}K is 35, 30, and 400 Bq kg^{-1}, respectively, United Nations Scientific Committee on the Effects of Atomic Radiation [UNSCEAR]).[1] The average concentration of ^{226}Ra is higher than the concentration of ^{232}Th; ^{40}K is lower than the Indian average values (29, 64, and 400 Bq kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K, respectively, UNSCEAR, 2000).

The measured activity concentrations of Cauvery River soil samples were compared with other river environs, which is presented in Table 2. The activity of ^{226}Ra in Cauvery River basin soil sample is less than the activity of ^{226}Ra in Netravathi River of Karnataka and is also below the world average value.[1] The activity concentration of Kallada River basin shows (343.4, 60.3, and 98.1 Bq kg^{-1} for ^{40}K, ^{226}Ra, and ^{232}Th, respectively) higher activity than Cauvery River activity concentration.[9] The activity concentration of Cauvery River is comparable with Wei River of China and Büyük Menderes River of Turkey.[13,17] The values are comparable with Chao Phraya, Thailand (364.49 and 42.95 Bq kg^{-1} for ^{40}K and ^{232}Th, respectively); Dez, Iran (289, 25, and 33 Bq kg^{-1} for ^{40}K, ^{226}Ra, and ^{232}Th, respectively); and Pra River, Ghana.[14,16,18] The activity concentration of ^{40}K was found to be higher in all river environments compared with this study.

The statistical analysis of ^{226}Ra, ^{232}Th, and ^{40}K activity is shown in Table 3. Figure 2 shows the frequency distribution of natural radionuclides in soil samples of Cauvery River. The kurtosis values which are positive indicate peaked distribution and negative kurtosis indicates the flat distribution. The skewness for a normal distribution is 0 and indicates the values are near to the mean value. Positive skewness indicates the few values are above the median values. The positive skewness and kurtosis values are in good agreement with the theoretical frequency distributions. The skewness and kurtosis values which are near to 0 indicate that there is no extreme deviation from the mean value.

To assess the radiologic risk of the river basin soil, various radiological hazard indices were computed and the results are shown in Table 4. The hazard indices were calculated using the following equations.

Absorbed dose rate (D)

To calculate the dose rate in the air using the activity concentrations and conversion factors of ^{226}Ra, ^{232}Th, and ^{40}K, the

Table 2. Comparison of ^{40}K, ^{226}Ra, and ^{232}Th activity in soil with other river environs.

ACTIVITY IN Bq KG^{-1}			RIVER	REFERENCE
^{40}K	^{226}Ra	^{232}Th		
182	34	19	Cauvery	Present study
588.9	58.2	39.3	Netravathi, India	Narayana et al[12]
343.4	60.3	98.1	Kallada, India	Venunathan et al[9]
643	—	42	Firtina Valley, Turkey	Kurnaz et al[13]
238.98	—	28.04	Pra River, Ghana	Adukpo et al[14]
353-958	—	11-71	Buyuk Menderes, Turkey	Akozcan[15]
289.57	25.21	19.76	Dez, Iran	Nasrabadi et al[16]
833.3	21.8	33.1	Wei River, China	Lu et al[17]
364.49	—	42.95	Chao Phraya, Thailand	Santawamaitre[18]
400	35	30	World average	UNSCEAR[1]

Abbreviation: UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation.

Table 3. Statistical data for the activities of ^{226}Ra, ^{232}Th, and ^{40}K in Cauvery River soils.

	PREMONSOON		
	^{40}K	^{226}Ra	^{232}Th
Min.	14	18	3
Max.	531	65	47
Mean	182	40	19
Median	170.9	31.2	17
SD	148	14	14
Skewness	0.95	0.91	0.70
Kurtosis	0.81	0.21	−0.28
Frequency	Normal	Normal	Uniform

absorbed dose rate (D in nGy h^{-1}) was calculated using the following equation[19,20]:

$$D = 0.462C_{Ra} + -0.604C_{Th} + 0.0147C_K \tag{4}$$

where C_{Ra}, C_{Th}, and C_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The mean dose rate was found to be 35 nGy h^{-1}, which is lower than the world (57 nGy h^{-1}) and Indian (56 nGy h^{-1}) average calculated by UNSCEAR.[1]

Annual effective equivalent dose

The annual effective dose rate for indoor and outdoor in units of μSv y^{-1} was calculated using the following formula[21]:

$$AEED\left(Outdoor\right)\left(\mu Sv\, y^{-1}\right) = Dose\ rate\left(nGy\, h^{-1}\right) \times 8760\, h \times 0.2 \times 0.7\, Sv\, Gy^{-1} \times 10^{-3} \tag{5}$$

$$AEED\left(Indoor\right)\left(\mu Sv\, y^{-1}\right) = Dose\ rate\left(nGy\, h^{-1}\right) \times 8760\, h \times 0.8 \times 0.7\, Sv\, Gy^{-1} \times 10^{-3} \tag{6}$$

where D is the dose rate. The mean annual effective equivalent dose (AEED) for outdoor and indoor was 43 and 170 μSv y^{-1}, respectively. The mean value of AEED is lower than the recommended value (outdoor: 70 μSv y^{-1} and indoor: 450 μSv y^{-1}).

Radium equivalent activity (Ra_{eq})

The radium equivalent is a single index or number which describes the total gamma output from the combination of ^{226}Ra, ^{232}Th, and ^{40}K radionuclides in the sample from individual location. The radium equivalent (Bq kg^{-1}) was calculated using the following equation (UNSCEAR, 2010)[22]:

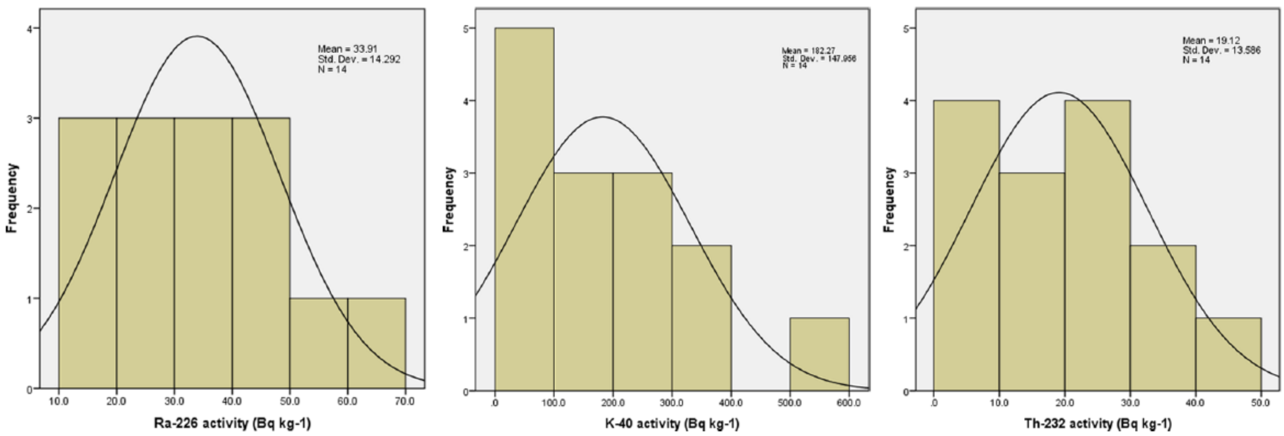

Figure 2. Frequency distribution of ^{226}Ra, ^{232}Th, and ^{40}K in monsoon soils of Cauvery.

Table 4. Radiological hazard indices of soil sample.

ABSORBED DOSE, nGy h^{-1}	AEED, µSv y^{-1}		$^{226}Ra_{eq}$, Bq kg^{-1}	HAZARD INDEX		AUI (I)	ELCR × 10^{-3} OUTDOOR	AGDE INDOOR
	OUTDOOR	INDOOR		H_{EX}	H_{IN}			
38.6	47.3	189.2	84.7	0.23	0.40	0.753	0.166	260.61
27.8	34.1	136.3	60.6	0.16	0.21	0.435	0.119	194.35
24.4	30.0	119.8	53.2	0.14	0.21	0.413	0.105	168.84
16.1	19.8	79.1	34.7	0.09	0.16	0.279	0.069	110.59
26.7	32.8	131.1	56.9	0.15	0.22	0.397	0.115	186.80
44.7	54.8	219.1	92.3	0.25	0.30	0.494	0.192	320.42
48.6	59.6	238.5	106.9	0.29	0.38	0.795	0.209	339.01
22.2	27.2	108.9	47.7	0.13	0.22	0.384	0.095	152.11
37.4	45.8	183.3	80.4	0.22	0.30	0.565	0.160	261.53
53.4	65.5	261.9	114.8	0.31	0.42	0.804	0.229	373.85
23.2	28.4	113.8	46.4	0.13	0.17	0.225	0.100	166.65
23.1	28.4	113.5	50.3	0.14	0.24	0.438	0.099	156.69
63.3	77.7	310.6	139.9	0.38	0.53	1.100	0.272	437.90
37.9	6.5	186.0	85.2	0.23	0.35	0.748	0.163	257.85

Abbreviations: AEED, annual effective equivalent dose; AGDE, annual gonadal dose equivalent; AUI, activity utilization index.

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (7)$$

where C_{Ra}, C_{Th}, and C_K are the activity of ^{226}Ra, ^{232}Th, and ^{40}K in Bq kg^{-1}, respectively. The mean radium equivalent value was found to be 75.28 Bq kg^{-1} for soils of Cauvery River basin, which is lower than the safety limit (370 Bq kg^{-1}).

Hazard index

The external and internal external hazard indices were calculated using the following equations[22,23]:

$$H_{ex} = \left(C_{Ra}/370 + C_{Th}/259 + C_K/4810\right) \le 1 \quad (8)$$

$$H_{in} = \left(C_{Ra}/185 + C_{Th}/259 + C_K/4810\right) < 1 \quad (9)$$

where C_{Ra}, C_{Th}, and C_K are the activity of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The mean external hazard index and internal hazard index values were 0.20 and 0.29, respectively.

Annual gonadal dose equivalent

The UNSCEAR has formulated equations to estimate the dose received by the body organs such as the thyroid, lungs, bone marrow, bone surface cell, and the gonads. The annual gonadal dose equivalent (AGDE) (µSv y^{-1}) was calculated using the following equation[23]:

Table 5. Radon activity and radon exhalation rate in Cauvery River soil samples.

SAMPLE LOCATIONS	C_{RA} mBq kg^{-1}	E_S mBq m^{-1} h^{-1}	E_M mBq kg^{-1} h^{-1}	RADON ACTIVITY, Bq m^{-3}
K1	50.80 ± 5.19	125.00 ± 26.41	46.35 ± 9.79	51.85 ± 10.96
K2	68.07 ± 6.00	167.41 ± 30.57	62.11 ± 11.34	69.44 ± 12.68
K3	50.14 ± 5.14	123.8 ± 26.29	45.75 ± 9.71	51.39 ± 10.91
K4	39.46 ± 4.57	97.10 ± 23.28	36.01 ± 8.63	40.28 ± 9.66
K5	259.54 ± 11.68	642.8 ± 59.90	236.80 ± 22.06	266.67 ± 24.85
K6	48.74 ± 5.07	120.53 ± 25.94	44.47 ± 9.57	50.00 ± 10.76
K7	104.75 ± 7.43	258.93 ± 38.01	95.58 ± 14.03	107.41 ± 15.77
K8	154.34 ± 9.03	380.58 ± 46.09	140.82 ± 17.05	157.87 ± 19.12
K9	394.02 ± 14.41	973.23 ± 73.70	359.49 ± 27.22	403.70 ± 30.57
K10	88.03 ± 6.83	216.52 ± 34.76	80.32 ± 12.90	89.81 ± 14.42
K11	156.04 ± 9.09	383.93 ± 46.29	142.37 ± 17.16	159.26 ± 19.20
K12	164.20 ± 9.31	405.14 ± 47.55	149.81 ± 17.58	168.06 ± 19.72
K13	61.52 ± 5.70	151.78 ± 29.1	56.13 ± 10.76	62.96 ± 12.07
K14	25.70 ± 3.68	63.61 ± 18.84	23.45 ± 6.95	26.39 ± 7.82

$$AGDE = 3.09C_{Ra} + 4.18C_{Th} + 0.31C_K \quad (10)$$

where C_{Ra}, C_{Th}, and C_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq kg^{-1}, respectively. The average value of AGDE is 241.94 μSv y^{-1}, which is lower than the world average value of 0.30 mSv y^{-1}.

Excess lifetime cancer risk

The mean value of the estimated excess lifetime cancer risk (ELCR) is 0.149 × 10^{-3}, which is lower than the world average 1.45 × 10^{-3}.[24] The ELCR was estimated using the following equation and is presented in Table 4:

$$ELCR = AEDE \times DL \times RF \quad (11)$$

where *AEDE* is the annual effective dose equivalent, *DL* is the duration of life (70 years), and *RF* is the risk factor (Sv^{-1}) fatal cancer risk per sievert. As per the International Commission on Radiological Protection (ICRP) recommendation, the risk factor for stochastic effect to the public is 0.05.

Activity utilization index (I)

In South India, the river soil is used as construction material for plastering. Therefore, the Cauvery River soil was also examined via calculating the activity utilization index (AUI) (*I*). The AUI (*I*) was calculated using the following equation[23]:

$$I = \left(C_{Ra} / 50\right) f_{Ra} + \left(C_{Th} / 50\right) f_{Th} + \left(C_K / 500\right) f_K \quad (12)$$

where C_{Ra}, C_{Th}, and C_K are the mean activity of ^{226}Ra, ^{232}Th, and ^{40}K in Bq kg^{-1} in the soil and f_{Ra}, f_{Th}, and f_K are the fractional contributions to the total dose rate of ^{226}Ra, ^{232}Th, and ^{40}K. The mean AUI is 0.55, which is below the recommended safety limit (<2). Therefore, the soils may be used as construction material.

Radon exhalation measurement

The radium activity and radon exhalation rates in the soil samples were measured using "Can technique" and the results are given in Table 5. The activity concentration of effective radium content (C_{Ra}) ranged from 25.7 to 394.02 mBq kg^{-1}, radon surface exhalation (E_S) ranged from 63.61 to 973.23 mBq m^{-1} h^{-1}, radon mass exhalation (E_M) ranged from 23.45 to 359.49 mBq kg^{-1} h^{-1}, and radon activity ranged from 26.39 to 403 Bq m^{-3}, respectively. It can be observed from the results that the radon exhalation rate varied appreciably from one sample to another. The high radium concentration in soil was found at locations K5 and K9. The mass exhalation rate and surface exhalation rate in soil samples were found to be high at K5 and K9. The variation in exhalation depends on the type of soil, emanation factor of radon from them, radium content, and diffusion coefficient of radon.

Conclusions

The measured activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the study were found to vary from place to place. The ^{40}K activity concentration dominates over ^{226}Ra and ^{232}Th. The radiological doses as absorbed dose (*D*), AEED, and AGDE were estimated using the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K and standard equations given by UNSCEAR and IAEA. The obtained results show that dose rates are lower than the world average. To assess the radiologic risk parameters, radium equivalent (Ra_{eq}), internal hazard index ($H_{in} < 1$), external hazard index ($H_{ex} < 1$), AUI (AUI < 2), and ELCR values were calculated. All the hazard indices are less than the safety limit. Hence, there is no significant radiologic risk to the population from soils in the study area. The radon exhalation rate was measured using the "Can technique." The results of this study show the variation in radon exhalation due to the variation in geological condition of the locations and properties of the soil. The soil is safe for its use as a construction material. This data will be helpful to establish new regulations and safety limits regarding the radiation dose and radon activity in Cauvery River basin.

Author Contributions

All the authors contributed equally and there is no conflict of interest.

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