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NATURAL RADIOACTIVITY IN EXTREME SOUTH OF BAHIA, BRAZIL USING GAMMA-RAY SPECTROMETRY

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ABSTRACT

The concentrations activity of natural radionuclides in beach sand in extreme south of Bahia-Brazil was measured using gamma-ray spectrometry. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in beach sand ranged from 14.5 to 8,318.4, 20.5 to 18,450.0, 15.4 to 3,109.0 Bq/kg, with a mean value of 1,078.2, 2,429.6, and 417.0 Bq/kg respectively. The values of radiation hazard indexes in sands of Alcobaça, Caraíva and Cumuruxatiba are higher than the limits preconized by Beretka and Mathew. The use of these sands may be not safe in building constructions. The results show that the absorbed dose rates range from 20.4-15,116.6 nGy/h with mean value of 1762.7. The highest value of gamma dose rates among the studied beaches was found in Cumuruxatiba (15,116.6 nGy/h). The annual effective dose varied between 0.028 and 18.539 mSv/year, with a mean of 2.162 mSv/year. Values of Alcobaça, Trancoso, Caraíva and Cumuruxatiba are higher than the worldwide average for outdoor annual effective dose, 0.07 mSv/year.

1. INTRODUCTION

Human beings have always been exposed to natural radiation, which is mainly due to the activity concentration of primordial radionuclides ^{238}U(^{226}Ra) series, ^{232}Th series and ^{40}K that present in the earth's crust, in building materials and in air, water and foods and in the human body itself [1]. As these radionuclides are not uniformly distributed, the knowledge of their distribution in soil, sand and rock play an important role in radiation protection and measurement.

Radionuclides are classified into four groups according to their origins: (1) cosmic-ray produced nuclides (such as ^{7}Be and ^{14}C); (2) artificially-produced nuclides (such as ^{137}Cs and ^{90}Sr); (3) primordial isotopes (such as ^{238}U and ^{40}K); and (4) natural decay products (such as ^{226}Ra and ^{222}Rn). The three naturally occurring radioactive decay chains include the ^{238}U, ^{235}U, and ^{232}Th, which decay through a series of radioactive elements up to stable Pb isotopes [2].

The study of the distribution of primordial radionuclides allows the understanding of the radiological implication of these elements due to the γ-ray exposure of the body and irradiation of lung tissue from inhalation of radon and its daughters [3]. In particular, it also is important to assess the radiation hazards arising due to the use of soil or sand samples in the construction of dwellings. Therefore, the assessment of gamma radiation dose from natural sources is of particular importance as natural radiation is the largest contributor to the external dose of the world population [1].

These dose rates vary depending upon the concentration of the natural radionuclides, ^{238}U, ^{232}Th, their daughter products and ^{40}K, present in soil, sands and rocks, which in turn depend upon the local geology of each region in the world. In certain beaches of Brazil, there are areas well known for their high background radiation [3].

In order to estimate the possible radiological hazards to human health, considerable attention has been paid in the last two decades to low-level exposure arising from members of the uranium and thorium decay chains and by ^{40}K in soils. A number of investigations have been performed in developed countries, but very few have been conducted in developing nations [4]. In Brazil along the Atlantic coast in the Espírito Santo state is a belt of monazite sand in Guarapari city. It has been exhaustively described by many authors and reviewed [3,2]. However, Cumuruxatiba in Bahia state is a place that has a large amount of monazite sand (4,500T) and less studied.

The present work investigates the concentrations of radioisotopes such as ^{232}Th, ^{226}Ra and ^{40}K, in beach sand samples from extreme south of Bahia state, an area in Brazil not so much studied, and estimate the radiological hazard. The radium equivalent activity, the external hazard index, the absorbed dose rate, and the effective dose rates were calculated and compared with internationally approved values.

2. MATERIALS AND METHODS

Beach sand samples were collected in the Porto Seguro (16°26'S 39°03'W), Arraial D'Ajuda (16°29'S 39°04'W), Trancoso (16°35'S 39°05'W), Caraíva (16°48'S 39°08'W), Cumuruxatiba (17°20'S 39°13'W), Prado (17°20'S 39°13'W), Alcobaça (17°33'S 39°11'W), and Caravelas (17°43'S 39°10'W), on May, 2008. The samples were collected at a depth of 10cm. Each sample was obtained from three sub-samples collected from an area corresponding to 1 m^2 and were homogenized in situ, and this sand mixture, weighing approximately 1.5kg, was considered representative of the profile. Figure 1 shows the region studied.

The samples were dried for about 48 h in an oven at 60°C and the analytical techniques used were gamma spectrometry for the gamma emitters isotopes ^{214}Bi(^{226}Ra) and ^{40}K, and Instrumental Neutron Activation Analysis (INAA) for ^{232}Th. The gamma lines represent ^{226}Ra was determined indirectly by ^{214}Bi. Table 1 shows the technique used for each radionuclide, the specific gamma line used and the detection limits.

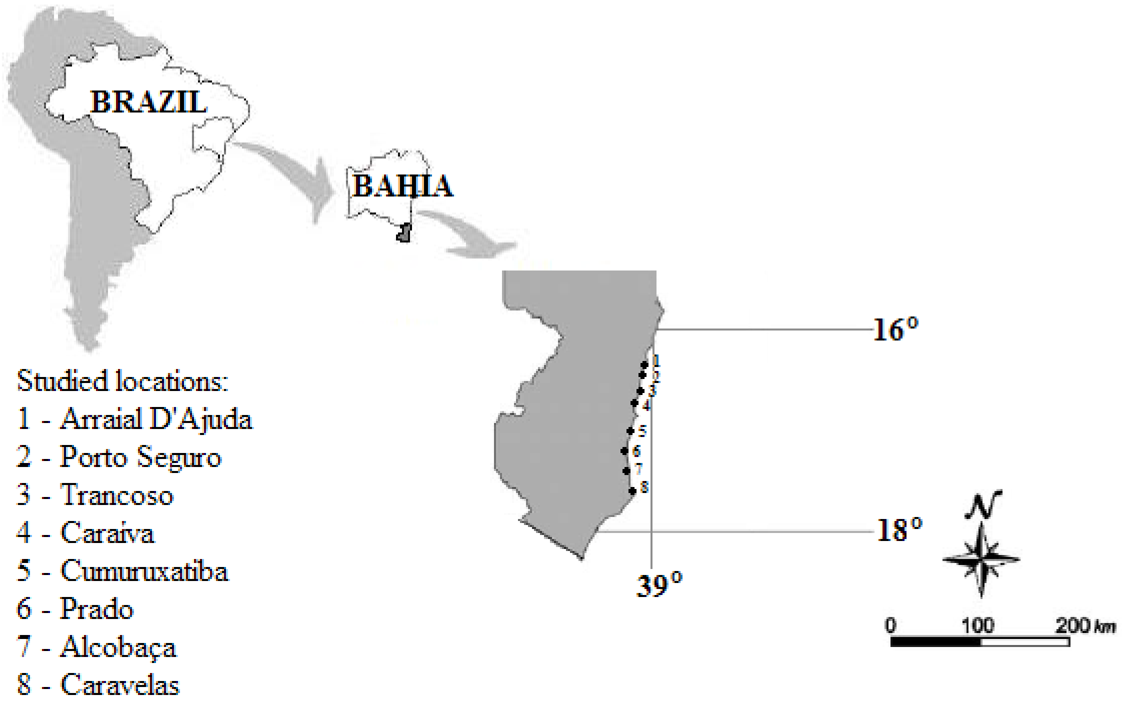

Figure 1. Map showing the region extreme south of Bahia state in Brazil and its geographic location. The numbers (1-8) indicate the location of the beaches studied.

Table 1. Radionuclides, respective analytical technique used and detection limits

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Radionuclide	Technique	Used gamma lines [energy (keV) - emission probability]	Detection limit (Bq/kg)
^{40}K	Gamma Spectrometry Analysis	1460.8 (10.7%)	1.0
^{214}Bi (^{226}Ra)	Gamma Spectrometry Analysis	609.3 (46.3%)	0.4
^{232}Th	INAA	^{233}Pa – 312 (100%)	4.0

2.1.1. Instrumental Neutron Activation Analysis

The activity of uranium and thorium were determined by using the TRIGA MARK I IPR-R1 Reactor, installed at *Centro de Desenvolvimento da Tecnologia Nuclear/Comissão Nacional de Energia Nuclear*, Belo Horizonte in Brazil and irradiated with flux of neutrons of 1.8 x 10^{11} n/cm^2/s and was followed a methodology of analysis itself. The samples were homogenized, weighed and irradiated in the pneumatic system, specially designed for this type of analysis. Thorium was analyzed by INAA, counting the gamma line of ^{233}Pa [5].

2.1.2. Gamma spectrometry

The specific activities of the radionuclides ^{226}Ra and ^{40}K in collected samples was determined using a system from *CANBERRA*, consisting of a hyper-pure germanium detector (HPGe), coaxial geometry, 15% efficiency, with data acquired and treated with *Genie 2000* software.

The samples after of weighed were transferred to Marinelli beakers (500mL). Each sample was hermetically and carefully sealed 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, all samples were subjected to gamma spectrometric analysis [6].

2.1.3. Dose calculation

The total air absorbed dose rate (nGy/h) 1 m above the ground due to the specific activities of ^{226}Ra, ^{232}Th and ^{40}K (Bq/kg) was calculated using the equation [1,3]

$$D = 0.0417A_K + 0.462A_{Ra} + 0.604A_{Th}, \tag{1}$$

Where D is the absorbed dose rate, A_K, A_{Ra} and A_{Th} are the activity concentrations for ^{40}K, ^{226}Ra and ^{232}Th respectively.

To estimate the annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose (0.7 Sv/Gy) and outdoor occupancy factor (0.2) proposed by [1] were used. The effective dose rate in units of mSv/year was calculated by following equation:

$$H_E = DTF, \tag{2}$$

where D is the calculated dose rate (in nGy/h), T is the outdoor occupancy time (24 h x 365.25 days x 0.2 = 1,753 h/year), and F is the conversion factor (0.7 Sv/Gy).

2.1.4. Radiation hazard indexes

According [3], sand beach minerals, rejected light sands and sea beach soils can be used in industries and building constructions, the γ-ray radiation hazards due to the specified radionuclides were assessed by three different indices.

When comparing the specific activity of samples containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K, it is necessary to introduce the term radium equivalent activity ($Ra_{eq)}$. It was defined as a single quantity that represents the combined specific activities of ^{226}Ra, ^{232}Th and ^{40}K, and develops a numerical indicator of an external dose to public and internal dose due to radon and its daughters. The following equation to calculate radium equivalent activity and stated the value of 370 Bq/kg as the maximum allowed value for public dose considerations according to [7].

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K, \quad (3)$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq/Kg respectively. Another radiation hazard index called the representative level index I_{yr} is defined as follows [8]

$$I_{yr} = (1/150)A_{Ra} + (1/100)A_{Th} + (1/1{,}500)A_K, \quad (4)$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq/Kg respectively.

The external hazard index (H_{ex}) is a radiation hazard index defined by [7] to evaluate the indoor radiation dose rate due to the external exposure to γ-radiation from the natural radionuclides in the 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/year [7] or 1.0mSv/year [3] based on the formula

$$H_{ex} = (A_{Ra}/370) + (A_{Th}/259) + (A_K/4810) \leq 1, \quad (5)$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq/Kg respectively. The maximum value of H_{ex} equal to unity corresponds to the upper limit of Ra_{eq} (370 Bq/kg).

3. RESULTS AND DISCUSSIONS

The activity concentrations of radionuclides measured in beach sand samples from extreme south of Bahia, Brazil are presented in Table 2. The naturally occurring ^{226}Ra, ^{232}Th and ^{40}K in beach sand ranged from 14.5 to 8,318.4, 20.5 to 18,450.0, 15.4 to 3,109.0 Bq/kg, with a mean value of 1,078.2, 2,429.6, and 417.0 Bq/kg respectively.

As shows in Table 2, the values of ^{226}Ra and ^{232}Th in Cumuruxatiba, Trancoso and Alcobaça are higher than the range of the corresponding typical world values [1] that are 50 and 50 Bq/kg respectively. The value of ^{232}Th in Caraíva and ^{40}K in Cumuruxatiba also has been higher than the range of the corresponding typical world values. The corresponding typical world value of ^{40}K is 500 Bq/kg.

Table 2. The activity concentration, in Bq/Kg, of beach sand samples

Radionuclides	^{226}Ra (^{238}U series)	^{232}Th (^{232}Th series)	^{40}K
Location			
Arraial D'Ajuda	28.2	41.0	15.4
Prado	14.5	26.2	16.6
Alcobaça	80.7*	405.9*	40.8
Porto Seguro	15.7	20.5	19.0
Trancoso	90.5*	168.1*	42.0
Caraíva	39.1	295.2*	17.8
Caravelas	38.4	29.9	75.4
Cumuruxatiba	8318.4*	18450.0*	3109.0*

* Values higher than the typical world value

In Table 3, the results obtained for the radium equivalent activity (Ra_{eq}), the representative level index (I_{yr}), external hazard index (H_{ex}), the total absorbed dose rate in air due to gamma radiation (D), as well as the outdoor annual effective dose rate (H_E) assessment for beach sand samples are presented.

Table 3. Characteristics of the radioactivity in sand samples collected from extreme south of Bahia, Brazil

Location	Radiation Hazard Indexes			Total air absorbed dose rate	Annual effective dose
	Ra_{eq}(Bq/kg)	I_{yr}(Bq/kg)	H_{ex}	D(nGy/h)	H_E(mSv/year)
Arraial D'Ajuda	87.9	0.61	0.24	38.4	0.047
Prado	53.2	0.37	0.14	23.2	0.028
Alcobaça	**664.0**	**4.62**	**1.79**	284.2*	0.349**
Porto Seguro	46.4	0.32	0.13	20.4	0.025
Trancoso	333.8	**2.31**	0.90	145.1	0.178**
Caraíva	**462.5**	**3.23**	1.25	197.1	0.242**
Caravelas	86.5	0.61	0.24	39.0	0.048
Cumuruxatiba	**34,919.5**	**242.03**	**94.36**	15,116.6*	18.539**

Values in bold represent use unsafe in building constructions
* Values higher than the world range (10-200 nGy/h)
**Values higher than the worldwide average (0.07 mSv/year)

The values of Ra_{eq}, I_{yr} and H_{ex} in Alcobaça and Cumuruxatiba are higher than the acceptable limits which are 370 Bq/kg, 1 Bq/kg and 1.5 Bq/kg [1,9,10]. Caraíva is values of Ra_{eq} and I_{yr} that suggest use unsafe in building constructions. Trancoso is value exceeded for I_{yr}.

As can be observed in Table 3 the values of Alcobaça and Cumuruxatiba are higher than the estimate of average global primordial radiation of 59 nGy/h and than the world range (10-200 nGy/h) [1].The results show that the absorbed dose rates range from 20.4-15,116.6 nGy/h with mean value of 1762.7. The highest value of gamma dose rates among the studied beaches was found in Cumuruxatiba (15,116.6 nGy/h), Areia Preta in Guarapari for example is ranged from 90 to 90,000 nGy/h [1,11].

The annual effective dose equivalent was determined as recommended by [1]. In studied locations, the annual effective dose varied between 0.028 and 18.539 mSv/year, with a mean of 2.162 mSv/year. Values of Alcobaça, Trancoso, Caraíva and Cumuruxatiba are higher than the worldwide average for outdoor annual effective dose, 0.07 mSv/year, reported by [1].

4. CONCLUSIONS

The analytical results confirmed that the samples of this particular area significantly contain three radioactive isotopes (^{226}Ra, ^{232}Th and ^{40}K). The values of activity concentrations of these elements (Bq/kg) in Cumuruxatiba were higher than others studied locations and than the typical world value, 8318.4, 18,450.0 and 3109.0 Bq/kg respectively. The values of total air absorbed dose rate of Alcobaça (284.2 nGy/h) and Cumuruxatiba (15,116.6 nGy/h) were higher than the estimate of average global primordial radiation of 59 nGy/h and than the world range (10-200 nGy/h). Values of annual effective dose in Alcobaça, Trancoso, Caraíva and Cumuruxatiba are higher than the worldwide average for outdoor annual effective dose, 0.07 mSv/year. The values of Ra_{eq}, I_{yr} and H_{ex} in Alcobaça, Caraíva and Cumuruxatiba are higher than the acceptable limits that are 370 Bq/kg, 1 Bq/kg and 1.5 Bq/kg. Therefore, these sands are unsafe for use in building constructions. The presence of magnetite, ilmenite and monazite may be the reason of black colorations of the sand of Cumuruxatiba, as is suggested by some studies.

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