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Determination of ^{226}Ra in cheese produced in the rural area of Pernambuco-Brazil

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Abstract This work is aimed at determining ^{226}Ra concentration in cheese produced by dairy manufacturers from Pedra and Venturosa, which are two districts in the rural region of Pernambuco-Brazil. Analyses showed concentrations of ^{226}Ra in cheese samples varying from 54 to 2,080 mBq kg^{-1} (wet matter). Using the cumulated risk method, the excess carcinoma and bone sarcoma were estimated for chronic ingestion of cheese containing this radioisotope. In this context, this report discusses the methodology of sample analyses as well as possible consequences of the ^{226}Ra intake for human health.

Keywords Radium ingestion · Cancer risk · Anomalous radioactivity

In 1975, during the Garanhuns Project, the Brazilian Nuclear Enterprise (NUCLEBRAS), in partnership with a Mineral Resource and Research Company (CPRM), carried out a radiogeological survey over 35,000 km^2 area involving the states of Pernambuco, Alagoas and Sergipe (Northeast from Brazil). Radioactive anomalies were detected on a farm located 8.7 km far from Venturosa (rural region of Pernambuco-Brazil). In this local rolled blocks of mafic rock were found, which presented a high percentage of U_3O_8 (Costa et al. 1976). At this place, the uranium mineral can be seen with the naked eye (emerging), as a greenish, yellow colored material that fills existent fractures in the blocks of rock. Although this project identified 263 points of radioactive anomalies, only one of them (Venturosa-Pernambuco) presented significant U_3O_8 content, where after some analyses of existing rocks revealed a maximum concentration 22,000 mg kg^{-1} (2.2%) of this compound (Costa et al. 1976). Ra-226, daughter of ^{238}U, is an alpha emitter with a half-life of 1,620 years, being produced by weathering of rocks rich in uranium and enters the soil and water from where it is transferred to plants, animals and eventually to humans (McDowell-Boyer et al. 1980). In the farms in Venturosa, as at most of the cities in the rural region of Pernambuco, the principal cultivated plants for feeding dairy cows are forage palm (*Opuntia* spp.), buffel (*Cenchrus ciliares*) and elephant grass (*Pennisetum purpureum*), and forage sorghum (*Sorghum bicolor*) (SEBRAE 2002). Analyses of ^{226}Ra concentration from samples of those plants showed concentrations varying from 96 to 7,990 mBq kg^{-1} in dry matter (Silva et al. 2004). Part of the ^{226}Ra ingested from the forage by a dairy cow is transferred to the milk (Watson et al. 1984). For dairy cattle raising and milk producing rural region of Pernambuco, the extensive and semi-intensive management systems practiced by 75% of the producers is predominant, 50% of the native pasture is the principal source of feeding (SEBRAE 2002). At the studied region, one would observe that 10 L of milk are employed to produce 1 kg of cheese (SEBRAE 2002). As a result of this, cheese consumption may represent an import via of internal contamination with ^{226}Ra. In terms of radiation protection, human intake of ^{226}Ra contaminated foodstuffs has usually being converted into dose to bone and soft tissues in order to estimate the risk of cancer in the population internally exposed to this radioisotope (Mays et al. 1985). After ingestion by the human being, ^{226}Ra is deposited in bones, in a similar way to calcium. The damage resulted of the assimilation of this radionuclide by human beings is the occurrence of cancer, where the most common types are

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bone cancer and carcinoma of the skull (Mays et al. 1985). In this context, the aim of this work was to determine the concentration of this radioisotope in the cheese produced in Venturosa municipality. The overall aim of this work was to carry out epidemiological studies into the level of cancer in a population potentially exposed to an internal contamination of ^{226}Ra, resulting from the systematic ingestion of cheese containing this radionuclide.

Materials and Methods

Ten samples of cheese were collected from three principal dairy good manufacturers in the Venturosa municipality, which are located near the mineralization of uranium. The selection criteria for choosing those manufacturers were the quantity of cheese produced and their proximity of the occurrence of uranium. The dairy industries chosen in the present study are the biggest producers of cheese in the studied region. Several farms nearby supply the three manufacturers of dairy products investigated in this research. All farms are located on the mineralization area of uranium. The samples were collected in May 2004. From each farm, 2 kg of cheese was collected in polyethylene containers. Dry matter was obtained after heating to 80°C, during 48 h. After that, the material was taken to the furnace, and the temperature was raised gradually up to 450°C, and then left for 48 h at this temperate until ashes were obtained. Following this step, 10 g of ashes were taken for digestion in concentrated nitric acid, left to evaporate and afterwards reheated to 450°C for 30 min. After cooling, concentrated nitric acid was added again and heated up to the boiling point. After cooling, the solution obtained was filtered and used for the determination of ^{226}Ra. For the determination of ^{226}Ra, 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 2 L glass beaker, then it was added: (a) 1 mL of barium carrier, (b) drops of methyl red, (c) 5 mL of citric acid and (d) NH_4OH slowly until reaching a pH between 4.5 and 5.0 (Godoy 1990). This solution is heated to boiling point, then H_2SO_4 (3 M) is added while agitating. The precipitate formed was dissolved in a solution of EDTA and NH_4OH, and heated until completely dissolved. Following this stage, the solution was transferred to a glass container (test tube) and the radon (^{222}Rn) residue was extracted by passing the old compressed air through the solution. Thus, test tubes are sealed so as to start the increase in ^{222}Rn (IRD 1983). After allowing sufficient elapse time to obtain an activity higher than 75% of the secular equilibrium, when ^{222}Rn produced by the decay of the ^{226}Ra is extracted from the test tube and stored in Lucas cell, where ^{226}Ra reaches secular equilibrium with its daughters, and then a gross alpha counting is performed (Goldin 1961). Alpha counting for ^{222}Rn and descendants have been 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). This technique has a detection limit of about 2.4 mBq L^{-1}. The concentrations of ^{226}Ra were obtained by the Eq. 1 (Godoy 1990).

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

Dairy manufacturer (DM) code	Concentration (mBq kg^{-1} wet matter) ($\overline{X} \pm S$)*
DM-1	62 ± 3
DM-1	58 ± 3
DM-1	56 ± 2
DM-1	329 ± 16
DM-2	90 ± 5
DM-2	223 ± 11
DM-2	54 ± 3
DM-3	63 ± 3
DM-3	65 ± 3
DM-3	2,080 ± 104

* Standard deviation (95% of confidence)

$$A_{^{226}Ra} = \frac{C_A - C_B}{\varepsilon V(1 - e^{-\lambda_1 t_1}) \times (e^{-\lambda_2 t_2})} \quad (1)$$

where

C_A = rate of alpha counting in the sample (cpm);

C_B = rate of background counting observed in Lucas cell (cpm);

ε = efficiency of Lucas cell counting (cpm/pCi);

V = volume of solution used in determination of ^{226}Ra (L);

λ_1 = decay constant of ^{222}Rn ($days^{-1}$);

λ_2 = decay constant of ^{222}Rn (h^{-1});

$\frac{1}{1-e^{-\lambda_1 t_1}}$ = correction factor for decay of ^{222}Rn in time t_1;

t_1 = time interval for the increase of the ^{222}Rn in test tube (days);

$\frac{1}{e^{-\lambda_2 t_2}}$ = correction factor for decay of ^{222}Rn in time t_2;

t_2 = time interval between emanation and start of counting (h).

Results and Discussion

Table 1 presents values of ^{226}Ra concentration in cheese samples from the investigated region. In the calculation of the standard deviation of the concentrations an error of 5% was adopted for the radiochemical analyses of the samples, in accordance with the National Intercomparison Program of the Institute of Radioprotection and Dosimeter (IRD 1983).

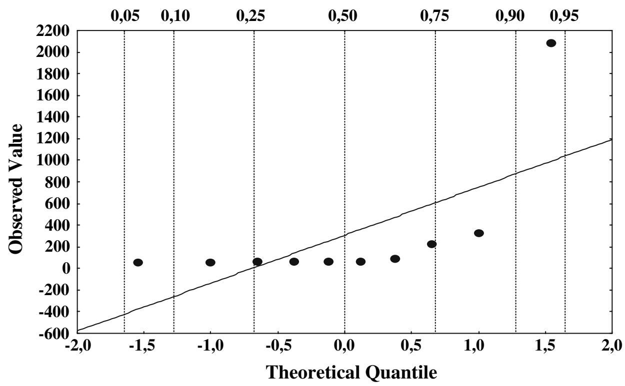

Fig. 1 Box-plot graphic of ^{226}Ra concentrations in cheese

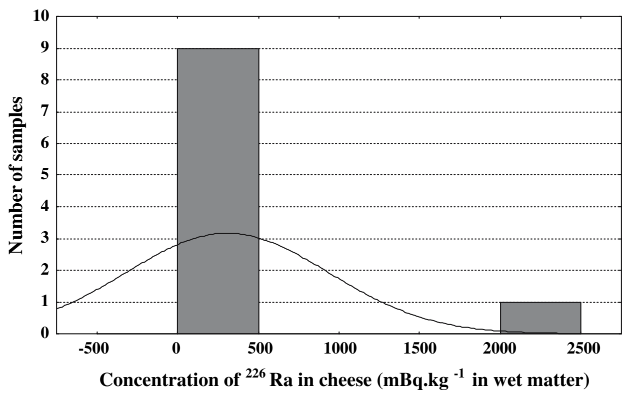

Fig. 2 Distribution of ^{226}Ra concentrations in cheese samples

It follows from Table 1 that there is an elevated variability in the results obtained. As dairy good manufacturers are supplied by several farms, in an intermittent process of milk delivery, the great variability of data obtained may be explained by considering that milk coming from different farms is employed at the same cheese process of production. Therefore, discrepant values can be interpreted as anomalous (Barnet and Lewis 1996). Any numeric value with a position in a box-plot above interquartile intervals of 1.5 can constitute an anomalous value (S-Plus 1997). Figure 1 shows a box-plot graphic of ^{226}Ra concentrations in cheese, which indicates the existence of anomalous values, very distant from the others. Thus, the concentrations 329 and 2,080 mBq L^{-1} were considered outliers. As a result, the distribution of contaminants, in typically anomalous locals, has an elevated skewness, which is caused by the outlier effects (Singh et al. 1997).

Figure 2 shows the elevated asymmetry of the set distribution presented in Table 1, induced by outliers' effects.

This type of distribution suggests that data obtained from Table 1 come from multiple populations. Indeed, according Singh et al. (1997), environmental data are a result of the mixture of several subpopulations. As seen in the Table 1, there are some ^{226}Ra anomalous concentrations, which are very significant in terms of public health. The daily cheese ingestion by the residents of the Venturosa city is about 0.3 kg. Thus, inhabitants of Venturosa are particularly interesting for studies involving radium-226 ingestion. When one considers the anomalous result of 2,080 mBq kg^{-1} (wet matter), the relation with the per capita amount of cheese consumed leads to 624 mBq ingested per day. Based on this result, an estimate of the cranium carcinoma induction risk and bone sarcoma risk to the population of the delimited region can be predicted. When ingested by the human being, ^{226}Ra is accumulated in bones in a similar way to calcium. The damage induced 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 (Mays et al. 1985). According to Mays et al. (1985), 20% of the total ^{226}Ra ingested by man is transferred to blood. To calculate the cranium carcinoma incidence rate resulting from ^{226}Ra presence in blood, Rowland et al. (1978) considered a latent minimum period of 10 years. According to their studies, the curve that best describes the risk for induction of cranium carcinoma is given by Eq. 2.

$$E_{CC} = 4.32 \times 10^{-10} \times A_{BLOOD} \quad (2)$$

where

E_{CC} = excess of annual cranium carcinoma incidence;

A_{BLOOD} = accumulated activity (in Bq) of ^{226}Ra in the blood during a lifetime.

For determining the bone sarcoma incidence rate, Rowland et al. (1978) verified that Eq. 3 is the one that best describes the risk, considering a minimum latent period of 5 years.

$$E_{BS} = 2.7 \times 10^{-10} \times A_{BLOOD} \quad (3)$$

where

E_{BS} = excess of annual bone sarcoma incidence;

A_{BLOOD} = accumulated activity in Bq of ^{226}Ra in the blood during lifetime.

On the basis of a life expectancy of the 68.6 years for the population of the region studied (IBGE 2003), only the transference of ^{226}Ra during 58.6 years is considered effective for inducing cranium carcinoma, considering a minimum of 10 years latency, for appearance of these kinds of tumors (Rowland et al. 1978).

Using the accumulated risk value, the number of cases of carcinoma in one million people per year is 0.02 per year. In contrast, the normal risk estimate is 375 cases of carcinoma are expected per 10^6 persons per year (FUSAM 2002). In the determination of bone sarcoma accumulated risk, the total ^{226}Ra activity transferred to blood in the period of 63.6 years is about 3.0×10^3 Bq, for 5 years latency (Rowland et al. 1978). Also using the accumulated risk value, the number of bone sarcoma cases in million people is 0.02 per year. In contrast, the normal risk estimate is 750 cases of sarcoma are expected per 10^6 persons per year (FUSAM 2002). This research presents the preliminary results obtained for evaluating ^{226}Ra concentrations from samples of cheese produced in the Venturosa municipality. Although anomalous concentrations of ^{226}Ra found in this study, the results of risk estimations indicate that the probability of appearance of cranium carcinoma and bone sarcoma in the residents, as a result of systematic ingestion of ^{226}Ra from cheese, is totally negligible.

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