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Gamma dose rates and distribution of natural radionuclides in sand beaches—Ilha Grande, Southeastern Brazil

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Abstract

In order to study the process of dispersion and the activity concentration of ^{232}Th and ^{238}U series and ^{40}K radionuclides in two island beaches in southeastern Brazil, analyses was made of sand samples of 0–10 cm profile, during a 12-month period. Moreover, the monthly variations of gamma dose rates were studied to determine the local environment absorbed dose rate. The average activity concentration of primordial radionuclides ^{232}Th, ^{238}U and ^{40}K at Preta beach, they were 239, 121 and 110 Bq kg^{-1}, while at Dois Rios beach they were 48, 39 and 412 Bq kg^{-1}, respectively. The absorbed dose rate in air, observed at 1 m above the ground, ranged from 54 to 228 nGy h^{-1} at Preta beach and from 39 to 110 nGy h^{-1} at Dois Rios beach. The annual effective dose equivalent corresponding to Preta beach is 0.15 mSv a^{-1} and to Dois Rios 0.08 mSv a^{-1}.

Keywords: Natural radionuclides; Gamma spectrometry; Brazil

1. Introduction

In certain beaches of Brazil and in the west coast of India, there are areas well known for their high background radiation. Some Brazilian beaches present high radiation due to the presence of monazite sand (B.A.S., 1997). Research in these places is of interest because monazite sand is considered an important geological material (Hassan et al., 1997), because it may contain 0.1–0.3% uranium and 5–7%

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thorium that are the main elements used in nuclear power plants (Alam et al., 1999). Around the world, several authors have been studying radionuclide concentrations in sand beaches in Kerala and Tamil Nadu coastal regions of India (Radhakrishna et al., 1993), in Bangladesh (Alam et al., 1999) and in the south-western Australia (de Meijer et al., 2001). Also in India, Kannan et al. (2002) analysed the distribution of natural and anthropogenic radionuclides in beach sand and soil from Kalpakkam area using gamma ray spectrometry. In spite of the high number of works carried out around the world, there is a lack of studies about radionuclides in the Brazilian beaches. Among the few studies, the recent work by Sachett (2002) stands out. It was carried out in Guarapari, a area in Brazil well-known for high background radiation, and presented results of gamma dose rates measured in some local beaches.

The Ilha Grande, shown in Fig. 1, is an island located on the coast of the State of Rio de Janeiro, near the city of Angra dos Reis, where the Nuclear Complex Almirante Alvaro Alberto—CNAAA is located. According to DePaula and Mozeto (2001), the bedrock of Ilha Grande is pre-Cambrian, with high to medium metamorphic grade rocks (charnockites, gneisses and migmatites), basic intrusives represented by diabase, basalt and gabbro dikes. Two beaches were studied in this island. The Preta beach (23° 07′ S 44° 10′ W), located on the continental side of Ilha Grande, is formed of deposits of dark sands, like other high background regions in the world such as Kerala and Tamil Nadu, India (Radhakrishna et al., 1993) and Guarapari, Brazil (Roser and Cullen, 1962). The other beach studied was Dois Rios (23° 10′ S 44° 11′ W), located on the ocean side of the island, which does not present dark sand deposits. The aim of this work is to investigate the variability in time of the concentrations of primordial radionuclides ^{40}K and the elements from series of ^{232}Th and ^{238}U in the sand and the consequent variability

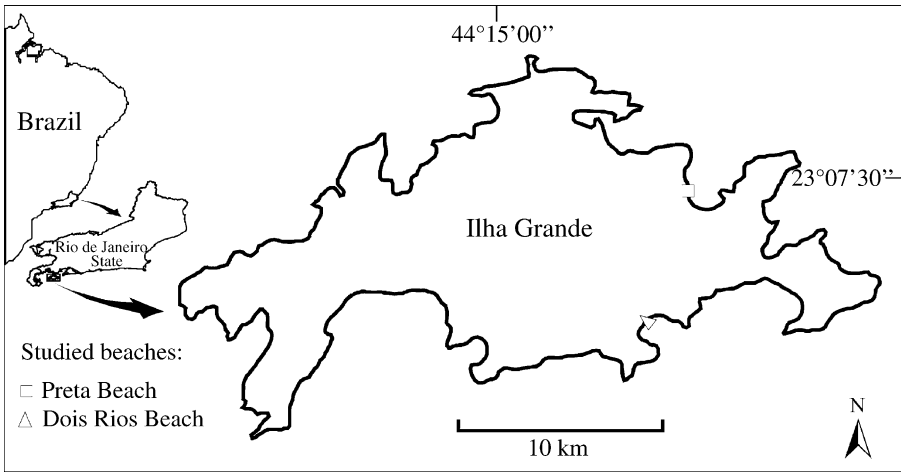

Fig. 1. Geographic location of Preta beach and Dois Rios beach in Ilha Grande, Rio de Janeiro State, Brazil.

of absorbed dose rate. These results may contribute to the determination of absorbed dose rate reference levels in areas near the CNAAA.

2. Materials and methods

2.1. Environment dose rate

The gamma dose rates were measured once a month at the two studied beaches during the period of June 2001–May 2002, using a portable digital environmental radiation detector, TRADOS 70046A VacuTec. The detector was calibrated at the Metrology Laboratory of "Laboratório de Ciências Radiológicas" (LCR) traceable to the National Standard of "Instituto de Radioproteção e Dosimetria/ Comissão Nacional de Energia Nuclear" (IRD/CNEN). Measurements were made using transects that cover the areas of interest in each beach. The gamma dose rate readings were recorded at 1 m above the ground level. For statistical purposes, 10 readings were taken at each point of reading and the average was recorded. The number of readings performed at the beaches, in each month, has direct relation with the tide level that covers the sand zone. On average, per month, 400 and 800 measurements were taken in Preta beach and Dois Rios beach, respectively.

2.2. Beach sand sampling

Each sand sample analysed was obtained from three sub-samples collected from the points with the highest gamma dose rate, based on in situ measurements, over an area corresponding to 1 m^2 and a depth of 10 cm. The three sub-samples were homogenized in situ and this sand mixture, weighing approximately 1.5 kg, was considered representative of the highest point of measurements. In the laboratory, all samples were dried at a temperature of 60 $^{\circ}C$ for 48 h, sieved through a 2 mm mesh, weighed and stored in cylindrical polyethylene containers. Each sample was hermetically and carefully sealed with aluminum foil to prevent the escape of gaseous ^{222}Rn and ^{220}Rn from the samples, which were kept aside for 30 days to allow radioactive equilibrium. After that, all samples of approximately 1.0 kg were subjected to gamma spectrometric analysis.

2.3. Gamma spectrometry

The activity concentration of primordial radionuclides ^{232}Th, ^{238}U and ^{40}K in the collected samples was determined using a high-resolution Ge detector (HPGe) gamma ray spectrometry system with 25% efficiency and a 3 keV resolution. It is composed of a p-type intrinsic germanium coaxial detector, vertically mounted and coupled to a digital high voltage source, an amplifier and a multichannel analyser. The detector was set inside a massive old lead shield, 10 cm thick, with an inner sheet of copper to reduce background radiation. This detector system was calibrated with a liquid standard cocktail made by AEA technology. The analysis of the gamma spectrum obtained was performed using the softwares Maestro II (MCA, EG&G ORTEC) and MCC (developed in the Instituto de Radioproteção

e Dosimetria/CNEN). All samples were counted over a period of 36,000 s. The γ-ray energies of ^{228}Ac (911.0 keV) and ^{214}Bi (609.3 keV) were used for the purpose of determining the activity of ^{232}Th and ^{238}U, respectively. The activity concentration of ^{40}K was measured directly from the 1460.8 keV γ-ray energy (IAEA, 1989).

The outdoor gamma dose rate was estimated using the equation (UNSCEAR, 1988)

$$D = 0.662\ S_{Th} + 0.427\ S_U + 0.043\ S_K$$

where D (in nGy h^{-1}) represents the total air absorbed dose rate due to the activity concentrations S_{Th}, S_U and S_K (in Bq kg^{-1}) for ^{232}Th, ^{238}U and ^{40}K, respectively. The background contribution from cosmic rays was determined using the same methodology as described by Selvasekarapandian et al. (2000). The background measurements were performed with the detector positioned on a small boat over the water in the studied area at a depth of about 12 m offshore. The average value from cosmic contribution measured in the studied area was added to the calculated gamma dose rate.

An ANOVA test was used to analyse the differences among the gamma dose rate in Preta beach and Dois Rios beach during the period of study.

3. Results and discussion

3.1. Terrestrial gamma dose rates

The graphic representation of gamma dose rate variation measured with the TRADOS detector at Preta beach and Dois Rios beach is shown in Fig. 2.

The average values of gamma dose rates for the study period in the Preta and Dois Rios beaches were 125 ± 11 and 63 ± 3 nGy h^{-1}, respectively. The minimum and maximum values found in Preta beach were, respectively, 58 nGy h^{-1} (May 2002) and 228 nGy h^{-1} (April 2002), while these values in Dois Rios beach were 39 nGy h^{-1} (June 2001) and 110 nGy h^{-1} (September 2001), respectively.

The values found at Preta beach are mainly due to spots of black sands observed in that beach. These spots were located approximately 5 m from the water line without great influence of local hydrodynamism. The black coloration of the sand may be due to the presence of magnetite, ilmenite and monazite, a characteristic of this kind of sand (Nagamalleswara-Rao, 1994).

According to Sachett (2002), the gamma dose rate in Areia Preta, a beach from Guarapari area, ranges from 75 to 14,400 nGy h^{-1}. The comparison of these values with those found for Preta beach in this work shows a large difference. The maximum value of gamma dose rate found in Preta beach was 63 times lower than the maximum value observed in Areia Preta, Guarapari.

An ANOVA test performed to compare the gamma dose rates in Preta and Dois Rios beaches, during the 12 months studied, shows that there is no significant statistical difference among the average values in both beaches.

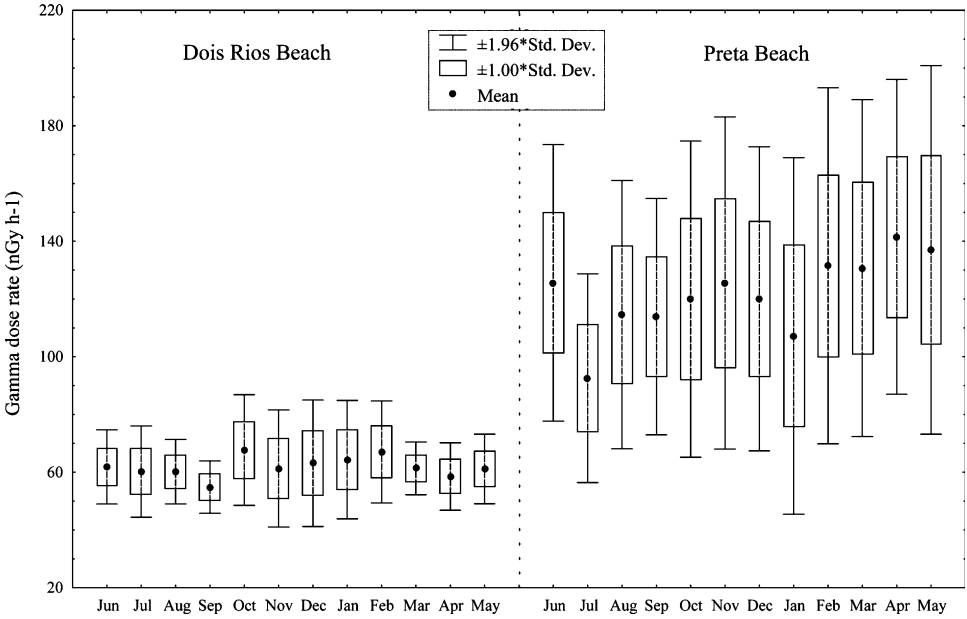

Fig. 2. Average values of gamma dose rates measured in Preta and Dois Rios beaches.

3.2. Natural radionuclides in sand beaches

Values corresponding to the activity concentration (Bq kg^{-1}) of ^{232}Th, ^{238}U and ^{40}K in dry weight, measured thin sand samples collected from points with the highest gamma dose rates in Preta and Dois Rios beaches are shown in Table 1. This table also shows the statistical data for the studied radionuclides. As shown in this table, the highest average activity concentration found in Preta beach, among the radionuclides analysed, was 239 Bq kg^{-1} for ^{232}Th, followed by 121 Bq kg^{-1} for ^{238}U and finally 110 Bq kg^{-1} for ^{40}K. In the Dois Rios beach, the highest average was for ^{40}K (412), followed by ^{232}Th (48 Bq kg^{-1}), and consequently the lowest activity concentration was found for ^{238}U (39 Bq kg^{-1}).

Fig. 3a–f shows the corresponding frequency distribution of activity concentrations for ^{232}Th, ^{238}U and ^{40}K in Preta and Dois Rios beaches. The activity concentrations corresponding to the ^{232}Th and ^{238}U radionuclides analysed in the two beaches were fitted to a normal curve. Only ^{40}K in Preta beach shows activity concentration fitted to a log-normal distribution.

According to UNSCEAR (2000), the world's average activity concentrations in surface soils from areas of normal radioactivity are 25, 25 and 370 Bq kg^{-1} for ^{232}Th, ^{238}U and ^{40}K, respectively. The activity concentrations of ^{232}Th, ^{238}U and ^{40}K for Dois Rios beach were 1.9, 1.6 and 1.1 times those reported by UNSCEAR (2000), while in Preta beach they were 9.5, 4.8 and 0.3 times, respectively.

Table 1
Activity concentrations and the respective statistical data of ^{232}Th, ^{238}U and ^{40}K (Bq kg^{-1}) in sand samples from Preta and Dois Rios beaches

	Preta beach			Dois Rios beach		
	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U	^{40}K
Jun 2001	128	133	116	23	23	473
Jul 2001	128	84	138	12	6	527
Aug 2001	287	113	85	26	17	489
Sep 2001	246	94	138	87	76	269
Oct 2001	232	125	47	46	33	366
Nov 2001	223	120	50	86	78	325
Dec 2001	265	116	70	85	72	349
Jan 2002	269	127	96	86	73	334
Feb 2002	271	158	103	50	41	421
Mar 2002	138	54	283	37	26	431
Apr 2002	349	180	100	14	10	510
May 2002	328	143	91	25	19	446
Average	239	121	110	48	39	412
S.D.	74	33	62	30	28	82
Median	255	122	98	42	30	426
Skewness	−0.4	−0.3	2.0	0.4	0.4	−0.2
Kurtosis	−0.7	0.7	6.0	−1.7	−1.6	−1.1
Freq. dist.	Normal	Normal	Log-normal	Normal	Normal	Normal

The enrichment of those minerals in Preta beach may be explained by inputs from the Abraão brook, which drains out on the beach. In time, the action of waves and wind causes variations in activity concentration of radionuclides.

The average contribution to the gamma dose rate at Preta beach was 72.9% from the ^{232}Th series, 24.5% from the ^{238}U series and 2.7% from ^{40}K. In Dois Rios beach, the contribution was 43.6% from the ^{232}Th series, 22.5% from the ^{238}U series and 33.9% from ^{40}K. The main contribution to the external gamma dose rate in Preta beach was due to the radionuclides of the ^{232}Th series, while in Dois Rios beach, the radionuclides of the ^{232}Th series and the radionuclide ^{40}K were the main contributors. These values result from the geological formation on Ilha Grande; according to Fernandes (2001), in the rocks from charnockitic, there is enrichment from elements like K, Rb, Ba, Th and Ce.

Table 2 represents a comparison of ^{232}Th, ^{238}U and ^{40}K activity concentration, from the present study, with those of other studies in different beaches of the world.

Comparing the values of ^{232}Th and ^{238}U for both Preta beach and Dois Rios beach with the values reported for Ullal and Kalpakkam (Radhakrishna et al., 1993; Kannan et al., 2002), it is possible to see which values from these two radionuclides in this study were smaller than the values reported for the beaches that

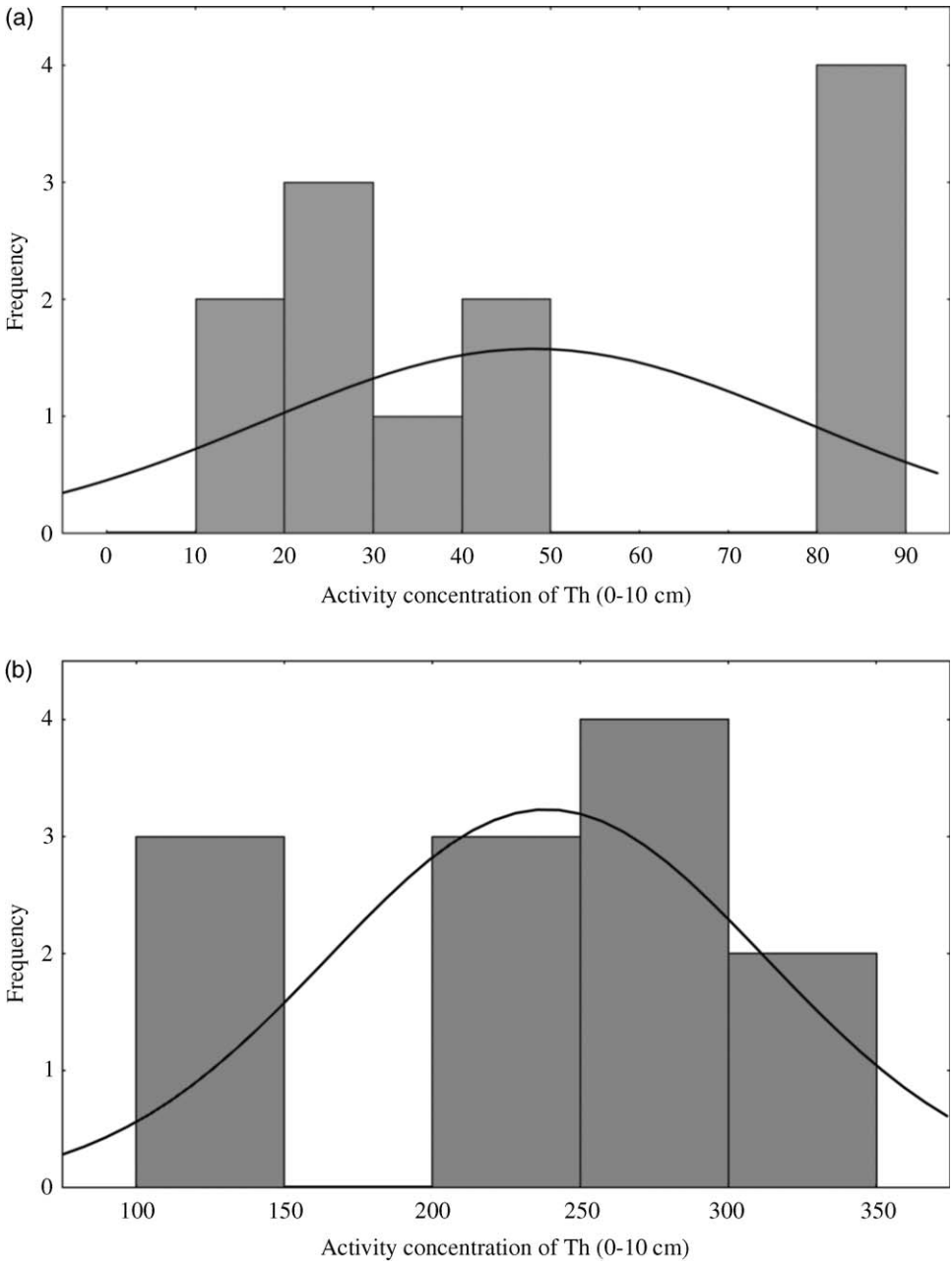

Fig. 3. Frequency distribution of activity concentration (Bq kg^{-1}) of ^{232}Th, ^{40}K and ^{238}U in Dois Rios (a, c, e) and Preta (b, d, f) beaches.

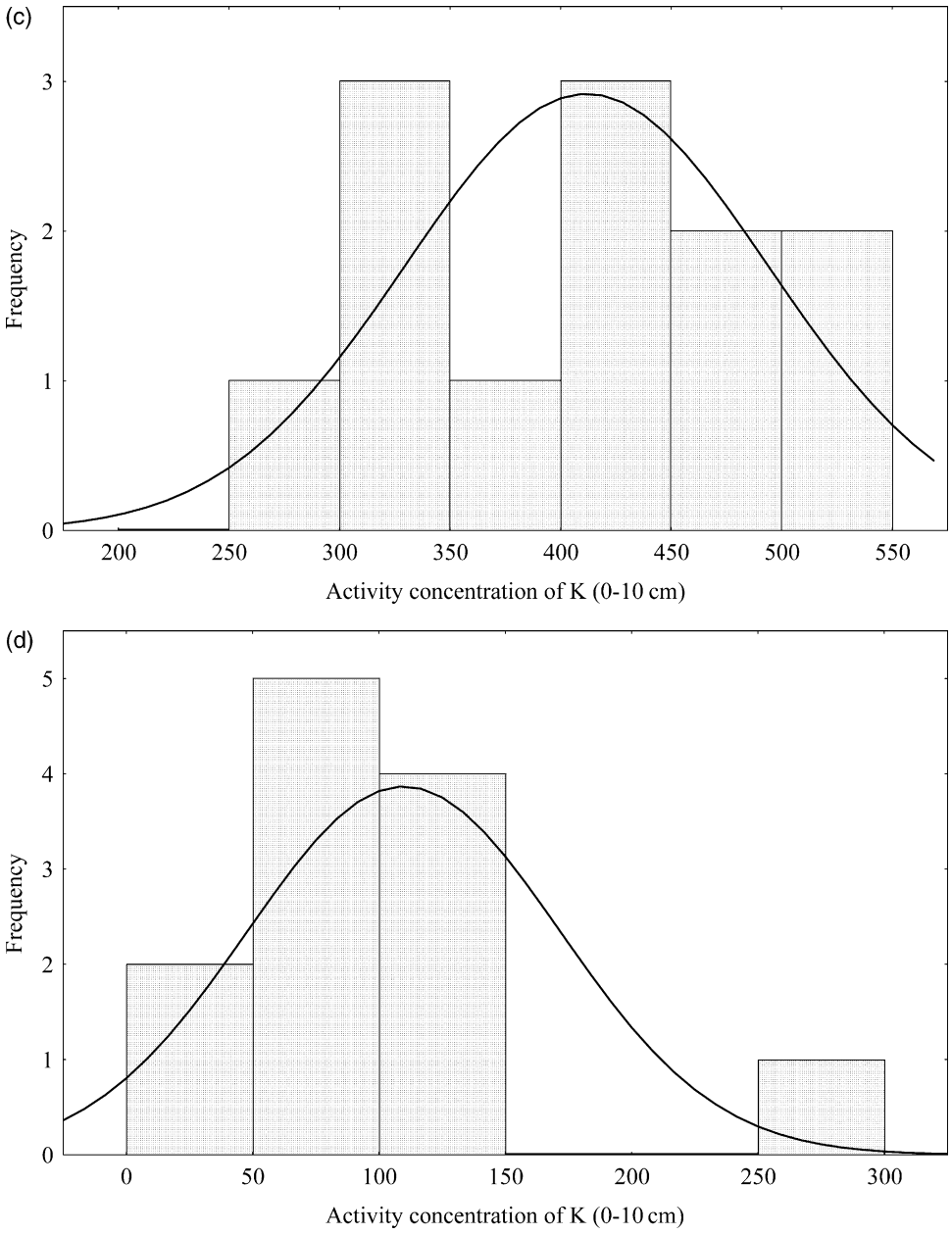

Fig. 3. (*Continued*).

were considered areas with high natural radiation. However, the values from ^{40}K, in Preta beach and, mainly, in Dois Rios beach were higher than those found in Ullal.

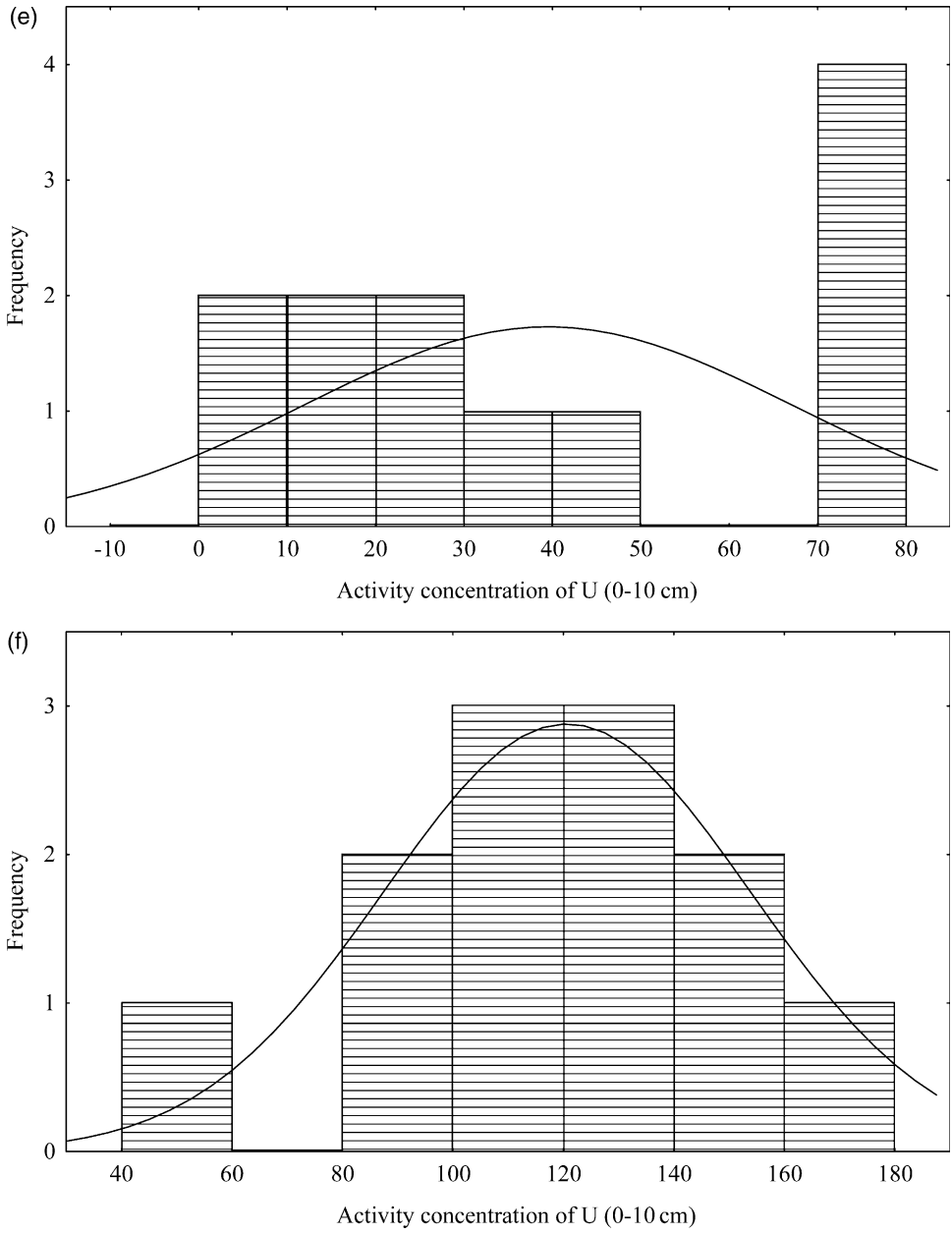

Fig. 3. (*Continued*).

Table 2
Concentrations of ^{232}Th, ^{238}U and ^{40}K (Bq kg^{-1}) in sand samples from Dois Rios and Preta beaches and other studies in different beaches of the world

Location	^{232}Th	^{238}U	^{40}K	Reference
Preta beach	128–349	54–180	47–283	Present study
Dois Rios beach	12–87	6–78	269–527	Present study
Visakhapatnam, India	300–600	100–400	–	Kalyani et al. (1990)
Northeast Coast, Spain	5–44	5–19	136–1087	Rosell et al. (1991)
Ullal, India	1842	374	158	Radhakrishna et al. (1993)
Valencia, Spain	1–11	4–16	30–253	Navarro and Roldan (1994)
Kalpakkam, India	352–3872	36–258	324–405	Kannan et al. (2002)

3.3. Dose computation

Using the Beck equation for converting the activity concentration of ^{232}Th, ^{238}U and ^{40}K into absorbed dose rate, the dose rates at Preta beach and Dois Rios beach were calculated. These values were added to the contribution of the cosmic radiation (36.0 nGy h^{-1}) measured in the studied area.

In the studied period of time, these values range from 162 to 348 nGy h^{-1} at Preta beach, with an average of 250 ± 58 nGy h^{-1}, while at Dois Rios beach, results range from 69 to 140 nGy h^{-1}, with an average of 102 ± 29 nGy h^{-1}.

The results of the absorbed dose rates calculated through the concentration of radionuclides in sand samples were compared with the highest gamma dose rate values measured in situ and are shown in Table 3.

As can be seen in the Table 3, there is a small difference between the calculated and measured values, for the two studied beaches. These small observed differences are probably due to the treatment of the samples before spectrometry. Factors

Table 3
Comparison between measured and calculated dose rate of Dois Rios and Preta beaches

	Gamma dose rate (nGy h^{-1})		Gamma dose rate (nGy h^{-1})	
Place	Preta beach		Dois Rios beach	
Month	Calculated	Measured	Calculated	Measured
Jun 2001	182	184	81	81
Jul 2001	162	172	69	92
Aug 2001	278	195	81	90
Sep 2001	245	191	138	109
Oct 2001	245	190	96	94
Nov 2001	237	190	140	101
Dec 2001	264	200	138	99
Jan 2002	272	190	139	105
Feb 2002	287	196	105	105
Mar 2002	162	184	90	94
Apr 2002	348	228	71	86
May 2002	318	215	79	93

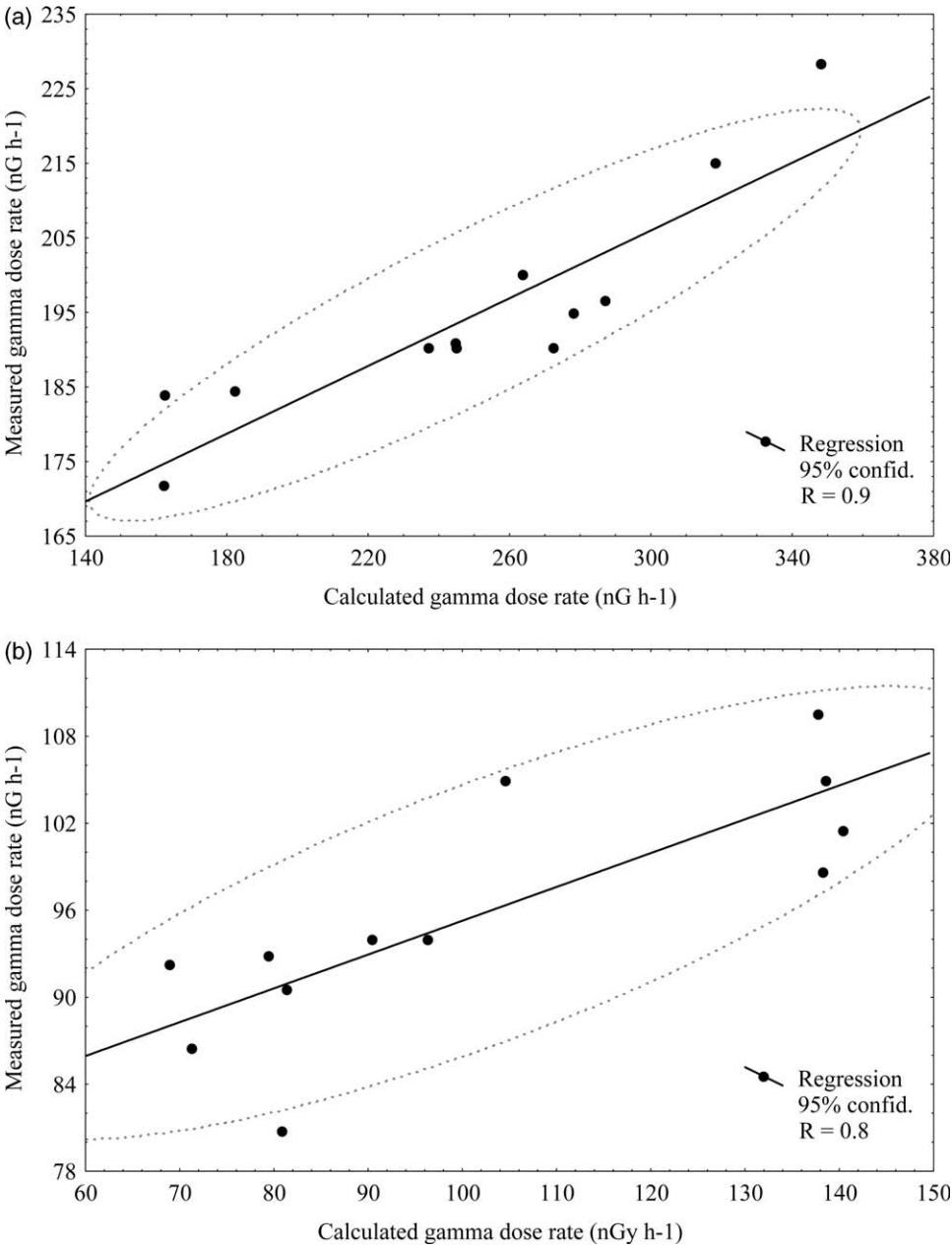

Fig. 4. Gamma dose rates calculated vs. measured in Preta beach (a) and Dois Rios beach (b).

such as density, humidity and compactness degree in situ are different from the dried samples.

According to Malanca et al. (1996), generally, there is no good correlation between calculated and measured gamma ray activities. However, the results in this study show a high correlation. In Preta beach, the correlation coefficient was $R = 0.9$ and at Dois Rios beach it was $R = 0.8$, as can be seen in Fig. 4(a,b), respectively.

The annual effective dose equivalent from the two beaches was determined as recommended by UNSCEAR (2000). At Preta beach, the annual effective dose varied between 0.07 and 0.28 mSv, with a mean of 0.15 ± 0.01 mSv, while at Dois Rios beach, it varied from 0.05 to 0.13 mSv, with a mean of 0.08 ± 0.01 mSv. The average values from both beaches and especially at Preta beach are higher than the worldwide average for outdoor annual affective dose, 0.07 mSv a^{-1}, reported by UNSCEAR (2000).

4. Conclusions

During the period of study, the average value of gamma dose rates in air at Preta beach was 125 nGy h^{-1} and at Dois Rios beach it was 63 nGy h^{-1}. These values correspond to an annual effective dose equivalent of 0.15 and 0.08 mSv a^{-1}, respectively. The analysed beaches are not considered to be radiological hazards for the local population.

In sand samples from Dois Rios beach, the average activity concentration for ^{232}Th (48 Bq kg^{-1}), ^{238}U (39 Bq kg^{-1}) and ^{40}K (412Bq kg^{-1}) was respectively 1.9, 1.6 and 1.1 times than the world average. In relation to the absorbed dose rate, these radionuclides contribute on average 43.6%, 22.5% and 33.9%, respectively. In sand samples from Preta beach, the mean activity concentration of ^{232}Th (239 Bq kg^{-1}), ^{238}U (121 Bq kg^{-1}) and ^{40}K (110 Bq kg^{-1}) was respectively 9.5, 4.8 and 0.3 times the world average. These radionuclides contribute on average 72.9%, 24.5% and 2.7% to the absorbed dose rate, respectively. During the period of study, in both beaches analysed, Preta and Dois Rios, there was no statistically significant variation in the terrestrial gamma dose rates determined by in situ measurements.

Weathering processes were the main factors related to the dispersion of minerals containing radioactive elements, which are carried out by the erosion process, throughout the water drainage system, and aerial and marine flux.

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