RADIATION HAZARDS EVALUATION FOR SELECTED SAND SAMPLES FROM CAMBURI BEACH, VITÓRIA, ESPÍRITO SANTO, BRAZIL

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ABSTRACT

In this work, a single location at Camburi beach, known to be a naturally high background region, was studied. Radiation hazards indexes and annual effective dose were evaluated from the ²²⁶Ra, ²³²Th and ⁴⁰K sands activities concentrations. Sand samples were monthly collected during 2011, dried, sealed in standard 100 mL HPDE polyethylene flasks and measured by high resolution gamma spectrometry after a 4 weeks in-growth period. The ²²⁶Ra concentration was determined from the weighted average concentrations of ²¹⁴Pb and ²¹⁴Bi. The ²³²Th concentration was determined from the weighted average concentrations of ²¹⁸Ac, ²¹²Pb and ²¹⁴Bi and the ⁴⁰K from its single gamma transition. The results, considering samples gamma-rays self-attenuation, show activities concentrations in the range from 6 Bq kg⁻¹ to 39 Bq kg⁻¹ for ²²⁶Ra, 13 Bq kg⁻¹ to 161 Bq kg⁻¹ for ²³²Th, and 7 Bq kg⁻¹ to 65 Bq kg⁻¹ for ⁴⁰K. The radium equivalent activity for the studied samples ranged from 26 Bq kg⁻¹ to 274 Bq kg⁻¹. The external and internal hazard indexes varied, respectively, from 0.07 to 0.74 and from 0.09 to 0.85. The annual effective dose values laid from 0.07 mSv.y⁻¹ to 0.72 mSv.y⁻¹. All values obtained in this work are below the radiological protection recommended limits.

1. INTRODUCTION

Throughout the history of life on Earth, gamma ray exposure from ⁴⁰K and radionuclides from the ²³⁸U and ²³²Th series occurs naturally in the environment. These radionuclides are present in rocks and soils and their distribution depends upon the local geology of each region in the world [1]. They are an important source that contributes to an increase in the dose received by humans. In order to determine the level of public exposures, the knowledge about activity concentration and distribution of natural radionuclides in environmental sites plays an important role.

Previous works show that Camburi beach, Espírito Santo State, is a naturally high background region in Brazil [2, 3, 4], whose sands are rich in silica minerals (SiO₂) and also monazitic ((Ce, La, Nd, Th)PO₄) and ilmenitic (FeTiO₃) minerals [5]. These accessory minerals contain traces of radioactive elements as 226 Ra (from the 238 U serie), 232 Th and 40 K. Beach sands are mineral deposits, remainders of geological varied formations, which are transported by wind, by tidal and by the actions of waves and currents [6]. In order to investigate the activity concentration of 226 Ra, 232 Th and 40 K and the respective radiation hazards in sands from Camburi beach, 11 selected points along the beach extension were

assessed during the 2011 year, with monthly collected samples. In this work, the follow-up of the concentrations activities and radiation hazards for one single beach location is presented.

2. MATERIALS AND METHODOLOGY

2.1. Study Area

Camburi beach is located in Vitória, capital of Espírito Santo State, Brazil, as shown in Fig. 1. Camburi is the only beach located in the mainland city, with an extension of 6 km. The city has approximately 300 thousand inhabitants [7] and the weather is warm almost the entire year, so the beach is constantly frequented both by locals and tourists.



Figure 1. (a) Brazil map (in red Espírito Santo State) [8]; (b) Espírito Santo State map (in red Vitória city) [9]; (c) Geographic location of Camburi beach on the coast of Espírito Santo State, Brazil [10].

2.2. Samples Collection and Preparation

The location selected (Fig. 2), with geographical coordinates 20°17'8.47''S 40°17'24.24''W, represents the most frequented region of Camburi beach, with the higher concentration of locals and tourists. A speckle pattern is visible in the region next the collecting point, due to the presence of ilmenite and monazite (Fig. 3).

The sand samples were collected every 15th of the month, from January to December 2011, almost at the beach surface, within a depth of about 2 cm, simulating a person sitting on the beach sand.

After manually removing macro-impurities (like shells, pebbles, food leftovers), the sand samples were dried on a stove and sealed in 100-mL HDPE flat-bottom cylindrical flasks with 52.5 mm plan screw cap and bubble spigot.



Figure 2. Camburi beach. "A" red bubble shows the Dante Micheline Avenue and the green arrow indicates the location of the collection point [10].



Figure 3. Camburi beach. A speckled pattern in the sand is visible next to sea line due to the presence of ilmenite and monazite. Photo: Personal archive.

2.3. Measurements

All samples were measured by high resolution gamma-ray spectrometry, after a 4 weeks ingrowth period, in order to allow secular equilibrium in the ²³⁸U and ²³²Th series [11].

The detection system is a 15% HPGe ORTEC EG&G detector with conventional electronics and a 919 ORTEC EG&G Spectrum Master 4k multichannel analyzer. The effective resolution for the 60 Co 1332 keV energy is 2.8 keV.

The background radiation was obtained with ultrapure water, in the same sample geometry. The efficiency calibration curve was performed with a multinuclide standard aqueous solution in the same geometry as all measured samples.

All spectra were analyzed with the InterWinner 6.0 software [12], considering samples gamma-rays self-attenuation.

The activity concentration of a single transition was calculated as [11]:

$$A_{i}(E_{\gamma i}) = \frac{C_{\gamma i}}{\varepsilon_{\gamma i} \mathbf{I}_{\gamma i} t_{l} . m} \times f(E_{\gamma i})$$
(1)

Where:

 $A(E_{\gamma i}) =$ activity of the considered gamma transition of the isotope X in the sample (Bq.kg⁻¹); $C_{\gamma i} =$ net area for the gamma transition with energy ($E_{\gamma i}$) emitted by X; $\epsilon_{\gamma i} =$ detector efficiency for the considered gamma transition;

 I_{yi} = probability of emission of the gamma transition, I_{yi} = probability of emission of the gamma transition with energy (E);

 t_1 = counting live time (s);

m = sample mass (kg);

 $f(E_{\gamma i}) =$ self-attenuation factor for the considered gamma transition.

The activity of ⁴⁰K was calculated through its single gamma transition of 1460.83 keV. The activity of ²²⁶Ra was determined by the weighted mean of the ²¹⁴Pb (295.21 keV and 351.92 keV) and ²¹⁴Bi (609.32 keV) gamma transitions and the activity of ²³²Th by the weighted mean of the ²¹²Pb (238.63keV and 300.09 keV), ²¹²Bi (727.33 keV) and ²²⁸Ac (911.07 keV and 968.90 keV) gamma ray transitions [12].

2.4. Self-Attenuation Factors

The high-resolution gamma-ray spectrometry is an effective method to analyze the activities of natural radionuclides in soils in order to assess their environmental impact. Low energy gamma rays have less penetrating ability and tend to interact more readily with matter, so, the gamma ray interaction is different for the sand samples and the aqueous standard solution [13]. The self-attenuation phenomenon depends on the sample composition, its density and the gamma emission energy.

Self-attenuation factors are needed because the sand samples from Camburi Beach have apparent densities in the range of 1.59 g/cm³ to 1.64 g/cm³ and the efficiency curve was

obtained with a standard aqueous solution [14]. The apparent density was determined dividing the mass in grams of each sample by the flask volume of 100 mL. The self-attenuation correction factors were experimentally determined by the Cutshall method [13]. The method is based on measuring the transmission of gamma-ray through the sand sample and an ultrapure water sample in the same geometry, with gamma transitions in the range of interest. Punctual standard gamma-ray sources of ⁶⁰Co, ¹³³Ba, ¹³⁷Cs and ¹⁵²Eu with gamma transitions ranging from 81 keV to 1408 keV were used, in order to cover the energies of the radionuclides used for the concentrations activities calculations.

2.5. Radium Equivalent Activity (Ra_{eq})

As the distribution of radioactivity in natural samples is not uniform, in order to assess the health effects from the natural radioactivity, the activity of ²²⁶Ra, ²³²Th and ⁴⁰K are converted into a single quantity termed Ra_{eq}, assuming that 370 Bq kg⁻¹ de ²²⁶Ra, 259 Bq kg⁻¹ de ²³²Th e 4810 Bq kg⁻¹ de ⁴⁰K produce an equal gamma ray dose rate [15].

The radium equivalent activity index is calculated using the equation [16]:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \tag{2}$$

where A_{Ra} , A_{Th} and A_K are the activities concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K, respectively.

The Ra_{eq} is a useful guide in assessing the safety standards of radiation protection for individuals of the public residing in buildings. For safe use as construction materials, it is recommended that the Ra_{eq} value does not exceed the limit of 370 Bq.kg⁻¹ [16].

2.6. External (Hext) and Internal (Hint) Hazard Indexes

The external (H_{ext}) and internal (H_{int}) hazard indexes are calculated, respectively, from equations (3) and (4) [17]. The H_{int} considers the risk of inhalation of radon accumulation and their decay products of short half-life.

$$H_{ext} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \le 1$$
(3)

$$H_{\rm int} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \le 1 \tag{4}$$

where A_{Ra} , A_{Th} and A_{K} are the activities concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K, respectively.

For safe use as construction materials, it is recommended that the value of H_{ext} and H_{int} does not exceed the limit of 1, which corresponds to the Ra_{eq} limit of 370 Bq kg⁻¹ [17] that must be observed.

2.7. Annual Effective Dose (E)

The Annual Effective Dose (E) is calculated from equation (5) [4]:

$$E = D.t.0, 7.10^{-6} \tag{5}$$

where D is the absorbed dose rate in the air (nGy h^{-1}), t is the time in hours of exposure per year (24 hours X 365 days X fraction of the day exposed to radiation) and 0.7 is the conversion factor to unit Sv.Gy⁻¹.

The ICRP 60 [18] recommended value is 1 mSv.y^{-1} for the general public.

The absorbed dose rate (D) in outdoor air at 1 m above the ground is given below (eq. 6) [1]

$$D = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \tag{6}$$

where A_{Ra} , A_{Th} and A_K are the activities concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K, respectively.

3. RESULTS AND DISCUSSION

3.1. Activity Concentration and Radium Equivalent Activity in Camburi Beach Sands

The activities concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K and the Radium Equivalent Activity (Ra_{eq}) of sand samples from the collection point (Fig. 2) on Camburi beach are presented in Fig. 4. The activities concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K were calculated by the equation (1) and the Radium Equivalent Activity by the equation (2).



Figure 4. Activities concentrations of ²²⁶Ra, ²³²Th, ⁴⁰K and Ra_{eq} in sand samples from the collection point (with geographical coordinates 20°17'8.47"S 40°17'24.24"W) at Camburi beach, ES, Brazil, throughout the year 2011.

The results, with self-attenuation correction, show activities concentrations in the range from 6 ± 2 Bq kg⁻¹ to 39 ± 2 Bq kg⁻¹ for 226 Ra, 13 ± 1 Bq kg⁻¹ to 161 ± 6 Bq kg⁻¹ for 232 Th and 7 ± 6 Bq kg⁻¹ to 65 ± 9 Bq kg⁻¹ for 40 K.

The Ra_{eq} values range from 26 Bq kg⁻¹ to 274 Bq kg⁻¹ calculated by the eq. 2.

All obtained results for Ra_{eq} are lower than the recommended limit of 370 Bq.kg⁻¹, for safe use as construction materials [16].

3.2. External (Hext) and Internal (Hint) Hazard Indexes in Camburi Beach Sands

The External (H_{ext}) and Internal (H_{int}) Hazard Indexes of sand samples from the collection point (Fig. 2) at Camburi beach are presented in Fig. 5. The H_{ext} and the H_{int} are calculated by the equations 3 and 4, respectively.



Figure 5. External (H_{ext}) and Internal (H_{int}) Hazard Indexes for sand samples from the collection point (with geographical coordinates 20°17'8.47"S 40°17'24.24"W) at Camburi Beach, ES, Brazil, throughout the year 2011.

The external hazard index (H_{ext}) varies from 0,07 to 0,74 and the internal hazard index (H_{int}) from 0,09 to 0,85.

All values obtained for the external and internal hazard index are below recommended limit of 1, for safe use as construction materials [17].

3.3. Annual Effective Dose (E) in Camburi Beach Sands

The Annual Effective Dose of sand samples from the selected location at the most frequented region of Camburi beach is calculated by equation 5, considering the worst case scenario of a 24-hours daily exposure all year round.

The results ranging from $0,07 \text{ mSv.y}^{-1}$ to $0,72 \text{ mSv.y}^{-1}$ are presented in Fig. 6. All values are lower than the limit of 1 mSv y^{-1} , recommended for ICRP 60 for the general public [18].



Figure 6. Annual Effective Dose of sand samples from the collection point (with geographical coordinates 20°17'8.47"S 40°17'24.24"W) at Camburi Beach, ES, Brazil, for the collection months of the year 2011 to entire day exposure.

The results show Annual Effective Dose in the range from 0,07 to 0,72 to entire day exposure throughout the year.

All the Annual Effective Doses obtained are lower than the limit of 1 mSv y^{-1} , recommended for ICRP 60 for the general public [18].

4. CONCLUSIONS

As expected, due to the high concentration of monazite, the concentration of 232 Th is higher than the concentration of 226 Ra and 40 K.

The activities concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K of sand samples from the collection point (with geographical coordinates 20°17'8.47''S 40°17'24.24''W) varied throughout the year 2011 on Camburi Beach, ES, Brazil.

The activities concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K for similar sands of regions with high natural background radiation level [2, 3, 4, 19, 20, 21] are in the range from 5 Bq kg⁻¹ to 1022 Bq kg⁻¹, 2,62 Bq kg⁻¹ to 7236 Bq kg⁻¹ and 11,6 Bq kg⁻¹ to 888 Bq kg⁻¹, respectively. All activities concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K of this work are within these ranges of literature values.

The variation of the activities concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K of sand samples from the collection point throughout the year 2011 should be related that the mineral composition of the superficial sands since weather conditions as wind, the tides and the actions of waves and currents influenced in the transport and deposition of sand.

All results of Ra_{eq} obtained are lower than the upper limit of 370 Bq.kg⁻¹, recommended for safe use in construction materials [16].

All the external (H_{ext}) and internal (H_{int}) hazard index obtained are lower than the limit of 1, recommended for safe use in construction materials [17].

All Annual Effective Doses obtained are lower than the limit of 1 mSv y^{-1} , recommended for ICRP 60 for the general public [18].

A complete assessment of the natural radioactivity of sand samples (2 cm depth) from the collection point on Camburi beach was done in this work. The Camburi beach do not present radiological risk to the population of Vitória, Espírito Santo, Brazil, due to the natural radioactivity of ²²⁶Ra, ²³²Th and ⁴⁰K, considering that all radiation hazards obtained in this work are bellow the recommended limits.

Further, the 226 Ra, 232 Th and 40 K activities concentrations variation will be assessed together with weather conditions such as wind and/or rain and behavior of tides, waves and ocean streams.

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REFERENCES

- 1. UNSCEAR, *Sources and effects of ionizing radiation*, United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations, New York (2000).
- L. F. Barros, B. R. S. Pecequilo and R. R. Aquino, "Gamma radiation measurements in select sand samples from Camburi Beach - Vitória, Espírito Santo, Brazil: Preliminary results." 2011 International Nuclear Atlantic Conference, Belo Horizonte, October 24-28 (2011).
- 3. L. F. Barros and B. R. S. Pecequilo, "Evaluation of Ra, Th, K and radium equivalent activity in sand samples from Camburi Beach, Vitória, Espírito Santo, Brazil." *IX Latin American IRPA Regional Congress on Radiation Protection and Safety*, Rio de Janeiro, April 15-19 (2013).

- R. R. Aquino, "Avaliação da radioatividade natural em areias das Praias da Grande Vitória - Espírito Santo". Thesis (Master Degree) - Instituto de Pesquisas Energéticas e Nucleares- IPEN-CNEN/SP, São Paulo, 2010. 79 p. <u>http://www.teses.usp.br</u> (2010).
- 5. F.B. Machado, C.A. Moreira, A. Zanardo, A.C. Andre, A.M. Godoy ; J.A. Ferreira, T.Galembeck, A.J.R. Nardy, A.C. Artur, M.A.F de Oliveira, *Enciclopédia Multimídia de Minerais*, <u>http://www.rc.unesp.br/museudpm (2012)</u>.
- 6. H. S Eissa, et al., "Radiation Dose Estimation of sand samples collected from different Egyptian beaches", *Radiation Protection Dosimetry*, Vol. 147, pp.533-540 (2010).
- 7. "IBGE: Instituto Brasileiro de Geografia e Estatística. Censo 2010," <u>http://www.ibge.gov.br/censo2010</u> (2012).
- 8. "Brazil map (in red Espírito Santo State)," <u>http://www.guianet.com.br/es/mapaes.htm</u> (2013).
- 9. "Espírito Santo State map (in red Vitória city)," <u>http://www.alunosonline.com.br/geografia/espirito-santo.html</u> (2013).
- 10. "Camburi beach map," https://maps.google.com.br/ (2013).
- 11. G. F. Knoll, *Radiation Detection and Measurement*, Third Printing, New York, United States (1999).
- 12. ORTEC, InterWinnerTM6.0 MCA Emulation, Data Acquisition and Analysis Software For Gamma and Alpha Spectroscopy, TOMCOM Software Ltd., (2004).
- 13. N. H. Cutshall, et al., "Direct analysis of ²¹⁰Pb in sediment samples: self-absorption corrections," *Nuclear Instruments and Methods in Physics Research*, Vol. 206, pp.309-312 (1983).
- 14. L. F. Barros and B. R. S. Pecequilo, "Self-Attenuation Factors in gamma-ray spectrometry of select sand samples from Camburi Beach, Vitória, Espírito Santo, Brazil.," *Radiation Physics and Chemistry*,

http://dx.doi.org/10.1016/j.radphyschem.2012.12.031 (2013).

- 15. E. Stranden, "Some aspects on radioactivity of building materials". *Physica Norvegica*., **Vol. 8**, pp. 167 (1976).
- 16. OECD. Organization for Economic Cooperation and Development. Exposure to radiation from natural radioactivity in building materials. Report by a Group of Experts of the OECD Nuclear Energy Agency, Paris, 1979.
- N. M. Hassan, et al., "Assessment of the natural radioactivity using two techniques for the measurement of radionuclide concentration in building materials used in Japan." J Radioanal Nucl Chem, Vol. 283 pp.15–21 (2010).
- 18. ICRP 60. International Commission on Radiological Protection, Recommendation of the ICRP, publication 60, Ed. Pergamosn Press, Oxford, 1990.
- 19. R. Veiga, et al, "Measurement of natural radioactivity in Brazilian beach sands", *Radiation Measurements*, Vol. 41, pp.189–196 (2006).
- 20. M. Sowmya, et al, "Some natural radioactivity and associated dose rates in soil samples from Kalpakkam, South India." *Radiation Protection Dosimetry*, **Vol.141**, pp. 239-247 (2010).
- Z. Korkulu and N. Özkan, "Determination of natural radioactivity levels of beach sand samples in the black sea coast Kocaeli (Turkey)" *Radiation Physics and Chemistry*, Vol.88, pp. 27-31 (2013).