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Natural radionuclide and ^{137}Cs activity concentration in soils of the Brazilian State of Sergipe and their correlation with environmental characteristics

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Abstract: Radionuclides occur naturally in the environment, exposing living organisms to ionizing radiation. In addition, human activities have introduced artificial radionuclides such as ^{137}Cs, among others, into the environment. To better understand the occurrence and behaviour of radionuclides in soil, several countries have developed radiological studies and constructed maps. An effort has been made in Brazil to assess the radioactivity in the soils of the country. In this study, the activity concentrations of natural radionuclides ^{40}K, ^{226}Ra and ^{228}Ra, and artificial ^{137}Cs were determined in samples at a depth of 20 cm collected from the soils of the Brazilian State of Sergipe, in a regular grid of 25 km x 25 km. Samples were analysed by gamma-ray spectrometry with the use of hyperpure germanium detectors. The results were correlated with the type of soil, geology, and climate characteristics of the State. The median activity concentrations of ^{40}K, ^{226}Ra, and ^{228}Ra were 210.4 $Bq.kg^{-1}$, 12.33 $Bq.kg^{-1}$, and 18.69 $Bq.kg^{-1}$, respectively. The results are lower than those reported for soils from other Brazilian States in the same region. The activity concentration of ^{137}Cs was lower than the minimum detectable activity. The activity concentration of ^{40}K in young soils of the semi-arid region was higher than that found in most weathered soils of the rainy regions due to the very low pluviometry of the semi-arid area. Higher activity levels of ^{226}Ra were found in Planosol and for ^{228}Ra it was found in the weathered soils (Acrisol) of the coastal zone. Maps of the distribution of the studied radionuclides were designed. The median value of the outdoor absorbed dose rate in air (Dnat) calculated for the soils of the State of Sergipe is lower when compared with other Brazilian States and the UNSCEAR's worldwide median value.

Keywords: natural radionuclides, soil radioactivity mapping, external dose.

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Concentração de atividade natural de radionuclídeos e ^{137}Cs em solos do estado brasileiro de Sergipe e correlação com características ambientais

Resumo: No meio ambiente, o radionuclídeo ocorre naturalmente, causando a exposição dos organismos vivos à radiação ionizante. Além disso, as atividades humanas introduziram no meio ambiente o radionuclídeo artificial ^{137}Cs, entre outros. Para melhor compreender a ocorrência e o comportamento dos radionuclídeos no solo, vários países desenvolveram estudos radiológicos e construíram mapas. Um esforço tem sido realizado no Brasil para avaliar a radioatividade nos solos do país. Neste estudo, as concentrações de atividade dos radionuclídeos naturais ^{40}K, ^{226}Ra e ^{228}Ra e ^{137}Cs artificiais foram determinadas em amostras de 20 cm de profundidade coletadas em solos do estado brasileiro de Sergipe, em uma grade regular de 25 km x 25 km. As amostras foram analisadas por espectrometria de raios gama com uso de detectores de germânio hiperpuro. Os resultados foram correlacionados com características de solo, geologia e clima do estado. As medianas da concentração de atividade de ^{40}K, ^{226}Ra e ^{228}Ra foram 210,4 Bq.kg^{-1}, 12,33 Bq.kg^{-1} e 18,69 Bq.kg^{-1}, respectivamente. Os resultados são inferiores aos reportados para solos de outros estados brasileiros da mesma região. A concentração de atividade de ^{137}Cs foi inferior à atividade mínima detectável. A concentração de atividade de ^{40}K em solos jovens da região semiárida foi superior àquelas encontradas nos solos mais intemperizados das regiões chuvosas devido ao baixíssimo índice pluviométrico da região semiárida. A concentração de atividade mais alta para ^{226}Ra foi encontrada para o Planossolo e para ^{228}Ra foi encontrada nos solos mais intemperizados da área costeira (Argissolo). Foram elaborados mapas de distribuição dos radionuclídeos estudados. O valor mediano da taxa de dose absorvida no ar (Dnat) em ambientes externos calculado para os solos do estado de Sergipe é menor quando comparado com outros estados brasileiros e com o valor mediano mundial da UNSCEAR.

Palavras-chave: radionuclídeos naturais, mapeamento da radioatividade no solo, dose externa.

1. INTRODUCTION

In the natural environment, there are unstable chemical elements that release energy or subatomic particles to become stable. These elements, known as radionuclides, can be found in rocks, soil and living organisms (vegetable, animal), such as ^{40}K. Other radionuclides, such as ^{137}Cs, can be produced artificially through processes such as nuclear reactions of nuclear weapons testing and nuclear power plants and are release into the environment through human activities [1].

Soil is a source and receptor of radionuclides, containing both man-made and naturally occurring radionuclides. The presence of radionuclides in soil can have environmental and health consequences [2]. The dose of ionising radiation received by humans from radionuclides in the soil varies greatly, depending on a number of factors, including the composition of the soil, the geology of the area, and the lifestyle of the people living there [1]. On average, exposure to ionising radiation from soil is relatively low and generally does not have significant impact on human health. It is worth noting that although exposure to ionising radiation is unavoidable, regulatory bodies set dose limits to protect people from the harmful effects of radiation [1,2].

The assessment of the radionuclides content of soils has been carried out in many parts of the world. Several countries have developed radiation maps of their soils and calculated the external doses to which their populations are exposed, e.g. [3–7]. In Brazil, most of the studies have been carried out in areas with high background levels, and assessments of radionuclides in Brazilian soils covering large areas of the country are scarce. Some authors have investigated the radionuclide levels in the states of Rio Grande do Norte, Alagoas, Pernambuco and Espírito Santo [8-11] and others assessed the public exposure in Brazilian inhabited areas [12, 13] . Some authors intended to set some values to act as a guide for the radionuclide content in Brazilian soils, as reference values [14-16]. In addition, there is a lack of knowledge about the concentration of natural radionuclides and fission products radionuclides in the largest part of the Brazilian territory and the radiation dose to which the population is exposed.

The aim of this study was to map the distribution of the natural radionuclides ^{40}K, ^{226}Ra, and ^{228}Ra and the anthropogenic ^{137}Cs in the soils of the Brazilian State of Sergipe, contributing to the Atlas of Radioactivity in Brazilian Soils, and to correlate the activity concentration of these radionuclides with the soil, geological, and climatic characteristics of the State. The external dose rates due to the exposure to the radionuclides studied was estimated and mapped.

2. MATERIALS AND METHODS

2.1. Study area

The State of Sergipe is situated in the Northeastern region of Brazil (figure 1), with an area of 22,925.4 km^2. The population of the State was estimated to be 2,318,822 inhabitants [17]. It has borders with the States of Alagoas in the north, Bahia in the south and west, and the Atlantic Ocean in the east. Its climate is classified into three mesoregions according to Koopen: the coastal zone known as "Zona da Mata," covering a strip along the entire coast of the State with a climate type As, with annual rainfall exceeding 800 mm; in the hinterlands, also known in Portuguese as "Sertão", the climate is classified as Bssh, with annual rainfall between 200 and 750 mm annually concentrated from May to July. Between these two areas is the midland known as "Agreste", classified as BSh [18].

The most prominent soils in the State according to the WRB/FAO classification are Acrisols, Leptosols and Planosols [19]. With respect to geology, the State is formed by granitic rocks, migmatites, sedimentary rocks, and granitoids [20].

Figure 1: Location of the Brazilian State of Sergipe.

2.2 Sampling and sample preparation

Forty-five (45) soil samples were collected by the Geological Survey of Brazil (SGB) throughout the entire State of Sergipe with a spacing of 25 km between collection points, in the depth of 20 cm. Figure 2 presents the distribution of sampling points in the state.

Samples were identified in the field and the geographical coordinates of each sample were recorded. All samples were sent to the Laboratory of Soil Preparation of the Institute of Radiation Protection and Dosimetry (IRD), where they were dried in an oven for 48 hours at 60°C, crushed, and sieved through a 2 mm (9 mesh) sieve. After these steps, the samples were placed in polyethylene pots with a capacity of 250 ml, then weighed, sealed, and wrapped in aluminium foil and stored for at least 40 days to allows secular equilibrium between ^{226}Ra and its radioactive decay products to be reached.

Figure 2: Distribution of soil sample collection points in the State of Sergipe.

2.3 Determination of radionuclides

For the determination of the natural radionuclides, the samples were analysed by gamma-ray spectrometry using hyper pure germanium detectors with 50% relative efficiency (model BE5030) and energy resolution for 1332 keV peak of ^{60}Co of 1.6 keV, in the Laboratory of Environmental Radiometry of the IRD/CNEN. The detector is housed in an ultra-low-background, cylindrical shield model 777, from Mirion Inc. (Meriden, USA), constructed of bulk lead and lined with a low background tin-copper liner.

The detection system was calibrated weekly of energy using a point source of ^{152}Eu and for efficiency using a radioactive cocktail of ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{203}Hg, ^{210}Pb, ^{113}Sn, ^{85}Sr, and ^{88}Y.

Background spectra were acquired monthly for 60,000 s, the same counting time as the samples, using the Genie 2000 Gamma Acquisition and Analysis software package from Mirion Inc. (Meriden, USA).

The natural radionuclides were determined using the following energy lines: ^{40}K at 1460.8 keV, ^{226}Ra from its decay products ^{214}Pb (351.9 keV and 295.2 keV) and ^{214}Bi (609.3 keV), and ^{228}Ra from its decay product ^{228}Ac (911.2 keV). ^{137}Cs was determined from its gamma-ray peak at 661.6 keV.

In order to assess the quality of the results, the Gamma-ray Spectrometry Laboratory has implemented a Quality Management System based on the ISO 17025:2017 standard and participated with satisfactory results in proficiency tests with the International Atomic Energy Agency (ALMERA-IAEA) and the Brazilian National Intercomparison Program (PNI), carried out by the National Laboratory of Metrology of Ionising Radiation (LNMRI/IRD), which is tracked to the *Bureau International des Poids et Mesures* (BIPM).

2.4 Statistical analysis and mapping

The results were entered into a spreadsheet and statistically evaluated using Minitab software version 22. Maps of the distribution of radionuclides in soils and interpretation of geographical data were carried out using QGIS software version 3.36.2. The statistical test used to compare medians was the Mann-Whitney's median test, which is used for data sets with non-parametric distributions and small sample sizes. The null hypothesis (H_0) asserts that the medians are not different, and the alternative hypothesis (H_1) states that the medians are different. The significance level adopted was 95%, thus, if the calculated p-value is lower than 0.05, the null hypothesis must be rejected and the alternative hypothesis that the distributions are different must be accepted.

The data interpolation method used to produce a dose and natural radionuclide map was kriging [21], a method that takes outliers values into account. These maps support the spatial interpretation of the data.

2.5 Outdoors absorbed dose rate (D_{nat})

The outdoors air absorbed dose rate (D_{nat}, in nGy.h^{-1}) is calculated using Equation 1 [22]:

$$D_{nat} = 0.0417C_{K40} + 0.462C_{Ra226} + 0.604C_{Ra228} \quad (1)$$

Where C is the activity concentration of the radionuclide (Bq/kg dry soil), and each radionuclide is multiplied by its respective factor to convert the kerma rate (nGy.h^{-1} per Bq.kg^{-1}): 0.0417 for ^{40}K, 0.462 for the decay series of ^{238}U and 0.604 for the ^{232}Th decay series [22].

3. RESULTS AND DISCUSSIONS

3.1 ^{137}Cs

For ^{137}Cs, none of the samples had an activity concentration higher than the minimum detectable activity concentration, which varied from 0.28 to 0.99 Bq kg^{-1}. This is probably due to the low pluviometry index of the State of Sergipe, and unlike the neighbouring State of Alagoas, as pointed out in [23], Sergipe does not have mountains high enough to form clouds and deposit the radionuclide by orographic rain (wet fallout). Furthermore, the dispersion of ^{137}Cs in the atmosphere resulted in low concentrations of the radionuclide in the soil close the equator line, as compared to higher latitudes [24]. These values are important for comparison if nuclear power plants are built on the banks of the São Franscisco River, the main river in Sergipe.

3.2 Natural radionuclides

The activity concentration of the natural radionuclides determined in the samples, with the median and range, are summarised in the Table 1. As the results did not show a normal frequency distribution, the median was the most representative measure to represent the data [8–10]. The median activity concentration values decreased in the following order: ^{40}K > ^{228}Ra>^{226}Ra.

Table 1: Comparison of the activity concentration (range and median, $Bq.kg^{-1}$) of results obtained in this study and other studies in Brazilian soils.

REFERENCES	^{40}K		^{226}Ra		^{228}Ra	
	RANGE	MEDIAN	RANGE	MEDIAN	RANGE	MEDIAN
Sergipe (present study)	9.16-1,128	210.4	3.8-28.7	12.33	7.86-90.2	18.69
Pernambuco [10]	<12 – 2,370	464	3.2-94	20	7.2-276	37
Alagoas [8]	6-2,160	420	5-63	29	54-157	43
Rio Grande do Norte [11]	56.4-1,972	517.2	10.3-137.6	25.1	12-191	36.5
Espírito Santo [9]	6.9-693	27.7	11-88	29	18-424	109

In comparison with the results reported by other studies in Brazil, the range is within the reported ranges, but the median activity concentration of ^{40}K obtained in this study was lower than those obtained in other Brazilian states in the same region (Alagoas [8] and Pernambuco [10]), but higher than those reported for the State of Espírito Santo [11], probably because Sergipe has a lower rainfall than Espírito Santo, which is located in a different region of Brazil.

The results indicated that the activity concentration of ^{228}Ra in the studied soils is higher than the activity concentration of ^{226}Ra, a typical behaviour in Brazilian soils that contain more thorium than uranium, as observed in the other studies on Brazilian soils [26].

3.3 Influence of climate, geology, and soil types on natural radionuclides concentration

Figure 3 presents the map of the distribution of ^{40}K in the soils of Sergipe. Figure 4 presents a box plot graphic comparing the occurrence of ^{40}K in the three climatic regions of the State.

Moving from the coast to the interior of Sergipe an increase in the median ^{40}K activity concentration can be observed. The coastal and midland areas had a lower median of ^{40}K activity concentration, and the hinterland region ("Sertão", semi-arid) had higher median activity concentration. However, the results of the Mann-Whitney's median test indicated that there was no difference between the medians of ^{40}K in the soils of the coastal zone and the midland (p-value 0.252) and between the midland and the hinterland (p-value 0.164), but there was a difference between the coastal zone and the hinterland (p-value 0.010), suggesting the influence of such different climates on the occurrence of ^{40}K.

Figure 3: Distribution of ^{40}K in Sergipe's soil ($Bq.kg^{-1}$).

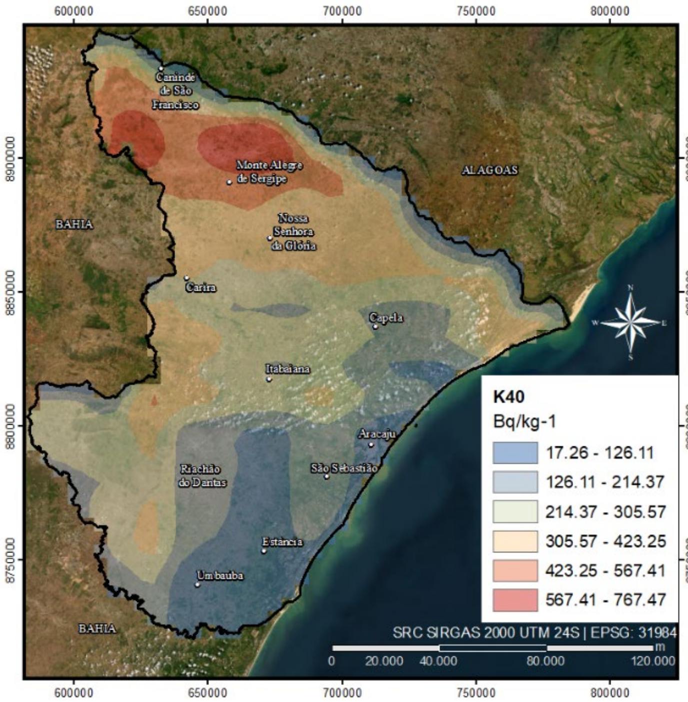
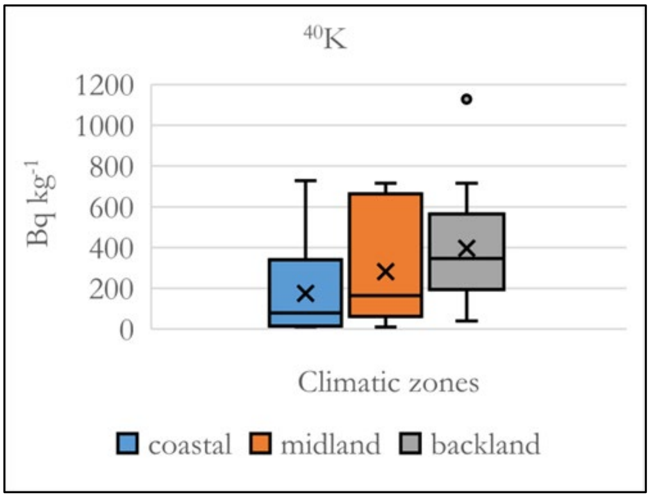
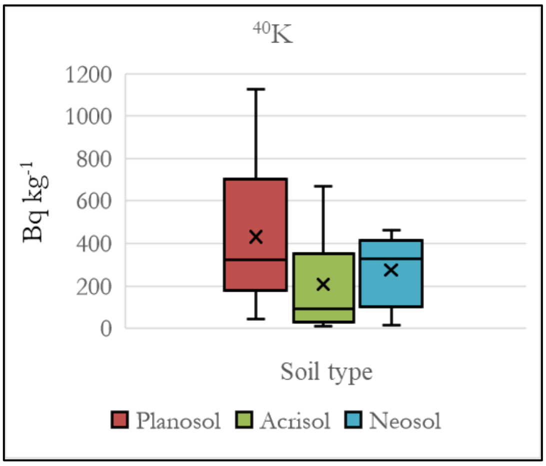


Figure 4: Comparison of the ^{40}K activity concentration among the climatic regions of Sergipe. The line inside the box represents the median, and X represents the arithmetic mean.



The association between dry weather and igneous rocks originated young soils, shallow and poorly developed, and with the presence of not weathered minerals. In these dry soils, ^{40}K that is a highly soluble radionuclide, tends to accumulate in the superficial layers. In contrast, the more weathered soils in the coastal zone showed a depletion of ^{40}K, mainly due to the higher pluviometry. A higher value of ^{40}K was found for Chernosols, but due to the small number of samples of Chernosols, this type of soil was excluded from the evaluation. The second soil type with high activity concentration of ^{40}K was Planosol. However, the median values of Planosol and Neosol were quite similar, which was also observed in the Mann-Withney's median test (p-value = 0.526). Both soils types are shallow and young soils, presenting less weathered minerals and higher adsorption capacity for cations. The lowest median activity concentration of ^{40}K was observed in the Acrisols (figure 5). The statistical test of median showed that there was a difference between Planosol and Acrisol in ^{40}K (p-value =0.042, lower than the 0.05 significance level).

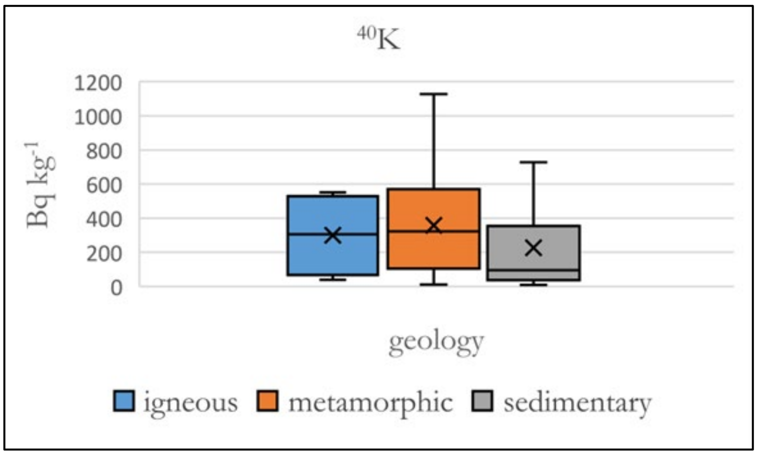
Figure 5: Comparison of ^{40}K activity concentration among the soils of Sergipe. The line inside the box represents the median, and X represents the arithmetic mean.



In terms of geology, samples from soils developed on both igneous and metamorphic rocks had similar activity concentrations, as can be seen in figure 6. Soils developed on the sedimentary rocks had a lower median, indicating potassium isotope leaching. Despite this difference, the Mann-Whitney test for median showed that there was no statistical difference among the medians for ^{40}K in the analyzed samples for soils developed according to the types of geology. Comparison of median with sedimentary and metamorphic rocks and igneous rocks could not be made because only 4 soil samples were collected over the igneous rocks.

Radium isotopes were better distributed in the soils of the state (Figures 7 and 8). The hinterland ("Sertão") had lower median concentrations of radium isotopes than the coastal and midland areas, which had similar medians (Figure 9). However, statistically no differences were observed when the Mann-Whitney's test of median was applied (for ^{226}Ra: coastal zone and midland p-value = 0.137, coastal zone and hinterland p-value= 0.197, midland and hinterland p-value = 0.828; for ^{228}Ra: coastal zone and midland p-value = 0.075, coastal zone and hinterland p-value = 0.066, midland and hinterland p-value = 0.270).

Figure 6: Comparison of ^{40}K activity concentration in soils according to the geology of Sergipe. The line inside the box represents the median, and X represents the arithmetic mean.



The higher concentration of radium isotopes in soils from the coastal zone can be explained by the fact that uranium and thorium, parent radionuclides of ^{226}Ra and ^{228}Ra, tend to concentrate in more developed soils with acid pH and rich in iron and aluminum oxides, such as those from the coastal zone. Other Brazilian authors [8-10] have also pointed out this behavior.

Figure 7: Map of the ^{226}Ra activity concentration distribution in the soils of Sergipe (Bq kg^{-1}).

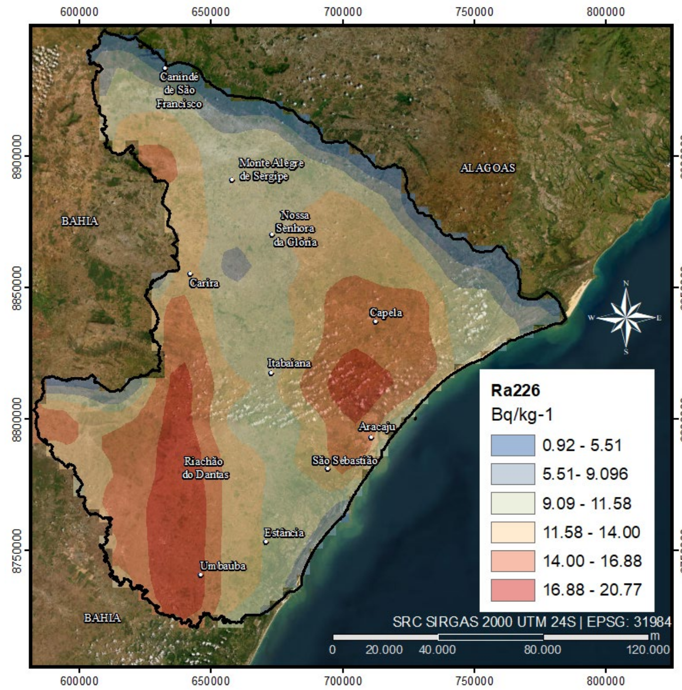


Figure 8: Map of the ^{228}Ra activity concentration distribution in the soils of Sergipe (Bq kg^{-1}).

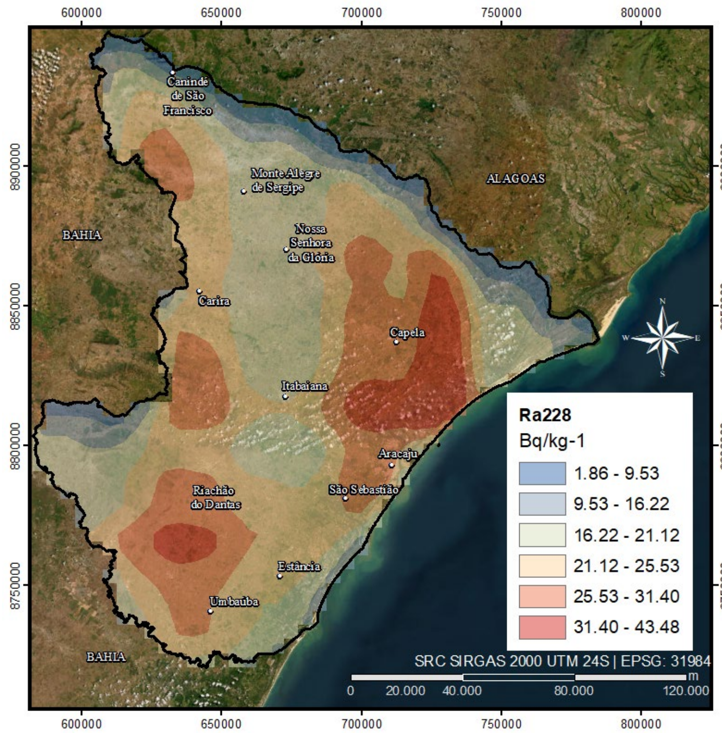
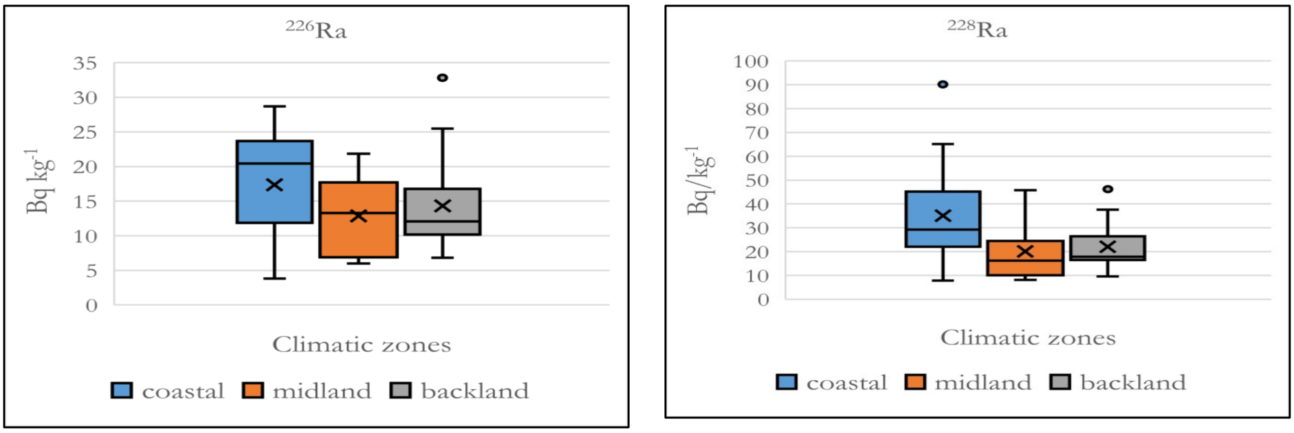


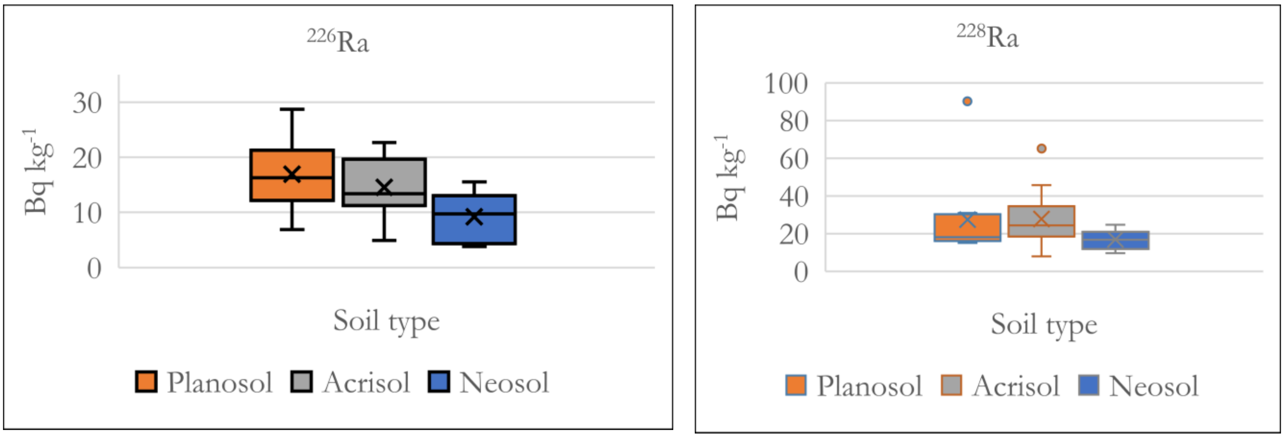
Figure 9: Comparison of the activity concentration of radium isotopes according to the different climatic types of Sergipe. The line inside the box represents the median, and X represents the arithmetic mean.



The influence of the soil type on the radium isotope in the soils of Sergipe can be seen in Figure 10. Planosol soil was also the soil type with the higher median value of ^{226}Ra. This result is different from those found in the neighboring states of Alagoas [23] and Pernambuco [27]. The lowest median radium isotope content was found in the Neosol. Comparing the median ^{226}Ra values, there was no significant difference between the Planosol and the Acrisol (p-valie = 0.401), but there was a significant difference between the Planosol and the Neosol (p-value = 0.022) and between the Acrisol and Neosol (p-value = 0,020)

For ^{228}Ra, a higher median value was observed in Acrisol. The median value was not statistically different (p-value = 0.260) from the Planosol, but different from the median of the Neosol (p-value = 0.043). A lower median value was observed for Neosol, similar to the ^{226}Ra results. This higher values of ^{228}Ra in the well-developed soils of the coastal and Agreste regions, mainly Acrisol, have also been observed by other authors in Brazilian states [24-27]. These soils are formed in an environment of sediment accumulation and are generally associated with the weathering of primary minerals containing traces of radioactive elements, including radium. As these minerals weather, they release radium into the soil.

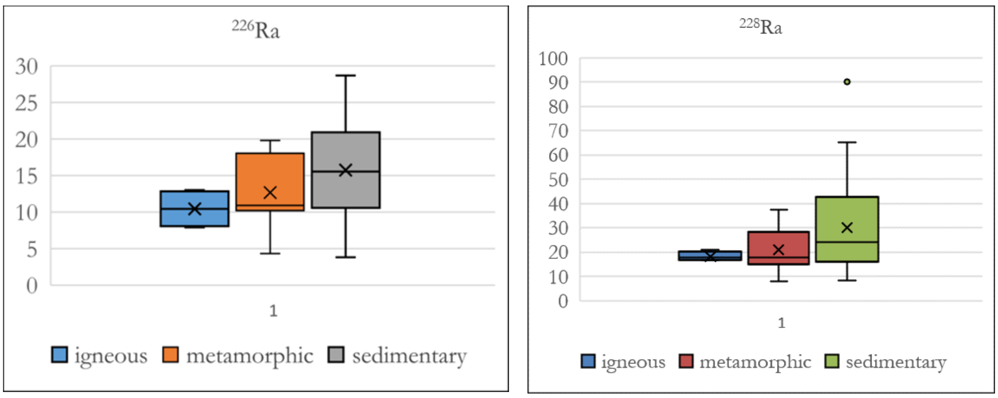
Figure 10: Comparison of radium isotopes activity concentration in different soil types of Sergipe. The line inside the box represents the median, and X represents the arithmetic mean.



In relation to geology, radium isotopes had higher activity concentrations in soils developed on sedimentary rocks, and ^{40}K had the highest concentration in soils developed on metamorphic rocks (Figure 11). Comparing the influence of geology on the abundance of radium isotopes in the soils of Sergipe, it can be seen a tendency to concentrate radium in soils developed on sedimentary rocks.

These results indicate a relationship between two factors that influence soil formation and consequently the occurrence of radionuclides: climate and the parent material, in the case of rocks.

Figure 11: Influence of the geology of the State of Sergipe and the radionuclides studied.



3.6 Comparison of medians among Brazilian States

Table 2 shows a comparison of the median values of the studied natural radionuclides in the Planosol and Acrisol for the three states of the Northeastern region of Brazil (Pernambuco [10], Alagoas [8] and Sergipe [this study]). The values varied widely for ^{40}K, and the two soils of Sergipe had lower median values, except for ^{40}K for Acrisol from Alagoas. For the radium isotopes, the values of median values are also lower than those from other states.

Table 2: Comparison of median activity concentrations (Bq kg^{-1}) of the studied natural radionuclides in Pernambuco, Alagoas and Sergipe.

SOIL TYPE	LOCATION	RADIONUCLIDE		
		^{40}K	^{226}Ra	^{228}Ra
Planosol	Pernambuco [10]	583	25	47
	Alagoas [8]	1132	31	59
	Sergipe	322	16.3	18
Acrisol	Pernambuco [10]	291	23	37
	Alagoas [8]	19	22	32
	Sergipe (this study)	57.6	13.4	26.3

3.7 Outdoors absorbed dose

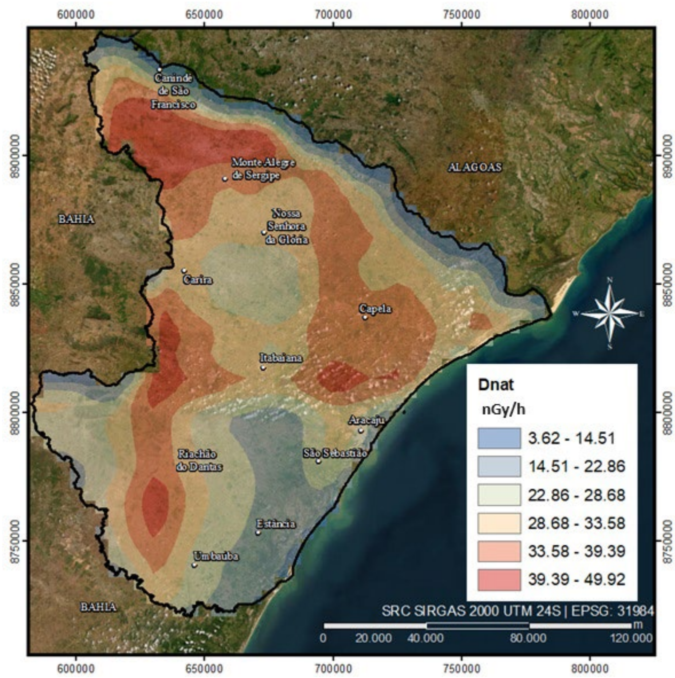
On the basis of the activity concentrations, the outdoors air absorbed dose (D_{nat}) was calculated (table 3). Comparing the values found in Sergipe with those reported for the State of Alagoas, it can be observed that the soils of Sergipe presented practically half of the values of other States, which are even lower than values reported by LEAL et al. [10] for the Brazilian State of Pernambuco. Also, comparing with the median value reported by UNSCEAR [1], the median value calculated for the State of Sergipe is lower.

Figure 12 shows the distribution of the absorbed dose rate outdoors (Dnat) calculated in this study. The areas with the greatest contribution to the external dose outdoors to the population were in the midland (“Agreste”) and hinterland (“Sertão”) regions of Sergipe.

Table 3: Outdoor Absorbed Dose Rate in air (D_{nat}, in $nGy.h^{-1}$) calculated for the soils of the State of Sergipe and compared with other Brazilian States and the UNSCEAR world median value.

Location	D_{nat} (nGy h^{-1})		Reference
	Range	Median	
Alagoas	8.9 - 167	70	[8]
Pernambuco	6.0 - 235	56	[10]
Sergipe	9.5 – 83.6	31.1	This study
UNSCEAR		58	[1]

Figure 12: Distribution of Air Absorbed Dose Rates Outdoors (D_{nat}) in the State of Sergipe ($nGy.h^{-1}$).



4. CONCLUSIONS

The activity concentrations of the natural radionuclides ^{40}K, ^{226}Ra, and ^{228}Ra were determined in soil samples from the State of Sergipe, Brazil, and the influence of environmental characteristics (soil and climate) was studied. The radionuclides studied showed a non-parametric frequency distribution of activity concentrations. The medians for ^{40}K, ^{226}Ra and ^{228}Ra, were 210.4 Bq kg^{-1}, 12.33 Bq kg^{-1} and 18.69 Bq kg^{-1}, respectively.

None of the samples showed an activity concentration of ^{137}Cs above the minimum detectable activity (which ranged from 0.28 to 0.99 Bq.kg^{-1}), indicating that the presence of the artificial radionuclide in the soils of the State is very low.

The activity concentration of ^{40}K is higher in the semi-arid region due to the high potassium content of the rocks that formed to the soil and the very low precipitation rate, which reduces the leaching of the element. For the radium isotopes (^{226}Ra and ^{228}Ra), the activity concentration was higher in the soils of the coastal zone and southwestern Sergipe. The soils of these areas are Acrisol, which are more developed, highly weathered and rich in mica and feldspar, minerals of igneous origin which are the original matter of Acrisols.

The highest activity values in ^{40}K and ^{226}Ra were found in the Planosol of the hinterland ("Sertão") and midland ("Agreste"), and in ^{228}Ra it was found in the Acrisol in the coastal area ("Zona da Mata"). Regarding geology, ^{40}K showed its highest values in igneous and metamorphic rocks. For radium isotopes, the highest median values were found in soils developed on sedimentary rocks. Differing from other studies on Brazilian soils, the soil that presented higher median of ^{226}Ra was Planosol.

The median value of the outdoor **air** absorbed dose rate (D_{nat}, in nGy.h^{-1}) calculated for the soils of the State of Sergipe is lower when compared to other Brazilian States and to the UNSCEAR world median value.

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