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Reference levels of natural radioactivity for the beach sands in a Brazilian southeastern coastal region

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Abstract

Gamma dose rates of ten beaches in Ilha Grande, an island of Brazilian southeastern cost, were measured in situ using an environmental radiation detector. The values ranged from 62 nGy h^{-1} at Dois Rios Beach to 126 nGy h^{-1} at Preta Beach. The activity concentrations of radionuclides ²³²Th series, ²³⁸U series and ⁴⁰K in three different sand profiles (0–10, 10–20 and 20–30 cm) were measured using gamma ray spectrometry and the result is shown in Bq kg⁻¹. In most of the cases, eight out of ten beaches, ⁴⁰K was the main contributor to the calculated local gamma dose rate. The annual effective doses were $0.15\pm0.03 \text{ mSv yr}^{-1}$ for Preta and $0.10\pm0.02 \text{ mSv yr}^{-1}$ for Caxadaço beaches, while in the others beaches the mean value of annual effective doses was 0.08 mSv yr^{-1} . None of the beaches analysed were considered a radiological risk. © 2004 Elsevier Ltd. All rights reserved.

Keywords: Natural radioactivity; Reference levels; Gamma dose rate; Beach sands; Brazil

1. Introduction

All living organisms of the planet are exposed to natural radiation, which is mainly due to the activity concentration of primordial radionuclides ²³²Th, ²³⁸U and their product of decay, in addition to the other natural radionuclide ⁴⁰K present in the earth' crust (UNSCEAR, 2000). According to Ramli (1997), one of the major interests in studies of natural background radiation is the need to establish reference levels, especially in areas where the risk of radioactive material being released to environment is high. There is, also a worldwide interest in identifying new areas with high natural radiation. Several authors have studied the levels of natural background radiation by in situ measurements or by analysis of radionuclides concentrations in soil or sand samples (e.g.

* Corresponding author. Tel.: +55 21 2568 9664. *E-mail address:* alencar@uerj.br (A.S. Alencar). Malanca et al., 1996; Selvasekarapandian et al., 2000; Navas et al., 2002; Mohanty et al., 2004). Two types of areas in Brazil are well known for their high background radiation: namely the region of volcanic intrusives in the State of Minas Gerais and the region of monazite sands along the Atlantic coast (Penna-Franca et al., 1965, B.A.S., 1997; Paschoa, 2000). Radiological studies have been made in sand beach locations, mainly in India, because along its coastline there are quite a few monazite sand bearing placer deposits causing natural high background radiation areas (UNSCEAR, 2000); in Kerala and Tamilnadu (Radhakrishna et al., 1993), in Kalpakkam (Kannan et al., 2002) and in recent work in the coast of Orissa (Mohanty et al., 2004). In Brazil stand out the studies at Guarapari, coastal region of Espirito Santo (Penna-Franca et al., 1965; Sachett, 2002) and in the Ilha Grande, Rio de Janeiro State (Freitas and Alencar, 2004).

The present work was carried out in ten beaches of Ilha Grande $(23^{\circ}08'S \ 44^{\circ}15'W)$, an island situated near to the

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city of Angra dos Reis—Rio de Janeiro, on the Brazilian Southeastern coast, where the Nuclear Complex Almirante Alvaro Alberto—CNAAA is located. The beaches studied are: Abraãozinho, Biquinha, Caxadaço, Crena, Dois Rios, Guaxuma, Julia, Parnaioca, Preta and Sobradinho; and their location in Ilha Grande is shown in Fig. 1 . The aim of this paper is to establish reference levels of natural radioactivity for the studied beaches of Ilha Grande, through the analysis of gamma dose rates by in situ measurements and by HPGe gamma ray spectrometry to determine the activity concentration of ²³²Th, ²³⁸U and ⁴⁰K in sand samples.

2. Methodology

2.1. In situ radiometric analysis

The environment dose rate measurements were performed between July 2001 and October 2001, using a portable environmental radiation detector, TRADOS 70046A VacuTec. The calibration of the detector was carried out for 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), through the use of a source of reference of ¹³⁷Cs. The use of this source of high activity associated with a series of lead attenuators allows to the variation of the air kerma rates to a fixed distance source-detector. All readings in the beaches were taken at 1 m above the ground level in the sand area through a transect which covers the area of interest. The number of readings taken at each beach is directly related to the size of the sand beach area. Ten readings were taken at each point of the transect and the average was recorded.

2.2. Sand sampling

All sand samples were collected from the spot showing highest gamma dose rate determined by in situ measurement. In order to analyse the variation of activity concentration for ²³²Th, ²³⁸U and ⁴⁰K, one sample was obtained for each of three different depth profiles (0-10, 10-20 and 20-30 cm). Each sample profile was obtained from three sub-samples collected from an area corresponding to 1 m^2 . The three sub-samples were homogenized in situ, and this sand mixture, weighing approximately 1.5 kg, was considered representative of the profile. In the laboratory, the samples were dried for about 48 h in an oven at 60 °C, sieved through a 2 mm mesh to remove the largest sand fraction, weighed and finally stored in PVC cylindrical containers. These containers were hermetically sealed with aluminium foil to prevent the escape of gaseous ²²²Rn and ²²⁰Rn from the samples and kept aside for about 30 days in order to ensure radioactive equilibrium. After that period, the samples were analysed using gamma spectrometry.

2.3. Gamma-ray analysis

A high-resolution hyper purity germanium (HPGe) gamma ray spectrometry system, with 25% relative efficiency, was used to determine the activity concentration of radionuclides ²³²Th, ²³⁸U and ⁴⁰K in the samples. This method uses a system that consists of a p-type intrinsic germanium coaxial detector mounted vertically and coupled to a 3 keV digital high-voltage source. The detector was housed inside a massive 10 cm thick old lead shield with an inner layer of copper to reduce background radiation. The efficiency curve for calibration was determined by a standard solution made using AEA technology. Each sand sample, of approximately 1.0 kg, was counted for 36 ks, and the analysis of the obtained gamma spectra was performed with the use of softwares Maestro II (MCA, EG&G OR-TEC) and MCC (developed by IRD/CNEN). The activity concentration, in Bq kg⁻¹, of 40 K was measured directly from the 1460.8 keV γ -ray energy, while activity concentrations of ²³⁸U and ²³²Th were determined through the photopeak energies of ²¹⁴Bi (609 keV) and ²²⁸Ac (911 keV). respectively (IAEA, 1989).

2.4. Dose calculation

Using the methodology described by Selvasekarapandian et al. (2000) the background contribution from cosmic rays was determined. The measurements were undertaken with the detector positioned above the water, at a point where the depth reached 12 m, close to the studied area. The calculated gamma dose rate in each beach was determined using the equation published in UNSCEAR (1988) plus the cosmic radiation contribution

$$D = (0.662S_{\rm Th} + 0.427S_{\rm U} + 0.043S_{\rm K}) + CR,$$
(1)

where D (in nGy h⁻¹) represents the absorbed dose rate due to the activity concentrations of S_{Th} , S_{U} and S_{K} (in Bq kg⁻¹) for ²³²Th, ²³⁸U and ⁴⁰K, respectively and CR represents the mean value from cosmic radiation. In order to achieve this, the sand sample collected from the depth profile of 0–10 cm was used.

3. Results and discussion

The mean value and standard deviation of the in situ radiometric analysis for the ten studied beaches was $69\pm14 \text{ nGy h}^{-1}$ for Abraãozinho, $65\pm9 \text{ nGy h}^{-1}$ for Biquinha, $79\pm14 \text{ nGy h}^{-1}$ for Caxadaço, $68\pm12 \text{ nGy h}^{-1}$ for Crena, $62\pm7 \text{ nGy h}^{-1}$ for Dois Rios, $68\pm12 \text{ nGy h}^{-1}$ for Guaxuma, $67\pm9 \text{ nGy h}^{-1}$ for Julia, $64\pm19 \text{ nGy h}^{-1}$ for



Fig. 1. Map of Ilha Grande and its geographic location on the cost of the Rio de Janeiro State, Brazil. The numbers (1-10) indicate the location of the beaches studied.

Parnaioca, 126 ± 24 nGy h⁻¹ for Preta and 67 ± 9 nGy h⁻¹ for Sobradinho. The higher values of gamma dose rates found at Preta beach are due to the great concentration of spots of black sands observed in this beach when compared with the other beaches studied. The presence of magnetite, ilmenite and monazite may be the reason of the black colouration of the sand (Freitas and Alencar, 2004). The values of gamma dose rates found in all beaches studied in this work are a lot of smaller than those found by Sachett (2002) in Areia Preta—Guarapari, that ranged from 75 to 14400 nGy h⁻¹.

The radiological contours maps and the estimated values of gamma dose rates measured at 1 m above ground level in the studied beaches are shown in Fig. 2. For each map there is a colour-code provided, which represents the variation of gamma dose rates measured in $nGvh^{-1}$. The rectangular area of maps represent the sand surface measured in each beach. In some beaches like, Biquinha, Caxadaço, Guaxuma and mainly in Parnaioca the presence of rocks in the sand surface are marked for "holes" in the perspective maps. The holes are due to the absence of measurements of gamma dose rate in these places. The numbers shown in the length and width of the maps represent the interval (in metres) in the transects, where the measurements of gamma dose rates were undertaken. These maps represent the reference levels of the environmental radiation measured in the ten studied beaches and its analysis allows to identify the "hot spots" from each beach during the period of study.

Fig. 3 shows the variation of gamma dose rates measured in situ and calculated through the activity concentrations of radionuclides 232 Th, 238 U and 40 K in samples collected in 0–10 cm sand profiles. The value of 36 nGy h⁻¹, from cosmic contribution measured in the studied areas, was added to the calculated gamma dose rate. The results of gamma dose rate measurement and calculated were compared. For this purpose, the maximum values, measured at 1 m above ground level, was used.

As can be observed in Fig. 1, the Preta beach shown the highest mean value $(126\pm24 \text{ nGy h}^{-1})$ of gamma dose rates measured among the studied beaches. The other beaches show mean values ranging from 62 to 79 nGy h⁻¹ as found in Dois Rios and Caxadaço beach, respectively.

As described before, the sand samples in all studied beaches were collected from the spots that showed the highest gamma dose rate determined by in situ measurement. Some beaches such as Caxadaço, Guaxuma, Parnaioca and Preta, showed different levels of dark sand deposits, probably due to the high concentration of minerals containing primordial radionuclides. This was corroborated by the values of calculated gamma dose rates (142, 105, 271 and $183 \, \text{nGy} \, \text{h}^{-1}$, respectively) in this region.

According to Malanca et al. (1996), usually, there is no good agreement between calculated and measured gammaray activity, however, this results show a high correlation (R = 0.85) between this gamma dose rates as can be seen in Fig. 4, indicating that the field and laboratory measurements are mutually corroborative.

Table 1 shows the corresponding values of activity concentrations, in Bq kg⁻¹, from ²³²Th, ²³⁸U and ⁴⁰K in the sand profiles of 0–10, 10–20 and 20–30 cm. The activity concentrations of radionuclides ²³²Th, ²³⁸U and ⁴⁰K did not show any pattern in relation to the sand profiles analysed. Only in the Sobradinho Beach, the three radionuclides showed the highest activity concentrations in the same profile (20–30 cm).

Fig. 5 represents the values of mean, standard deviation and standard error from activities concentration of



Fig. 2. Radiological contours maps and the perspective viers of gamma dose rates measured in the beaches studied. The colour-coding shown in the left side of the maps represents the variation values of gamma dose rate measured in $nGyh^{-1}$.

these primordial radionuclides in all the beaches studied. The 40 K shows the highest contribution for the local gamma dose rate in eight studied beaches. Only

in two beaches, Preta and Parnaioca, the main contributors for the gamma dose rate were $^{232}\mathrm{Th}$ and $^{238}\mathrm{U},$ respectively.



Fig. 2. (continued).

These results reflect the geological formation of the island where the beaches are located. The island is rich in elements such as K, Th, Rb, Ba, and Ce (Fernades, 2001). The results of annual effective dose rate $(mSv yr^{-1})$ calculated from the data of the gamma dose rate measured in each studied beach were determined as



Fig. 3. Variations of measured and calculated gamma dose rates $(nGyh^{-1})$ in the beaches studied. MGDR Measured gamma dose rates; CGDR calculated gamma dose rates.



Fig. 4. Correlation between measured and calculated gamma dose rate in the beaches studied.

recommended by UNSCEAR (2000) and are shown in Table 2.

The two highest mean values of annual effective dose rates were found in Preta and Caxadaço beaches with values of $0.15 \pm 0.03 \,\mathrm{mSv} \,\mathrm{yr}^{-1}$ and $0.10 \pm 0.02 \,\mathrm{mSv} \,\mathrm{yr}^{-1}$, respectively. The other beaches show the same mean value $(0.08 \,\mathrm{mSv} \,\mathrm{yr}^{-1})$ of annual effective dose, and did not show significant statistical variation. In all the beaches studied, especially Preta and Caxadaço, the mean values of annual effective doses were higher than the worldwide average $(0.07 \,\mathrm{mSv} \,\mathrm{yr}^{-1})$ for outdoors annual affective doses published in UNSCEAR (2000). Nevertheless, none of the studied beaches was considered a radiological hazard, because

of the low values of gamma dose rates found in the most of analysed beaches.

4. Conclusions

Among the beaches studied, the Preta beach (126 nGy h^{-1}) shown the highest average value of gamma dose rates measured in situ, followed by Caxadaço beach (79 nGy h^{-1}) . Consequently, the highest annual effective doses were found in these beaches with 0.15 mSv yr^{-1} for Preta beach and 0.10 mSv yr^{-1} for Caxadaço beach. While in the others beaches studied, the average value of annual effective

Table 1 Activity concentrations, in $Bqkg^{-1}$, of ^{232}Th , ^{238}U and ^{40}K in sand samples profiles (0–10, 10–20 and 20–30 cm) of beaches studied	
Activity concentration $(Bqkg^{-1})$	

Beaches	Activity concentration (Eq.Kg)									
	²³² Th			²³⁸ U			⁴⁰ K			
	0–10	10-20	20-30	0-10	10–20	20-30	0–10	10-20	20-30	
Abraãozinho	51	81	58	18	44	31	841	769	886	
Biquinha	38	56	43	53	80	57	335	229	280	
Caxadaço	115	23	33	56	11	19	147	254	304	
Crena	15	30	27	19	40	41	1114	624	1006	
Dois Rios	23	29	20	23	32	17	473	406	399	
Guaxuma	22	22	12	38	31	18	884	1080	1082	
Julia	14	7	10	12	7	13	512	483	472	
Parnaioca	235	240	213	176	193	171	102	97	111	
Preta	128	122	92	133	167	116	116	92	101	
Sobradinho	2	18	55	5	14	42	633	641	1219	



Fig. 5. Activity concentrations, in $Bqkg^{-1}$, of ^{232}Th , ^{238}U and ^{40}K in all beaches studied.

doses was 0.08 mSv yr⁻¹. In spite of the high value of the gamma dose rate found in Preta Beach (2.14 times the world average), comparing with the other beaches, none of the studied beaches were considered a radiological hazard, and that is more evident when the studied beaches are compared to the beaches with high value of the gamma dose rate found around of the world, as Areia Preta in Guarapari. In eight beaches the radionuclide ⁴⁰K was the main contributor to the local gamma dose rate. Differences observed in the activity concentrations from the radionuclides ²³²Th, ²³⁸U and ⁴⁰K in relation to the sand profiles analysed, can be due to different sand properties, like density, humidity and porosity, in each beach.

Further radiological investigations must be carried out in the others beaches of Ilha Grande and the results may be used as radiometric reference levels, owing to its proximity to CNAAA, an area with a potential risk of radiological accident.

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Table 2 Statistics data of annual effective dose rate $(mSv yr^{-1})$ in the analysed beaches

	Annual effective dose							
	Mean	SD	Min.	Max.				
Abraãozinho	0.08	0.02	0.05	0.16				
Biquinha	0.08	0.01	0.06	0.11				
Caxadaço	0.10	0.02	0.06	0.14				
Crena	0.08	0.01	0.05	0.13				
Dois Rios	0.08	0.01	0.05	0.10				
Guaxuma	0.08	0.01	0.05	0.12				
Julia	0.08	0.01	0.05	0.13				
Parnaioca	0.08	0.02	0.05	0.21				
Preta	0.15	0.03	0.09	0.23				
Sobradinho	0.08	0.01	0.06	0.11				

SD; Standard deviation.

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