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Natural radioactivity distribution in geological matrices around Kaiga environment

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The activity and absorbed dose rate of the naturally occurring radionuclides, viz. ^{238}U, ^{232}Th and ^{40}K were determined in soil and rock samples collected around Kaiga site. The mean activity levels (Kaiga soil) of naturally occurring ^{232}Th are comparable with that in worldwide soil, while concentrations of ^{238}U and ^{40}K are lower than those in worldwide soil. The absorbed dose rate in outdoor air ranged 20–58 nGy·h^{-1} with a mean of 33.3 nGy·h^{-1}, which is below the world average of 60 nGy·h^{-1}. The total effective dose rate in outdoor air for soils ranged 25.6–74.4 μSv·y^{-1} with a mean of 43.0 μSv·y^{-1}. The estimated dose rate at Kaiga is comparable with that estimated at Kakrapar and Rawatbhata and much less than that estimated at coastal sites of India.

Introduction

Radioactive elements constitute a part of the Earth's spheres. Their composition and distribution are subject to the same natural laws as the other non-radioactive elements. Natural radioactivity is composed of the cosmogenic and primordial radionuclides. Cosmogenic radionuclides, such as ^{3}H, ^{7}Be, ^{14}C and ^{22}Na are produced by the interaction of cosmic-ray particles in the Earth's atmosphere. Primordial radionuclides are formed by the process of nucleosynthesis in stars. The most common terrestrial radionuclides that produce gamma-rays are ^{238}U, ^{232}Th daughter products and ^{40}K. The intensity of the terrestrial natural radioactivity varies by an order of magnitude for different regions due to geological factors. The external radiation exposure arises mainly from cosmic rays and from terrestrial radionuclides occurring at trace levels in all geological matrices. The cosmic ray induced γ field generally contributes one-third to one-half of the total, the rest is from terrestrial radioelements.[1] Therefore, major spatial variations in natural γ-radioactivity are caused by variations in the abundance and distribution of U, Th and K. The terrestrial background radiation is related to the types of rock from which the soil originates. The higher concentrations of uranium, thorium and potassium are associated with soil developed from acid magmatic rocks and clay[2] and probably, the majority of uranium is associated with the phosphatic sands and clays of these formations.[3] The study of natural terrestrial radiation is useful for various reasons as has been reported by many authors.[4–10]

The primary objective of the present study was to determine the activity of ^{238}U, ^{232}Th and ^{40}K in geological matrices such as soil, rock and its contribution to dose rate around Kaiga environment.

Experimental

Site description

This study was carried out at Kaiga site, located east of the coastal town Karwar in Karnataka on the southwest coast of India (latitude 14.86 °N and longitude 74.44 °E), as shown in Fig. 1. Kaiga is identified as the site for six nuclear power stations (Two 220 MWe PHWR type stations operational, two similar units under construction and two more units proposed). The site is surrounded by tropical forest populated by a variety of plants and animals.

Sample collection and counting

A total of 24 rock samples and 16 soil samples have been collected from various locations at Kaiga site. Soil samples were dried in an oven at 100 °C for 24 hours, and then crushed, ground to fine powder and homogenized by passing through 180 μm test sieve. The rock samples were dried and crushed.

The samples were sealed and stored in a Marinelli beaker to prevent the escape of radiogenic gases ^{222}Rn and ^{220}Rn, and to allow the attainment of radioactive equilibrium in the decay chain. Natural radionuclides of relevance for this work are mainly gamma-ray emitting nuclei in the decay series of ^{232}Th and ^{238}U, and single occurring ^{40}K. ^{40}K was measured directly by its own gamma-rays (1461 keV). Decay products of ^{238}U (^{214}Pb: 352 keV; and ^{214}Bi: 609 keV) and ^{232}Th (^{228}Ac: 911 keV; ^{212}Pb: 238 keV; ^{212}Bi: 727 keV; and ^{208}Tl: 583 keV) were used by assuming the decay series to be in secular equilibrium. After attainment of secular equilibrium between ^{232}Th, ^{238}U and their daughter products, the samples were subjected to gamma-ray

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spectrometric analysis. A high purity germanium detector (HPGe) of coaxial type having dia 50.2 mm and length 49.5 mm coupled to a PC aided 4K MCA was used for analysis. The system was calibrated for energy and efficiency using a Marinelli beaker containing spiked soil sample and liquid standards containing ^{137}Cs, ^{60}Co, ^{133}Ba and ^{40}K. The efficacy of the sample counting was verified by the analysis of certified reference materials IAEA-375 Soil and IAEA-156 Clover provided by IAEA. The results agreed within ±5% with the certified values.

Derivation of gamma dose rates in air outdoors: If naturally occurring nuclides are uniformly distributed in the ground, dose rates at 1 m above the ground surface were calculated by:[11]

$$\text{Dose rate (nGy·h}^{-1}\text{)} = \\ = \text{Concentration of radionuclide (Bq·kg}^{-1}\text{)} \cdot \\ \cdot \text{Conversion factor (nGy·h}^{-1}\text{ per Bq·kg}^{-1}\text{)} \quad (1)$$

The absorbed gamma dose rate in air was mainly determined from ^{212}Pb (photon energy 238 keV), ^{208}Tl (583 keV), ^{212}Bi (727 keV) and ^{228}Ac (911 keV) for the ^{232}Th series, and from ^{214}Pb (352 keV) and ^{214}Bi (609 keV) for the ^{238}U series. Due to the small fractional yield and relatively low energy of the emitted photons, radionuclides such as ^{226}Ra and ^{224}Ra were neglected.[12] TZORTZIS et al.[13] reported the dose rate conversion factor for ^{232}Th and ^{238}U series and for ^{40}K as 0.52813, 0.38919, 0.03861, respectively, and these values were used in the present work.

Finally, in order to make a rough estimate for the annual effective outdoor dose, one has to take into account the conversion coefficient from absorbed dose in air to effective dose and the outdoor occupancy factor. In the UNSCEAR reports (1993, 2000),[14,15] the committee used 0.7 Sv·Gy^{-1} as the conversion coefficient from absorbed dose in air to effective dose received by adults, and 0.2 for the outdoor occupancy factor. Effective outdoor dose rate (μSv·y^{-1}) was calculated by:

$$\text{Effective dose rate (μSv·y}^{-1}\text{)} = \\ = \text{Dose rate (nGy·h}^{-1}\text{)} \cdot \\ \cdot 24\text{ (h)} \cdot 365\text{ (d)} \cdot 0.2\text{ (occupancy factor)} \cdot \\ \cdot 0.7\text{ Sv·Gy}^{-1}\text{ (conversion coefficient)} \times 10^{-3} \quad (2)$$

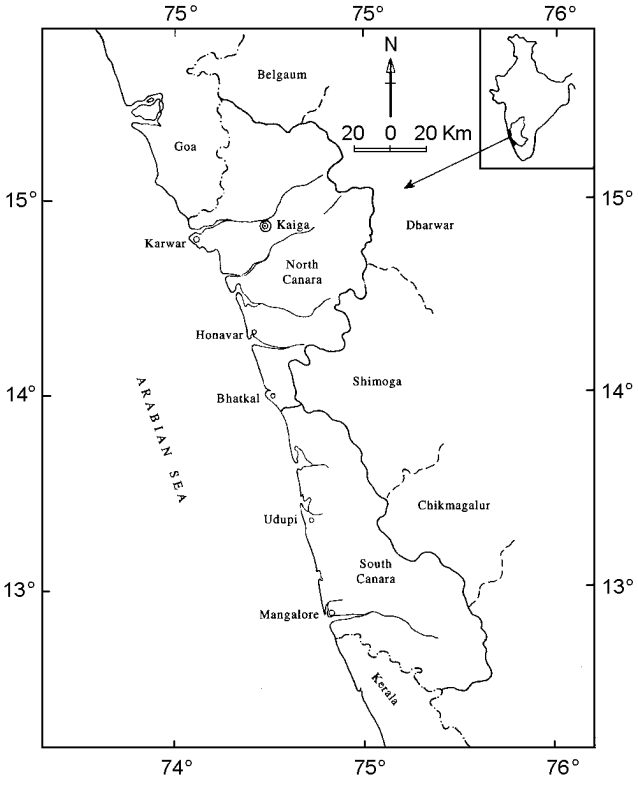

Fig. 1. Location of Kaiga

Results and discussion

Distribution of natural radioactivity in rocks

The activity of ^{232}Th, ^{238}U and ^{40}K in the rock samples collected around Kaiga site is shown in Table 1. The activity of ^{232}Th in the rock ranged from 1.2–14.2 Bq·kg^{-1} with a mean of 8.1 Bq·kg^{-1}, ^{238}U ranged from 0.5–11.5 Bq·kg^{-1} with a mean of 4.3 Bq·kg^{-1} and ^{40}K ranged from 14.8–866.2 Bq·kg^{-1} with a mean of 349.6 Bq·kg^{-1}, respectively. The ^{232}Th/^{238}U ratio in the rock ranged from 0.5–4.2 with a mean of 2.2. It is observed that the ^{40}K activity in the rocks is higher than that of ^{232}Th and ^{238}U. MOHANTY et al.[16] reported the ^{232}Th, ^{238}U and ^{40}K in the bulk sand samples in Erasama beach (eastern coast of Orissa, India) as 900–4700 Bq·kg^{-1}, 150–500 Bq·kg^{-1}, 100–250 Bq·kg^{-1}, respectively. Figure 2 shows the normal distribution of ^{238}U, ^{232}Th and ^{40}K content in rock samples.

Distribution of natural radioactivity in soil

The activity of ^{232}Th, ^{238}U and ^{40}K in the soil samples collected around Kaiga site is shown in Table 2. The activity of ^{232}Th in the soil ranged from 19.8–45.3 Bq·kg^{-1} with a mean of 31.7 Bq·kg^{-1}, ^{238}U ranged from 12.8–42.2 Bq·kg^{-1} with a mean of 24.0 Bq·kg^{-1} and ^{40}K ranged from 135.8–344.6 Bq·kg^{-1} with a mean of 201.4 Bq·kg^{-1}, respectively. The ^{232}Th/^{238}U ratio in the soil ranged from 0.9–1.9 with a mean of 1.4. SIDDAPPA et al.[17] reported ^{232}Th/^{238}U in soil samples in the Kaiga region as 0.8–5.1. It is observed that the ^{40}K activity in the Kaiga soil is higher than that of ^{232}Th and ^{238}U.

Table 1. Natural radioactivity in rocks around Kaiga (in Bq·kg^{-1})

Sample ID	^{232}Th	^{238}U	^{40}K	^{232}Th/^{238}U
Rock-1	4.6 ± 0.5	1.4 ± 0.3	14.8 ± 3.0	3.3
Rock-2	4.3 ± 0.5	1.9 ± 0.3	107.0 ± 5.8	2.3
Rock-3	11.8 ± 0.6	5.4 ± 0.3	232.2 ± 6.5	2.2
Rock-4	11.6 ± 0.8	5.7 ± 0.4	337.2 ± 9.4	2.0
Rock-5	6.1 ± 0.4	11.5 ± 0.4	317.6 ± 5.9	0.5
Rock-6	7.0 ± 0.4	6.4 ± 0.3	318.2 ± 5.1	1.1
Rock-7	7.5 ± 0.5	3.7 ± 0.3	309.5 ± 6.9	2.0
Rock-8	8.6 ± 0.6	4.9 ± 0.4	364.0 ± 9.4	1.8
Rock-9	14.2 ± 0.9	5.5 ± 0.4	866.2 ± 15.3	2.6
Rock-10	8.1 ± 0.7	5.8 ± 0.5	617.7 ± 12.4	1.4
Rock-11	9.1 ± 0.7	3.3 ± 0.4	376.0 ± 9.7	2.8
Rock-12	10.3 ± 0.7	4.2 ± 0.4	544.1 ± 11.0	2.5
Rock-13	14.0 ± 0.7	4.8 ± 0.4	315.6 ± 7.3	2.9
Rock-14	11.1 ± 0.9	5.0 ± 0.4	351.2 ± 10.1	2.2
Rock-15	7.2 ± 0.4	4.7 ± 0.3	232.9 ± 5.5	1.5
Rock-16	10.4 ± 0.7	6.1 ± 0.5	324.3 ± 8.6	1.7
Rock-17	8.0 ± 0.4	4.1 ± 0.3	295.8 ± 5.3	2.0
Rock-18	2.6 ± 0.3	1.5 ± 0.2	22.1 ± 2.5	1.7
Rock-19	2.0 ± 0.3	0.9 ± 0.2	529.9 ± 7.8	2.2
Rock-20	2.5 ± 0.5	0.6 ± 0.3	501.2 ± 11.9	4.2
Rock-21	1.2 ± 0.4	0.5 ± 0.1	416.7 ± 9.1	2.6
Rock-23	12.5 ± 0.7	4.2 ± 0.3	352.2 ± 7.7	1.8
Rock-22	6.2 ± 0.5	3.4 ± 0.3	280.9 ± 6.6	3.0
Rock-24	12.5 ± 0.6	7.1 ± 0.3	363.0 ± 7.1	1.8
Range:	1.2–14.2	0.5–11.5	14.8–866.2	0.5–4.2
Mean:	8.1	4.3	349.6	2.2
Std. Dev.:	3.8	2.5	181.4	0.8

Table 2. Natural radioactivity in soil around Kaiga (in Bq·kg^{-1})

Sample ID	^{232}Th	^{238}U	^{40}K	^{232}Th/^{238}U
Soil-1	38.9 ± 2.6	38.1 ± 1.6	217.7 ± 13.8	1.0
Soil-2	45.3 ± 2.3	42.2 ± 1.5	266.5 ± 12.2	1.1
Soil-3	37.5 ± 2.3	31.5 ± 1.5	229.4 ± 12.8	1.2
Soil-4	38.5 ± 2.5	26.4 ± 1.5	226.6 ± 14.7	1.5
Soil-5	24.4 ± 2.0	26.7 ± 1.5	189.4 ± 13.0	0.9
Soil-6	23.2 ± 2.0	18.4 ± 1.2	169.3 ± 14.1	1.3
Soil-7	25.1 ± 1.7	17.6 ± 1.0	189.5 ± 12.2	1.4
Soil-8	19.8 ± 1.4	16.7 ± 0.8	154.0 ± 9.6	1.2
Soil-9	19.9 ± 1.4	17.6 ± 0.9	154.5 ± 9.7	1.1
Soil-10	34.1 ± 2.0	20.3 ± 1.2	179.2 ± 12.4	1.7
Soil-11	27.4 ± 1.1	18.5 ± 0.8	172.9 ± 8.0	1.5
Soil-12	45.0 ± 2.9	24.1 ± 1.6	238.3 ± 17.9	1.9
Soil-13	28.6 ± 1.8	21.9 ± 1.1	151.5 ± 11.4	1.3
Soil-14	33.6 ± 3.0	23.6 ± 1.9	344.6 ± 23.6	1.4
Soil-15	41.8 ± 2.1	27.4 ± 1.2	202.9 ± 12.4	1.5
Soil-16	23.3 ± 1.6	12.8 ± 0.9	135.8 ± 10.2	1.8
Range:	19.8–45.3	12.8–42.2	135.8–344.6	0.9–1.9
Mean:	31.7	24.0	201.4	1.4
Std. Dev.:	8.8	8.0	52.7	0.3

It is interesting to compare the natural radioactivity of Kaiga soil with the soil worldwide. RAMLI et al.[18] studied the natural radioactivity around Palong area in the Segamat district, northern part of Johor state, Malaysia and the concentration of ^{238}U and ^{232}Th in the soil ranged from 58.8–484.8 Bq·kg^{-1}, 59.6–1204 Bq·kg^{-1}, respectively. TZORTZIS et al.[13] reported the ^{232}Th, ^{238}U and ^{40}K in the soil ranged from 1.3–52.8 Bq·kg^{-1}, 0.9–90.3 Bq·kg^{-1}, 13–894 Bq·kg^{-1}, respectively. The worldwide revised median values of ^{232}Th, ^{238}U and ^{40}K concentrations in soil is reported as 30, 35 and 400 Bq·kg^{-1}, respectively.[15] This reveals that the mean concentration levels measured in Kaiga soil from naturally occurring radioisotopes such as ^{232}Th is comparable with worldwide soil, ^{238}U and ^{40}K concentrations are lower than the corresponding values obtained worldwide. Figure 3 shows the normal distribution of ^{238}U, ^{232}Th and ^{40}K content in soil samples. Good correlation was observed between ^{232}Th and ^{238}U in soil samples ($R^2 = 0.90$), ^{232}Th and ^{40}K ($R^2 = 0.78$) as shown in Figs 4 and 5.

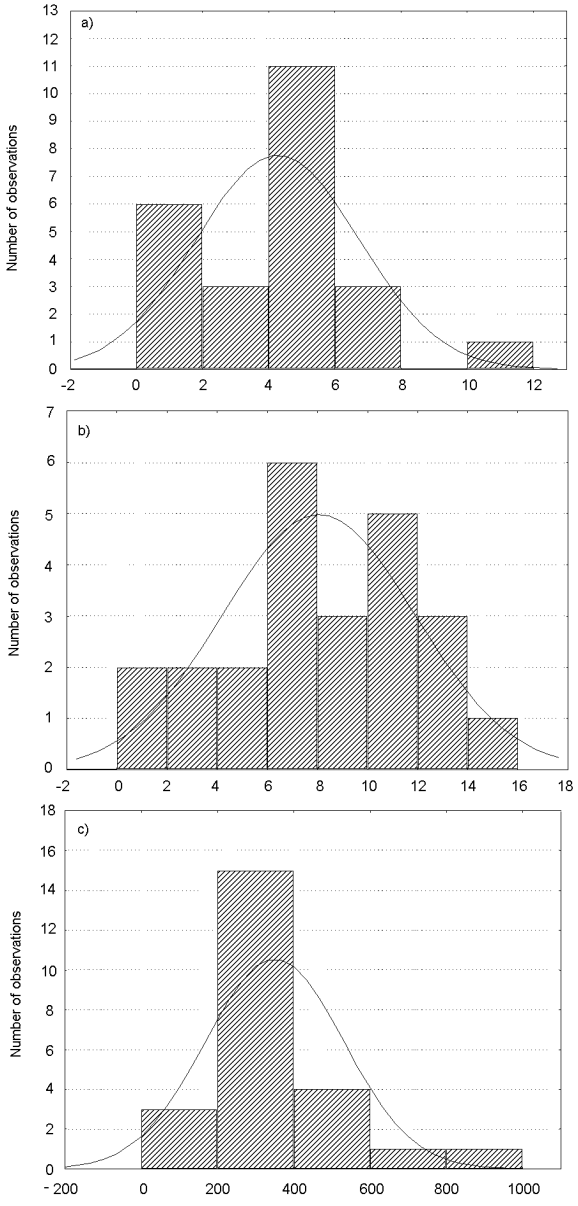

Fig. 2. Normal distribution of ^{238}U (a), ^{232}Th (b) and ^{40}K (c) content in rock samples

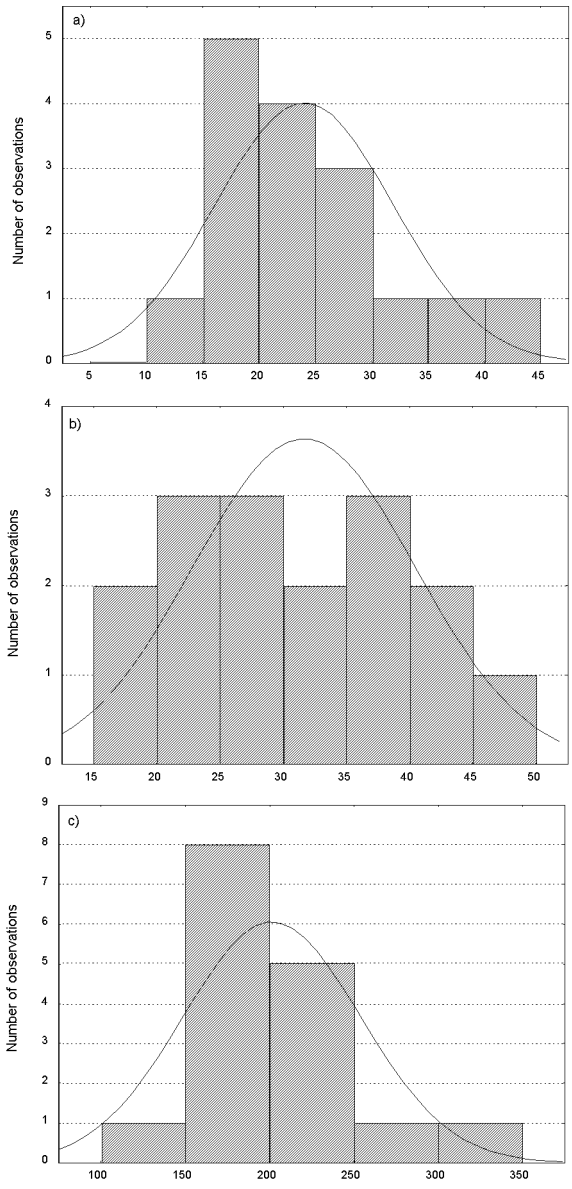

Fig. 3. Normal distribution of ^{238}U (a), ^{232}Th (b) and ^{40}K (c) content in soil samples

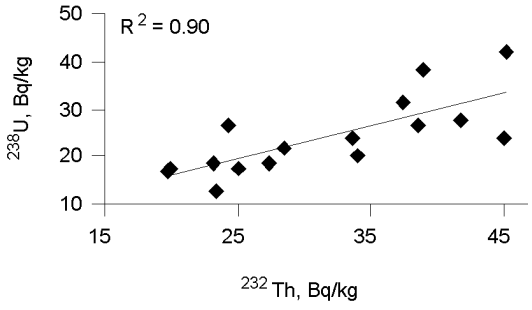

Fig. 4. Correlation between ^{232}Th and ^{238}U activity in soil

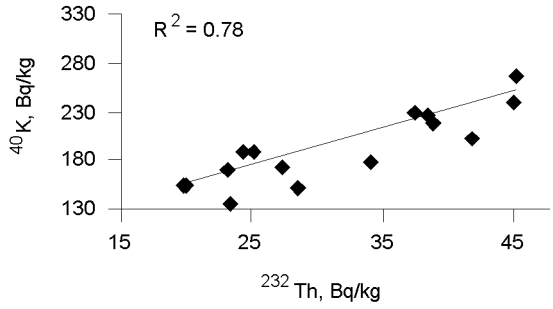

Fig. 5. Correlation between ^{232}Th and ^{40}K activity in soil

Comparison of ^{238}U, ^{232}Th and ^{40}K in rocks and soils

Table 3 shows the comparison of ^{238}U, ^{232}Th and ^{40}K concentration in rocks and soil around Kaiga site. It is observed that the ratio of soil/rock in the case of ^{238}U, ^{232}Th and ^{40}K is 5.6, 3.9, 0.58, respectively. WOLLENBERG and SMITH[19] reported the ratio of soil/rock in the case of ^{238}U and ^{232}Th as 0.71, 0.72, respectively. WOLLENBERG and SMITH[19] also reported that K/U and K/Th ratios may be used to differentiate between low radioactivity terrains (carbonates, ultrabasics, and basic igneous rocks) and between high radioactivity terrains (acid-igneous and alkali feldspathoidal rocks).

Gamma dose rate

The dose rates in outdoor air were calculated from concentration of radionuclides of ^{232}Th and ^{238}U series, and of ^{40}K using Eq. (1) and are tabulated in Table 4. In the soil samples, the computed values were for ^{232}Th series (^{228}Ac: 10.5–23.9 nGy·h^{-1} with a mean of 16.7 nGy·h^{-1}, ^{212}Bi: 10.3–32.4 nGy·h^{-1} with a mean of 18.7 nGy·h^{-1}, ^{208}Tl: 1.8–9.0 nGy·h^{-1} with a mean of 5.2 nGy·h^{-1}, ^{212}Pb: 11.1–38.0 nGy·h^{-1} with a mean of 19.1 nGy·h^{-1}), for ^{238}U series (^{214}Pb: 7.0–20.5 nGy·h^{-1} with a mean of 11.8 nGy·h^{-1}, ^{214}Bi: 5.0–16.4 nGy·h^{-1} with a mean of 9.3 nGy·h^{-1}) and for ^{40}K: 5.2–13.3 nGy·h^{-1} with a mean of 7.8 nGy·h^{-1}, respectively. Figure 6 illustrates the measured relative contributions to total absorbed dose in outdoor air due to ^{232}Th and ^{238}U decay products and ^{40}K content in soil. The relative contribution to dose due to ^{40}K was 27%, followed by the contribution due to ^{232}Th and ^{238}U series elements as 43% and 30%, respectively. The absorbed dose rate in outdoor air were found to be in the range of 20–58 nGy·h^{-1} with a mean of 33.3 nGy·h^{-1}, which is below the corresponding population-weighted (world-average) value of 60 nGy·h^{-1}.[13] The total

effective dose rates in outdoor air estimated according to Eq. (2) for soils ranged 25.6–74.4 μSv·y^{-1} with a mean of 43.0 μSv·y^{-1}. Table 5 shows the comparison of radiation dose rate of Kaiga, Karnataka with different parts of India.[9,20–26] It is observed that the estimated dose rate at Kaiga from ^{238}U, ^{232}Th and ^{40}K content in soil is comparable with that estimated at Kakrapar and Rawatbhata and much less than that estimated at coastal sites such as Kalpakkam, Ullal, Bhimilipatanam, Chhatrapur, Kerala coast, Tamilnadu coast of India.

Table 3. Comparison of ^{238}U, ^{232}Th and ^{40}K in rock and soil

Type of matrix	^{238}U	^{232}Th	^{40}K	^{40}K/^{238}U	^{40}K/^{232}Th
Rock	0.5–11.5 (4.3)	1.2–14.2 (8.1)	14.8–866.2 (349.6)	81.3	43.2
Soil	12.8–42.2 (24.0)	19.8–45.3 (31.7)	135.8–344.6 (201.4)	8.4	6.4
Ratio soil/rock	5.6	3.9	0.58		

Figures in parenthesis indicate the arithmetic mean.

Table 4. Activity, dose and effective dose rates assessment for the ^{232}Th and ^{238}U series and ^{40}K

DRCF, nGy·h^{-1} per Bq.kg^{-1}	Nuclide	Activity range, Bq·kg^{-1}	Dose rate outdoors range, nGy·h^{-1}	Effective dose rate outdoors range, μSv·y^{-1}	Total effective dose rate outdoors range, μSv·y^{-1}
Thorium series 0.52813	^{228}Ac (911 keV)	19.8–45.3 (31.7)	10.5–23.9 (16.7)	12.8–29.3 (20.5)	10.3–31.7 (18.3)
	^{212}Bi (727 keV)	19.5–61.4 (35.4)	10.3–32.4 (18.7)	12.6–39.8 (22.9)	
	^{208}Tl (583 keV)	3.5–17.1 (9.9)	1.8–9.0 (5.2)	2.3–11.1 (6.4)	
	^{212}Pb (238 keV)	21.1–72 (36.2)	11.1–38.0 (19.1)	13.7–46.6 (23.5)	
Uranium series 0.38919	^{214}Pb (352 keV)	17.9–52.7 (30.4)	7.0–20.5 (11.8)	8.5–25.2 (14.5)	7.3–22.6 (13.0)
	^{214}Bi (609 keV)	12.8–42.2 (24.0)	5.0–16.4 (9.3)	6.1–20.1 (11.4)	
Potassium 0.03861	^{40}K (1461 keV)	135.8–344.6 (201.4)	5.2–13.3 (7.8)	6.4–16.3 (9.5)	7.9–20.0 (11.7)
Total					25.6–74.4 (43.0)

Values in parenthesis indicate arithmetic mean.

Table 5. Comparison of radiation dose rate of Kaiga, Karnataka with different parts of India

Location in India	Characteristics of area	Absorbed dose rate in air, nGy·h^{-1}	Reference
Kalpakam (Tamilnadu)	Monazite sands	3500	KANNAN et al.[20] (2002)
Ullal (Karnataka)	Monazite sands	2100	RADHAKRISHNA et al.[9] (1993)
Kerala coast	Monazite sands	200–4000	SUNTA et al.[21] (1982)
Tamilnadu coast	Monazite sands	200–4000	SUNTA et al.[22] (1993)
Bhimilipatanam (Andhra Pradesh)	Monazite sands	200–3000	PAUL et al.[23] (1998)
Chhatrapur (Southern Orissa)	Monazite sands	375–5000	MOHANTY et al.[24] (2004)
Kakrapar (Gujrat)	Soil	7–32	RAMKUMAR et al.[25] (2001)
Rawatbhata (Rajasthan)	Soil	21–94	VERMA et al.[26] (2002)
Kaiga (Karnataka)	Soil	20–58	Present study

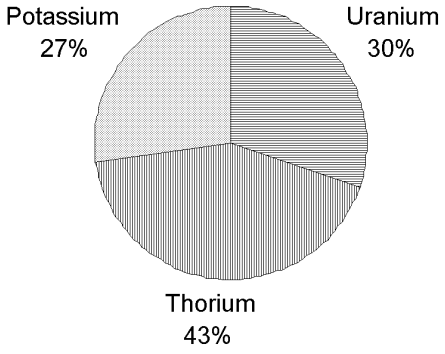

Fig. 6. Relative contributions to total absorbed dose in outdoor air due to ^{232}Th and ^{238}U decay products and ^{40}K content in soil

Conclusions

The mean activity levels in Kaiga soil due to naturally occurring ^{232}Th is comparable with those of worldwide soil but that of ^{238}U and ^{40}K concentrations are lower than in the worldwide soil. The ratio of soil/rock in the case of ^{238}U, ^{232}Th and ^{40}K was 5.6, 3.9, 0.58, respectively. The absorbed dose rate in outdoor air was found to be in the range of 20–58 nGy·h^{-1} with a mean of 33.3 nGy·h^{-1}, which is below the corresponding population-weighted (world-average) value of 60 nGy·h^{-1}. The estimated dose rate at Kaiga is comparable with that estimated at Kakrapar and Rawatbhata and much less than that at coastal sites of India. The total effective dose rate in outdoor air for soils ranged 25.6–74.4 μSv·y^{-1} with a mean of 43.0 μSv·y^{-1}. The relative contribution to dose due to ^{40}K is 27%, followed by the contribution due to ^{232}Th and ^{238}U series radionuclides as 43% and 30%, respectively.

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