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Radium-228 as an Indicator of Thorium-232 Presence in a Soil in Pernambuco, Brazil

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Abstract Radiometric measurements were taken in a small area of Pernambuco, Brazil, with the objective of monitoring the radium of the soil. For this, 78 soil samples were collected. The gamma analyses of the samples were carried out using HPGe. The values obtained for the ^{226}Ra varied from 14 to 367 Bqkg^{-1} and for the ^{228}Ra from 73 to 429 Bqkg^{-1}. The ratio ^{228}Ra/^{226}Ra varied from 1.0 to 7.0. Therefore, it is an indicator of an additional radioactivity source in this soil, maybe ^{232}Th, which will be further investigated in future studies.

Keywords Radium · Environment radionuclides · Gamma-spectrometry

Natural radioactivity may represent a great source of exposure to living beings. In soils, these levels are associated with the mineral composition of the rocks in each area and the concentrations of radionuclides present in the rocks that originated the soil. In general, more elevated levels of radiation are associated with igneous rocks and lower levels with sedimentary rocks (Tzortzis and Tsertos 2004).

Radium is present in all of the natural series of radioactive decay and is usually found in nature in low concentrations (in soil, a mean of 0.8 ppt) (Arafa 2004). However, greater concentrations of radium are found in minerals that contain uranium and thorium. The mean activity of radium estimated on the soil surface of normal areas is nearly 26 Bqkg^{-1} (Malanca et al. 1993). From a biological and radiological point of view, the ^{226,228}Ra isotopes, are considered the most important radionuclides of the ^{238}U and ^{232}Th series, respectively. When in the human body, the radioisotopes behave chemically and physiologically like calcium and tend to concentrate basically in the bones and teeth. In the environment they form soluble compounds and can contaminate underground reservoirs, soils, plantations, food sources and consequently the human food chain.

In the northeastern region of Brazil, uranium anomalies were identified during radiometric monitoring studies carried out in the period of 1974–1975 in the municipality of Pedra, in the state of Pernambuco. In this area, measurements above 15,000 cps, carried with a scintillation counter, were found in bodies of a rock (Santos Júnior et al. 2006). However, despite the knowledge about these uranium anomalies, no monitoring studies have been conducted so far with respect to the presence of ^{226,228}Ra isotopes in the soil. This is a relevant question, since these isotopes may enter the human food chain in the milk produced by cattle raised in this area, which is sold in many cities in the state, including Recife, which is the capital of Pernambuco. Therefore, this work was conducted with the purpose of determining ^{226}Ra and ^{228}Ra activity in the soil, as well as their spatial distribution, around the main uranium anomaly in the municipality of Pedra, in Pernambuco.

Materials and Methods

About 78 soil samples were collected in the study area, above and around one of the main anomalies of uranium in an amphibolitic calcium-silicatic rock. The sampling was

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carried out at intervals of 25 m at a mean depth of 35 cm, on the C horizon. In the laboratory, the samples were dried at 100°C, then homogenized and sieved at a granulometry lower than 63 μm, and 200 g of each sample were packed in polyethylene containers of 210 cm^3 volumetric capacity. After this, the samples were hermetically sealed for a minimum period of 30 days, to establish the radioactive equilibrium between the short half-life radionuclides before measurements with the HPGe. In order to avoid any Rn loss, the internal part of the polyethylene receptacle was covered with aluminum foil, so as to cover the entire soil sample.

The measurements were carried out with a gamma spectrometry system, which allows to obtain the activities of these isotopes, to verify the environmental contamination levels. The measuring system is composed of a Canberra™ GX2518 (HPGe) hyper pure germanium detector, type BeGe, with an active volume of 41.1 cm^3. The detector presents a resolution of 1.77 keV for the 1,332 keV energy of the ^{60}Co, intrinsic efficiency of 27.7%, and is coupled to a multichannel analyzer (MCA) with 8,192 channels and Canberra™ Genie-2000 software. The detector is wrapped in a lead cover of ~6.5 cm thickness to reduce the background (BG) interface effects. For analyzing the ^{226}Ra, the gamma photopeaks of ^{214}Pb ($E_\gamma = 352$ keV; $\gamma_\% = 37.10\%$) and ^{214}Bi ($E_\gamma = 609$ keV; $\gamma_\% = 46.09\%$) were used, as well as the gamma photopeaks of ^{228}Ac ($E_\gamma = 911$ keV; $\gamma_\% = 29.00\%$ and $E_\gamma = 969$ keV; $\gamma_\% = 17.46\%$) for ^{228}Ra.

In gamma spectrometry, the counting efficiency is considered the most difficult problem to be solved, because the final result depends directly on a good efficiency curve. To obtain this, a liquid standard of ^{152}Eu certified by the IRD (Institute of Radioprotection and Dosimetry—Rio de Janeiro) was used. To reduce the effects of gamma auto-absorption, an aliquot of 50 mL of standard was added to 200 g of a previously analyzed soil matrix with a density similar to the samples with unknown activities. To prepare this standard sample, the same conditions as in the analysis of soil samples were used, such as: density, granulometry, mass, geometry of the container, geometry of counting (detector positioning) and counting time.

By using an statistical software, it was possible to obtain a better adjustment for the efficiency curve and consequently Eq. 1.

$$\varepsilon = \frac{a}{a-b}\left(e^{-bE_\gamma} - e^{-aE_\gamma}\right). \quad (1)$$

where ε is the counting efficiency of the considered gamma photopeak, a and b are statistical parameters for curve correction (0.0016 and 0.04757, respectively), and E_γ the gamma energy for which the counting efficiency is to be obtained.

For calculation of the radionuclide activities, Eq. 2 (Tzortzis et al. 2003), based on the established efficiency curve for ^{152}Eu, was employed:

$$A = \frac{C}{\varepsilon \times t \times \gamma \times m} \quad (2)$$

where A is the activity ($Bqkg^{-1}$); C is the total net area of the radionuclide photopeak at the considered energy (counts); ε is the counting efficiency for the considered specific energy; t is the counting time (s); γ the gamma abundance of the considered radionuclide; and m the sample mass (kg).

To determine the ^{226}Ra activities, the mean of the activities obtained for ^{214}Pb ($E_\gamma = 352$ keV; 37.10%) and for ^{214}Bi ($E_\gamma = 609$ keV; 46.09%), was used; for ^{228}Ra it was used the mean of the ^{228}Ac activities ($E_\gamma = 911$ keV; 29.00% and $E_\gamma = 969$ keV; 17.46%). The sample masses were standardized in 200 g and the gamma counting time in 43,200 s (12 h).

For the detection limit (LD) calculations, a soil sample from an area of low radioactivity and Eq. 3 were used:

$$\mathrm{LD} = \frac{4.66\sqrt{C}}{\varepsilon \times \gamma \times m \times t} \quad (3)$$

where C is the net counting; ε the counting efficiency; γ the gamma abundance or gamma emission probability of the considered radionuclide; m the mass (kg); t the counting time (s), and the value 4.66 is the factor that corresponds to 95% confidence.

Results and Discussion

The values obtained for the radionuclides activities of the anomalous rock (amphibolitic calcium-silicatic) varied from 14,018 to 83,567 $Bqkg^{-1}$ for the ^{226}Ra (Santos Júnior et al. 2006). On the other hand, the values for ^{228}Ra stayed below the detection limit (<10 $Bqkg^{-1}$).

Table 1 shows statistical parameters for $^{226,228}Ra$ and for the $^{228}Ra/^{226}Ra$ ratio, based on the same mean activity calculations. It is observed in this table that the ^{226}Ra activities vary from 14 to 367 $Bqkg^{-1}$ (mean of 58 $Bqkg^{-1}$), while for

Table 1 Statistical parameters for $^{226–228}Ra$

Parameter	Activity ($Bqkg^{-1}$)		
	^{228}Ra	^{226}Ra	$^{228}Ra/^{226}Ra$
Measure of interval	73–429	14–367	0.3–6.9
Arithmetic mean	223	58	4.6
Median	211	49	5.3
Mode	160	41	5.7
Frequency	Tendency to log-normal		

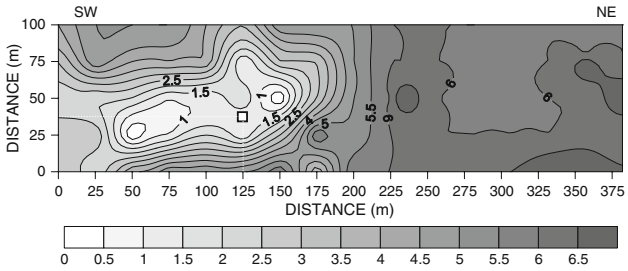

Fig. 1 $^{228}Ra/^{226}Ra$ isolines. Uranium anomaly in rock

^{228}Ra the values vary from 73 to 429 $Bqkg^{-1}$ (mean of 223 $Bqkg^{-1}$). Based on these data, it was possible to observe that in 96% of the samples the ^{228}Ra activities are higher than those of ^{226}Ra. For the $^{228}Ra/^{226}Ra$ ratio, it was possible to see that the radiological concentrations of ^{228}Ra are on average five times greater than those of ^{226}Ra, with values varying from 0.3 to 7 (mean of 5).

The limits of detection (LD) for the ^{226}Ra were $\sim$2 $Bqkg^{-1}$ for 352 keV photopeak of the ^{214}Pb and 3 $Bqkg^{-1}$ for the 609 keV photopeak of the ^{214}Bi. For ^{228}Ra the LD values were 8 and 12 $Bqkg^{-1}$ for the 911 and 969 keV photopeaks of the ^{228}Ac, respectively.

Figure 1 shows the isolines for the $^{228}Ra/^{226}Ra$ ratio. The values obtained show what was observed previously, i.e., near the anomaly the $^{228}Ra/^{226}Ra$ ratio is <1, while the values increase in the NE direction of the area.

These evidences show that ^{228}Ra concentrations are not directly correlated with the ^{226}Ra concentrations, it being obvious that ^{228}Ra found in the soil is not migrating from the uraniferous anomalous rock, but from another radioactive source that still has not been analyzed. The isorades show that the uranium anomalous rock does not present associated ^{232}Th, since the ^{228}Ra concentrations are greater in the northeastern portion of this area (contrary to the uranium anomaly). Therefore, as said above, we conclude that there is another natural radioactive source to be analyzed, very likely the granite that occurs in this area. From these results, new studies will be developed to identify the source of ^{232}Th supply, and so to warrant a better radiological control of the area, thus minimizing the risks associated with this radionuclide.

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