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Natural Radioactivity measurement and Radiological hazards of Sand Samples used as building material in Tiruvannamalai district, Tamilnadu, India

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Abstract

Objectives: To study the naturally occurring radioactivity level of the sand samples and its radiological hazards while it is used as a building construction material.

Materials and methods: Fifteen sand samples were collected from different sand quarries from Tiruvannamalai district for the purpose of investigation of natural radioactivity measurement and their associated radiation hazards using gamma-ray spectrometry system.

Results: The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in sand samples were measured using NaI (Tl) detector and it is found that mean activity concentrations that ^{226}Ra is lower by a factor of 7.66 and ^{40}K is lower by a factor of 1.03 while ^{232}Th is higher by a factor of 3.98 ^{226}Ra, ^{232}Th and ^{40}K when compared with world average values. The radiological hazards associated with the sand samples were computed using radiation indices to assess its suitability for building constructions. Results showed that the investigated sand can be used in construction of dwellings in the study area.

Conclusion: The studied sands in this works are recommended for building constructions without any radiological sequences. This analysis will be very much useful to assess the radiation hazards in sand used as building material in dwelling and it is an initiate step to obtain database as well as radiological map of the area at stake.

Key words: Gamma-ray spectrometry, activity concentration, radiation hazards

1. Introduction

Human population is always exposed to ionizing radiation from natural sources. Natural radioactivity is widespread in the earth environment and it exists in various geological formations such as earth crust, rocks, soils, plants, water, air and also traces level in construction materials, which may contribute significantly towards an increased radiation dose received by human beings [1-2].

Building materials are the main source of indoor gamma radiation, besides terrestrial and cosmic radiation [2]. Measurement of natural radioactivity in different types of building materials was reported by many authors in different parts of the world [1-7].

Among the various building materials, sand (riverbed) is one of the essential and important materials for building constructions in different parts of the world and also in India. In Tamilnadu it is frequently used as binder in cement for construction purposes. A considerable amount of natural radionuclides can be found in sand as the end result of fertilizer washing, industrial activities and human activities [8].

Building and industrial materials like sand contribute to environmental radioactivity in two ways. First, by gamma radiation mainly due to ^{226}Ra, ^{232}Th, ^{40}K and their progenies to a whole body dose and in some cases by beta radiation to a skin dose, and secondly by releasing the noble gas radon, its radioactive daughters, which are deposited in the human respiratory tract [9].

Based on the above discussion, it is expected that ionizing radiation from sand due to the natural radionuclides. Knowledge of natural radioactivity present in sand (river beds) enables one to assess any possible radiological hazard to mankind. The main goal of the present work is to study the naturally occurring radioactivity level in sand samples and its relevant dose and radiological hazards while it is used as a building construction material.

2. Materials and Methods

2.1. Sample collection and preparation

Fifteen sand samples of different locations in and around Tiruvannamalai Dist of Tamilnadu were collected from sand quarries and river beds and were prepared by a standard method of IAEA Technical Report 295 [10]. The location map of the study area is shown in Fig 1. Samples were cleaned and grounded into powder form using a ball mill grinder machine. Samples then were dried in the oven at 110ºc until the sample weight became constant. Samples were left to cool at room temperature and fine quality was obtained using a scientific sieve of 150 micron mesh to obtain uniform size particles. All the samples were sealed in radon-impermeable plastic containers. The samples were then stored for more than 40 days to bring ^{222}Rn and its short-lived daughter products into equilibrium with ^{226}Ra [11].

2.2. Gamma ray Spectroscopic Analysis

All the selected samples were subjected to gamma spectral analysis with a counting time of 20,000 secs. A 3" x 3" NaI(Tl) detector was employed with adequate lead shielding which reduced the background by a factor of about 95%. The concentrations of various nuclides of interest were determined in Bq Kg^{-1} using the count spectra. The samples were then counted in the same source-to-detector geometry used for the establishment of the efficiency calibration. The spectra were acquired for 20,000 sec and the photo peaks were evaluated by the MCA software.

The gamma- ray photo peaks corresponding to 1.46 MeV (^{40}K), 1.76 MeV (^{214}Bi) and 2.614 MeV (^{208}Tl) were considered in arriving at the activity of ^{40}K, ^{226}Ra and ^{232}Th in the samples. The detection limit of NaI (Tl) detector system for ^{40}K, ^{226}Ra and ^{232}Th are 8.5, 2.21 and 2.11 in Bq Kg^{-1} respectively for a counting time of 20,000 secs. [6].

3. Results and Discussion

3.1. Specific activity concentration

The activity concentrations of the detected radionuclides of ^{226}Ra ^{232}Th and ^{40}K in 15 sand samples from different locations of Tiruvannamalai district are tabulated in 2nd, 3rd and 4th column of Table 1. The highest values observed for the specific activities of ^{226}Ra, ^{232}Th and ^{40}K are 15.33 (SA-6), 358.56 (SA-1) and 633.94 (SA-1) in Bq Kg^{-1} respectively while the lowest values of the same radionuclides are ≤ 2.21 (BDL), 12.99 (SA-12) and 262.11 (SA-10) in Bq Kg^{-1} respectively. It was also found that the arithmetic mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K are 4.57, 119.42 and 388.78 in Bq Kg^{-1} respectively. To compare the values of mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K of the present study with the world average values (which are 35, 30 and 400 in Bq Kg^{-1}) [12], ^{226}Ra is lower by a factor of 7.66 and ^{40}K is lower by a factor of 1.03 while ^{232}Th is higher by a factor of 3.98.

3.2. Radium equivalent activity (Ra_{eq})

The distribution of ^{226}Ra, ^{232}Th and ^{40}K in the sand samples was not uniform. With respect to the radiation exposure, the radioactivity has been defined in terms of radium equivalent activity Ra_{eq} in Bq Kg^{-1} to compare the specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K.

The Ra_{eq} is related to the external gamma dose and the internal dose due to radon and its daughters [13]. It was calculated through the following relation [14].

$$Ra_{eq}\,(Bq\,Kg^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad \text{-- (1)}$$

Where A_{Ra}, A_{Th}, and A_K are the activity concentrations in Bq Kg^{-1} of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. It may be noted that ^{238}U has been replaced with decay product ^{226}Ra because there may be disequilibrium between ^{238}U and ^{226}Ra. While defining Ra_{eq} activity according to the above equation, it has been assumed that 10 Bq of ^{226}Ra, 7 Bq of ^{232}Th, and 130 Bq of ^{40}K produce the same gamma doses [15].

The estimated Ra_{eq} activity for all investigated sand samples is given in the 5th column of Table 1. The lowest and higher values of Ra_{eq} are 59.11 (SA-12) and 561.55 (SA-1) in Bq Kg^{-1} with an average of 205.27 Bq Kg^{-1}.

The values of Ra_{eq} in building materials must be less than the 370 Bq Kg^{-1} for safe use [16-18]. All the values of Ra_{eq} for the present study (except SA-1) showed lower than recommended safety value and do not pose any radiological hazard when used for the construction of buildings.

Table 3 is used to compare the activity concentrations and radium equivalent activities (Bq Kg^{-1}) of sand samples between the present study and different areas of the world.

3.3. The absorbed gamma dose rate (D_R)

The outdoor absorbed dose rate (nGy h^{-1}) in air from terrestrial gamma radiation at 1m above the ground is calculated after applying the conversion factors (in nGy h^{-1} per Bq Kg^{-1}) to transform specific activities A_{Ra}, A_{Th} and A_K into absorbed dose rate. According to the formula provided by UNSCEAR[12] and European Commission [19], the absorbed dose rate is calculated.

In the UNSCEAR and European Commission reports, the resulting dose coefficients were found to be 0.92 nGy h^{-1} per Bq Kg^{-1} for ^{226}Ra, 1.1 nGy h^{-1} per Bq Kg^{-1} for ^{232}Th and 0.080 nGy h^{-1} per Bq Kg^{-1} for ^{40}K and the formula for the absorbed gamma dose rate can be written as:

$$D_R(nGyh^{-1}) = 0.92A_{Ra} + 1.1A_{Th} + 0.0807A_K \quad \text{-- (2)}$$

Where A_{Ra}, A_{Th} and A_K are the activities of ^{226}Ra, ^{232}Th and ^{40}K respectively in the units of Bq Kg^{-1}.

The results of absorbed dose rate for the present study are given in the 6th column of Table 1. Its values are ranging from 56.40 (SA-12) to 445.13 (SA-1) with an average of 166.67 in nGy h^{-1}. The obtained mean value of D_R in the studied samples is greater than the world average (populated-weighted) indoor absorbed gamma dose rate of 84 nGy h^{-1}. This is may be due to high activity concentrations of ^{232}Th and ^{40}K in the samples.

3.4. The annual effective dose rate (H_R)

To estimate the annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose (0.7 $svGy^{-1}$) and outdoor occupancy factor (0.2) proposed by UNSCEAR (2000) [12] were used. The effective dose rate in units of mSv y^{-1} was calculated according to Arafa, (2004) [20] using the following formula.

H_R (mSv y^{-1}) = D_R (nGy h^{-1}) × 24 h × 365.25 d × 0.2 (outdoor occupancy factor) × 0.7 Sv Gy^{-1} (conversion factor) × 10^{-6}

$$H_R(mSv\,y^{-1}) = D_R(nGy\,h^{-1}) \times 8766 \times 0.2 \times 0.7 \times 10^{-6} \qquad \text{(OR)}$$

$$H_R(mSv\,y^{-1}) = D_R\,(nGy\,h^{-1}) \times 0.00123 \quad \text{-- (3)}$$

Where D_R (nGy h^{-1}) is given by equation (2). The obtained values of annual effective dose rate for all sand samples are listed in the 7th column of Table 1. Its values varied between 0.0694 (SA-12) and 0.547 (SA-1) with a mean value of 0.205 in mSv y^{-1}. The estimated mean value of H_R is lower than the world average value which is 0.45 mSv y^{-1} [12]. It indicated that sand samples can be used safely for building constructions.

3.5. Criteria formula

Based on models suggested by Krisiuk et al., 1971[21] and Stranden, 1976 [22], a value of 1.5 mGy was obtained by Kreiger, 1981 [23] when evaluating the annual external radiation dose inside dwellings constructed of building materials with a Ra_{eq} value of 370 Bq Kg^{-1}. These authors later corrected their calculations by taking into consideration a wall of finite thickness and applying a weighing factor of 0.7 [24] to account for the presence of windows and doors.

Their results can be used as a criterion to limit the annual radiation dose from building materials based on the formula

$$CF = \frac{A_{Ra}}{740\ Bq/kg} + \frac{A_{Th}}{520\ Bq/kg} + \frac{A_K}{9620\ Bq/kg} \quad \text{---(4)}$$

Where A_{Ra}, A_{Th} and A_K are the activities of ^{226}Ra, ^{232}Th and ^{40}K respectively in units of Bq Kg^{-1}.

The calculated CF values from the sum of the three quotients for the annual radiation dose associated with the studied samples are given in the 5th column of Table 2. The criteria formula values are ranging from 0.0797 (SA-12) to 0.7554

(SA-1) with an average of 0.2762. All the studied samples are well below the recommended maximum value (<1). This indicates that the gamma activities in the studied sand samples do not exceed the proposed criterion level and the sand can be used for constructions purpose.

3.6. Radiation Hazard Indices

Beretka and Mathew, (1985) [13] defined two indices that represent (i) the external radiation hazard, H_{ex}, and (ii) the internal radiation hazard, H_{in}, which are discussed in the following section. The prime objective of these indices is to limit the radiation dose to a dose equivalent limit of 1 mSv y^{-1}.

3.6.1. External radiation hazard (H_{ex})

The external hazard index is defined as exposure of gamma rays emitted from radionuclides and also to assess the radiological suitability of a material. Beretka and Mathew (13) introduced a hazard index for the external gamma radiation dose from building materials as given below:

$$H_{ex} = \frac{A_{Ra}}{370\ Bq/kg} + \frac{A_{Th}}{259\ Bq/kg} + \frac{A_k}{4810\ Bq/kg} \leq 1 \quad \text{--- (5)}$$

Where A_{Ra}, A_{Th} and A_K are the activities of ^{226}Ra, ^{232}Th and ^{40}K respectively in the units of Bq kg^{-1}.

The calculated external hazard is listed in 6^{th} column of Table 2 and its minimum and maximum values are ranging from 0.1596 (SA-12) to 1.5162 (SA-1) with the mean value of 0.5543. All the values of H_{ex} are less than unity except (SA-1) indicating that the radiation hazard to be negligible and the studied samples can be used for construction purposes.

3.6.2. Internal radiation hazard (H_{in})

The internal hazard index (H_{in}) gives the internal exposure to carcinogenic radon and its short-lived progeny and it is given by the following formula [13, 17]:

$$H_{in} = \frac{A_{Ra}}{185\ Bq/kg} + \frac{A_{Th}}{259\ Bq/kg} + \frac{A_k}{4810\ Bq/kg} \leq 1$$

The value of H_{in} must also be less than unity to have negligible hazardous effects of radon and its short-lived progeny to the respiratory organs [12]. The calculated values of this index are given in7^{th} column of Table 2.

It was found that the internal hazard index values are varied between 0.1596 (SA-12) to 1.5162 (SA-1) with the mean value of 0.5666. The hazard indices (H_{in}) in most of the samples are less than unity (permissible level) [25]. This indicated that studied samples can be used as a construction material in the building of houses.

Conclusion

The average specific activity concentration of ^{226}Ra and ^{40}K is lower and ^{232}Th is higher in sand samples when compared with world average value. The potential radiological hazards associated with sand samples were identified by computing radiation indices. All the radiation indices are well below their recommended limits. Hence the investigated sand in this study can be recommended for safe usage for dwelling construction.

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Table. 1. Different located sand samples with activity concentration and other calculated radiological parameters

Sample ID	Activity concentrations (Bq Kg^{-1})			Radium equivalent Ra_{eq} (Bq Kg^{-1})	Absorbed dose rate D_R (nGyh^{-1})	Annual effective dose rate H_R (mSv y^{-1})
	^{226}Ra	^{232}Th	^{40}K			
SA-1	BDL	358.56	633.94	561.55	445.13	0.5475

SA-2	BDL	121.37	439.73	207.42	168.69	0.2075
SA-3	BDL	93.70	322.16	158.80	128.84	0.1585
SA-4	7.96	33.58	300.87	79.15	68.33	0.0840
SA-5	7.47	27.09	293.21	68.79	60.13	0.0740
SA-6	15.33	196.71	339.29	322.75	257.63	0.3169
SA-7	13.58	187.42	303.26	304.94	242.92	0.2988
SA-8	10.3	184.9	282.35	296.45	235.45	0.2896
SA-9	8.71	83.49	302.68	151.41	124.07	0.1526
SA-10	5.2	28.08	262.11	65.54	56.64	0.0697
SA-11	BDL	68.84	351.78	125.53	103.87	0.1278
SA-12	BDL	12.99	526.42	59.11	56.40	0.0694
SA-13	BDL	94.64	395.56	165.79	135.75	0.1670
SA-14	BDL	121.25	554.11	216.05	177.70	0.2186
SA-15	BDL	178.66	524.27	295.85	238.47	0.2933
MEAN	**4.57**	**119.42**	**388.78**	**205.27**	**166.67**	**0.2050**

Table. 2. Different located sand samples with activity concentration and other calculated radiological parameters

Sample ID	Activity concentrations (Bq Kg^{-1})			Criteria formula	Hazard indices	
	^{226}Ra	^{232}Th	^{40}K	CF	H_{ex}	H_{in}
SA-1	BDL	358.56	633.94	0.7554	1.5162	1.5162
SA-2	BDL	121.37	439.73	0.2791	0.5600	0.5600
SA-3	BDL	93.70	322.16	0.2137	0.4288	0.4288
SA-4	7.96	33.58	300.87	0.1066	0.2137	0.2352
SA-5	7.47	27.09	293.21	0.0927	0.1857	0.2059
SA-6	15.33	196.71	339.29	0.4343	0.8715	0.9129
SA-7	13.58	187.42	303.26	0.4103	0.8234	0.8601
SA-8	10.3	184.9	282.35	0.3988	0.8004	0.8283
SA-9	8.71	83.49	302.68	0.2038	0.4088	0.4324
SA-10	5.2	28.08	262.11	0.0883	0.1770	0.1910
SA-11	BDL	68.84	351.78	0.1690	0.3389	0.3389
SA-12	BDL	12.99	526.42	0.0797	0.1596	0.1596
SA-13	BDL	94.64	395.56	0.2231	0.4476	0.4476
SA-14	BDL	121.25	554.11	0.2908	0.5833	0.5833
SA-15	BDL	178.66	524.27	0.3981	0.7988	0.7988
MEAN	**4.57**	**119.42**	**388.78**	**0.2762**	**0.5543**	**0.5666**

Table. 3. Comparison of activity concentrations and radium equivalent (Bq Kg^{-1}) for sand samples in different areas of the world

Country	^{226}Ra Bq Kg^{-1}	^{232}Th Bq Kg^{-1}	^{40}K Bq Kg^{-1}	Ra_{eq} Bq Kg^{-1}	Reference
Australia	3.7	40	44.4	65.3	[13]
China	39.4	47.2	573	151	[14]
Hong Kong	24.3	27.1	841	128	[14]
Brazil	14.3	18	807	102	[26]

Netherland	8.1	10.6	200	38.6	[27]
USA	37	33.3	18.5	86	[28]
Cuba	17	16	208	55	[29]
Egypt	9.2	3.3	47.3	16.6	[30]
Turkey	6.7	6.7	882.9	84.3	[31]
Pakistan	21.5	31.9	520	107	[32]
Pakistan	20	29	383	91	[1]
India	43.7	64.4	455.8	170.8	[33]
India/Haryana	63	96	824	263	[34]
World	35	30	400	-	UNSCEAR (2000)[12]
India/ Tiruvannamalai	**4.57**	**119.42**	**388.78**	**205.27**	**Present work**

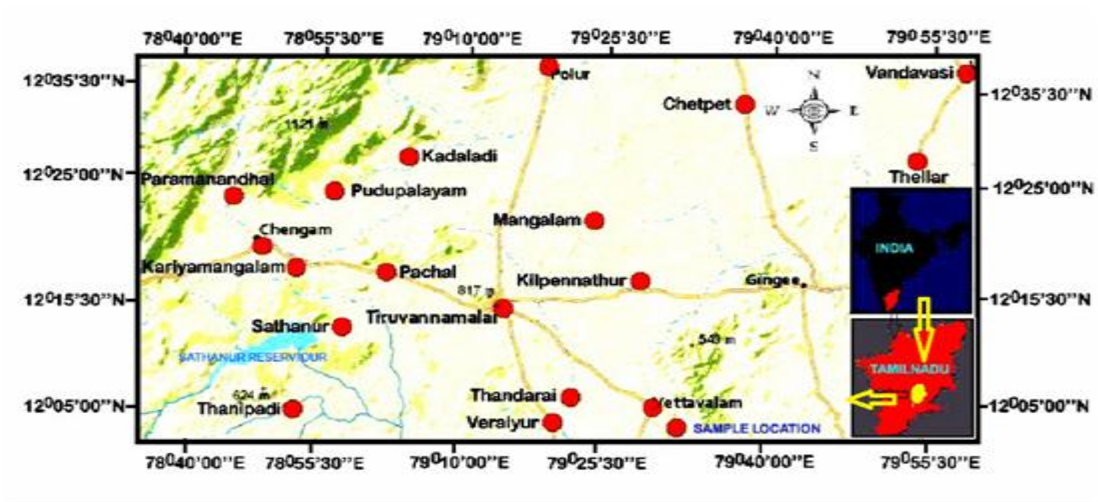

Fig. 1. Location map of Tiruvannamalai district