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ORIGINAL PAPER

Assessment of radiation hazard indices due to naturally occurring long-life radionuclides in the coastal area of Barra de Valizas, Uruguay

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Abstract The Uruguayan east coast has several mineral resources, which include black sand ores in the Barra de Valizas–Aguas Dulces area. Cancer in Uruguay shows non-homogeneous geographical distribution, with the highest standardized mortality ratio (SMR) in the northeast and east region, which includes the aforementioned area and the town of Barra de Valizas. The activity concentration of natural radionuclides (^{226}Ra, ^{232}Th and ^{40} K) in Barra de Valizas´soil was determined by gamma spectrometry in order to evaluate the radiological hazard for inhabitants and tourists. The outdoor annual effective dose (AEDE), excess lifetime cancer risk (ELCR), and annual gonadal dose equivalent (AGDE) were evaluated for inhabitants with a life expectancy of 77.7 years, a 0.2 and 0.5 occupancy factor, and using the conversion coefficients recommended by United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). The annual effective dose was also evaluated for both summer and fortnight tourists. The radiological hazard indices for Barra de Valizas inhabitants are higher than the worldwide mean and recommended values. This may contribute to Rocha's higher SRM value, although a direct correlation cannot be assured with the epidemiological information currently available. Social, medical and anthropological studies will be carried out in future to provide data and verify this correlation.

Keywords Environmental radioactivity · Radiological risk · Gamma spectrometry · Uruguay

Introduction

It is widely recognized that human beings are exposed to radioactivity from a variety of background sources. Radionuclides belonging to ^{238}U and ^{232}Th series and ^{40} K are the majority of naturally occurring radionuclides present in the Earth's crust and, together with the interaction of cosmic rays in the earth's atmosphere, are responsible for the 85% of the annual exposure dose received by the world population (United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 2008). The effective dose due to natural sources averages 2.4 mSv per year, ranging from 1 to 13 mSv, with populations whose values exceed 13 mSv. Those areas where exposure to natural sources exceeds values that can be considered normal background radiation are called high background radiation areas (HBRAs) (United Nations Scientific Committee on the Effects of Atomic Radiation

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(UNSCEAR), 2008). The presence of these radionuclides and their radiological impact has been reported in many places worldwide. For instance, there are reports from the Mediterranean coast (Abbasi & Mirekhtiary, 2020), Egypt (Aziz et al., 2020; Tawfic et al., 2021), Cyprus (Abbasi & Mirekhtiary, 2020), Malaysia (Khandaker et al., 2018), Iran (Ghiassinejad et al., 2002), Poland (Fornalski & Dobrzyński, 2012), China (Morishima Et Al., 2000; Omori et al., 2015; Zhou et al., 2020), and India (Monica et al., 2016; Srinivasa et al., 2019). Related to this study, reports of similar deposits in Brazil are of special relevance because of the country's geographical proximity to Uruguay. In Brazil, naturally occurring long-life radionuclides have been found in Guarapari (Veiga et al., 2006), Espíritu Santo (Dutra Garcêz et al., 2020), and Río de Janeiro (Freitas & Alencar, 2004; Ribeiro et al., 2017) (approximately 1700 km from Barra de Valizas, Uruguay).

The incidence of radiation on cancer has been widely studied and reported since the mid-nineteenth century. When ionizing radiation passes through cells, it ionizes or excites the atoms and may change the DNA, inducing injuries. Depending on the type and energy of the incident radiation, it can interact with human DNA either directly or indirectly. Direct incident radiation may disrupt the molecular structure of DNA due to single-strand breaks (SSB), double-strand breaks (DSB), and base lesions, and there may be clustered DNA damage (Elgazzar, 2015; Mohan & Chopra, 2022). Indirect interactions take place when the radiation interacts with non-critical target atoms or molecules present in the cell, generally the water molecules, and hydrolysis of water molecules occurs, producing ions as well as free radicals (which are highly reactive). The biological effects of radiation depend on the type and energy of radiation, dose and exposure rate, and radiosensitivity of the cells. The classification of effects could be based on the nature of the effect, somatic and hereditary effects, and based on the timing of the effect, which are categorized as deterministic and stochastic effects. Deterministic effects are result of intense radiation exposure and their intensity is very much dependent on the radiation dose, whereas stochastic effects are probabilistics (Elgazzar, 2015; Mohan & Chopra, 2022). These last effects are associated with lower doses of radiation and do not have a threshold; the main stochastic effect is cancer (Magill & Galy, 2005).

Likewise, alpha and gamma emitters are classified by the International Agency for Research on Cancer as carcinogenic to humans (Group 1); meaning that there is sufficient evidence of carcinogenicity in humans (International Agency for Research on Cancer (IARC) 2021). The correlation between the presence of radionuclides in the environment and cancer has been and continues to be the subject of extensive radiological and epidemiological studies worldwide for both anthropogenic (Abylkassimova et al., 2000; Auvinen et al., 1994; Balonov et al., 2007; Bauer et al., 2006; Eidemüller et al., 2008, 2010; Grant et al., 2017; Haylock et al., 2018; Hjalmars et al., 1994; Kossenko et al., 2005; Leuraud et al., 2017; Qu et al., 2018) and natural radionuclides (Aliyu & Ramli, 2015; Boice et al., 2007; Chaudhury et al., 2023; David et al., 2021; Dobrzyński et al., 2015; Hendry et al., 2009; Hosoda et al., 2021; Mosavi-Jarrahi et al., 2005; Noguchi et al., 1986; Zlobina et al., 2022).

Some of the most representative studies about radiological hazards in HRBAs have been carried out in Kerala, India, where studies have reported a high incidence of genomic pathologies and an increased incidence of birth defects (Jaikrishan et al., 2013; Kochupillai et al., 1976). However, some studies in the same area have concluded that other factors may contribute to these effects, and results from different studies in Kerala and other high radiation areas have been contradictory (Aliyu & Ramli, 2015; Goldberg & Lehnert, 2002; Hendry et al., 2009).

Veiga and Koifman (2005) conducted epidemiological studies of Poços de Caldas, Araxa, and Guarapari, Brazil, using ten years of mortality data (1991–2000) related to cancer and other causes. The study showed that expected cancer mortality was higher in Poços de Caldas and Guarapari compared to the control area, but lower in Araxa (Veiga & Koifman, 2005). However, it is important to note that the study was based on preliminary investigations, and not all deaths related to cancer may be associated with radiation exposure. Additionally, the higher cancer incidence in Guarapari despite lower radiation levels suggests that other factors such as socioeconomic factors, diet, smoking, and exposure to pesticides and agricultural activities may have influenced the results. In Araxa, cancer mortality was lower than expected, but deficiencies in data may have led to confounding and biased results (Aliyu & Ramli, 2015). Meanwhile, Barcinski et al. (1975) conducted a cytogenetic survey of Guarapari residents to investigate possible biological effects of chronic exposure to natural radiation. They observed an increase in chromosomal aberrations in residents of the HBRAs compared to the nearby normal background radiation area, which they attributed to elevated radiation caused by the presence of monazite in coastal beach sands (Barcinski et al., 1975).

According to the Zlobina et al., (2022) study in Belokurikha and Kolyvan in Russia, Zhuhai in China and Echassieres in France, the mortality was clearly higher in HBRAs for non-transmissible diseases and cancers. This work also found a positive and linear correlation of radiation exposures with morbidity rates with a reasonable statistically significance level (Zlobina et al., 2022).

It is important to note that these studies have limitations—mostly regarding epidemiology data and amount of population—and that more research is needed to fully understand the relationship between radiation exposure and cancer risk in high natural radiation regions. In the case of Uruguay, previous studies along the strip coast of the 290 RAMSAR site indicate that the Barra de Valizas—Aguas Dulces region has the highest activity concentrations of ^{238}U (determined via ^{226}Ra, radionuclide belonging to its progeny and theoretically in secular equilibrium) and ^{232}Th in the department of Rocha (Noguera et al., 2018). However, no systematic study of radiological hazards due to terrestrial radionuclides in soils around homes has been carried out, in either the town of Barra de Valizas or Aguas Dulces. In addition, the incidence on cancer of those natural radionuclides has not yet been study. In light of this, this work is a first attempt to search correlations between that area and the cancer incidence and mortality ratios registers for such area.

The study area

Geographic and geologic setting

Uruguay is a country in the southeastern region of South America, with an area of 176,215 km^2 and a population of 3.3 million inhabitants, according to the last performed population census in 2011 (Instituto Nacional de Estadística, 2011). It is divided into 19 administrative regions, called departments. Though small in area, Uruguay has a rich in biodiversity, especially in areas with low human interaction. The 290 Ramsar site (coordinates − 33.8, − 53.8, area 407,408.0 ha) is located in the eastern region of Uruguay, spanning over three departments (Rocha, Treinta y Tres and Cerro Largo), and extending from the Atlantic Ocean to the country's border with Brazil. This site includes wetlands, brackish and freshwater lagoons and rivers. In the coastal area, it includes large extensions of sand (some as shifting dunes, others fixed by vegetation), beaches with big waves and strong winds, and rocky outcroppings (Altamirano, 2002). The site was included in the List of Wetlands of International Importance due to the high incidence of species of flora and fauna, many of which were endemic or classified as vulnerable, who found favorable conditions for their habitats (Ramsar Sites Information Service RAMSAR, 2023). This area's rich biodiversity makes it and its surroundings very attractive for tourists from around the world. Each year, Rocha is one of the departments most popularly visited by tourists during the summer months. It has several beach villages such as Barra de Valizas, a typical coastal sand village located on the coast of the Atlantic Ocean. This town has a permanent population of about 400 inhabitants according to the last performed population census in 2011 (Instituto Nacional de Estadística, 2011) and increases exponentially during the summer months due to a high influx of tourists.

In addition to the stunning landscapes and rich biodiversity, the Valizas site is interesting from a geological standpoint. The sandy deposit on which Valizas is located consists of a strip 1.5 to 3 km wide, between the Atlantic Ocean and a rocky paleo-coastline. The sandy deposit rests unconformably on a relatively compact clayey substrate (named Barra del Chuy formation), which is 5 to 10 m under the current coast line (Ferrando et al., 2003). Morphologically, the deposit has an homogeneous nature, without significant geological variations, and is a mixing of sand and heavy minerals (mixing: black sands). The deposit was accumulated by marine intrusions whose waves generated an ancient ravine coast. The lithologies of the Barra del Chuy Formation were affected, generating unevenness of up to 20 m. The accumulation of the aforementioned sediments took place at a former relative maximum of sea level.

This is demonstrated by the fact that all profiles show sequences of decreasing black sand grades toward the base of each cycle, and since the most important tenors tend to register in the highest parts of each profile (Ferrando et al., 2003). The deposition process of these sands turns out to be equivalent to that proposed by Almagro, (2001) for the Barra del Chuy Formation in the area between the towns of Chuy and La Coronilla. Additionally, the geometry of the deposit responds perfectly to the model proposed by Willwock and Tomazelli, (1998) for the evolution of the Atlantic coast of Rio Grande do Sul (Brazil) during the Quaternary. The age of this accumulation has yet to be established with certainty due to the data about fossils are scarce. It seems to be Pleistocene because it reached the level + 10 with respect to the current level in the area of the deposit, since, according to data from Martínez et al., (2001), in Argentina this ingression occurred 130,000 years ago. Therefore, the genesis of the deposit is considered to have been determined by the littoral deposits of the Atlantic Ocean, at an indeterminate moment between 130,000 and 5000 years ago (Ferrando et al., 2003). The different pulses of ingression and regression of the sea level caused the concentration of dense minerals in the coastal deposits. In turn, the action of the wind on the surface of the land caused the elimination of part of the light fraction, increasing the concentration of heavy minerals. The heavy minerals were evaluated in 7 million tons in the Aguas Dulces-Barra de Valizas area and have been determined to have a mean composition of 50% of ilmenite, 20% of magnetite, 10% of epidote, 9% of garnet, 5% of zircon, 4% of amphibole, 1% of rutile and 1% of monazite (Bossi & Navarro, 2000; Dirección Nacional de Minería y Geología (DINAMIGE) 2002; Ferrando et al., 2003).

For a deposit of the characteristics considered to be produced, the existence of basic igneous rocks on the continent is necessary, since ilmenite can be found in them in significant concentrations (Ferrando et al., 2003). The best conditions for this to happen are generated when the area is subject to climatic alternation with periods that favor intense weathering (warmth and humidity) and periods where erosion predominates. In warm periods, there is intense decomposition of silicates and ilmenite is relatively more resistant. In the arid periods, the vegetation is scarce, the rains are torrential and concentrated, and this generates active erosion and intense dragging toward the watercourses and through them to the ocean. This alteration of climatic conditions has occurred on a very basis in the geological evolution of Uruguay and especially during the Tertiary and Quaternary (last 65 Ma), associated with glaciations and interglacials (Ferrando et al., 2003).

Environmental health considerations

As previously mentioned, the heavy minerals of the deposit contain monazite, which in turn contains Thorium and Uranium evaluated as 4,75% ThO_2 and 0,18% UO_2 (Bossi and Navarro, 2000; Dirección Nacional de Minería y Geología (DINAMIGE) 2002). The existence of Uranium and Thorium, that means, naturally occurring ^{238}U and ^{232}Th radionuclides, and all the radionuclides of their radioactive series, make the sand of the area a source of radioactivity. As both radioactive series have alpha, beta and gamma emitters, all of them theoretically contribute to the radioactive dose. However, when the external dose is considered, gamma radiation is the most dangerous (International Agency for Research on Cancer (IARC) 2000) and will thereby be the radiation factor taken into account in the rest of this work. Therefore, the purpose of estimating radiation hazard indices due to the naturally occurring long-life radionuclides in the coastal area of Barra de Valizas, this work will consider only the contribution of gamma emitters of the deposit, as with common indices used for this purpose in the scientific community (Caridi et al., 2022; Filgueiras et al., 2020; Hannan et al., 2015; Hilal & Borai, 2018; Jallad, 2016; Miller & Voutchkov, 2016; Taskin et al., 2009; Tawfic et al., 2021).

In Uruguay, as in the rest of the world, cancer is one of the leading causes of death, accounting for a quarter of all deaths recorded in the country. Each year 15,000 new cases are reported, corresponding to a standardized rate (excluding non-melanoma skin cancer) of 298 cases per 100,000 people. While Uruguay exhibits comparable values of incidence rates with more developed countries, it displays notably higher mortality rates. Nationally, lung cancer is considered to be the leading cause of death in men and the third highest in women (Barrios & Garau, 2017). These authors report that lung cancer in Uruguay shows non-homogeneous geographical distribution, with the highest standardized mortality ratio (SMR) in the northeast and east which includes the departments of Rocha and Treinta y Tres. The reasons behind these high rates have been scarcely studied, and not yet identified. While behavioral factors such as high tobacco consumption may contribute statistically, environmental factors cannot be ruled out without prior study. It is well known that smoking is the leading cause of cancer and of death from cancer, but a wide variety of other chemical and physical agents such as metals, arsenic, and ionizing radiation are potential factors in the high incidence of cancer. Figure 1 exhibits the standardized incidence ratio (SIR) (Barrios et al., 2020) and the standardized mortality ratio (SMR) (Barrios et al., 2020) reported by the "Comisión de Lucha Honoraria contra el Cáncer" (Honorary Commission to Fight Cancer) for general cancer, whereas Fig. 2 shows the SIR for lung cancer, for each Uruguayan department (Barrios et al., 2020). In both figures, the location of the town Barra de Valizas is shown. The department of Rocha shows the highest Uruguayan SIR and SRM ratios. It is noteworthy that while the Uruguayan health system registers cancer patients according to the health center where they receive care, and the department to which they belong, it does not record the town where they reside.

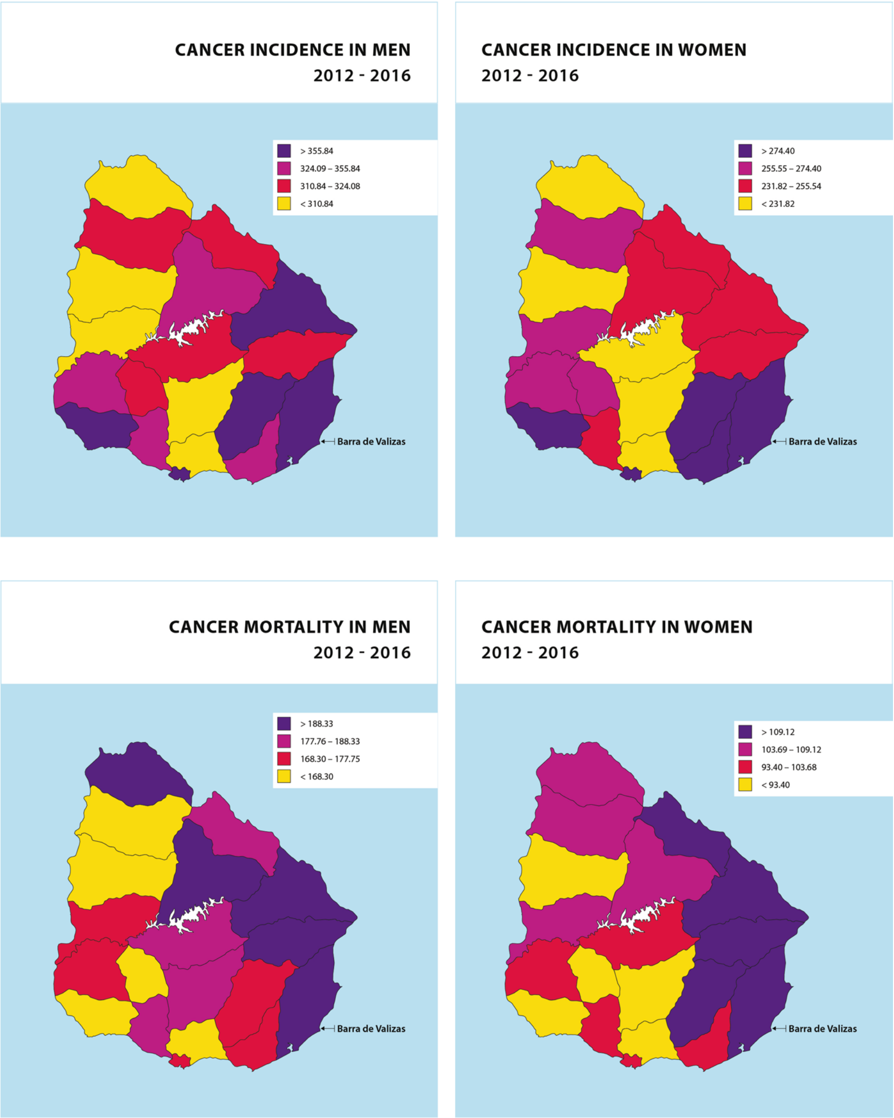

Fig. 1 Standardized mortality ratio (SMR) and standardized incidence ratio (SIR) reported by the "Comisión de Lucha Honoraria contra el Cáncer" (Honorary Commission to fight Cancer), Uruguay, and Barra de Valizas location. Adapted from Barrios et al., (2020) and Barrios et al., (2022)

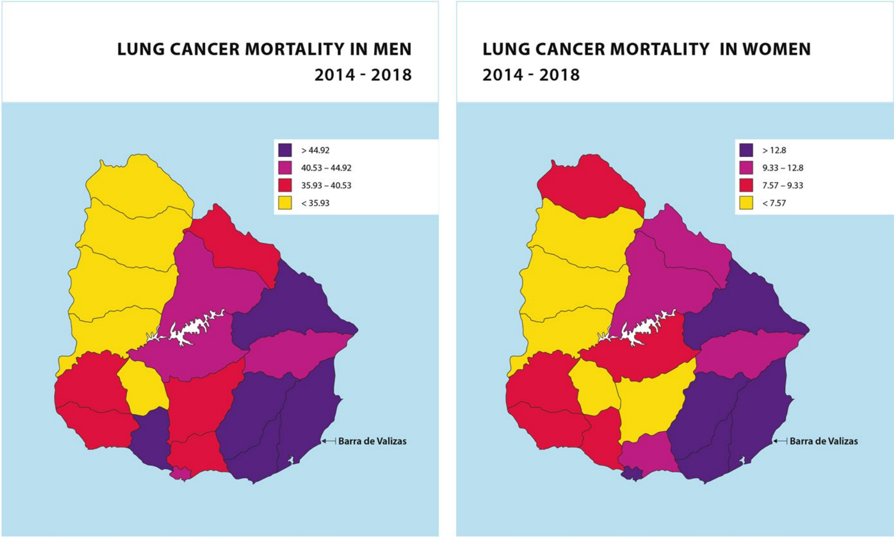

Fig. 2 Lung standardized mortality ratio (SMR) reported by the "Comisión de Lucha Honoraria contra el Cáncer" (Honorary Commission to fight Cancer), Uruguay, and Barra de Valizas location. Adapted from Barrios et al., (2020) and Barrios et al., (2022)

Considering the background detailed in the preceding paragraphs, the aim of this work was to evaluate the population's radiological risk due to natural long-life radionuclides in town of Barra de Valizas, and overall, to determine if the presence of natural long-life radionuclides in the area might be one of the causes of the highest standardized mortality ratio (SMR) in the department of Rocha.

Materials and methods

Sample collection and preparation

According to reports in the literature, the sampling was done randomly (Hannan et al., 2015; Jallad, 2016; Khandaker et al., 2018; Margineanu et al., 2014), at a rate ratio of 25 samples per square kilometer, that is higher than the one used in bibliography. Figure 3 shows the geographical distribution of the sampling zone (−34.336111, 53.794444), the specific geographic location for each sample is shown in supplementary information. Ten subsamples of sand and soil samples were gathered using hand auger, 9.8 cm^2 cross section area, at a typical depth of 20 cm from the top surface layer. After removing stones and other materials, each sample was dried at a temperature of 60 °C until a constant weight was reached (two weeks) and passed through a standard 2-mm mesh size. The homogenized samples were milled and hermetically sealed into 500-mL Marinelli flasks and stored for four to six weeks; four weeks are needed in order to achieve radioactive secular equilibrium.

Naturally occurring radionuclides determination

The activity concentrations of ^{232}Th, ^{226}Ra and ^{40} K were determined by measuring some of their decay products by gamma spectrometry, using a high-resolution gamma-ray spectrometry system with a low background High Pure Germanium Detector AMETEK-ORTEC GMX35P4-76-RB associated with an ORTEC Dspec jr 2.0 multichannel analyzer, and with a shield specifically designed for environmental measurements, 10 cm aged lead, with inner liner of copper and tin layers for the suppression of lead X-rays. The relative efficiency of the detector and the effective energy resolution for the 1.33 MeV photopeak of ^{60}Co are 35% and 1.75 keV, respectively. International Atomic Energy Agency (IAEA) reference materials RGU-1, RGK-1 and RGTh-1 in the same geometry of the samples were used for efficiency calibration. Each sample, reference material and the background were measured for 150,000 s, with a dead time lower than 1%. ^{226}Ra was studied by the photopeak of ^{214}Bi (609.3 keV, Emission probability ($I\gamma$) 45.49%), ^{232}Th was evaluated by the photopeak of ^{228}Ac (911.1 keV, Emission probability ($I\gamma$) 25.80%), and ^{40} K was evaluated by its own photopeak of 1460.8 keV, Emission probability ($I\gamma$) 10.66%. The activity concentration (A) for each radionuclide was calculated through

Fig. 3 Study zone of Barra de Valizas and geographical distribution of sample points

Eq. (1) where N is the count rate onto the selected photopeak of the sample, B is the background count rate, t is the counting time (s), $P\gamma$ is the emission probability, ε is the detector efficiency, and m is the mass (Kg) of the sample.

$$A = \frac{N - B}{t\,x\,P_\gamma x\,\varepsilon\,x\,m} \tag{1}$$

The activity concentration uncertainty (σ) was calculated as follows Eq. (2), where σN is the sample counting uncertainty, σB is the background counting uncertainty, $\sigma\varepsilon$ is the efficiency uncertainty, σm is the mass measurement uncertainty, and $\sigma P\gamma$ is the gamma line energy uncertainty (Abbasi & Mirekhtiary, 2020)

$$\sigma = A\sqrt{\left(\frac{\sigma N}{N}\right)^2 + \left(\frac{\sigma B}{B}\right)^2 + \left(\frac{\sigma\varepsilon}{\varepsilon}\right)^2 + \left(\frac{\sigma m}{\mathrm{Nm}}\right)^2 + \left(\frac{\sigma P\gamma}{P\gamma}\right)^2} \tag{2}$$

The minimum detectable activity for each sample was calculated using the following relation Eq. (3), where F is the Compton background area of the selected gamma-ray spectrum region with a 96% confidence.

$$\mathrm{MDA} = \frac{4.66\sqrt{\mathrm{FC}}}{t\,x\,P_\gamma x\,\varepsilon\,x\,m} \tag{3}$$

The mean minimum detectable activity (MDA) was determined as 0.24 Bq kg^{-1} for ^{226}Ra, 0.21 Bq kg^{-1} for ^{232}Th and 1.0 Bq kg^{-1} for ^{40} K (Currie, 1968; Turner et al., 2012). All spectra were analyzed with ORTEC Gamma Vision software version 6.09 for Windows.

Radiological hazard risk evaluation

Annual effective dose equivalent (AEDE)

The annual effective dose equivalent (AEDE) Eq. (4) was determined according to the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) recommendation (UNSCEAR, 2008), taking into account the absorbed dose rate (D) in air at 1 m above the ground's surface Eq. (5), the conversion factor (0.7 Sv.Gy^{-1}) and the outdoor occupancy factor for adults (f).

$$\mathrm{AEDE}\left(\mu\mathrm{Sv}.y^{-1}\right) = D\left(\mathrm{nGy}.h^{-1}\right).f.0.7\left(\mathrm{Sv}.\mathrm{Gy}^{-1}\right)x10^{-3} \tag{4}$$

$$D\left(\mathrm{nGy}.h^{-1}\right) = \left(0.462A_{\mathrm{Ra}} + 0.604A_{\mathrm{Th}} + 0.0417A_{K}\right) \tag{5}$$

In Eq. (5) the terms A_{Ra}, A_{Th} and A_K are the activity concentrations of each radionuclide in Bq kg^{-1}, and the dose coefficients, in nGyh^{-1} per Bq kg^{-1}, are those recommended by UNSCEAR as well.

The standard occupancy factor of 1752 h yr^{-1}—which implies 20% of the year time— was used for comparison with international reports. Most of Valizas´s inhabitants have outdoor professional activities, so they have an outdoor occupancy factor higher than the mean world population. Taking into account the actual behavior, the AEDE for an occupancy factor of 4380 h yr^{-1}—which implies 50% of the year's time—was also evaluated. Many houses in Barra de Valizas are built directly on beach dunes with people living directly on the black sand deposit where the AEDE occupancy factor is even higher than that previously mentioned.

Finally, it is important to point out that Uruguay´s Atlantic coast is a popular tourist destination, one whose visitors can be classified into two types of tourists: those who vacation during the entire summer season, and those who visit during the first fortnight of the summer. The AEDE for both types of tourist is also reported here, with an occupancy factor of 0.336 h yr^{-1} and 0.084 h yr^{-1}, respectively.

Annual gonadal dose equivalent (AGDE)

The gonads are particularly sensitive to ionizing radiation; hence, they are considered by the International Commission on Radiological Protection (ICRP) as organs of interest. The factor by which equivalent dose in a tissue or organ is weighted to represent the relative contributions of that issue or organ to the total detriment resulting from uniform irradiation is the tissue weighting factor. ICRP assigns to gonads the highest tissue weighting factor (0.20) (Eckerman et al., 2018) making necessary to determine the equivalent doses received by these organs; in order to evaluate the incidence of long-life naturally occurring radionuclides in the gonadal dose, the annual gonadal dose equivalent (AGDE) index was evaluated using Eq. (6), where the terms A_{Ra}, A_{Th} and A_K are the activity concentrations of each radionuclide in Bq kg^{-1}, and the dose coefficients, in nGyh^{-1} per Bq kg^{-1}, are those recommended by UNSCEAR for this case.

$$\mathrm{AGDE}\left(\mu\mathrm{Sv}.y^{-1}\right) = \left(3.09A_{\mathrm{Ra}} + 4.18A_{\mathrm{Th}} + 0.314A_{K}\right) \tag{6}$$

Excess lifetime cancer risk (ELCR)

The excess lifetime cancer risk (ELCR), which is a plausible upper bound estimate of the probability that a person may develop cancer sometime in his lifetime following exposure to a specific contaminant, was evaluated. In this work, the contaminants evaluated were the natural radionuclides ^{226}Ra, ^{232}Th and ^{40} K, and for these the whole body ELCR was calculated using Eq. (7):

$$\mathrm{ELCR} = \mathrm{AEDE}\left(\mu\mathrm{Sv}\cdot y^{-1}\right)\cdot \mathrm{DL}(y)\cdot \mathrm{RF}\left(\mu\mathrm{Sv}^{-1}\right) \tag{7}$$

where DL corresponds to the mean duration of life (77.7 years in Uruguay), and RF corresponds to the risk factor of contracting a fatal cancer per Sievert (Sv^{-1}) received, 0.05 for the public according to ICRP 103 (Tsapaki et al., 2007). The occupancy factors of 1752 h yr^{-} and 4380 h yr^{-1}, which implies 20% and 50% of the year time, were used for AEDE determination.

Results and discussion

Table 1 summarizes ^{226}Ra, ^{232}Th and ^{40} K activity concentrations measured in this study and used to calculate the radiological hazard indices. Detailed information can be found in supplementary material. As can be appreciated in Fig. 4, radionuclide's activity concentration does not fit normal distribution, so the median is the best parameter of the central data tendency; however, the mean values are also presented to allow comparison with worldwide values. The quantile–quantile plot is shown in supplementary material. This is also true for radiological hazard indices. The Bootstrap percentile confidence intervals (0.05 level of significance) were calculated using package 'boot' version 1.3–28.1 for R 3.6.1 software (R Core Team, 2018).

The skewness is a measure of symmetry and if it is zero it means that the tails on both sides of the

Table 1 ^{226}Ra, ^{232}Th and ^{40} K mean, median, and range of activity concentration in Barra de Valizas compared to worldwide mean values (reported by United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR, 2008)

	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40} K (Bq kg^{-1})
Mean	38.5	79.4	235.8
Median	16.3	50.0	191.3
Min	3.22 ± 0.25	6.80 ± 0.53	81.4 ± 2.5
Max	512.3 ± 6.6	799.0 ± 9.2	937 ± 67
N	25	25	25
Percentile confidence Interval	15.0–80.7	41.5–143.9	169–316
Kurtosis	22.14	21.52	8.34
Skewness	4.55	4.44	2.26
Worldwide mean UNSCEAR, (2008)	35	30	400

mean balance out overall; a positive value indicates these data are skewed wright, while a negative value indicates a leftward skew. The kurtosis parameter is a measure of peakedness, if the kurtosis value is positive, the curve is more peaked than the normal curve (leptokurtosis) (Abbasi et al., 2020). Activity concentration value for ^{232}Th, ^{226}Ra and ^{40} K does not fit a normal distribution according to Shapiro–Wilk—p values for α 0.05 were 2.0×10^{-09}, 6.8×10^{-10} and 1.6×10^{-05}, respectively—so there were analyzed using Spearman´s Product Correlation coefficients between two elements.

Regression analysis between individual radionuclides is shown in Fig. 5. A significant correlation was observed between ^{232}Th and ^{226}Ra ($r = 0.92$, $p = 1.6 \times 10^{-6}$), a negative weak correlation was observed between ^{40} K and ^{232}Th ($r = -0.18, p = 0.4$), and no correlation was found between ^{40} K and ^{226}Ra ($r = -0.022$, $p = 0.92$). The Spearman's correlation coefficients matrix of ^{226}Ra, ^{232}Th and ^{40} K are presented in Table 2.

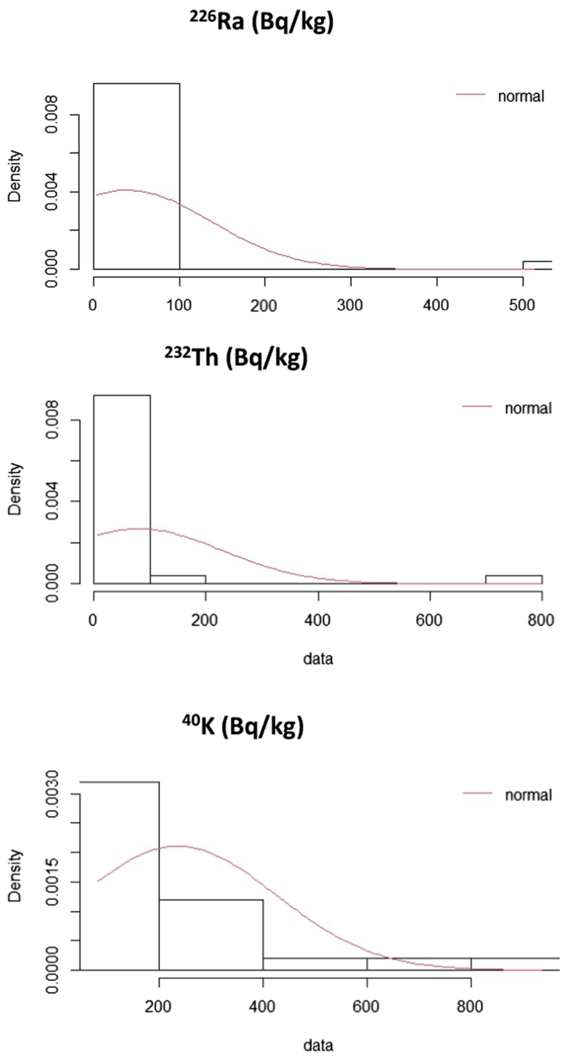

Fig. 4 Frequency distribution of ^{226}Ra, ^{232}Th and ^{40} K in the 25 samples from Barra de Valizas sands and soils, and theoretical normal density function

Table 1 indicates that Barra de Valizas shows higher ^{232}Th activity concentration than the worldwide mean (30 Bq kg^{-1} reported by United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR, 2008), whereas ^{226}Ra activity concentration is similar to the worldwide mean (35 Bq kg^{-1} reported by United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR, 2008), and ^{40} K activity concentration is below the worldwide mean value (400 Bq kg^{-1} reported by United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR, 2008). These results confirm previously reported data from Rocha´s coast (Noguera et al., 2018) and agree with the composition reported for the black sand ore in the area, considering secular equilibrium between ^{238}U and ^{226}Ra. As it was mentioned before, the black sand ore includes ilmenite, magnetite, zircon, rutile, and monazite, this last containing mean concentrations of

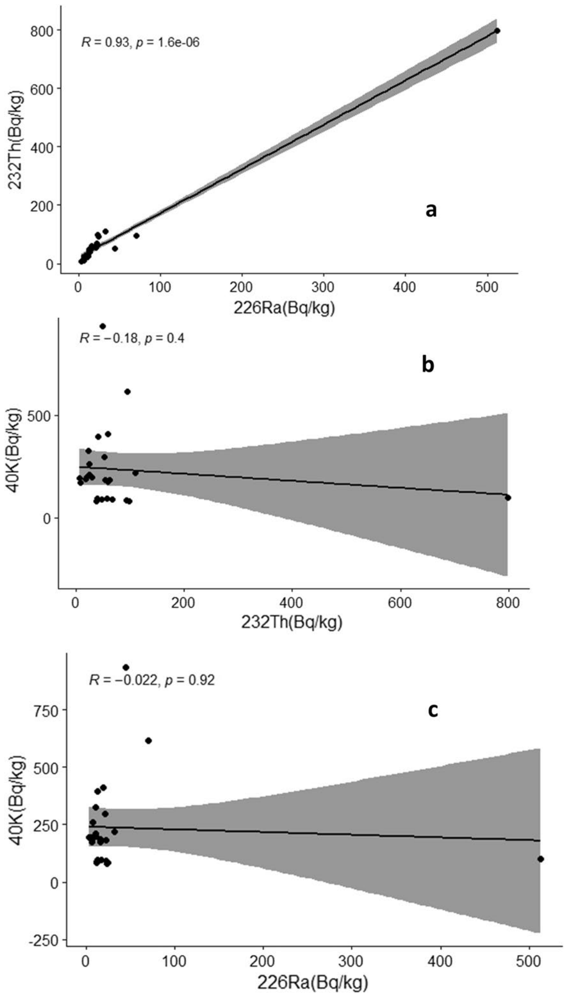

Fig. 5 Regression analysis between radionuclides activity concentration **a** Correlation between ^{232}Th and ^{226}Ra, **b** correlation between ^{40} K and ^{226}Ra and **c** correlation between ^{40} K and ^{232}Th

Table 2 The Spearman's correlation coefficients matrix of ^{226}Ra, ^{232}Th and ^{40} K

Parameters	^{226}Ra	^{232}Th	^{40} K
^{226}Ra	1.000	0.928	−0.022
^{232}Th	0.928	1.000	−0.175
^{40} K	−0.022	−0.175	1.000

4.75% ThO_2 and 0.18% of UO_2 (Dirección Nacional de Minería y Geología (DINAMIGE), 2002; Ferrando et al., 2003), therefore with a higher ^{232}Th activity concentration than ^{238}U ones. The ^{226}Ra and ^{232}Th activity concentrations are also comparable with the previous reports for South American soils, Buenos Aires, Argentina (Montes et al., 2016), Rio de Janeiro (Ribeiro et al., 2017), and Espiritu Santo (Dutra Garcêz et al., 2020). The activity concentrations for natural radionuclides are lower than the ones reported for high natural radiation areas, such as Egypt—215.43 Bq kg^{-1} for ^{226}Ra, 131.26 Bq kg^{-1} for ^{232}Th and 822.76 Bq kg^{-1} for ^{40} K (Tawfic et al., 2021), Guarapari, Brazil (4043 Bq kg^{-1} for ^{226}Ra, 55,537 Bq kg^{-1} for ^{232}Th) (Veiga et al., 2006), and Kerala, India (296.5 Bq kg^{-1} for ^{226}Ra, 1087 Bq kg^{-1} for ^{232}Th) (Monica et al., 2016).

Table 3 shows a summary of radiological indices for inhabitants and tourists in Barra de Valizas. Long-life naturally occurring radionuclides activity concentrations and radiological hazard indices show high variability between samples, demonstrating similar behavior to results previously reported for the department of Rocha (Noguera et al., 2018, 2022). The mean values for all radiological indices are higher than the worldwide mean and those recommended for inhabitants. These values are accentuated when considering the real situation of occupation in Barra de Valizas, described in previous sections, and consider the real occupation factor of 4380 h yr^{-1} (AEDE (50)). In this case, the mean ELCR is 9×10^{-4}, which triples the recommended value 2.9×10^{-4} (United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 2008). The annual effective dose equivalent for tourists is below the recommended value (70 µSv.yr^{-1}, United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 2008) for both, summer-season and fortnight-length tourists.

The high value of the Excess Lifetime Cancer Risk (ELCR) index, higher than 2.9×10^{-4} recommended by United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR (2008), may be a factor to consider when explaining the high Standardized Mortality Ratio (SMR) and Standardized Incidence Ratio (SIR) previously mentioned for this Uruguayan region (Barrios et al., 2022). However, the lack of data about actual residence of cancer patients in this region made it impossible to establish a local correlation between ELCR and SMR for Barra de Valizas.

Table 4 compares the radiological hazard indices for inhabitants in Barra de Valizas with values calculated

Table 3 Statistical values for radiological indices for population and tourists in Barra de Valizas

Inhabitants hazard indices

	AEDE (20) μSv.yr^{-1}	AEDE (50) μSv.yr^{-1}	AGDE μSv.yr^{-1}	ELCR (20) × 10^{-4}	ELCR (50) × 10^{-4}
Mean	92.7	232	523	3.6	9.0
Median	58.8	147	335	2.2	5.7
Minimum	16.80 ± 0.35	42.0 ± 1.6	99.4 ± 3.9	0.653 ± 0.014	1.630 ± 0.034
Maximum	887 ± 49	2217 ± 19	4953 ± 44	34.5 ± 1.9	86.20 ± 0.48
Percentile confidence interval	50.8–165.0	126 ± 414	291–926	1.9–6.4	5.0–16.0
Worldwide mean UNSCEAR (2008)	70		300	2.9	

Tourist hazard indices

	AEDE (μSv.yr^{-1}) summer	AEDE (μSv.yr^{-1}) fortnight
Mean	25.4	6.4
Median	16.1	4.0
Minimum	4.60 ± 0.10	1.151 ± 0.024
Maximum	243 ± 14	60.8 ± 3.4
Percentile confidence interval	13.9–45.3	3.5–11.2

Worldwide mean AEDE, AGDE and ELCR values reported by the United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR, (2008)

from previously reported natural radionuclide activity concentrations in Uruguay, and with indices reported in other parts of the world. The mean value for excess lifetime cancer risk is similar to that calculated for Cerro Largo (Eastern region) and Paysandú (Western region) and higher than the ELCR calculated in Salto (Western region), all in Uruguay. Despite this, it does not show a clear geographical distribution between the eastern and western regions of the country. The outdoor annual effective dose (AEDE) and the annual gonadal dose equivalent (AGDE) show similar distributions. If radiological hazard indices are compared with other countries ones, they are higher than those from Jamaica, Kuwait, Vietnam, Iraq, USA, the coastal zone of Alagoas (Brazil) and China, and lower than those from blacklands in Alagoas, Brazil, Turkey, Malaysia, India, Cyprus and Egypt.

Conclusions

Radiological hazard indices for Barra de Valizas inhabitants and tourists were evaluated. The outdoor annual effective dose (AEDE), excess lifetime cancer risk (ELCR) and annual gonadal dose equivalent (AGDE) are higher than the recommended values (70 μSv.yr^{-1} for AEDE, 300 μSv.yr^{-1} for AGDE and 2.9 × 10^{-4} for ELCR, (United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) 2008) for inhabitants with both occupation factors (0.2 and 0.5 total time). The radiological risk for summer and fortnight tourists is not significant. The excess lifetime cancer risk was evaluated and may contribute to Rocha´s higher SRM value, although a direct correlation cannot be assured with the available epidemiological information. To evaluate the real hazard of the assessed value on the population, reliable and standardized mortality and morbidity statistics of Barra de Valizas are needed. Social, medical and anthropological studies must be carried out in future in order to corroborate any correlation between ELCR and SMR and have been already recommended to the Uruguayan Honorary Commission to Fight Cancer.

Table 4 The outdoor annual effective dose (AEDE), excess lifetime cancer risk (ELCR) and annual gonadal dose equivalent (AGDE) for Barra de Valizas inhabitants compared with values obtained for other regions of Uruguay and other places in the world

	ELCR × 10^{-3}	AEDE (μSv.yr^{-1})	AGDE (μSv.yr^{-1})	References
Barra de Valizas	0.36	93	525	This work
Cerro Largo, Uruguay	0.47 ± 0.17	120 ± 44	703 ± 43	Calculated from Montes and Desimoni, (2011); Odino, (2010)
Salto, Uruguay	0.065 ± 0.007	16.8 ± 1.8	96.5 ± 9.0	Calculated from Montes and Desimoni, (2011); Odino, (2010)
Paysandú, Uruguay	0.29 ± 0.06	73 ± 16	422 ± 25	Calculated from Montes and Desimoni, (2011); Odino, (2010)
Alagoas (Coastal), Brazil	0.217 ± 0.0095	62 ± 2.7		Filgueiras et al., (2020)
Alagoas (Agreste), Brazil	0.34 ± 0.013	98 ± 3.7		Filgueiras et al., (2020)
Alagoas (Blackland), Brazil	0.43 ± 0.16	124 ± 4.6		Filgueiras et al., (2020)
Kajaran, Armenia	0.21–0.77	60–220		Belyaeva et al., (2019)
Kapan, Armenia	0.08–0.31	20–90		Belyaeva et al., (2019)
Jamaica	0.0016–0.792		69.28–2018.25	Miller and Voutchkov (2016)
Kuwait	0.063–0.305		96–457	Jallad, (2016)
Ho Chi Minh City, Vietnam	0.088–0.273			Ba et al., (2019)
Kirklareli, Turkey	0.10–1.20	144		Taskin et al., (2009)
Langkawi Island, Malaysia	1.81–8.56	490–2300		Khandaker et al., (2018)
Kerala, India	4.45–14.14			Monica et al., (2016)
Hassan, India		120		Srinivasa et al., (2019)
China	0.19 ± 0.02	55.20 ± 5.22		Zhou et al., (2020)
Nigeria	0.39 ± 0.03	111.74 ± 8.71	635.75 ± 79.03	Gbadamosi et al., (2018)
Egypt	0.40–1.46	230	131	Hilal and Borai (2018)
Iraq	0.19	60		Mohammed and Ahmed (2017)
Texas, USA		59.3 ± 15.2		Hannan et al., (2015)
Cyprus			669.43 ± 136.48	Abbasi et al., (2020)
Worldwide mean	0.29	70	300	United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), (2008)

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Declarations

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