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^{226}Ra in milk of the dairy cattle from the rural region of Pernambuco, Brazil

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Studies performed by the Brazilian Nuclear Corporation (NUCLEBRAS), in collaboration with the Geological Survey Company of Brazil (CPRM), identified high levels of natural uranium in the districts of Pedra and Venturosa, in the rural region of the state of Pernambuco (PE) – Brazil, where the maximum value found in rocks was 22,000 mg·kg^{-1}. The raising of dairy cattle is one of the principal activities in these districts and adjacent areas. ^{226}Ra is released by weathering of rocks rich in uranium, entering the soil and water from where it is transferred to plants, animals and humans. The aim of this research was to determine ^{226}Ra concentrations in the milk produced in farms located in the two districts and nearby areas. The methodology used to determine and quantify ^{226}Ra was based on the ^{222}Rn emanation classical technique. The results of ^{226}Ra in milk samples varied from 4 to 500 mBq·l^{-1}. The consequences of the ^{226}Ra intake on human health are discussed.

Introduction

The so-called "Cow's Milk Basin", situated in the rural area of Pernambuco state, is the only tropical cow's milk basin in the world. In the physiographical regions of Pernambuco, this rural area is responsible for 60% of state lactic production (574,000 l/day) and has an average productivity per cow of around 800 l/year.[1]

Pedra and Venturosa, two cities in the "Cow's Milk Basin", together contribute with about 10% of the state production. The market of milk and derived products from the "Cow's Milk Basin" has a predominantly local destination (the state of Pernambuco itself), where they are commercialized mainly in supermarkets and bakeries.[1]

Radioactive anomalies were detected on a farm situated 8.7 km from Venturosa, in the proximity of the Ipanema River (Fig. 1). In this region local mafic rock was found, with a high uranium concentration.[2] The mineralization of uranium is localized in rolled blocks, found near the porfiric granite contact with the migmatites, which are two regional emergent units.[2] The analyses carried out on the existing rocks, revealed a maximum value of 22,000 mg·kg^{-1} (2.2%) of U_3O_8.[2] Figure 2 shows the specific activities of ^{238}U in the soil around these anomalous rocks.[3]

Among the radionuclides, ^{226}Ra (descendant of ^{238}U) is one of the most abundant in the environment. The radium dissolved from the rocks enters the food chain, primarily through water, plants, subsequently animals and finally reaching man.[4]

On the farms in the municipalities of Pedra and Venturosa, as most of the cities in the rural region of Pernambuco, the main crops cultivated and used for feeding dairy cows are forage palm (*Opuntia* spp.), buffel grass (*Cenchrus ciliares*), elephant grass (*Pennisetum purpureum*) and forage sorghum (*Sorghum bicolor*).[1] The determination of ^{226}Ra carried out in these plants showed concentrations varying from 96 to 7,990 mBq·kg^{-1} in dry matter. The forage palm specifically has elevated amounts of this radionuclide, varying from 1,490 to 7,990 mBq·kg^{-1} in dry matter.[5]

In the dairy cattle raising and milk producing rural region of Pernambuco, the extensive and semi-intensive management systems are predominant (75% of the producers), 50% using native pasture as the main source of feeding.[1]

Part of the ^{226}Ra ingested from the forage by a dairy cow is transferred to the milk.[6] In this context, the objective of this work was to determine the concentration of ^{226}Ra in milk of dairy cows in the Pedra and Venturosa municipalities. The overall aim was to carry out epidemiological projections about the degree of cancer in a population potentially exposed to an internal contamination with ^{226}Ra, resulting from the systematic ingestion of milk containing this radionuclide.

The analyses of the samples were carried out in the Environmental Monitoring Laboratory of the Nuclear Energy Department of the Federal University of Pernambuco.

Experimental

Criteria for selection of samples

Eighteen samples of milk were collected, six from control points. Milk samples were collected from nine farms located near the mineralization of uranium, anomalous rock (Fig. 1). The selection criteria were: the proximity of the farms to the occurrence of uranium and the quantity of milk produced. The farms chosen in the

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present study are the biggest producers of milk in the region studied, which have a daily production of approximately 6,000 l. The samples were collected in June, 2002 (rainy period), December, 2003 (dry period) and May, 2004 (rainy period).

Treatment of samples

From each farm, five liters of milk were collected from the storage tank of the daily milk production. The samples were collected in polyethylene containers, and immediately 5 ml of acetic acid and 10 ml of 37% formaldehyde were added for conservation.

Dry material was obtained after heating to 80 °C during 48 hours. Next, the material was taken to a furnace, the temperature was raised gradually up to 450 °C, and then left for 48 hours at this temperate until ashes were obtained. After these procedures, 10 g of ashes were taken for digestion in concentrated nitric acid, left to evaporate and afterwards reheated to 450 °C for 30 minutes. After cooling, concentrated nitric acid was added again and heated to the boiling point. The solution obtained was filtered and used for the determination of ^{226}Ra.[7]

Determination of ^{226}Ra

For the determination of ^{226}Ra in the samples, 1 l of solution was prepared from the solution formed by digestion of the ashes. In this case, the quantity of 1 l was transferred to a 2-l glass beaker, then adding: (a) 1 ml of barium carrier (20 mg $Ba^{2+}\cdot ml^{-1}$), (b) drops of methyl red, (c) 5 ml of citric acid and (d) NH_4OH slowly until a pH between 4.5 a 5.0 was obtained.[7]

The solution obtained was heated to boiling point, then 3M H_2SO_4 was added while agitating. The precipitate formed was dissolved in a solution of EDTA and NH_4OH, being heated until totally dissolved. After this stage, the solution was transferred to a glass container (test tube) and the radon (^{222}Rn) residue was extracted by passing old compressed air through the solution. Then the test tube was sealed so as to start the increase in ^{222}Rn.[7] After allowing sufficient time to obtain an activity higher than 75% of the equilibrium activity, the ^{222}Rn produced by the decay of the ^{226}Ra was extracted from the test tube and stored in Lucas cell. At this stage, the ^{226}Ra reached equilibrium with its daughters, and then a total alpha-counting was carried out.[8]

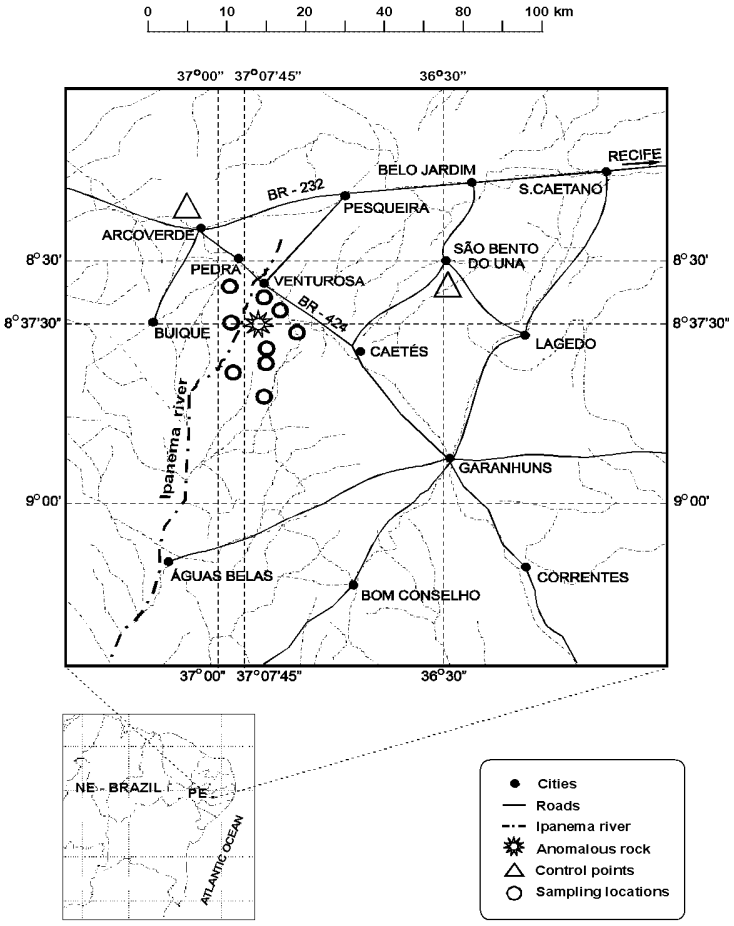

Fig. 1. Study area showing the sampling locations and anomalous rock

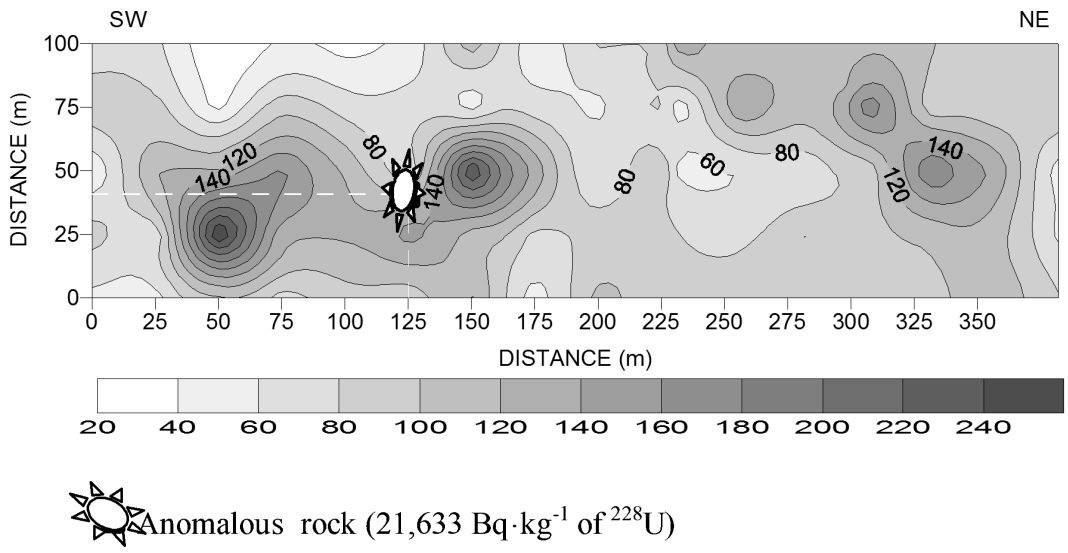

Fig. 2. Specific activities of ^{238}U in the soil around anomalous rock[3]

The alpha-counting for ^{222}Rn and descendants was carried out by coupling a Lucas cell to the photocathode of a photomultiplier, connected to a conventional modular electronic system (preamplifier, amplifier, discriminator and a counter with a timer).

The Lucas cells were calibrated using a standard solution of ^{226}Ra supplied by the Institute of Radioprotection Dosimetry (IRD, Brazil). The Environmental Monitoring Laboratory of the Nuclear Energy Department of the Federal University of Pernambuco participate of the National Intercomparison Program, whose objective is to evaluate the accuracy of the determination of ^{226}Ra by the method used in this work.

Through internal intercomparisons, the reliability of the method used was assessed for determining the concentration of ^{226}Ra. Standard samples were prepared, using a certified material provided by IRD. In this case, 10 g of ashes from four samples with known concentration values of ^{226}Ra were taken, adding 21 mBq of standard solution activity of ^{226}Ra, with the objective of determining the efficiency for the standard concentration during the normal analyses procedures. The obtained results showed an average efficiency of 80% of standard concentration in the method.

Risk of induction of cranium carcinoma and bone sarcoma

When ingested by human being, the ^{226}Ra is accumulated in bones in a similar way to calcium. The damage caused by the assimilation of this radionuclide by human beings is the occurrence of cancer, where the most common types are bone cancer and carcinoma of the skull.[9]

To calculate the cranium carcinoma incidence rate resulting from ^{226}Ra presence in blood, ROWLAND et al.[10] considered a latent minimum period of 10 years. According to these authors, the curve that best describes the risk for induction of cranium carcinoma is given by:

$$E_{CC} = 4.32 \cdot 10^{-10} \cdot A_{Blood} \quad (1)$$

where E_{CC} is the excess of annual cranium carcinoma incidence; and A_{Blood} is the accumulated ^{226}Ra activity (Bq) in the blood during a lifetime.

For determining the bone sarcoma incidence rate, ROWLAND et al.[10] verified that Eq. (2) is the one that best describes the risk, considering a minimum latent period of 5 years:

$$E_{BS} = 2.7 \cdot 10^{-10} \cdot A_{Blood} \quad (2)$$

where E_{BS} is the excess of annual bone sarcoma incidence; and A_{Blood} is the accumulated ^{226}Ra activity (Bq) in the blood during lifetime.

Results and discussion

^{226}Ra concentration in milk

In Table 1 are presented the values of ^{226}Ra concentration in cow's milk from the studied region. In the calculation of the standard deviation of the concentrations, a 5% error was adopted for the radiochemical analysis of the samples, in accordance with the National Intercomparison Program of IRD.[11] It follows from data in Table 1 that there is an elevated variability in the results obtained. Therefore, discrepant values can be interpreted as anomalous values.

Any numeric value with a position in a box-plot above interquarterly intervals of 1.5 can constitute an anomalous value.[12] Figure 3 shows a normal qq-plot of ^{226}Ra concentrations in milk, which indicates the existence of anomalous values, very distant from the others, i.e., they deviate from the distribution of the other values. The interquarterly parameters obtained from the box-plot graphic analysis are given in Table 2.

Analyzing Table 2, one finds: $1.5A_4 = 1.5 \times 36 = 54$ mBq·l^{-1}, which provides, for close values, the interval $m_e - 1.5A_4 < x < m_e + 1.5A_4$, including all the group values. Therefore: $14.5 - 54 < x < 14.5 + 54$. Any result of ^{226}Ra concentration (mBq·l^{-1}) in milk that does not belong to the interval [–39.5; 68.5] is an anomalous value. Thus, the concentrations 82 and 500 mBq·l^{-1} are considered anomalous values.

Table 1. Concentration of ^{226}Ra in the milk samples

Farm code	Latitude	Longitude	Altitude, m	Year of collection	Concentration, mBq·l^{-1}
F-1	8º 49' 16"	37º 00' 24"	482	2002	11 ± 1*
F-1	8º 49' 16"	37º 00' 24"	482	2003	5 ± 1
F-2	8º 48' 30"	37º 00' 51"	474	2003	4 ± 1
F-2	8º 48' 30"	37º 00' 51"	474	2004	500 ± 25
F-3	8º 55' 9"	37º 00' 59"	461	2003	13 ± 1
F-4	8º 49' 9"	37º 00' 42"	494	2003	10 ± 1
F-5	8º 44' 16"	37º 00' 41"	468	2004	16 ± 1
F-6	8º 2' 14"	37º 00' 32"	486	2003	40 ± 2
F-6	8º 2' 14"	37º 00' 32"	486	2004	22 ± 1
F-7	8º 39' 10"	37º 00' 19"	469	2004	25 ± 1
F-8	8º 39' 20"	37º 00' 18"	472	2004	6 ± 1
F-9	8º 53' 39"	37º 00' 44"	487	2004	5 ± 1
F-10	8º 27' 14"	37º 10' 21"	682	2002	30 ± 2
F-10	8º 27' 14"	37º 10' 21"	682	2003	43 ± 2
F-10	8º 27' 14"	37º 10' 21"	682	2004	56 ± 3
F-11	8º 29' 30"	36º 43' 9"	582	2002	10 ± 1
F-11	8º 29' 30"	36º 43' 9"	582	2003	4 ± 1
F-11	8º 29' 30"	36º 43' 9"	582	2004	82 ± 4

* Standard deviation (95% confidence).

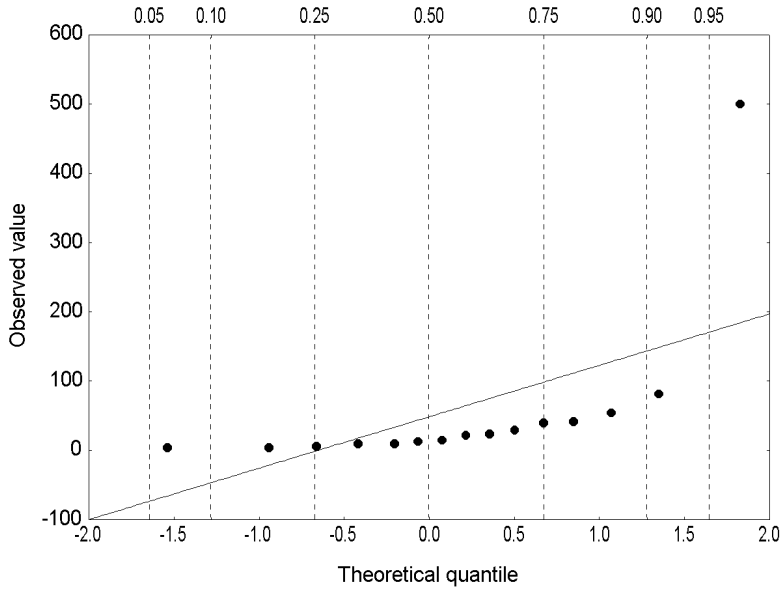

Fig. 3. Normal qq-plot of ^{226}Ra concentrations in milk

Table 2. Interquarterly parameters obtained from the box-plot graphic analysis

First quantile ($q_{1/4}$), mBq·l^{-1}	Median (m_e), mBq·l^{-1}	Third quantile ($q_{3/4}$), mBq·l^{-1}	Interquarterly amplitude (A_4), mBq·l^{-1}	Interquarterly interval, mBq·l^{-1}
6	14.5	40	36	[–39.5; 68.5]

Estimate of daily ^{226}Ra ingestion

Anomalous results of ^{226}Ra in milk are very significant from the point of view of public health. The daily milk ingestion by the residents of the Pedra and Venturosa cities is 0.5 l. Specifically considering the anomalous result of 500 mBq·l^{-1}, 250 mBq could be the daily ingested activity. Based on this, an estimate of the cranium carcinoma induction risk and bone sarcoma risk to the population of the delimited region for study was made.

Estimate of the cranium carcinoma risk

Allowing for a life expectancy of the 68.6 years for the population of the region studied,[13] only the transference of ^{226}Ra during 58.6 years is considered effective for inducing cranium carcinoma, considering a minimum latent time of 10 years for the appearance of these kinds of tumors.[10]

Taking the daily ingestion value of 250 mBq and the period of 58.6 years, $5.3\cdot10^6$ mBq is obtained,[13] which is the ^{226}Ra activity ingested during all the lifetime. According to MAYS et al.,[9] 20% of the total ^{226}Ra ingested by man is transferred to the blood. Therefore, 20% of $5.3\cdot10^6$ mBq is equal to $1.1\cdot10^6$ mBq. Transforming this value to Bq, we have $1.1\cdot10^3$ Bq as a value to be introduced into Eq. (1), given as result $5\cdot10^{-7}$. Therefore, the accumulated risk up to end of life will be $5\cdot10^{-7}$/year. Based on the accumulated risk value, the number of cases of carcinoma in one million people was $(5\cdot10^{-7}/\text{year})\times10^6 = 0.5/\text{year}$. In contrast, the normal risk estimate is 375 cases of carcinoma expected for each 10^6 persons per year.[14]

Estimate of bone sarcoma induction risk

In the determination of bone sarcoma accumulated risk, the total ^{226}Ra activity transferred to the blood in the period of 63.6 years, for a latent time of 5 years,[10] was $1.2\cdot10^3$ Bq. Substituting this value into Eq. (2), the risk $3\cdot10^{-7}$/year was obtained. Based on the accumulated risk value, the number of bone sarcoma cases in million people was 0.3/year. In contrast, the normal risk estimate is 750 cases of sarcoma for each 10^6 persons per year.[14]

Conclusions

According to the results obtained for the ^{226}Ra concentrations in the cow's milk consumed in Pedra and Venturosa cities, the highest anomalous value showed that there are no sufficient indications of elevation of risks for cranium carcinoma and bone sarcoma in the residents, although it was considered to be a systematic ingestion of ^{226}Ra from milk.

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