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Distribution of natural and anthropogenic radionuclides in soil and beach sand samples of Kalpakkam (India) using hyper pure germanium (HPGe) gamma ray spectrometry

V. Kannan[a,*], M.P. Rajan[a], M.A.R. Iyengar[a,1], R. Ramesh[b]

[a] *Environmental Survey Laboratory, Health Physics Division, BARC, Kalpakkam–603 102, India*
[b] *Institute for Ocean Management, Anna University, Chennai-600 025, India*

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Abstract

Pre-operational survey at Kalpakkam coast, indicated elevated gamma background radiation levels in the range of 100–4000 nGy h^{-1} over the large tracts of the coastal sands due to the presence of pockets of monazite mineral in beach sands. In view of the prevalence of monazite, a systematic gamma spectrometric study of distribution of natural radionuclides in soil and beach sand samples collected from the terrestrial and coastal environment of Kalpakkam was performed and concentrations of primordial radionuclides such as ^{238}U, ^{232}Th and ^{40}K and anthropogenic radionuclide ^{137}Cs were determined. The concentrations of ^{238}U, ^{232}Th and ^{40}K in soil samples were 5–71, 15–776 and 200–854 Bq kg^{-1} dry, respectively. In beach sand samples, ^{238}U, ^{232}Th and ^{40}K contents varied in the range of 36–258, 352–3872 and 324–405 Bq kg^{-1} dry, respectively. The total absorbed gamma dose rate in air due to the presence of ^{238}U, ^{232}Th and ^{40}K in Kalpakkam soil samples varied between 24 and 556 nGy h^{-1} with a mean of 103 nGy h^{-1}. The contribution to the total absorbed gamma dose rate in air in the decreasing order was due to the presence of ^{232}Th (76.4%), followed by ^{40}K (16.9%) and ^{238}U (6.7%) in Kalpakkam soils. However, in beach areas of Kalpakkam, the presence of ^{232}Th in beach sand contributed maximum (94.0%) to the total absorbed gamma dose rate in air followed by ^{238}U (4.7%) and minimum contribution was by ^{40}K (1.3%). ^{137}Cs in Kalpakkam soils ranged from $\leqslant$1.0 to 2.8 Bq kg^{-1} dry, which was 1–3 order of magnitude less than the concentration of primordial radionuclides in soil.

Keywords: Natural and anthropogenic radionuclides in soil and beach sand; HPGe gamma spectrometry; ^{238}U; ^{232}Th; ^{40}K; and ^{137}Cs; Absorbed gamma dose rate in air

1. Introduction

Radiation is present in every environment of the Earth's surface, beneath the Earth and in the atmosphere. According to UNSCEAR (1993), about 87% of the radiation dose received by mankind is due to natural radiation sources and the remaining is due to anthropogenic radiation. There are few areas such as Austria, Brazil, China, France, India and Iran in the world, where the background radiation levels were found to be high, varying over an order of magnitude depending upon the site-specific terrestrial radioactivity (Roser and Cullen, 1964; Brazilian Academy of Sciences, 1977; Hanson and Komarov, 1983; Sunta et al., 1982). Cosmic radiation, and terrestrial radiation arising out of earth's crust and building materials used for construction of houses and buildings give rise to the external exposure and inhalation or ingestion of natural/man-made radionuclides that are present in the environment forms due to internal exposure. Therefore, at the site of any nuclear

*Corresponding author. Fax: +91-4114-803-16.
E-mail address: vkannan@igcar.ernet.in (V. Kannan).
[1] Former address (now retired).

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power station, knowledge of the distribution pattern of both natural and anthropogenic radionuclides is an essential pre-requisite for evaluation and control of public exposures. A predominant part of the radioactivity in the upper layers of the soil is derived from the decay of the primordial radionuclides, ^{238}U, ^{232}Th, ^{40}K and ^{87}Rb. Significant amounts of man-made radionuclides, ^{137}Cs and ^{90}Sr, are also present in the soil as a result of nuclear weapon tests and nuclear accidents. An attempt is made in this paper to determine the concentration of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in soil and beach sand samples collected from Kalpakkam using HPGe gamma ray spectrometer and to compute the total absorbed gamma dose rate in air due to the presence of ^{238}U, ^{232}Th and ^{40}K in the samples.

2. Area of study

Fig. 1 shows a map of Kalpakkam located about 65 km south of Chennai city, on the east coast of peninsular India, which is a major nuclear complex comprising of Madras Atomic Power Station (MAPS), a Fast Breeder Test Reactor (FBTR), a Centralized Waste Management Facility (CWMF), a Reprocessing and Development Laboratory (RDL), Kalpakkam Reprocessing Plant (KARP) and a host of allied laboratories. A pre-operational survey conducted in 1974 in the beach area of Kalpakkam revealed comparatively elevated levels of ambient gamma radiation background in the range of 100–4000 nGy h^{-1}. The gamma spectrometric analysis and mineralogical

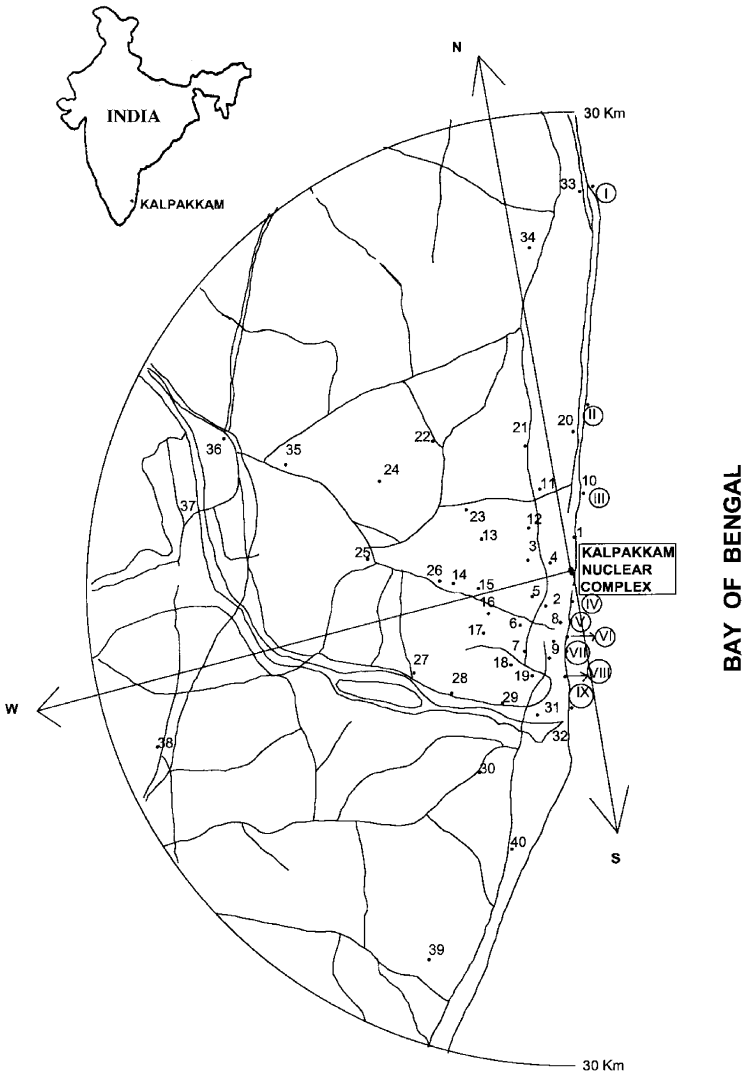

Fig. 1. Map of Kalpakkam environment with sampling locations.

analysis led to the conclusion that the beach sand at Kalpakkam contained monazite mineral (~0.6%) (Iyer et al., 1974).

3. Materials and methods

Surface soil samples were collected from 40 locations, believed to be undisturbed, in Kalpakkam and its neighbouring environments covering unto 30 km of radius (marked with nos. 1–40 in Fig. 1). Beach sand samples collected from nine locations on the seashore of Kalpakkam covering a distance of 20 km stretch on either side of MAPS are shown in Fig. 1 (marked with nos. I–IX). One beach sand sample was also collected from Marina beach, Chennai, located about 65 km north of Kalpakkam. Each sample (soil/beach sand) was collected by employing a template method from 1 m × 1 m area upto a depth of 5 cm, pooling the whole sample together and an aliquot of about 2 kg sample was collected after mixing thoroughly. Ambient gamma radiation level was measured at each sampling location, while collecting the samples, using a scintillometer (ECIL brand-SM-141D), which comprised NaI (1.75″ × 2″) detector with a reading range of 0–10000 nGy h^{-1}. The scintillometer was calibrated at regular intervals using standard ^{137}Cs and ^{60}Co sources. All the readings were taken at 1 m above ground level.

After collection, each sample was dried in an oven at 100–110°C for about 24 h and sieved through a 2-mm mesh-sized sieve to remove stone, pebbles and other macro-impurities. The homogenised sample was placed in a 250 ml airtight PVC container. The inner lid was placed in and closed tightly with outer cap. The container was sealed hermetically and externally using a cellophane tape and kept aside for about a month to ensure equilibrium between ^{226}Ra and its daughters and ^{228}Ra and its daughters before being taken for gamma spectrometric analysis.

The concentrations of primordial radionuclides (^{238}U, ^{232}Th and ^{40}K) and anthropogenic radionuclide ^{137}Cs in the sample were determined by employing a high-resolution hyper pure germanium (HPGe) gamma ray spectrometer system consisting of a p-type intrinsic germanium coaxial detector (type: IGC 30; volume 133 cc; PGT make) mounted vertically and coupled to a 4 K multichannel analyser (ORTEC MODEL 7450). The detector was housed inside a massive lead shield to reduce the background of the system. It was calibrated using a standard solution of ^{226}Ra in equilibrium with its daughters (obtained from NBS, USA), mixed with simulated soil matrix and counted in the same geometry as that of the soil samples. Three IAEA standard reference materials (a standard soil of known radioactivity—Soil-6, a Uranium ore sample—RGU1 and a Thorium ore sample—RGTh1) were also used for checking the calibration of the system. The energy resolution of 2.0 keV and relative efficiency of 33% at 1.33 MeV was achieved in the system.

Each sample, after the equilibrium, was kept on top of the HPGe detector and counted for period of 50000 s. The activity of ^{238}U was evaluated from the gamma ray 609 keV of ^{214}Bi peak, while 911 keV gamma line of ^{228}Ac peak was used to determine ^{232}Th. ^{40}K activity was determined from ^{40}K peak at 1461 keV and ^{137}Cs was determined using te 662 keV peak. The activity of each radionuclide in the sample was determined using the total net counts under the selected photopeaks after subtracting appropriate background counts, and applying appropriate factors for photopeak efficiency, gamma intensity of the radionuclide and weight of the sample. The analysis of the gamma spectra obtained was performed using the dedicated software Microsoft Excel. The minimum detectable activity (MDA) for each radionuclide was determined from the background radiation spectrum for the counting time of 50,000 s and was estimated (3σ) to be 3 Bq kg^{-1} for ^{238}U, 5 Bq kg^{-1} for ^{232}Th, 38 Bq kg^{-1} for ^{40}K and 1 Bq kg^{-1} for ^{137}Cs.

4. Results and discussion

4.1. Primordial radionuclides in soil

Data on activity concentration of primordial radionuclides such as ^{238}U (assuming secular equilibrium between ^{238}U, ^{226}Ra and their progenies), ^{232}Th and ^{40}K in soil (all values reported as Bq kg^{-1} dry) collected from each location of the terrestrial environment of Kalpakkam are given in Table 1. The associated statistical error, at 68% confidence level, was $<10\%$ for ^{238}U and ^{232}Th and $<20\%$ for ^{40}K and ^{137}Cs. The lowest concentration of ^{238}U (5 Bq kg^{-1}) was observed at Arambakkam and the highest (71 Bq kg^{-1}) was recorded at Devneri. Similarly, the lowest (15 Bq kg^{-1}) and highest (776 Bq kg^{-1}) levels of ^{232}Th were found at Maduranthagam and Meyyur, respectively. In most locations, both ^{238}U and ^{232}Th concentrations in soil varied in the range of 5–20 and 15–94 Bq kg^{-1}, respectively. However, significantly higher levels of ^{238}U (43–71 Bq kg^{-1}) and ^{232}Th (591–776 Bq kg^{-1}) activities were found in the soil samples collected from the locations, Kokkilimedu, Meyyur, Sadras and Devneri, which are very close to the beach and hence there is a possibility of inclusion of monazite-bearing beach sand with the soil collected from these locations. ^{40}K activity in soil varied widely between 200 and 854 Bq kg^{-1} due to heterogeneous soil characteristics, the lowest being at Amaipakkam and the highest being at Keelakalani. In the case of ^{40}K, the data did not indicate any particularly special distribution trend as observed in the case of ^{238}U and ^{232}Th activities in soil samples.

Table 1
Activity of primordial radionuclides in the soil from Kalpakkam environment

Sr. no.	Location	Activity ($Bq\,kg^{-1}$)			Absorbed gamma dose rate ($nGy\,h^{-1}$) due to						
		^{238}U	^{232}Th	^{40}K	^{238}U	^{232}Th	^{40}K	Total	Cosmic ray contribution[a]	Aggregate total[b]	Total observed by scintillometer
1	Kokkilimedu	61	724	225	26	479	10	515	32	547	400
2	Keelankalani	10	37	854	4	25	37	66	32	98	100
3	Manamai	10	54	826	4	36	36	76	32	108	100
4	Kunnathur	13	63	326	5	41	14	61	32	93	130
5	Arambakkam	5	37	300	2	24	13	39	32	71	60
6	Vengampakkam	9	23	275	4	15	12	31	32	63	100
7	Poonthandalam	13	41	577	6	27	25	58	32	90	160
8	Meyyur	59	776	403	25	514	17	556	32	588	400
9	Sadras	43	591	255	18	391	11	421	32	453	380
10	Mahabalipuram	14	94	321	6	62	14	82	32	114	200
11	Poonjeri	19	85	486	8	56	21	85	32	117	100
12	Kadambadi	13	29	309	6	19	13	38	32	70	70
13	Melakkuppam	8	55	262	4	36	11	51	32	83	100
14	Kariacheri	8	41	272	3	27	12	42	32	74	80
15	Amaipakkam	9	65	200	4	43	9	56	32	88	110
16	Neikuppi	12	64	511	5	42	22	69	32	101	180
17	Perambakkam	6	33	209	3	22	9	33	32	65	60
18	Vitilapuram	13	87	659	6	57	28	91	32	123	120
19	Pudupattinam	13	75	505	5	50	22	77	32	109	110
20	Devneri	71	761	323	30	504	14	548	32	580	400
21	Paiyanur	14	45	348	6	30	15	50	32	82	80
22	Manamathi	14	53	350	6	35	15	56	32	88	100
23	Kulpanthandalam	10	88	266	4	58	11	74	32	106	100
24	Oragadam	15	50	290	6	33	12	52	32	84	100
25	Tirukalikundram	10	57	338	4	38	15	56	32	88	120
26	Mullikolathur	11	68	309	5	45	13	63	32	95	80
27	Nerumbur	15	63	612	6	41	26	74	32	106	150
28	Panankattu cheri	14	43	521	6	28	22	57	32	89	100
29	Ayapakkam	16	60	472	7	40	20	67	32	99	100
30	P. Managalam	11	63	567	5	42	24	71	32	103	100
31	Voyalur	9	43	423	4	28	18	51	32	83	150
32	Kadalur	9	55	477	4	36	20	61	32	93	120
33	Kovalam	6	20	390	3	13	17	33	32	65	90
34	Thaiyur	12	52	371	5	34	16	56	32	88	100
35	Nemmeli	7	33	241	3	22	10	35	32	67	60
36	Chingleput	15	47	460	7	31	20	58	32	90	100
37	Mamandur	9	32	318	4	21	14	39	32	71	80
38	Maduranthagam	10	15	231	4	10	10	24	32	56	50
39	Cheyyur	20	69	593	8	46	25	79	32	111	100
40	Mugaiyur	10	62	555	4	41	24	69	32	101	120

[a] Sea-level cosmic background radiation value at Chennai as given by Nambi et al. (1987).
[b] Including cosmic ray component.

From the activities given in Table 1, activity ratios of ^{232}Th and ^{238}U were computed for all the soil samples. These ratios ($^{232}Th/^{238}U$) varied between 1.5 and 13.7 (mean: 5.5) for all the soil samples indicating that the concentration of ^{232}Th is greater than that of ^{238}U. However, $^{232}Th/^{238}U$ ratio was found to be 1.5–9.1 (mean: 4.8) when the data belonging to the above-mentioned four locations that are closer to the beach were excluded. The correlation between ^{232}Th and ^{238}U in soil samples (after excluding the data from the above-mentioned four locations) is found to be weak ($R = 0.47$) as shown in Fig. 2a indicating that the presence of monazite mineral in soil samples is less likely.

Table 2 lists the statistical data (range, arithmetic mean with standard deviation, geometric mean, GM, GSD, median, mode, skewness, kurtosis co-efficient and the type of frequency distribution) for these radio-

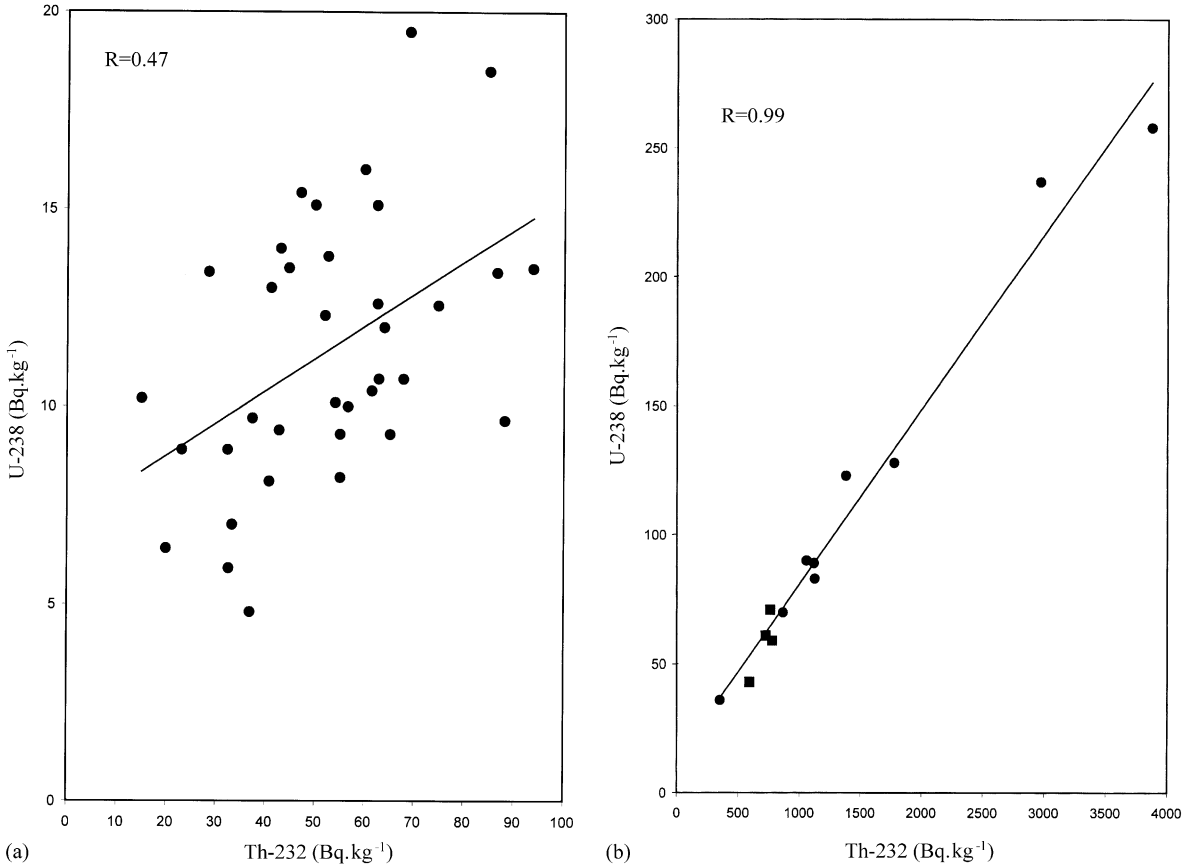

Fig. 2. (a) Correlation between ^{232}Th and ^{238}U in Kalpakkam soils by gamma-ray spectrometry. (b) Correlation between ^{232}Th and ^{238}U in Kalpakkam beach sand samples by gamma-ray spectrometry.

Table 2
Statistical data for radioactivity concentrations of ^{238}U, ^{232}Th and ^{40}K (Bq kg^{-1}) in surface soil samples from the Kalpakkam environment

	^{238}U	^{232}Th	^{238}U*	^{232}Th*	^{40}K
Range	5–71	15–776	5–20	15–94	200–854
Arithmetic Mean (AM)	16	119	11	53	406
Standard Deviation (SD)	15	203	3	19	160
Geometric Mean (GM)	13	64	11	49	377
Geometric Standard Deviation (GSD)	1.7	2.2	1.4	1.5	1.4
Median	12	55	11	53	349
Mode	9	55	9	55	348
Skewness	2.8	2.8	0.3	0.2	1.1
Kurtosis	6.8	6.3	−0.0001	−0.30	0.82
Frequency distribution	Log-normal	Log-normal	Normal	Normal	Log-normal

Data with * mark were calculated after excluding values for the four locations, Kokkilimedu, Meyyur, Sadras and Devneri, which were showing high activities.

nuclides in the soil samples. Among the three natural radionuclides studied in the soil samples (Table 2), the mean concentration observed was the highest for ^{40}K (406 Bq kg^{-1}), followed by ^{232}Th (119 Bq kg^{-1}) and the lowest mean activity was observed for ^{238}U (16 Bq kg^{-1}). It can be observed from Table 2 that the values of skewness and kurtosis co-efficients for all the three radionuclides are not closer to the null value, indicating

the non-existence of normal distribution. The positive values of kurtosis coefficient given for these radionuclides in Table 2 also indicate that the distributions are higher and narrower than normal. Table 2 also include statistical data calculated for ^{238}U and ^{232}Th activities in soil samples after excluding those data belonging to the four locations, Kokkilimedu, Meyyur, Sadras and Devneri, which are very close to the beach and whose composition of the collected soil from these locations contained huge amount of beach sand. It can also be observed that the values of skewness coefficient and kurtosis co-efficient for ^{238}U and ^{232}Th activities are closer to the null value, after the exclusion of those above-mentioned locations, indicating the existence of normal distribution and the activity distribution is practically symmetrical. This is also confirmed from symmetrical figures shown in Fig. 3a and b. The activity corresponding to ^{40}K fitted to lognormal distribution is shown in Fig. 3c.

Table 3 compares the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in soil samples observed Figs. 3a, b, c in the present study with those by other investigators in different locations of the world. From Table 3, it can be observed that the mean activity of ^{238}U (11 Bq kg^{-1}) for Kalpakkam soil is about 0.4 times lower than that of the world average (25 Bq kg^{-1}), whereas ^{232}Th activity of Kalpakkam soil (53 Bq kg^{-1}) in the present study is about 2 times higher than that of the world average (25 Bq kg^{-1}) as reported by UNSCEAR (1988). ^{40}K activity of Kalpakkam soil (406 Bq kg^{-1}) is found to be almost in the similar level as with that of the world average value of 370 Bq kg^{-1} (UNSCEAR, 1988). While the mean ^{238}U activity of (11 Bq kg^{-1}) in Kalpakkam soils observed in the present study is comparable with the all-India average value of 14.8 Bq kg^{-1}, the mean of ^{232}Th activity (53 Bq kg^{-1}) in the same soils is found to be about three times higher than that of the all-India average value of 18.3 Bq kg^{-1} (Mishra and Sadasivan, 1971). The data of both ^{238}U and ^{232}Th activities observed in the present study, however, are about 1–2 orders of magnitude lower than those collected at Ullal, Karnataka (Radhakrishna et al., 1993).

4.2. Anthropogenic radionuclides in soil

Anthropogenic radionuclide ^{137}Cs, measured in Kalpakkam soils in all the locations given in Table 1 by gamma spectrometry, was found to be in the range of $\leqslant$1.0–2.8 Bq kg^{-1} dry. The concentration of ^{137}Cs in soil is presumed to be the result of fallout of radioactivity from the atmospheric tests conducted around the world till 1962. In soil, the concentration of ^{137}Cs is found to be 1–3 orders of magnitude less than the concentrations of primordial radionuclides observed in the present study. ^{137}Cs in soil found in the present study is comparable with that in the soil at Chittagong (Bangladesh) as reported by Nurul Alam et al. (1990). However, ^{137}Cs is found to be comparatively lower than (Table 3) that reported by Miah et al. [18] for Dhaka city (Bangladesh) soils.

4.3. Primordial radionuclides in beach sand

Table 4 gives ^{238}U and ^{232}Th activity data in beach sand samples collected from different beach locations of the coastal environment of Kalpakkam that were in the range of 36–258 and 352–3872 Bq kg^{-1}, respectively. It can be observed from Table 4 that ^{238}U and ^{232}Th activities in beach sand are an order of magnitude higher than those observed in soil samples collected from Kalpakkam. This clearly indicates the presence of monazite in beach sand samples in a significant amount than in soil samples. However, as observed in Table 4, activity concentrations of ^{238}U, ^{232}Th and ^{40}K in the beach sand sample collected from Marina beach, Chennai (6, 37 and 322 Bq kg^{-1}, respectively), indicates insignificant presence of monazite in the beach sand sample. A narrow range of ^{40}K activity was observed in beach sand samples (324–405 Bq kg^{-1}), which was comparable with that observed in soil samples (200–854 Bq kg^{-1}). Higher ratio of ^{232}Th/^{238}U was observed in beach sand samples varying in the range of 9.8–15.0 with a mean of 12.5. A strong correlation exists between ^{232}Th and ^{238}U in beach sand samples ($R = 0.99$), as shown in Fig. 2b, indicating the presence of monazite mineral in beach sand samples. The data of soil samples collected from Kokkilimedu, Meyyur, Sadras and Devneri were also plotted in Fig. 2b (indicated by • marker style) and it was found that the ratios of ^{238}U to ^{232}Th were very close to the line indicating that the nuclides in these soil samples essentially originated from monazite. By introducing these soil data in Fig. 2b, the correlation coefficient also remained as 0.99 without any change. Comparatively similar range of concentrations of ^{238}U, ^{232}Th and ^{40}K were observed by many authors (Kalyani et al., 1990; Radhakrishna et al., 1993) in the beach sand samples with an exception of that collected in the beach sand samples from Spain [33,34], where the observed values were significantly lower (Table 3).

5. Dose calculation

UNSCEAR (1988) has given the dose conversion factors for converting the activity concentrations of ^{238}U, ^{232}Th and ^{40}K into doses (nGy h^{-1} per Bq kg^{-1}) as 0.427, 0.662 and 0.043, respectively. Using these factors, the total absorbed gamma dose rate in air at 1 m above the ground level is calculated as given in the equation below:

$$D = (0.427C_{\mathrm{U}} + 0.662C_{\mathrm{Th}} + 0.043C_{\mathrm{K}})\ \mathrm{nGy\,h^{-1}},$$

Table 3
Comparison of activity concentrations of ^{238}U, ^{232}Th, ^{137}K and ^{137}Cs in surface soil samples of Kalpakkam, India and different areas in the world

Location	^{238}U Bq kg^{-1} dry	^{232}Th	^{137}K	^{137}Cs	Reference
SOIL					
India					
Kalpakkam, Tamilnadu	5–71 (16)	15–776 (119)	200–854 (406)	⩽1.0–2.8	Present study
Kalpakkam, Tamilnadu[a]	5–20 (11)	15–94 (53)			Present study
Bhubaneswar, Orissa	18–30[b]	33–80	213–247	—	Vijayan and Behera (1999)
Coonoor (Ooty), Tamilnadu	BDL-49	4–224	14–731	—	Selvasekarapandian et al. (1999a)
Gudalore, Tamilnadu	17–62	19–272	78–596	—	Selvasekarapandian et al. (2000)
Narora, Uttar Pradesh	32–65	46–90	469–756	—	Verma et al. (1998)
Rawatbhata,Rajasthan	17–40	27–67	127–549	—	Verma et al. (1998)
Udagamandalam (Ooty) taluk, Tamilnadu	0–88	26–226	96–444	—	Selvasekarapandian et al. (1999b)
Ullal, Karnataka	546	2971	268	—	Radhakrishna et al. (1993)
Uttar Pradesh	12–25	20–25	538–1018	—	Mishra and Sadasivan (1971)
All India	*14.8*	*18.3*	—	—	Mishra and Sadasivan (1971)
Other Than India					
Antartica	6–12[b]	6–18	149–302	—	Baeza et al. (1994)
California, USA					Ingersoll (1983)
China	20–520	2–440	12–2190		UNSCEAR (1993)
Chittagong, Bangladesh	15–81	7–39	128–610	1–4	Nurul Alam et al. (1990)
Dhaka city, Bangladesh	21–43[b]	9–22	402–750	3–10	Miah et al. (1998)
Hong Kong	119	146	352	—	Yu et al. (1992)
Ireland	37	26	350	—	Mc-Aulay and Moran (1988)
Japan	32	54	794	—	Ching-Jiang Chen et al. (1993)
Nile Delta, Egypt	17	19	316		Ibrahiem et al. (1993)
Lake Nasser, Egypt	4–41	9–50	16–487	⩽0.01–16.6	Ibrahiem et al. (1995)
Rio Grande do Nore, Brazil	10–138	12–191	56–1972	—	Malanca et al. (1993)
Spain	8–310[b]	5–258	31–2040		Quindos et al. (1994)
Taiwan	30	44	431	—	Yu-Ming Lin et al. (1987)
Tripoli	10.5	9.5	270	—	Shenber (1997)
USA	9–1558	4–126	—	—	Myrick et al. (1983)
USA	4–140	4–130	100–700		UNSCEAR (1993)
Venezuela	15–37[b]	24–141	325–822	—	Palacios et al. (1998)
Venezuela	27	31	357	—	LaBrecque (1994)
World Average	*25*	*25*	*370*	—	UNSCEAR (1988)
BEACH SAND					
Kalpakkam, India	36–258 (124)	352–3872 (1613)	324–405 (358)		Present Study
Ullal, India	374	1842	158		Radhakrishna et al. (1993)
Visakhapatnam, India	100–400	300–3600	—	—	Kalyani et al. (1990)
Northeast Coast, Spain	5–19	5–44	136–1087	⩽0.1–1.0	Rosell et al. (1991)
Valencian Community, Spain	4–16[c]	1–11[d]	30–253	—	Navarro and Roldan (1994)

[a] —after excluding values for the four locations, Kokkilimedu, Meyyur, Sadras and Devneri, which were showing high activities.
[b] Data at different depths (30–200 m).
[c] —^{214}Bi
[d] —^{228}Ac
( ) figures in brackets are arithmetic means.

where C_U, C_{Th} and C_K are the activity concentrations (Bq kg^{-1}) of uranium, thorium and potassium in soil, respectively. It can be observed from Table 1 that the calculated total absorbed gamma dose rate due to the presence of ^{238}U, ^{232}Th and ^{40}K in soil varied between 24 (Maduranthagam) and 556 nGy h^{-1} (Meyyur). The

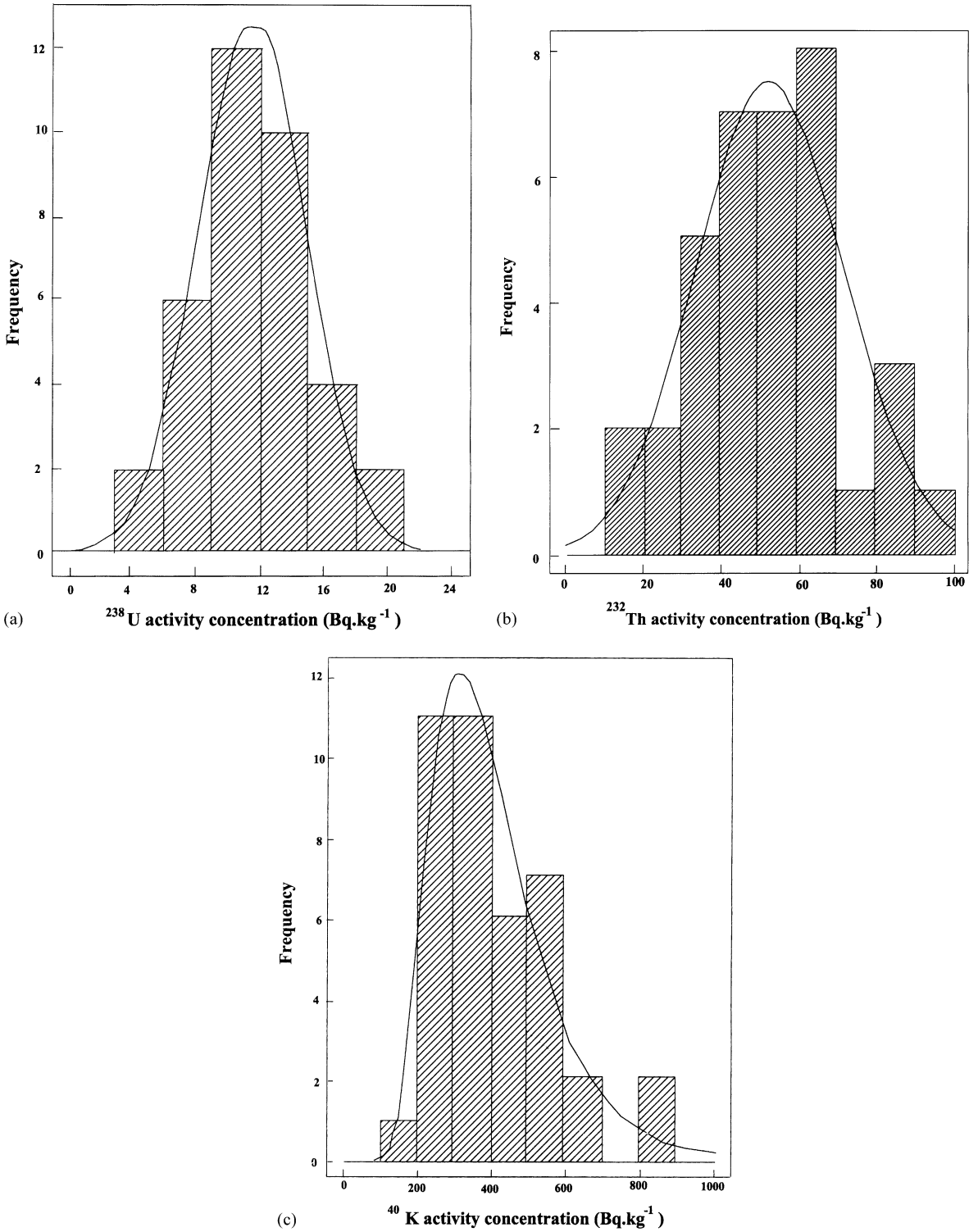

Fig. 3. (a) Frequency distribution of ^{238}U activity concentration in Kalpakkam soils. (b) Frequency distribution of ^{232}Th activity concentration in Kalpakkam soils. (c) Frequency distribution of ^{40}K activity concentration in Kalpakkam soils.

Table 4
Activity of primordial radionuclides in beach sand from the Kalpakkam environment

Sr. no.	Location	Activity ($Bq\,kg^{-1}$)			Absorbed gamma dose rate ($nGy\,h^{-1}$) due to						
		^{238}U	^{232}Th	^{40}K	^{238}U	^{232}Th	^{40}K	Total	Cosmic ray contribution[a]	Agregate total[b]	Total observed by scintillometer
I	Kovalam	36	352	352	15	233	15	264	32	296	240
II	Devneri	123	1381	332	53	914	14	981	32	1013	800
III	Mahabalipuram	237	2969	324	101	1965	14	2081	32	2113	2250
IV	DAE main gate	83	1126	330	35	745	14	795	32	827	750
V	Old Presteel qtrs	258	3872	373	110	2563	16	2689	32	2721	2000
VI	Meyyur kuppam	70	866	368	30	573	16	619	32	651	800
VII	Sadras Township	90	1059	345	38	701	15	754	32	786	850
VIII	Pudupattinam township	128	1776	405	55	1176	17	1248	32	1280	1200
IX	Kendriya vidyalaya-1	89	1120	389	38	741	17	796	32	828	800
X	Marina Beach, Chennai	6	37	322	3	24	14	41	32	73	100

[a] Sea-level cosmic background radiation value at Chennai as given by Nambi et al. (1987).
[b] Including cosmic ray component.

computed mean dose rate ($103\,nGy\,h^{-1}$) was found to be about 2.5 times higher than the world average ($43\,nGy\,h^{-1}$) as reported by UNSCEAR (1988). The contribution by each of the radionuclides ^{238}U, ^{232}Th and ^{40}K to the total external radiation dose rate was 6.7% ($7\,nGy\,h^{-1}$), 76.4% ($79\,nGy\,h^{-1}$) and 16.9% ($17\,nGy\,h^{-1}$), respectively. However, after excluding the values for four locations, Kokkilimedu, Meyyur, Sadras and Devneri, which were showing high activities, the range of calculated total absorbed gamma dose rate varied between 24 and $91\,nGy\,h^{-1}$ with a mean of $58\,nGy\,h^{-1}$ and the contribution by each radionuclide ^{238}U, ^{232}Th and ^{40}K to the total external radiation dose rate was 8.4% ($5\,nGy\,h^{-1}$), 60.5% ($35\,nGy\,h^{-1}$) and 31.1% ($18\,nGy\,h^{-1}$), respectively.

The total absorbed gamma dose rate in air in the beach areas of Kalpakkam is computed by applying the same conversion factors given by UNSCEAR (1988) and is found to be in range of 264–2689 $nGy\,h^{-1}$ (Table 4) with a mean of $1136\,nGy\,h^{-1}$. However, in the beach areas of Kalpakkam, the presence of ^{232}Th in beach sand contributed a maximum of 94.0% ($1068\,nGy\,h^{-1}$) to the total absorbed gamma dose rate in air followed by ^{238}U of 4.7% ($53\,nGy\,h^{-1}$) and the minimum contribution was by ^{40}K (1.3%, i.e. $15\,nGy\,h^{-1}$). The absorbed gamma dose rate computed for the beach area of Marina Beach, Chennai was found to be 1–2 orders of magnitude less than that observed for Kalpakkam beach areas.

The in situ gamma dose rate at 1 m above the ground has also been measured using the scintillometer in each location where soil sample was collected and are included in Table 1. It can be observed from the table that the maximum dose was obtained at Kokkilimedu and Meyyur ($400\,nGy\,h^{-1}$) and the minimum at Madur anthagam ($50\,nGy\,h^{-1}$). In general, scintillometer dose rates given in Table 1, compare well with the total gamma dose rates calculated by adding the individual dose due to ^{238}U, ^{232}Th and ^{40}K and the contribution by cosmic ray components (by applying the reported sea level cosmic background radiation value of $32\,nGy\,h^{-1}$ at Chennai by Nambi et al. (1987)) for Kalpakkam samples since Kalpakkam also is at sea level and closer to Chennai).

6. Conclusion

The concentrations of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in Kalpakkam soils were 5–71, 15–776, 200–854 and $\leqslant 1.0$–$2.8\,Bq\,kg^{-1}$ dry, respectively. The lowest concentration of ^{238}U ($5\,Bq\,kg^{-1}$) was observed in Arambakkam and the highest ($71\,Bq\,kg^{-1}$) was seen in Devneri. Similarly, the lowest ($15\,Bq\,kg^{-1}$) and highest ($776\,Bq\,kg^{-1}$) levels of ^{232}Th were found in Maduranthagam and Meyyur, respectively. Significantly higher levels of ^{238}U (43–$71\,Bq\,kg^{-1}$) and ^{232}Th (591–$776\,Bq\,kg^{-1}$) activities were found in the soil samples collected from the four locations, Kokkilimedu, Meyyur, Sadras and Devneri, which are very close to the beach and hence, the composition of the collected soil from these locations contained huge amounts of sand. After excluding those data belonging to these four locations, skewness and kurtosis co-efficients for ^{238}U and ^{232}Th activities were computed and were found to be closer to the null value indicating normal distribution and the activity distribution was practically symmetrical. The ratio of $^{232}Th/^{238}U$ varied in the range of 1.5–13.7 (mean: 5.5) in Kalpakkam soils. However, $^{232}Th/^{238}U$ ratio was found to be 1.5–9.1 (mean: 4.8) when the data belonging to the above-mentioned four locations, which are closer to the beach, were excluded. ^{238}U and ^{232}Th activities in beach sand samples are an order of magnitude higher than those of observed in soil samples, which can be traced to the presence of monazite in significant amounts in beach sand samples of Kalpakkam. A

higher activity ratio of $^{232}Th/^{238}U$ was observed in beach sand samples varying in the range of 9.8–15.0.

The total absorbed gamma dose rate calculated due to the presence of ^{238}U, ^{232}Th and ^{40}K in Kalpakkam soil samples varied from 24 to 556 nGy h^{-1} with a mean of 103 nGy h^{-1}. The highest contribution to total external radiation dose rate was due to ^{232}Th (76.4%) followed by ^{40}K (16.9%) and ^{238}U (6.7%). However, after excluding the values for four locations, Kokkilimedu, Meyyur, Sadras and Devneri, which were showing high activities, the range of calculated total absorbed gamma dose rate varied between 24 and 91 nGy h^{-1} with a mean of 58 nGy h^{-1} and the contribution by each of the radionuclides ^{238}U, ^{232}Th and ^{40}K to the total external radiation dose rate was 8.4% (5 nGy h^{-1}), 60.5% (35 nGy h^{-1}) and 31.1% (18 nGy h^{-1}), respectively. The total gamma dose rate in beach sand samples was found to be in range of 264–2689 nGy h^{-1}. In beach areas of Kalpakkam, the presence of ^{232}Th in beach sand contributed a maximum of 94.0% (1068 nGy h^{-1}) to the total absorbed gamma dose rate in air, followed by ^{238}U of 4.7% (53 nGy h^{-1}) and the minimum contribution was by ^{40}K (1.3%, i.e. 15 nGy h^{-1}).

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