X Congreso Regional Latinoamericano IRPA de Protección y Seguridad Radiológica
"Radioprotección: Nuevos Desafíos para un Mundo en Evolución"
Buenos Aires, 12 al 17 de abril, 2015
SOCIEDAD ARGENTINA DE RADIOPROTECCIÓN

NATURAL OCCURRING RADIONUCLIDES IN NOVEL SAND BEACHES FROM ESPÍRITO SANTO, STATE, BRAZIL.

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ABSTRACT

Novel locations in Espírito Santo State, southeast Brazil, from the mouth of Itapemirim River (south of the state) to the mouth of the river Cricaré (upstate) were collected and the natural radioactivity in the beach sand samples was determined by measuring the ^{226}Ra, ^{232}Th and ^{40}K concentration. In these sands, the minerals from the silica group (white up to yellow) represent the major concentration and the tonal variation is a strong indication of an anomalous radioactive region. Sand samples tightly sealed in standard 100 mL HDPE flasks were measured by high resolution gamma spectrometry after a resting time of approximately 4 weeks before counting, in order to ensure secular equilibrium of natural uranium and thorium series. The ^{226}Ra concentration was determined as the weighted mean from the average concentrations of ^{214}Pb and ^{214}Bi, the ^{232}Th concentration was determined as the weighted mean from the average concentrations of ^{228}Ac, ^{212}Pb and ^{212}Bi and the ^{40}K concentration by its single radiation of 1460 keV. The results show the higher concentrations for the south region (Marataizes), namely 860±70 Bq·kg^{-1} for ^{226}Ra, 2300±260 Bq·kg^{-1} for ^{232}Th and 270±40 Bq·kg^{-1} for ^{40}K. For Great Vitoria central metropolitan region, the higher values were 3870±290 Bq·kg^{-1} for ^{226}Ra 11070±1960 Bq·kg^{-1} for ^{232}Th and 270±40 Bq·kg^{-1} for ^{40}K. For the upstate region (Guriri), the higher values were 60±5 Bq·kg^{-1} for ^{226}Ra, 80±10 Bq·kg^{-1} for ^{232}Th and 110±10 Bq·kg^{-1} for ^{40}K.

1. INTRODUCTION

The main external source of irradiation to the human body are the natural occurring radionuclides elements in the soil and rock, namely ^{40}K and the radionuclides from ^{238}U and ^{232}Th series present throughout the Earth's crust [1].

Brazil is among the countries with the largest and most important deposits of these minerals on Earth. The largest anomalies in the concentration of thorium in soil in Brazil are deposits of monazite sands in Guaraparí, Municipality of metropolitan region of Espírito Santo (southeast of Brazil) [2, 16].
Favored by geographical conditions and ancient sedimentary processes, the Epírito Santo estate has the constitution of their sands, minerals such as ilmenite ($FeTiO_4$), monazite ([Ce, La, Nd, Th]PO_4), rutile (TiO_3) and zirconite ($ZrSiO_4$), natural sources of minerals such as ^{40}K, ^{232}Th and ^{238}U, responsible for the activity detected and increased background radiation [2-3].

Three sampling locations were selected considering the tonal differentiation of the sands and geographic position in relation a local river mouth (the sedimentary and cumulative processes may be favored by the rivers): Guriri beach in São Mateus Municipality (upstate) nearby of

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Cricaré River, Curva da Jurema Beach in central region of Vitória (several river mouths) and Marataízes beach in Marataízes Municipality, nearby of Itapemirim River (Figure 1).

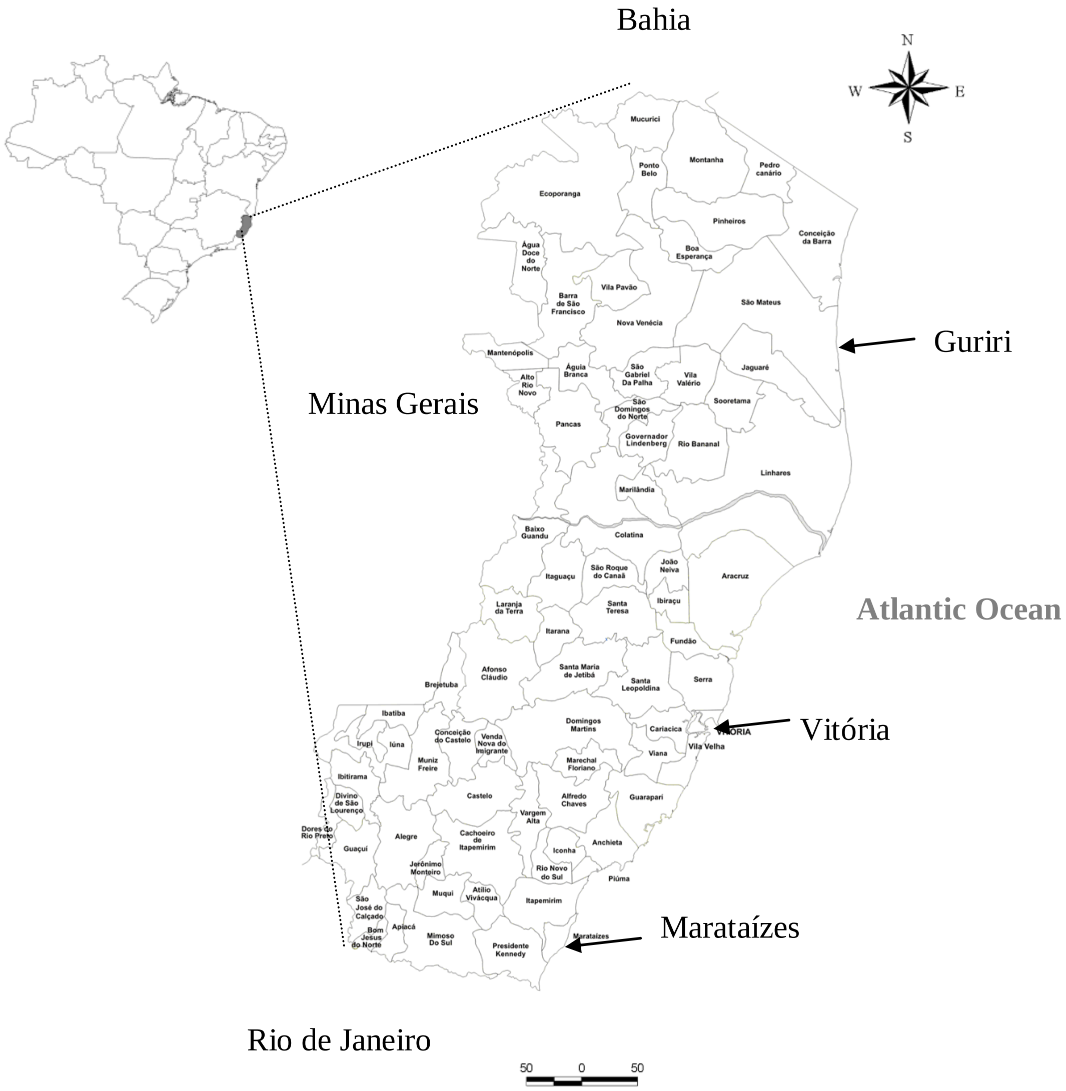

Figure 1. Sampling locations.

In this work, novel locations in Espírito Santo state, southeast Brazil, from the mouth of Itapemirim river (south of the state) to the mouth of the river Cricaré (upstate) were collected and the natural radioactivity in the beach sand samples, was determined by measuring the ^{226}Ra, ^{232}Th and ^{40}K concentration activities, establishing a comparison with other regions already studied in the state of Espírito Santo, comparing with Brazilian and world averages.

2. MATERIALS AND METHODOLOGY

2.1. Sampling Collection and Preparation

In the sampling process, the location was selected considering the apparent tonality variation, which suggest concentration of black sands (ilmenitics mineral) and red to brown sands (monazitics sands).

Superficial black sands samples were collected with a 2 cm a depth on black spots. After manually removing macro-impurities (like shells, pebbles, food leftovers), the sands were dried at 80°C for 24h, and therefore sealed in 100-mL HDPE flat-bottom cylindrical flask and stored for approximately 4 weeks before counting, in order to allow the reaching of secular equilibrium in the ^{238}U and ^{232}Th series [4]. For each location, the samples were prepared in triplicate.

2.2. Measurements

All samples were measured by high resolution gamma spectrometry with a coaxial high-purity germanium detector (HPGe) of 15% relative efficiency with conventional electronics and a 919 ORTEC EG&G Spectrum Master 4k multichannel analyzer. The measured resolution for the ^{60}Co 1332.5 keV is 2.9 keV. The HPGe detector and the samples were placed inside a conventional lead shield, with 10 cm of thickness and the counting time for all samples was 1000 s. All spectra were analyzed with the WinnerGamma software [5].

The activity concentration of a single transition is classically calculated as in Expression 1[4]:

$$A(x) = \frac{C(E)}{P_\gamma(E) \cdot \varepsilon(E) \cdot m \cdot t} \qquad (1)$$

Where A(x) is the activity of the considered gamma transition of the isotope “x” in the sample (Bqkg^{-1}), C(E) is the net number of counts obtained for the gamma transition with energy (E) emitted by “x”, Pγ(E) is the probability of emission of the gamma transition with energy (E), ε(E) is the detector efficiency for the considered gamma transition, m is a sample mass (kg) and t is the counting time in seconds. The radionuclides and respectively gamma transitions are shower in the Table 1.

Table 1. Energy and gamma transitions applied.

Nuclide		E (Energy) [keV]	Pγ(E) [%]
^{226}Ra	^{214}Pb	295.21	18.70
		351.92	35.80
	^{214}Bi	609.32	45.00
^{232}Th	^{212}Pb	238.63	43.50
		300.09	3.25
	^{212}Bi	727.33	6.64
	^{228}Ac	911.07	27.80
		968.90	16.74
^{40}K	^{40}K	1.460.83	10.67

The activity concentration Ac in $Bqkg^{-1}$ is determinate by de pondered mean applied to the values of activity concentration for a single transition in according with the Expression 2:

$$Ac = \frac{\sum_{i=1}^{n} \frac{A_i(x)}{\sigma_i^2}}{\sum_{i=1}^{n} \frac{1}{\sigma_i^2}} \qquad (2) \qquad (2)$$

Where σ_i is the uncertainty associated with activity concentration for a single transition in $Bqkg^{-1}$. The associated activity concentration σ_c is determinate in according with the Expression 3;

$$\sigma_c = \frac{1}{\sqrt{\sum_{i=1}^{n} \frac{1}{\sigma_i(x)^2}}} \qquad (3) \qquad (2)$$

Where $\sigma_i(x)$ is an uncertainty associated with activity concentration in $Bqkg^{-1}$.

The background radiation was determined by measuring a high purity water sample in the same geometry as the sand samples.

The detector efficiency curve was determined with a standard multi-radionuclide aqueous solution in the same geometry as all measured samples. The high-resolution gamma spectrometry methodology and the detector efficiency curve are regularly verified through proficiency tests [5].

All nuclides activities are given with uncertainty statistics at $\pm 1\sigma$ confidence level. Detections limits are given at $\pm 2\sigma$ confidence level with the GTN5 formulae.

3. RESULTS AND DISCUSSION

The results show the higher concentrations for the south region (Marataizes), namely 860±70 Bq·kg⁻¹ for ^{226}Ra, 2300±260 Bq·kg⁻¹ for ^{232}Th and 270±40 Bq·kg⁻¹ for ^{40}K. For Great Vitoria central metropolitan region, the higher values were 3870±290 Bq·kg⁻¹ for ^{226}Ra 11070±1960 Bq·kg⁻¹ for ^{232}Th and 270±40 Bq·kg⁻¹ for ^{40}K. For the upstate region (Guriri), the higher values were 60±5 Bq·kg⁻¹ for ^{226}Ra, 80±10 Bq·kg⁻¹ for ^{232}Th and 110±10 Bq·kg⁻¹ for ^{40}K.

The measured values and other literature comparative values to the concentrations values of ^{40}K, ^{232}Th and ^{226}Ra, together with their standard deviations, are represented in Table 2.

Table 2. Measured results and literature comparative values for activity concentrations.

Beach Location	Municipality	^{40}K	^{232}Th	^{226}Ra	Ref.
Present Work*		Bq·kg⁻¹	Bq·kg⁻¹	Bq·kg⁻¹	
Guriri Beach	São Mateus	110±10	80±10	60±5	*
Marataises Beach	Marataises	270±40	2300±260	860±70	*
Curva da Jurema Beach	Vitória	970±120	11070±1960	3870±290	*
Other locations in Espírito Santo State					
Preta Beach (Black Spot)	Guarapari	4320±570	37000±2600	2880±215	[11]
Camburi Central	Vitória	380±20	4160±260	755±40	[11]
Ilha do Boi Beach	Vitória	150±20	1100±80	297±76	[11]
Ilha do Frade Beach	Vitória.	2200±300	19800±1300	3220±200	[12]
Average values for other locations World					
Cuba		188±10	15.6±1.1	16.7±1.2	[6]
Egyptian		385±8.9	27±4.2	33±3.7	[7]
China		809.8±207.1	35.5±11.0	23.0±6.6	[8]
India		210.1-607.0	37.0-163.0	9.6-53.0	[9]
Japan		611.2-1176.0	10.2-33.7	29.2-46.3	[10]
Brazil		**51-809**	**7,1-213**	**10,2-116**	**[13-15]**
World		**400**	**30**	**35**	**[1]**

** Measurements with 68% (± 1σ) confidence level, k=1

The measured values in upstate, central e south regions of the Espírito Santo State are very high in relation of other places in the World.

For comparative effect, the measured values and the Brazilian and World averages are shown in a Figure 2:

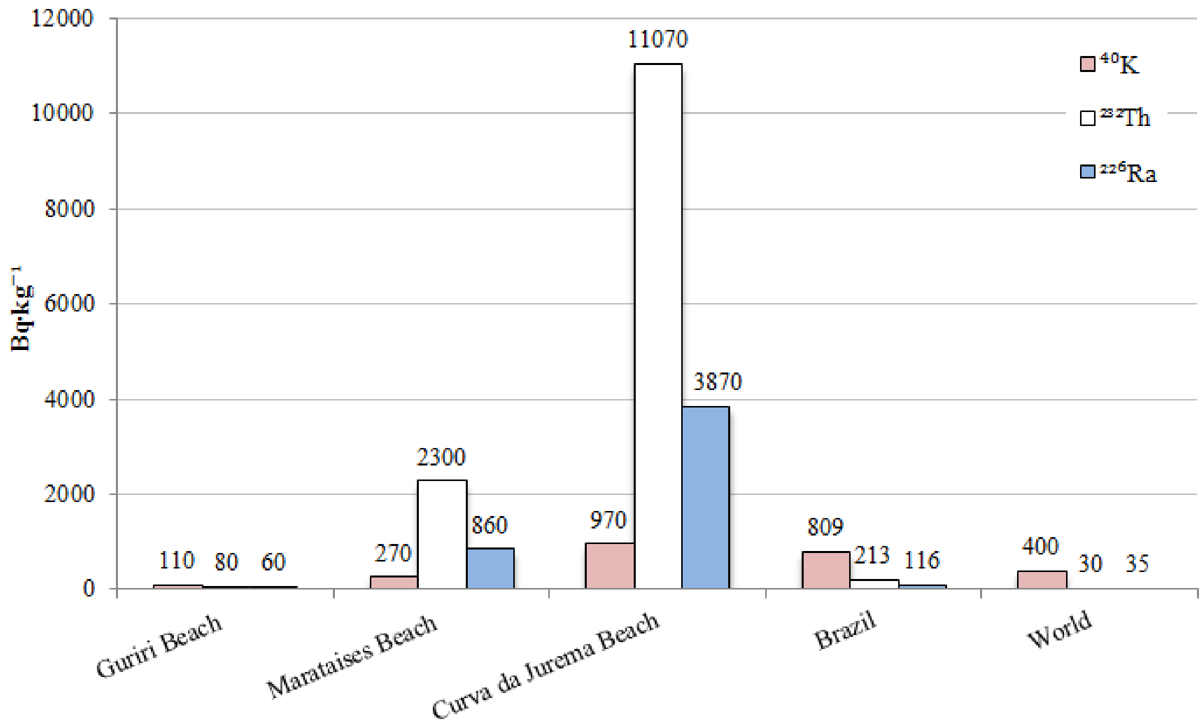

Figure 2. Measured values and comparative average values.

The activity concentration in Curva da Jurema Beach (central region of Espírito Santo State) for ^{232}Th is 50 times higher than a Brazilian average and, 370 times higher in relation a World average for this radionuclide. For the activity concentration in a same region to the ^{226}Ra, is 33 times higher in relation a World average and 110 times higher in relation a Brazilian average for this radionuclide. For the ^{40}K, the values are around 2.5 times highs in relation the World and Brazilian averages. For all reasons the values are very high for a natural background radiation in the considered region.

For the Guriri Beach (upstate region of Espírito Santo State), the results showed 3 times higher in relation the World average for the ^{232}Th, 2 time higher for the ^{226}Ra and 0.5 times high. In relation a Brazilian average, these values are approximately equals varying around 0.1 and 0.5 times more than the considered literature average.

In Marataizes Beach (south region of the Espírito Santo State) the values determined to ^{232}Th in relation a Brazilian average is a 10 times higher and 77 times higher in relation a World average. The ^{226}Ra in relation a Brazilian average is a 7 times higher and 25 timer higher in relation of the World average.

4. CONCLUSIONS

The activities concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Marataízes Beach, Guriri Beach and Curva da Jurema Beach in Espírito Santo state, southeast of Brazil, were evaluated by high resolution gamma-ray spectrometry.

The highest activity concentration has been observed for Curva da Jurema Beach in central region of the Espírito Santo State. The measured values for this location are high considering the world average values [1].

The values determined for activity concentrations of ^{232}Th in the sand samples showed that a major mineral constituent in the analyzed samples is the monazite sand.

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