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NATURAL RADIOACTIVITY AND ASSOCIATED DOSE RATES IN SOIL SAMPLES FROM KALPAKKAM, SOUTH INDIA

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The activity concentration of naturally occurring radioactive elements such as ^{226}Ra, ^{232}Th and ^{40}K were measured for 46 soil samples collected in the vicinity of the Madras atomic power station, Kalpakkam, South India using gamma-ray spectroscopy. The average activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in soil samples were found to be 22.6 ± 12.6, 92.8 ± 44.3 and 434.1 ± 131.1 Bq kg^{-1}, respectively. The activity concentration of natural radionuclides is higher than the world average except for ^{226}Ra. The external absorbed gamma dose rates due to ^{226}Ra, ^{232}Th and ^{40}K are observed to be 74.6 ± 30.8 nGy h^{-1} with a corresponding annual effective dose of 91.5 ± 37.8 μSv y^{-1}, which are also above the world average. The values of radium equivalent activity and external hazard index are less than the world average. Whereas, the values of the radioactivity level index (I_γ) and the total gamma dose rate were found to be above the required criterion.

INTRODUCTION

The natural radioactivity present in the environment is the main source of radiation exposure for humans and constitutes the background radiation level(1). Natural radionuclides in soil generate a significant component of the background radiation exposure to the population and gamma-radiation intensity in a region depends on soil and geomorphology(2, 3). The main natural contributors to external exposure from gamma rays are ^{226}Ra, ^{232}Th and ^{40}K. Since these radionuclides are not uniformly distributed, the knowledge of their distribution in soil and rocks play an important role in radiation protection and measurement(4). It is important to determine the sources and their individual contributions to the total radiation dose(5). Therefore, measurements of natural radioactivity in soil are of a great interest for many researchers throughout the world, which has led to worldwide national surveys in the last two decades(2, 6). There have been many surveys to determine the background levels of radionuclides in soils, which can in turn be related to the absorbed dose rates in air(7). All spectrometric measurements indicate that three components of the external radiation field, namely from the gamma-emitting radionuclides such as ^{226}Ra, ^{232}Th and ^{40}K made approximately equal contributions to the externally incident gamma-radiation dose to individuals in typical situations both outdoors and indoors(8).

In the present study, an attempt was made to identify and determine the natural radionuclide activity concentration in soil samples and to estimate the radiological effects and gamma-ray absorbed dose rate in the vicinity of Madras atomic power station (MAPS), Kalpakkam, South India.

MATERIALS AND METHODS

Study area

Kalpakkam (12°33′N and 80°11′E) is located about 65 km south of Chennai city, on the east coast of India (Figure 1). It is India's one of major nuclear complexes comprising the MAPS, a Fast Breeder Test Reactor, a Centralised Waste Management Facility, a Reprocessing and Development Laboratory, Kalpakkam Reprocessing Plant and a host of allied laboratories(7). The MAPS has twin units of capacity 220 MWe each. The area is bounded by the Bay of Bengal on the East, the Edaiyur Backwaters on the North and the Buckingham Canal to the West. The topography of the study area is slightly undulating with sand bars and depressions. The regional gradient is towards the eastern side. The altitude varies between <1 and 13 m above MSL(9). The geology of the area consists of Archean basement at the bottom, which is made up of charnockites which is overlain by recent alluvium. The depth of hard rock varies from 12 to 20 m below the ground. Lenses of clays and clay pockets are generally encountered in the alluvial formations. The soil in the area is essentially sand with sandy clay and soft disintegrated rock. The thickness of the sandy formations varies from 3 to 12 m(10). Owing to the fertile nature of the soil, agriculture is

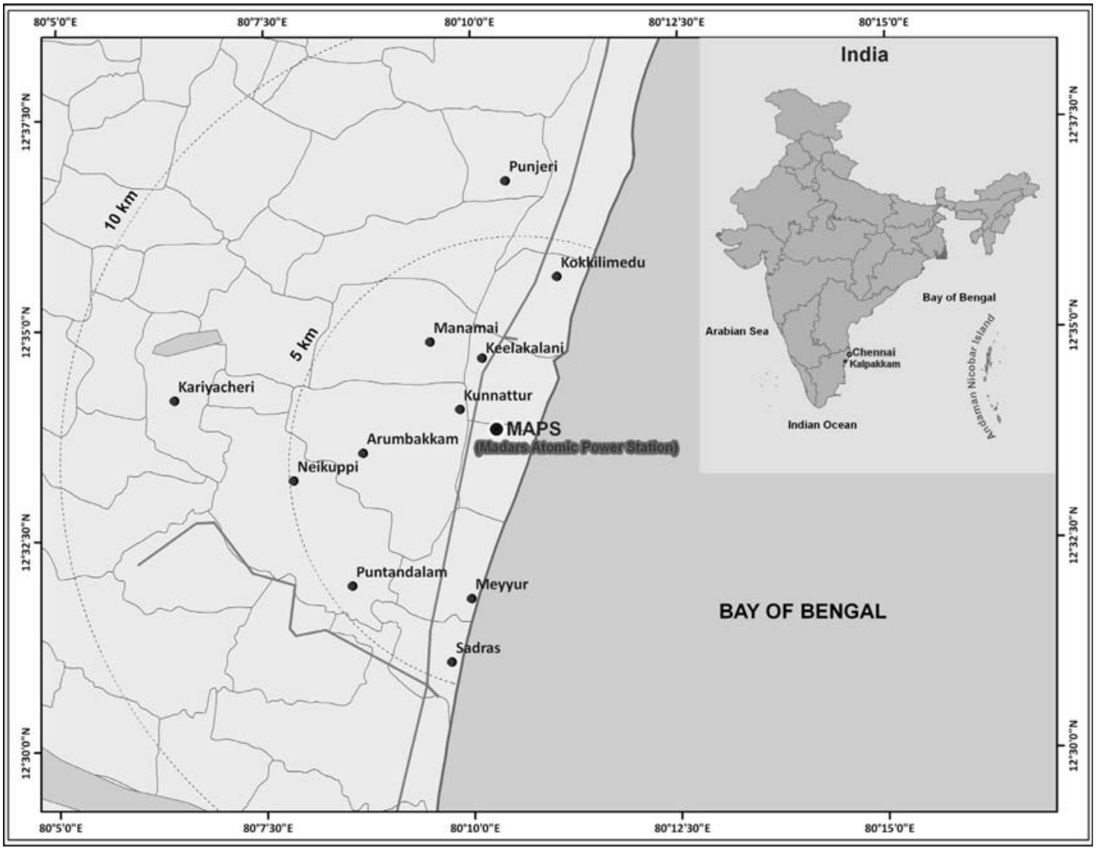

Figure 1. Map of Kalpakkam showing the sampling locations.

the primary landuse and fishing is predominant along this coast.

Gamma absorbed dose in air

The outdoor gamma-radiation levels were measured at 152 sites for 11 locations above the 1 m ground level using a Scintillometer (SM-141D). It is a battery operated portable instrument used to measure environmental radiation levels in the range between 0.025 and 10 μGy h^{-1} (2.5–1000 μR h^{-1}). It consists of a NaI (TI) crystals (size: $1.75' \times 2'$) coupled to a photomultiplier tube. The instrument is calibrated by using ^{137}Cs and ^{60}Co sources. In the present study, environmental radiation levels prevailing up to 7.5 km radius in the vicinity of the Madras Atomic Power Plant was measured.

Sampling of surface soil

In order to measure the radioactivity levels in soil, 46 surface soil samples were collected from uncultivated locations in the vicinity of MAPS, Kalpakkam, South India. At each location, the ground was cleared of stones, pebbles, vegetation and roots and about 1–2 kg of material from the first 10 cm of topsoil was collected and placed in a labelled polythene bag. The samples were dried in an oven at about 100°C for 24 h to remove the moisture content and then pulverised, homogenised and sieved through 1-mm mesh. Then, a sample of 250 g was weighed and finally, a split of the prepared sample was packed in standard plastic container (12×6.5 cm^2 of diameter) and then sealed with teflon tape. The plastic container were left for 4 weeks in order to maintain radioactive equilibrium between ^{226}Ra and its daughters before counting by gamma-ray spectrometry.

Gamma-ray detection system

The concentration of natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in soil samples were determined using a higher resolution hyper pure germanium

(HPGe) gamma-ray spectrometer system at the Environmental Survey Laboratory, Health Physics Division, Babha Atomic Research Centre, Kalpakkam, India. It consists of a p-type intrinsic germanium coaxial detector (type: IGC 30; volume 133 cc; PGT make) mounted vertically and coupled to an 8 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 FSoil-6, Uranium ore sample (RGU1) and 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 a period of 50 000 s. The activity of ^{226}Ra was evaluated from the gamma line 609 keV of ^{214}Bi peak, whereas the 911.1 keV gamma line of ^{228}Ac peak and 583.2 keV of ^{208}TI were used to determine ^{232}Th. ^{40}K activity was determined from ^{40}K peak at 1461.8 keV. 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 for each radionuclide was determined from the background radiation spectrum for the counting time of 50 000 s and was estimated to be 3 Bq kg^{-1} for ^{226}Ra, 5 Bqkg^{-1} for ^{232}Th and 38 Bq kg^{-1} for ^{40}K.

Calculation of radiological effects

In order to assess the health effects from the radioactivity of the earth's surface materials containing ^{226}Ra, ^{232}Th and ^{40}K, the activity of these nuclides is converted into a single quantity termed the radium equivalent activity (Ra$_{eq}$).The Ra$_{eq}$ is an index that has been introduced to represent the specific activities of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazards associated with them and the maximum value of Ra$_{eq}$ must be <370 Bq kg^{-1}. This index can be calculated according to Beretka and Mathew[(11)] as follows:

$$\mathrm{Ra_{eq}} = C_{\mathrm{Ra}} + 1.43\, C_{\mathrm{Th}} + 0.077\, C_{\mathrm{K}}, \qquad (1)$$

where C_{Ra}, C_{Th} and C_{K} are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. The external outdoor absorbed gamma dose rates [D (nGy h^{-1})] due to terrestrial gamma rays at 1 m above the ground level are calculated from the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K. The conversion factor described by UNSCEAR[(5)] was adopted and the gamma absorbed dose rates were calculated using the equation followed by Lu and Xiaolan[(12)] as given below.

$$D = (0.604\, C_{\mathrm{Th}} + 0.462\, C_{\mathrm{Ra}} + 0.0417\, C_{\mathrm{K}}). \qquad (2)$$

The annual average effective dose equivalent is calculated using a conversion factor of 0.7 Sv Gy^{-1}to convert the absorbed dose rate to the effective dose equivalent and 0.2 for the outdoor occupancy factor[(5)]. The first factor has been recommended by the UNSCEAR[(5)] which suggests that from the absorbed dose in air to the effective dose received by adults and considering that people in India, on the average, spent ~20 % of their time outdoors, the annual effective doses are calculated[(4, 7)]. This factor suits the pattern of life in the study area, yielding the outdoor effective dose rates. The effective dose rate (μSv y^{-1}) due to natural activity in the soil was calculated by:

$$\text{Effective dose} = \text{Dose rate}\,(\mathrm{nGy\,h^{-1}}) \times 8760\,(\mathrm{hy^{-1}}) \times 0.2 \times 0.7\,\mathrm{Sv\,Gy^{-1}} \times 10^{3}. \qquad (3)$$

The external hazard index (H_{ex}) is another radiation hazard index defined by Beretka and Mathew[(11)] to evaluate the indoor radiation dose rate due to the external exposure to gamma radiation from the natural radionuclides in construction building materials of dwellings. This index value must be less than unity to keep the radiation hazard insignificant, i.e. the radiation exposure due to the radioactivity from construction materials to be limited to 1.5 mSv y^{-1} based on the following criterion[(12)]:

$$H_{\mathrm{ex}} = \frac{C_{\mathrm{Ra}}}{370} + \frac{C_{\mathrm{Th}}}{259} + \frac{C_{\mathrm{K}}}{4810} \leq 1, \qquad (4)$$

where C_{Ra}, C_{Th} and C_{K} are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively.

The radioactivity level index, I_{γ}, can be used to estimate the gamma-radiation hazard levels typically for those associated with the natural radionuclides. The representative level of radiation hazard index can be estimated according to the equation adopted by Sroor *et al.*[(13)].

$$I_{\gamma} = \frac{C_{\mathrm{Ra}}}{100} + \frac{C_{\mathrm{Th}}}{150} + \frac{C_{\mathrm{K}}}{1500} \leq 1, \qquad (5)$$

The total gamma-radiation dose rate is modified to include the contributions from natural radionuclides and cosmic radiation according to the following equation[14]:

$$D_{\gamma} = 0.604\, C_{\mathrm{Th}} + 0.462\, C_{\mathrm{U}} + 0.0417\, C_{\mathrm{K}} + 34, \quad (6)$$

where C is the activity concentration of radionuclides measured. The number 34 is a factor included to ensure that the effects of cosmic rays are implemented.

RESULTS AND DISCUSSION

Radionuclide content in soil

The activity concentration of radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in soil samples collected from 11 locations around Kalpakkam is given in Table 1. The activity concentration of the ^{226}Ra range from 9.6 to 53.0 Bq kg^{-1} with an average value of 22.6 $\pm$ 12.6 Bq kg^{-1} and that of ^{232}Th ranges from 37.0 to 163.0 Bq kg^{-1} with an average value of 92.8 $\pm$ 44.3 Bq kg^{-1}. The activity concentration of ^{40}K ranges from 210.1 to 607.0 Bq kg^{-1} with an average value of 434.1 $\pm$ 131.1 Bq kg^{-1} and was found to be higher than that of both ^{226}Ra and ^{232}Th. The results obtained in this study for ^{232}Th and ^{40}K are slightly higher than the worldwide average concentration of these radionuclides in soils reported by UNSCEAR[5] which are 35, 30 and 400 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The associated statistical errors, at 69 % confidence level (1σ), was <10 % for ^{226}Ra and ^{232}Th and <20 % for ^{40}K.

From Table 1, it is also observed that the lowest concentration of ^{226}Ra (9.6 Bq kg^{-1}) and ^{232}Th (37. Bq kg^{-1}) was in Poonjeri and highest was recorded in Naikuppi (^{226}Ra 53.0 Bq kg^{-1}and ^{232}Th 163.0 Bq kg^{-1}). Significantly, higher levels of ^{226}Ra and ^{232}Th activities was found in soils collected from Kokkilamedu, Meyyur and Sadras is due to the fact that these locations are in close proximity to the beach and hence the composition of the collected soil from these areas contained significant quantities of monazite bearing beach sand[7, 15]. High activity of ^{40}K was observed in locations near the beach and lower activity was observed in the interior locations as observed in the case of ^{226}Ra and ^{232}Th. These results are also substantiated by statistical data for the natural radionuclide concentration in soil samples. It can be observed that the skewness and kurtosis coefficients for all radionuclides are not closer to the null value indicating asymmetric distribution of radionuclides (Table 2 and Figure 2).

Table 1. Average radioactivity concentration (in Bq kg^{-1}; n=46) in soil samples collected in Kalpakkam, South India.

Location	Distance from atomic power plant (km)	n	^{226}Ra	^{232}Th	^{40}K
Meyyur	4.0	5	18.6	70.8	290.3
Keelankalani	2.1	4	12.9	59.8	317.1
Kunnathur	2.5	3	29.3	148.6	607.0
Sadras	5	5	10.1	46.9	395.0
Manamai	2.5	3	12.1	64.1	210.1
Naikuppi	4.0	4	53.0	163.0	588.7
Kokkilimedu	5	4	28.0	78.0	474.0
Poonjeri	5.5	4	9.6	37.0	438.1
Poonthandalam	4.7	5	21.7	83.5	607.0
Arambakkam	2.1	5	26.8	143.4	411.7
Kariacheri	7.1	4	26.9	125.6	435.9

Table 2. Statistical data for radioactivity concentration of ^{226}Ra, ^{232}Th and ^{40}K and the absorbed dose rate for surface soil samples.

	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	Dose rate (nGy h^{-1})
Minimum	9.6	37.0	210.1	41.2
Maximum	53.0	163.0	607.0	131.9
Average	22.6	92.8	434.1	74.6
Standard deviation	12.6	44.3	131.1	30.8
Geometrical mean	19.9	83.3	414.3	69.1
Skewness	1.4	0.5	−0.1	0.6
Kurtosis	2.7	−1.4	−0.8	−0.8
Frequency distribution	Normal	Normal	Normal	Normal

Dose rate in air using scintillometer

The average outdoor gamma absorbed doses in air (n=152) at the height of 1 m above the ground level are given in Table 3. The values ranged from 4.7 to 9 μR $h^{-1}$with an average value of 6.5 ± 1.14 μR h^{-1}. The maximum value

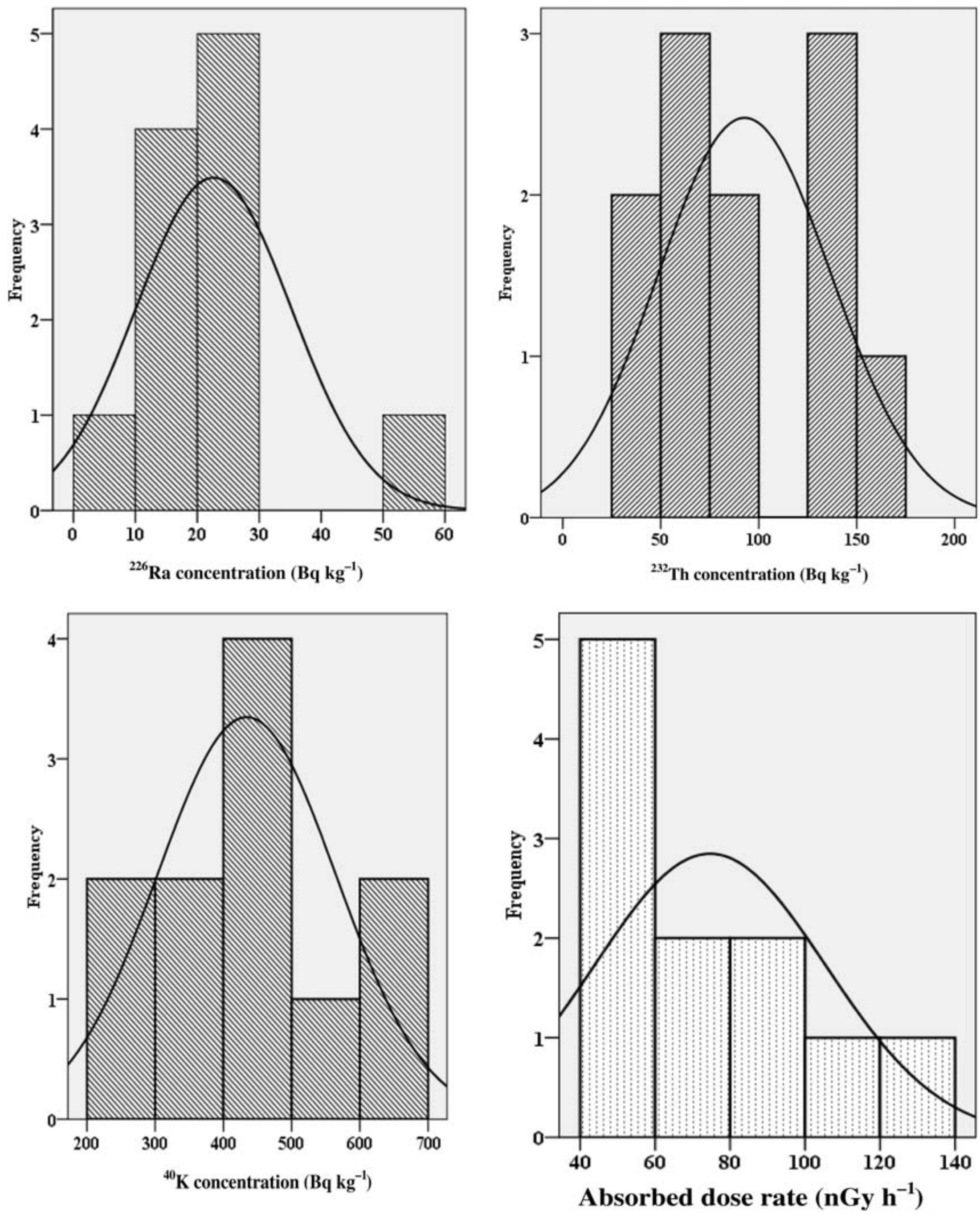

Figure 2. Frequency distributions of the activities of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1} dry weight) and relative frequency of distribution of the total gamma dose at 1 m above ground (nGy h^{-1}).

Table 3. The observed outdoor gamma absorbed doses in air using scintillometer (n=152) and the corresponding annual effective doses for the locations in Kalpakkam, South India.

Location	Latitude/ longitude	n	μR h^{-1}	Standard deviation	Minimum	Maximum	Dose rate (nGy h^{-1})	Effective dose (μSv y^{-1})
Meyyur	N12°37′377″/ E80°06′319″	12	7.1	2.0	4.1	10.0	61.6	75.6
Keelankalani	N12°34′739″/ E80°09′526″	22	5.6	1.6	3.0	9.0	49.4	60.6
Kunnathur	N12°34′163″/ E80°09′232″	10	6.8	2.2	4.0	10.0	59.2	72.6
Sadras	N12°31′550″/ E80°09′353″	12	6.3	2.2	3.0	9.0	55.1	67.6
Manamai	N12°35′403″/ E80°08′856″	30	7.5	2.1	3.0	15.0	65.5	80.3
Naikuppi	N12°33′390″/ E80°07′784″	13	9.0	3.5	4.0	18.0	78.3	96.0
Kokkilimedu	N12°38′205″/ E80°05′356″	22	5.9	1.3	4.0	8.0	51.6	63.3
Poonjeri	N12°36′588″/ E080°10′017″	12	4.7	2.1	3.0	9.0	51.6	63.3
Poonthandalam	N12°38′205″/ E80°05′358″	12	5.6	2.3	4.0	13.0	51.6	63.3
Arambakkam	N12°33′430″/ E80°08′419″	20	6.3	2.1	3.0	10.0	54.3	66.6
Kariacheri	N12°33′496″/ E80°07′669″	10	6.8	2.3	5.0	10.0	59.2	72.6

Table 4. The dose rate, effective dose rate, Ra_{eq}, radioactivity level index, total gamma-radiation dose rate and H_{ex} of the soil samples.

Location	Dose rate (nGy h^{-1})	Effective dose rate (μSv y^{-1})	Ra_{eq} activity (Bq kg^{-1})	I_γ (Bq kg^{-1})	D_γ	H_{ex}
Meyyur	56.1	68.7	142.2	0.85	90.1	0.38
Keelankalani	48.6	59.6	122.8	0.74	82.6	0.33
Kunnathur	111.7	136.9	288.5	1.69	145.7	0.78
Sadras	44.2	54.2	107.6	0.68	78.2	0.29
Manamai	45.7	56.0	119.9	0.69	79.7	0.32
Naikuppi	131.9	161.7	331.4	2.01	165.9	0.89
Kokkilimedu	72.7	89.2	176.0	1.12	106.7	0.48
Poonjeri	41.2	50.5	96.3	0.64	75.2	0.26
Poonthandalam	77.0	94.4	187.9	1.18	111.0	0.51
Arambakkam	99.6	122.2	263.6	1.50	133.6	0.71
Kariacheri	92.4	113.4	240.0	1.40	126.4	0.65
Minimum	41.2	50.5	96.3	0.64	75.2	0.26
Maximum	131.9	161.7	331.4	2.01	165.9	0.89
Average	74.6	91.5	188.8	1.13	108.6	0.51
Standard deviation	30.8	37.8	80.7	0.47	30.8	0.22

of 9 μR h^{-1} was observed in Naikuppi, which corresponds to a location with a relatively higher soil radioactivity. When the readings in μR h^{-1} are converted to nGy h^{-1} using the conversion factor of 8.7 nGy μR^{-1}[1], the average absorbed gamma dose for the study area is calculated to be 57.9 ± 8.4 nGy h^{-1} which is lower than the world average value of 60 nGy h^{-1}[5]. The gamma absorbed doses in nGy h^{-1} can further be converted to annual effective doses in mSv y^{-1} using appropriate conversion factors[5]. The calculated values of the annual effective dose due to gamma-radiation range from 60.6 to 96 μSv y^{-1} with an annual average of 71.1 ± 10.3 μSv y^{-1}, which is also lower than the world average of 80 μSv y^{-1}[5].

Dose rates and gamma-ray hazard indices

The values of Ra_{eq} for the soil samples are given in Table 4. The Ra_{eq} values for the soil samples varied from 96.3 to 331.4 Bq kg^{-1} with an average value of 188.8 ± 80.7 Bq kg^{-1}. All the Ra_{eq} values are less than the safe limit as recommended by the Organisation for Economic Cooperation and Development[16]. Accordingly, any Ra_{eq} concentration that exceeds 370 Bq kg^{-1} may pose radiation hazards.

The determined absorbed dose rates in air at 1 m above the ground surface due to the concentration of ^{226}Ra, ^{232}Th and ^{40}K in the soil samples are presented in Table 4. The outdoor gamma absorbed dose rate in air due to soil radioactivity ranged between 41.2 and 131.9 nGy h^{-1} with an average value of 74.6 ± 30.8 nGy h^{-1} was observed, which is higher than the world average value of 60 nGy h^{-1}[5]. The differences are considered to be due to the geology and landuse patterns, which vary from one place to another and from one locality to another in the same zone. The mean dose rate is important for determining radiation detriment to the population as a whole, but some members of the population may receive higher doses due to high concentration of radionuclides. A common feature in any environmental radiation measurement is the considerable variation in soil radioactivity with the location depending on soil physio-chemical parameters.

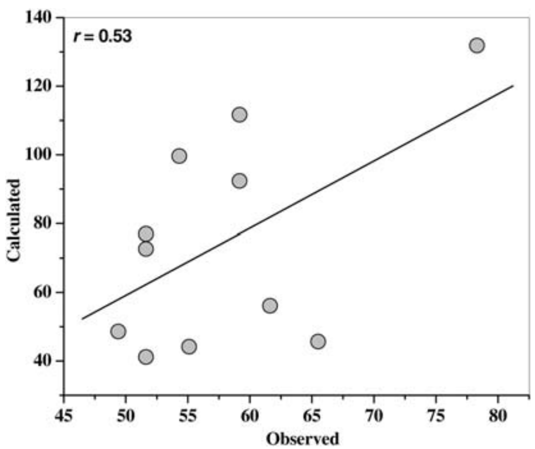

Figure 3. The correlation between calculated and observed dose rates (nGy h^{-1}).

The average relative contribution by individual components of natural radioactivity is 12.3 % from ^{226}Ra, 66.2 % from ^{232}Th and 21.4 % from ^{40}K. Therefore, the largest contribution from natural radionuclides in soil samples to the absorbed doses in air is due to ^{232}Th. The relationship between observed and calculated dose rates exhibit a positive correlation ($r = 0.53$) and the calculated dose rate are slightly lower than the *in situ* measurements in most of the sites. This small difference is to be expected, as the scintillometer is also responsive to high-energy beta particles and X-rays (Figure 3).

The corresponding outdoor annual effective dose rates range from 50.5 to 161.7 μSv y^{-1} with an average value of 91.5 ± 37.8 μSv y^{-1} was calculated, which is above the world average of 80 μSv y^{-1}[5]. Thus, the results are higher than the average worldwide limits. Indoor dose rates were not evaluated because the essential data on the average build-up of radon gas in the indoor atmosphere were not available.

The radioactivity level index I_γ is determined to estimate the gamma-radiation hazard associated with the natural radionuclides in the soil samples. The calculated values for most of the sampling locations were higher than the international values ($I_\gamma \geq 1$) and in the remaining areas the values are close to the upper limit (Table 4).

The calculated results of H_{ex} for the soil samples range from 0.26 to 0.89 with an average of 0.51 ± 0.22 (Table 4). These values are far below the criterion limit ($H_{ex} \leq 1$) as per the European Commission on Radiation Protection reports[14] and the terrestrial soils around MAPS has no high

Table 5. Comparison of activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in surface soil samples in the vicinity of MAPS, Kalpakkam with other parts of India.

Location	^{226}Ra	^{232}Th	^{40}K	Reference
Kalpakkam, South India	9.6–53	37.0–163.0	210.1–607.0	Present study
Kalpakkam, Tamilnadu	5–71	15–776	200–854	Kannan *et al.*[7]
Bhubaneswar, Orissa	18–30	33–80	213–247	Vijayan and Behera[17]
Coonoor (Ooty), Tamilnadu	BDL-49	4–224	14–731	Selvasekarapandian *et al.*[18]
Gudalore, Tamilnadu	17–62	19–272	78–596	Selvasekarapandian *et al.*[19]
Ullal, Karnataka	546	2971	268	Radhakrishna *et al.*[20]
All India	14.8	18.3	—	Mishra and Sadasivan[21]
World average	25	25	370	UNSCEAR[22]

exposure for the inhabitants and it can be used as a construction material without posing any significant radiological threat to the population.

The calculated total gamma-radiation dose rates [D_γ=(nGy h^{-1})] of natural radionuclides and cosmic radiation are presented in Table 4. As it can be seen from the table, the results of total gamma-radiation dose rates ranged between 75.2 and 165.9 nGy h^{-1} with an overall average value of 108.6 $\pm$ 30.8 nGy h^{-1}, which is above the world average values.

CONCLUSION

This study intends to bring out the background radiation levels in the vicinity of MAPS, Kalpakkam through various radioactivity measurements. The results indicate that the average concentration of ^{226}Ra is relatively lower than the concentration of ^{232}Th and ^{40}K. The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K are comparable to the other studies conducted by different authors in India (Table 5). The calculated dose rates and the average effective dose equivalent were higher than the estimated global average values. The natural radioactivity and gamma-absorbed dose rates due to the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in soil and air reveal that the vicinity of MAPS, Kalpakkam can be regarded as a region with normal natural background radiation level.

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