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Natural radioactivity and external hazard index in Brazilian sands

M. A. Guazzelli[1] · **N. H. Medina**[2] · **V. A. P. Aguiar**[2]

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Abstract
The distribution of natural radiation from Brazilian beach sands was studied using gamma-ray spectrometry. While in most of the regions studied the dose due to external exposure to gamma-rays, proceeding from natural terrestrial elements, are within the values 0.3 and 1.0 mSv/year, some sand samples from Bahia, Rio de Janeiro and Sao Paulo states present higher radioactivity levels due to the presence of monazite and zircon, exceeding the world average value for external exposure due to naturally occurring radionuclides.

Keywords Natural radioactivity · Terrestrial external radiation · Effective dose · Sands · Gamma spectrometry

Introduction

Soils and rocks contain naturally occurring radioactive material (NORM) results which are an unavoidable feature of life on Earth, and are present in its crust since its origin [1–5]. Primordial radionuclides with long half-lives, such as ^{40}K and the elements of the series of ^{238}U and ^{232}Th, decaying to stability and emitting ionizing radiation in the process, are found in most soils. The gamma radiation from ^{40}K, which represents only 0.0117% of natural potassium in nature, comes from electron capture decay (10.7% of the cases) to stable ^{40}Ar, emitting a gamma-ray of 1460 keV (the other 89.3% β-decays, producing the stable nucleus ^{40}Ca). The lifetime of ^{40}K is 1.25×10^9 years [6]. The decay-chains of ^{232}Th and ^{238}U are sequences of unstable nuclei that decay by emitting gamma-rays, alpha or beta particles until the series stop at the stable nuclei ^{208}Pb and ^{206}Pb, respectively [6].

In most regions, the natural radiation activity varies only within narrow limits [5], but in some places there are wide deviations from world average values due to high concentrations of radioactive minerals. There are a few regions in the world which are known for being high background radiation areas (HBRAs) due to geological formation and geochemical effects [5]. The higher concentration of radionuclides in the Earth's crust is due to minerals such as monazites and zircons [7]. These HBRAs are found at Guarapari, in the coastal region of Espírito Santo and the Morro do Ferro in Minas Gerais, Brazil [5, 8–11]; Yangjiang, in China [5, 12]; southwest and east coast of India [5, 7, 13]; Ramsar and Mahallat in Iran [5, 14]; in the United States and Canada [15], and in some other countries [5]. In Refs. [16, 17], there are a few measurements of the activity concentration in sand used for civil construction in the Brazilian states of São Paulo, Rio Grande do Norte and Espírito Santo. They present values of 51 to 809 Bq/kg due to the primordial radionuclide ^{40}K, 10.2 to 24.1 Bq/kg from the uranium series, and 12.6 to 180 Bq/kg from the thorium series. The radiation exposure of the human body from external terrestrial sources is mainly due to gamma rays coming from the natural radionuclides. According to Ref. [5], about 2.0 mSv of human exposure is due to natural radiation, including radon (1.15 mSv), and including also 0.29 mSv due to ingestion exposures [5]. The resulting worldwide average of the annual effective dose is 0.48 mSv (0.41 mSv for indoors and 0.07 mSv for outdoors), with typical range between 0.3 mSv and 1.0 mSv values which are the typical values depending on the radionuclide composition of the soil [5, 16]. The study of the radionuclide distribution in soils allows us to understand the radiological implications relative to the exposure of the humans to ionizing radiation, the knowledge of the geographic region of the radioactive series [2, 8, 17–30], and for geochronology applications. Natural background radiation studies are

✉ M. A. Guazzelli
marcilei@fei.edu.br

[1] Centro Universitário da FEI,
São Bernardo do Campo 09850-901, Brazil

[2] Instituto de Física da Universidade de São Paulo,
São Paulo 05508-090, Brazil

needed also to establish reference levels, especially in areas where the risk of radioactive exposure may be higher. As the materials used in civil construction can present natural radioisotopes among their constituents, some studies have been carried out with the aim of identifying and quantifying these nuclides [31, 32].

The present work investigated fourteen Brazilian sand beaches in order to analyze the dose from total terrestrial external radiation. We have applied the gamma-ray spectrometry technique to measure the activities from the radionuclides ^{40}K and the elements from the series of ^{238}U and ^{232}Th, and the associated effective dose present in each area studied. In order to verify the minerals responsible for the high activity found in one specific site, the sand samples were analyzed with a scanning electron microscope, using Energy-Dispersive X-Ray Spectroscopy (EDS) microanalysis.

Methodology

Regions studied and sample processing

For this study 100 samples were collected from a sandy region near the Brazilian coast and fourteen Brazilian beaches: 13 of them are located in São Paulo state and one on the island Ilha Grande, Rio de Janeiro state, as shown in Fig. 1. Three samples from Itacaré beach in Bahia were analyzed. The principal features of these regions are cliffs, reefs and beaches of quartz sands. The region under study is dominated by the mountain range, *Serra do Mar*, and the coast is very rugged with many bays and small coves.

As previous studies showed no changes in radionuclide concentration at different depths [30], samples were obtained from a single depth (15–25 cm), except on Preta Beach, Ilha Grande (an island near the city of Angra dos Reis, Rio de Janeiro), which showed a difference in color depending on the depth. In this case, a sample was collected for each of the three depths examined (surface, 15–25 cm and 35–45 cm). All the samples were dried at about 100ºC for 24 h. After sifting materials lower than 1.2(5) mm in diameter, the samples were packed in cylindrical polyethylene boxes. In order to reach secular equilibrium, they were stored for approximately 28 days [5, 15].

Activity and effective dose determination

The activity concentrations of ^{40}K and the series of ^{238}U, ^{232}Th were measured with gamma spectrometry analysis. To detect the emitted gamma rays, we used a measurement system available at the Centro Universitário FEI, São Bernardo do Campo, São Paulo, Brazil, consisting of a 3×3-in^2 NaI(Tl) scintillation detector placed inside a 7.0 cm thick lead shield and a spectrometer unit (Canberra, USA) [30]. The data acquisition and spectra were analyzed using GENIE-2000 software.

The activity calibration was performed by using the IAEA quality assurance reference materials: RGU-238, RGTh-232 and RGK-1, prepared in the same geometry of the soil samples so that the detection geometry remained the same. The determination of effective dose due to external natural radiation was based on the assumption of secular equilibrium and measurements of photopeak gamma radiation from ^{214}Bi decay (1764.5 keV) in the ^{238}U decay series, from ^{208}Tl decay (2614.5 keV) in the ^{232}Th decay series and from

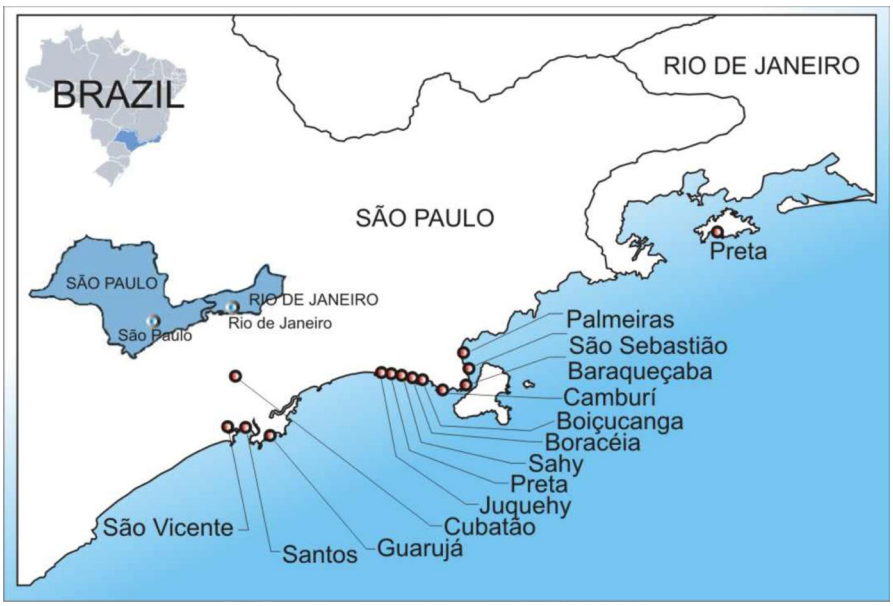

Fig. 1 Map of Southeastern Brazilian coast indicating the main sites studied in this research

primary decay of ^{40}K (1460.8 keV). The spectrum acquisition time of the collected sand samples was set to 8 h.

The absorbed gamma-ray dose rates in air, in Gy/h, were calculated according to Reference [5]:

$$D = \left[\left(0.462 \times A_U\right) + \left(0.604 \times A_{Th}\right) + \left(0.0417 \times A_K\right)\right]$$

where A_U, A_{Th} and A_K are the activities of ^{40}K and the series of ^{238}U and ^{232}The respectively, in Bq/kg. The conversion factors to transform specific activities in absorbed dose rate at 1 m above ground were estimated by Monte Carlo simulation [5].

It is important to note that, if there is any disequilibrium between ^{226}Ra and ^{238}U, the dose estimation is not affected, because about 98% of the external dose from ^{238}U series is delivered by ^{226}Ra sub-series [4, 5]. To estimate annual effective doses, the conversion coefficient must be considered from absorbed dose in air to effective dose received by adults that is 0.7 Sv/Gy. The outdoor and indoor occupancy factors, that are 0.8 and 0.2, respectively, can also be considered to estimate the effective dose [5]. In this paper total external terrestrial radiation was considered, which is the sum of both contributions. Therefore, the annual effective dose, indoors and outdoors, is calculated using the following formula:

$$E_f = D \times 0.7 \times 8760$$

where E_f is the annual effective dose due to total external terrestrial radiation, D is the absorbed dose rate in nGy/h and 8760 is the number of hours for the period of one year.

Since mineral sand can be used in industries and building construction, it is important to know the external gamma-ray hazard index due to ^{40}K, and the elements of the ^{232}Th and ^{238}U series, which considers only the external exposure risk due to gamma-rays. This index value must be less than unity in order to keep the radiation hazard negligible. The radiation exposure due to the radioactivity from construction materials is limited to 1.0 mSvy^{-1}. The external hazard index can be defined as [32, 33]:

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}$$

where A_U, A_{Th} and A_K are the specific activities of ^{238}U, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively.

Results and discussions

The activity, in Bq/kg, of ^{40}K and the elements of the ^{232}Th and ^{238}U series are shown in Table 1. In Fig. 2 total terrestrial external radiation from São Paulo, Rio de Janeiro and Bahia sands are shown. For all regions studied it was found that the main contribution of the effective external radiation dose is due to the elements of the series of ^{232}Th, with a smaller contribution from the isotopes ^{40}K and elements of the ^{238}U chain [34]. Samples collected in Sahy beach showed very different values of natural radioisotope concentrations among each other, and therefore were analyzed separately, namely Sahy I and Sahy II. In this case an average value does not represent the value attributed to natural radiation from the site. This occurrence explains the difficulty of finding an average value representative of a specific region, especially when the soil of the study area is composed of sediments of different mineral origins. Although most of the values found in this work are within the annual typical range (0.3–1.0 mSv) indicated by the United Nations Scientific Committee on the Effects of Atomic Radiation, the results show high doses in Preta Beach in Ilha Grande, Rio de Janeiro, Preta Beach and Sahy in São Paulo, Itacaré in Bahia, exceeding more than twice the international average value for the external exposure due to natural elements. Preta Beach in Ilha Grande, Rio de Janeiro, was already known to present high activity concentration [17, 18]. Itacaré was intensively studied showing high concentration of natural radionuclides [34].

To better understand the high value of the total effective dose found on some beaches, samples of Preta Beach, RJ, were collected at three different depths, since they showed different colors and, after the data analysis, we have found different concentrations of ^{238}U. The gamma ray spectrometry analysis showed that the uranium activity is higher in

Table 1 The activity concentration of naturally occurring radionuclides in the Brazilian sandy sites

Sites	^{40}K (Bq/kg)	^{238}U (Bq/kg)	^{232}Th (Bq/kg)
Itacaré, Ba	< LLD	448 (18)	610 (25)
Sahy I	< LLD	89 (10)	330 (20)
Preta, RJ	36 (25)	112 (15)	235 (6)
Preta, SP	18 (5)	27 (7)	251 (16)
Baraqueçaba	145 (11)	< LLD	< LLD
Cubatão	143 (11)	37 (8)	94 (8)
Juquehi	42 (6)	22 (4)	146 (9)
Palmeiras	312 (20)	< LLD	11 (4)
Boiçucanga	138 (10)	11 (5)	14 (4)
São Sebastião	93 (17))	10 (4)	13 (6)
Santos	145 (25)	38 (4)	6.7 (8)
Camburi	192 (13)	< LLD	< LLD
São Vicente	92 (5)	5.3 (27)	3.6 (18)
Sahy II	41 (7)	< LLD	< LLD
Boracéia	9 (6)	< LLD	< LLD
Guarujá	14 (6)	< LLD	< LLD
Boqueirão	53 (12)	< LLD	< LLD

LLD is defined as 3σ above the background

LLD lower limit of detection

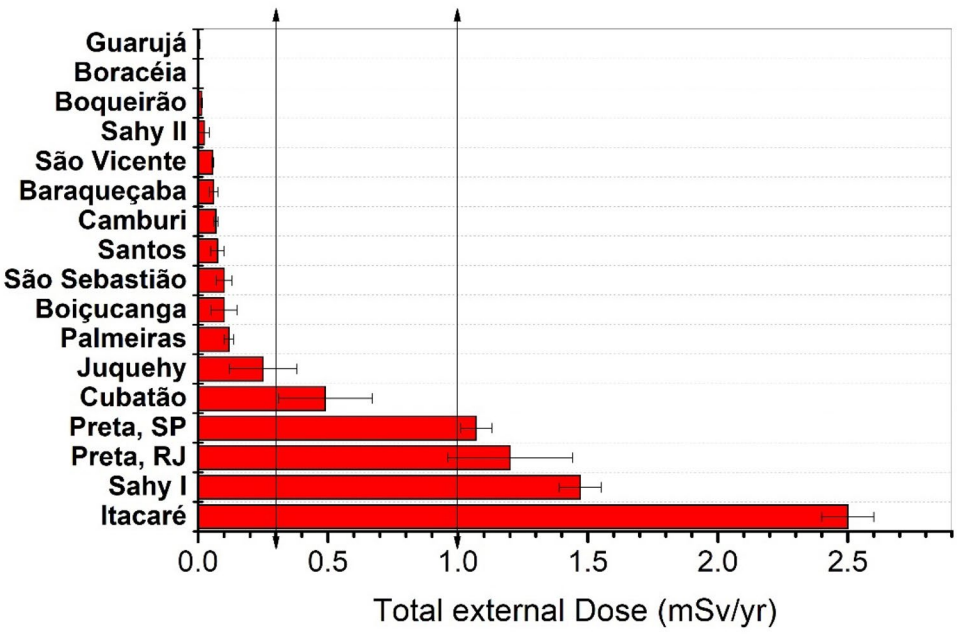

Fig. 2 Terrestrial external radiation from the natural radionuclide for the sandy regions in São Paulo, Bahia and Rio de Janeiro. The arrows indicate the typical range to annual effective dose by Reference [16]

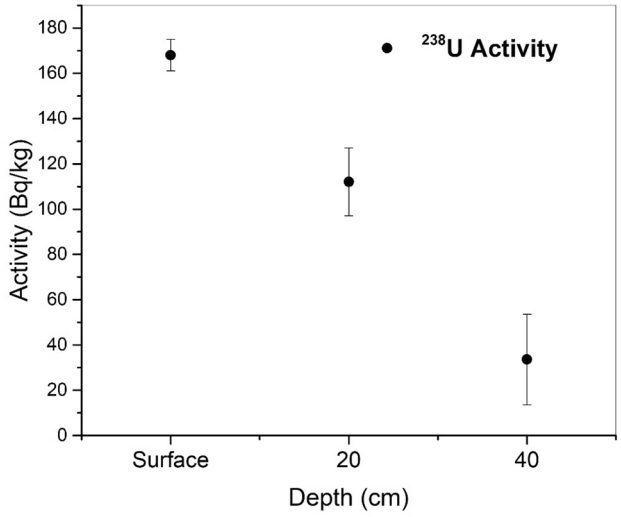

Fig. 3 ^{238}U activity, as a function of the sample depth collected in Preta Beach, Ilha Grande, RJ

the samples collected near the surface. In Fig. 3 ^{238}U activity at Preta Beach for three depths are shown. Thorium and potassium concentrations are about nearly constant in the samples collected at the same depths.

In Fig. 4 the external hazard indices, calculated for all sandy samples in this work are shown. The values from the sand samples from Itacaré, Ba, Sahy I, SP, Preta Beach, SP, and Preta Beach, RJ, are greater than unity, which is the external hazard limit.

In order to identify the mineral responsible for the high activity at Preta Beach, RJ, the sand samples from all depths were separated in three fractions: a highly magnetic fraction, composed by ferromagnetic grains attracted to a common magnet, a weakly magnetic fraction, composed by paramagnetic grains, attracted using a current of 0.4 A and a standard configuration in a Frantz Magnetic Separator [35], and a non-magnetic fraction. The current of the separator was chosen to be close to the lower limit of monazite separation range, while keeping the non-magnetic fraction free of opaque minerals. These fractions were analyzed with the gamma-ray spectrometry technique to verify the amount of radionuclides present in each of them. In Fig. 5 the percentage of magnetic concentration as a function of the sample depths collected at Preta Beach, RJ, is shown. It is possible to observe that magnetic particles are present mainly in the surface corresponding to about 95%, 75% at 20 cm depth and only 60% at 40 cm depth. This fact indicates that the high radioactivity is probably connected with the presence of the magnetic materials and this assumption is reinforced by the gamma-ray spectrometry qualitative analysis on the separated fractions, in accordance with the results presented in Fig. 5.

To understand which minerals are contributing to different amounts of radioactivity and magnetic particle concentration, we have studied eleven samples with a *Leo 440i* scanning electron microscope. Backscattered electron signals showed that in the highly magnetic fraction there are some differences of mean atomic number. Energy-Dispersive X-Ray (EDS) microanalysis using an Oxford Si(Li) detector revealed the chemical composition of the mineral types encountered. In the highly magnetic fraction of the sediments we confirmed the presence of inclusions composed of cerium and phosphorus indicating the presence of monazite, a mineral commonly associated with thorium and uranium,

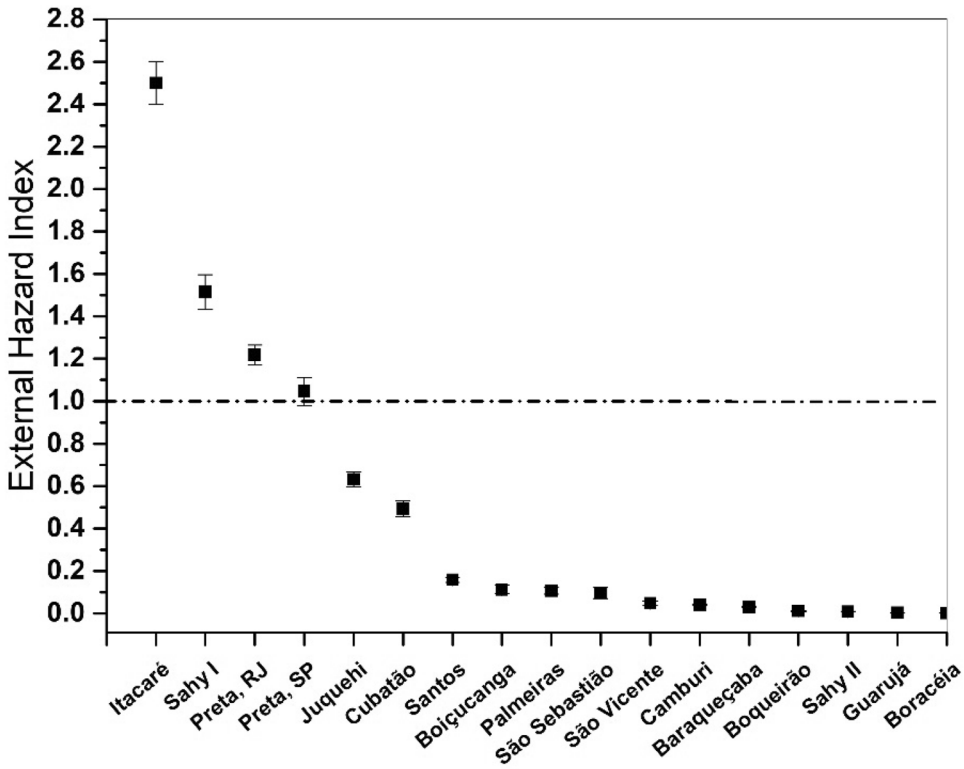

Fig. 4 External hazard indices calculated for all sandy regions. The dotted line represents the external hazard limit

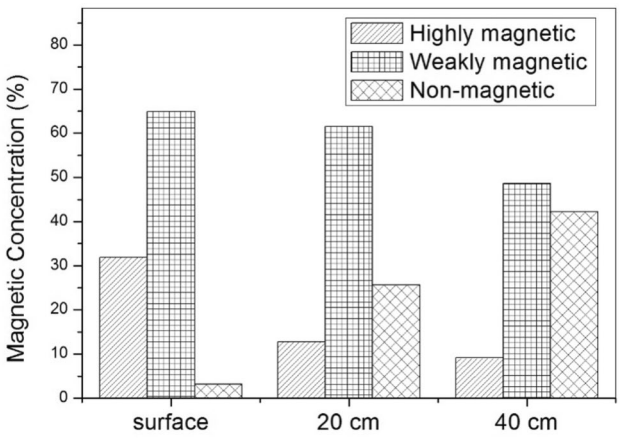

Fig. 5 Percentage of magnetic concentration as a function of the three sample depths collected at Preta Beach, Ilha Grande, Rio de Janeiro

as indicated by the X-ray spectrum, shown in Fig. 6 [36, 37]. With EDS microanalysis we have found a few grains composed of rare earth elements with a large amount of thorium. One of these grains is in evidence in the left side of Fig. 7. The X-ray spectrum shown in the right side of Fig. 7 confirms the presence of these elements.

The same analysis performed in the non-magnetic fraction suggested that it is composed only of quartz/feldspar, minerals with no U/Th radioactive content, and zircon, which commonly has a small content of uranium and thorium [36, 37]. In the right part of Fig. 8 it is possible to observe two X-ray spectra showing the presence of potassium, which indicates that it may be a feldspar (upper right) and showing the presence of zirconium which indicates that this is a zircon mineral (lower right). In the weakly magnetic fraction, no radioactive material was identified.

Summarizing, in the non-magnetic fraction of the sediments there is a low concentration of uranium and thorium present, probably in zircon. In the highly magnetic fraction a high concentration of thorium and rare earth elements was found. The presence of cerium and phosphorus in this highly magnetic fraction indicate the presence of monazite, a mineral commonly associated with thorium and uranium, although the monazite grains, as a non-magnetic mineral, are in this case found in association with ilmenite and magnetite, suggesting that the presence of highly magnetic fractions may play an important role in background radiation, as long as the conditions the minerals were formed allowed the growth of Th/U-rich inclusions. Some single monazite grains were also found, indicating also that the magnetic separation procedure was not able to completely remove contributions from other fractions. In the weakly magnetic fraction there were no indications of radioactive materials [37].

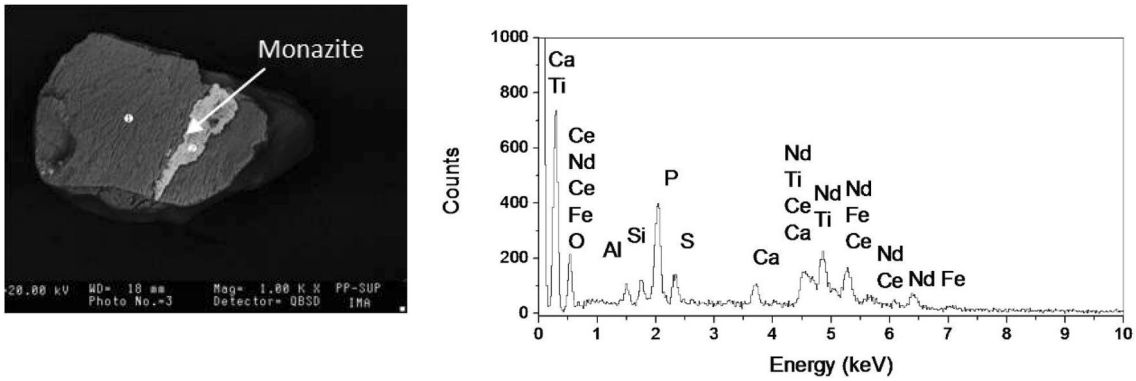

Fig. 6 Image of the highly magnetic fraction analyzed with a scanning electron microscope (left). The EDS spectrum, shown in the right part of the figure, indicates the presence of rare earth elements and phosphorus, suggesting the presence of monazite crust (right)

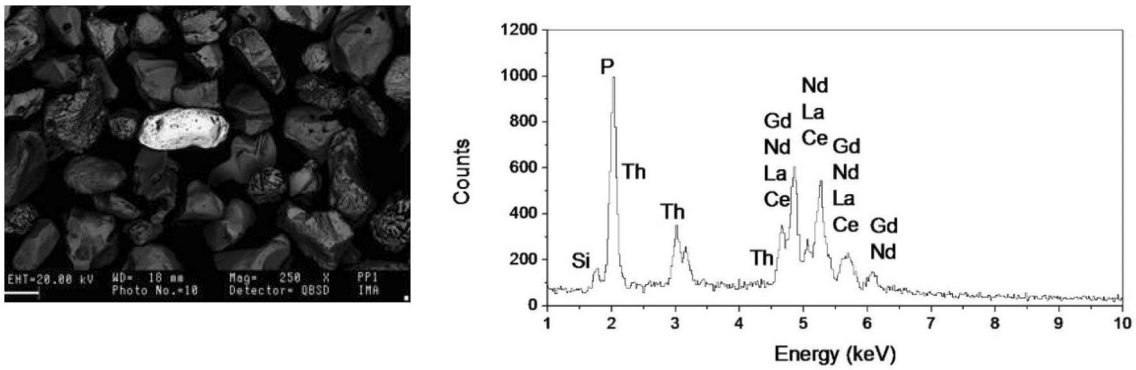

Fig. 7 The EDS microanalysis shows that the highly magnetic grain in evidence (left) is composed of rare earth elements and a large amount of thorium. The EDS spectrum, shown in the right part of the Figure, indicates the presence of rare earth elements and phosphorus, suggesting the presence of monazite crust (right)

Conclusion

In this work, the sand of fourteen beaches in Brazil was investigated using gamma-ray spectrometry. The main contribution of the radiation dose is due to the elements of the series of ^{232}Th, with a smaller contribution from the isotope ^{40}K and the elements of the series of ^{238}U. Eleven beaches present a total external radiation per year lower than 0.6 mSv/year, which is the typical upper value indicated by the United Nations Scientific Committee on the Effects of Atomic Radiation. In all beach samples we found a low concentration of ^{40}K. However, in Itacaré, Ba; in Preta Beach on Ilha Grande, RJ; Preta Beach and Sahy I in SP, the results show high quantities of the elements from the series decay of ^{238}U and ^{232}Th, exceeding more than twice the international average value for external exposure to the natural elements: 2.5(5) mSv/year for Itacaré, Ba, 1.47(8) mSv/year for Sahy I, SP, 1.20(24) mSv/year Preta Beach, RJ and 1.06(7) mSv/year Preta Beach, SP.

As expected, the values of the external hazard index determined from the sand radioactivity are lower than the external hazard limit for most of the studied sandy regions. Itacaré in Ba, Sahy I Beach, Preta Beach in RJ and Preta Beach in SP presented values above this limit.

With the magnetic separation procedure, it was possible to correlate the magnetic concentration with the high activity found in the beach surface. Energy-Dispersive X-Ray Spectroscopy (EDS) microanalysis revealed the probable chemical composition of the mineral types encountered in Preta Beach, RJ. The presence of cerium and phosphorus in the highly magnetic fraction of the sediments indicate the presence of monazite, a mineral commonly associated with thorium and uranium. High concentrations of heavy minerals were also found in these sediments. The non-magnetic fraction analysis suggested that it is composed only of quartz and feldspar, which are minerals with no radioactive content, and zircon, which commonly have a small content of uranium and thorium. More analysis must be performed in order to determine if there is a contribution of thorium

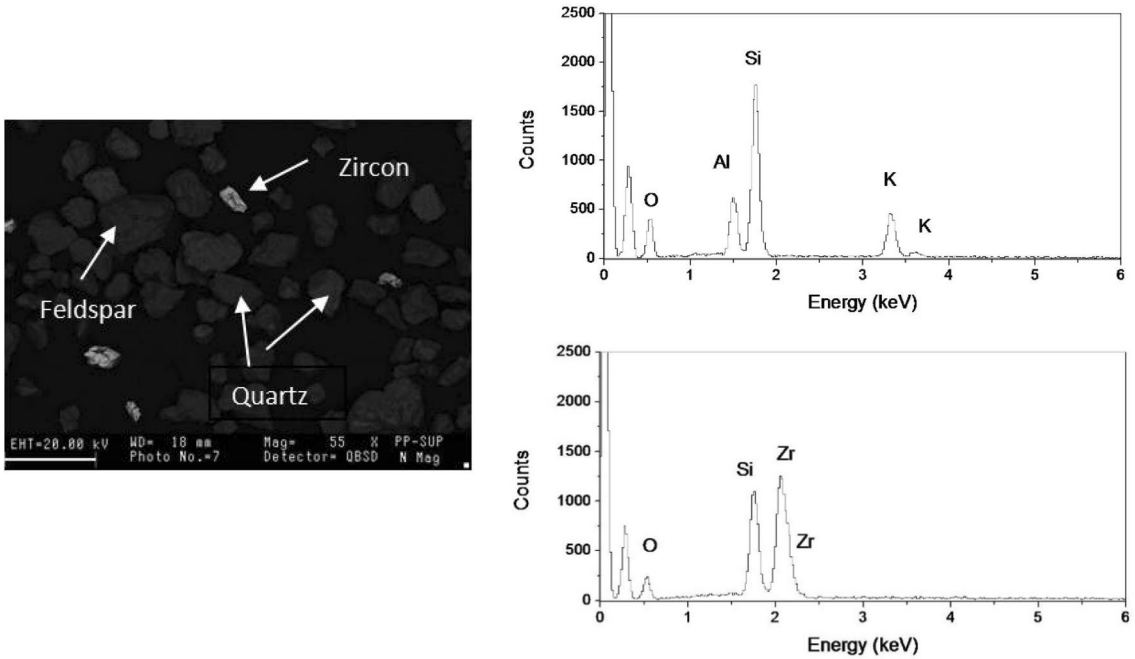

Fig. 8 Image of the non-magnetic fraction (left), containing feldspar (upper right spectrum), quartz and zircon (lower right spectrum)

impurities in magnetite and ilmenite that adds to the total external radiation. The results obtained in this work may be used to collaborate in the development of reference levels of natural radioactivity along the coast of Brazil.

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