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Radium-226 in Sugar Cane, *Saccharum officinarum*, Products in the State of Pernambuco, Brazil

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Abstract The objective of this study was to determine the ^{226}Ra concentrations in samples of sugar cane juice, which is sold for human consumption in the city of Recife, capital of the state of Pernambuco, Brazil. The sample collections were carried out in seventeen commercial establishments where high quantities of the juice are usually sold. The methodology used to determine the concentrations of ^{226}Ra in the juice was based on ^{222}Rn emanation classical technique. Concentrations of ^{226}Ra in the samples varied from 18 to 89 mBqL^{-1}.

Keywords Sugar cane · ^{226}Ra · Risk of cancer

Radioecological studies have been conducted in different parts of the world to evaluate the transference of ^{226}Ra in the soil-plant system and also to study the presence of this radioelement in foodstuffs for human beings (Amaral et al. 1988; Bettencourt et al. 1988, Lima and Penna-Franca 1988). In Pernambuco, Brazil, sugar cane (*Saccharum officinarum*) is predominantly cultivated in the humid coastal region (known as Zona da Mata). Since the majority of the soils in this region are well developed (Ultisols and Oxisols) and, therefore, present low phosphorus availability, there is intensive use of phosphate fertilizers for the cultivation of sugar cane. As a result, the amount of ^{226}Ra in the soils has been increasing. A great amount of the sugar cane cultivated in Pernambuco is destined to the production of alcohol and sugar. However, part of the sugar cane is also used for the manufacture of food products, such as the "rapadura" (an edible brazilian toffee) and also for obtaining the "garapa" (sugar cane juice) popularly known as "caldo de cana". The juice is consumed in natura by the local population in all of the state, mainly in the metropolitan region of Recife, the capital of Pernambuco. It is possible that the ingestion of sugar cane juice could constitute an important pathway of ^{226}Ra contamination in the food chain of the population. Once incorporated into the human body the radium is metabolized in a similar way to calcium. The main possible harm to the human health, as a result of the ^{226}Ra assimilation, is the occurrence of cancer, particularly bone sarcoma and carcinoma of the cranium. In this context, this research was conducted to with the objectives of determining the ^{226}Ra concentrations in samples of sugar cane juice sold in strategic commercial localities in the city of Recife and to discuss the possible consequences of the ^{226}Ra intake to the consumers of the juice.

Materials and Methods

Sugar cane juice sampling was carried out in seventeen commercial establishments that usually sell high amounts of juice. For this, five liters of juice were collected from each establishment in December 2004. The samples were collected in polyethylene containers, where 10 mL of 37% formaldehyde were immediately added to preserve the juice. The samples were then heated at 80°C during 48 h in order to obtain dry material. After that, the material was taken to the furnace, the temperature was raised gradually up to 450°C, and then left for 48 h at this temperature until ashes were obtained. After these procedures, 10-g

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sub-samples of the ashes were taken for digestion in concentrated nitric acid (65%), left to evaporate and afterwards reheated to 450°C for 30 min. After cooling, 20 mL of concentrated nitric acid was added again to each sample and heated to the boiling point. After cooling, the solution obtained was filtered and used for the determination of ^{226}Ra (IRD 1983).

For the determination of ^{226}Ra in the samples, 1 liter of solution was prepared from the solution formed by the digestion of the ashes. In this case, the quantity of 1 L was transferred to a 2-liter glass beaker, followed by the addition of: (a) 1 mL of barium carrier (20 mg Ba^{+2} 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 (IRD 1983). After this, the solution obtained was heated up to the boiling point, kept under stirring and then H_2SO_4 3M was added. The precipitate formed was dissolved in a solution of EDTA and NH_4OH and heated until it had been 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 through the solution aged compressed air, which was compressed for a period longer than 30 days, in order to guarantee the decay of any ^{222}Rn present in the air. Then the test tube was sealed so as to start the increase in ^{222}Rn (IRD 1983). After allowing sufficient time to elapse 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 a Lucas cell (Goldin 1961). At this stage, the ^{226}Ra reached equilibrium with its daughters, and then a total alpha counting was carried out. 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). The Environmental Monitoring Laboratory of the Nuclear Energy Department of the Federal University of Pernambuco is a member of the National Intercomparison Program, whose objective is to evaluate the accuracy of the determination of concentrations of ^{226}Ra using the method described in the present study. By internal intercomparisons, an evaluation of the reliability of the method of analysis used was assessed for determining the concentration of ^{226}Ra. Standard samples were prepared, using certified material provided by IRD. In this case, 10 g of ashes from four samples were taken with known concentration values of ^{226}Ra, adding 21 mBq of standard solution activity of ^{226}Ra with the objective of determining the efficiency of the standard concentration during the normal analyses procedures. The results obtained showed an average efficiency of 80% of standard concentration in the method. This technique has a detection limit of about 2.4 $mBq.L^{-1}$. The concentration of ^{226}Ra was determinated using the Eq. 1 (Godoy 1990).

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

where C_A = rate of alpha counting in the sample (cpm); C_B = rate of Background counting observed in the Lucas cell (cpm); ε = efficiency of Lucas cell counting (cpm/pCi); V = volume of solution used in the determination of ^{226}Ra (L); λ_1 = decay Constant of ^{222}Rn (days); λ_2 = decay constant of ^{222}Rn (hours);

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

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

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

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

Results and Discussion

The values of ^{226}Ra concentration in the sugar cane juice from the metropolitan region of Recife are presented in Table 1. 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 the Institute of Radioprotection and Dosimetry (IRD 1983). The median of the values presented in Table 1 was 45 $mBq.L^{-1}$. The interquarterly parameters are shown in Fig. 1.

The average daily sugar cane juice ingestion per person in the metropolitan region of Recife is 0.3 L (IBGE 2003). Considering the result of 89 $mBq.L^{-1}$, 26.7 mBq could be the daily ingested activity derived from ^{226}Ra. Based on this, an estimate of the head carcinoma induction risk and bone sarcoma risk to the population of the study region for was calculated. Therefore, based on the life expectancy of the 68.6 years for the population of the study region (IBGE 2003), the transference of ^{226}Ra during 58.6 years would be sufficient to induce head carcinoma, allowing a minimum latent time of 10 years for the appearance of this kind of tumor (Rowland et al. 1978). On the other hand, for the daily ingestion of 26.7 mBq and a period of 58.6 years, the activity ingested, due to ^{226}Ra during a lifetime would be approximately 5.7×10^5 mBq. According to Mays et al. (1985), 20% of the total ^{226}Ra ingested by humans is transferred to the blood. Therefore, 20% of 5.7×10^5 mBq is equal to 1.1×10^5 mBq. Transforming this value to Bq,

Table 1 Concentration of radium in samples of sugar cane juice collected in commercial establishments of Recife, Brazil

Sampling localities	Date of collection	Concentration ($mBq.L^{-1}$)
Cidade Universitária	15-12-04	25 ± 3[a]
Torrões	16-12-04	40 ± 5
Várzea	17-12-04	49 ± 6
Engenho do Meio	17-12-04	56 ± 8
Boa Vista	20-12-04	38 ± 3
Avenida Caxangá	21-12-04	35 ± 4
Avenida Caxangá	23-12-04	33 ± 7
Avenida Caxangá	23-12-04	89 ± 11
Várzea	23-12-04	53 ± 9
Várzea	23-12-04	18 ± 2
Cidade Universitária	27-12-04	78 ± 12
Várzea	27-12-04	67 ± 7
Várzea	27-12-04	26 ± 3
Centro do Recife	27-12-04	45 ± 4
Centro do Recife	27-12-04	44 ± 8
Centro do Recife	27-12-04	75 ± 12
Centro do Recife	27-12-04	63 ± 10

[a] Standard deviation (95% of confidence)

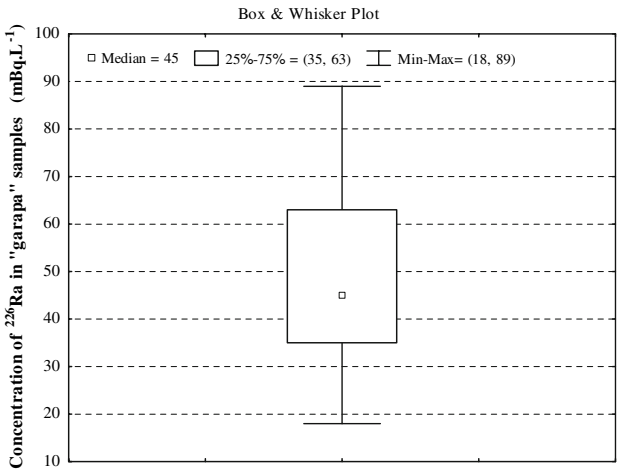

Fig. 1 Box-plot graphic of ^{226}Radium concentrations in the samples of sugar cane juice

this is equivalent to 1.1×10^2 Bq. If this value is substituted in equation 1, it results in 4.8×10^{-8}. Therefore, the accumulated risk, up to the end of the human life will be 4.8×10^{-8}/year. Based on the accumulated risk value, the estimated number of cases of carcinoma was 0.05/year per one million people. In contrast, the normal risk estimate is 375 cases of carcinoma per 10^6 persons per year (FUSAM 2002). 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 (Mays et al. 1985), was 1.2×10^2 Bq. Based on this, the risk obtained was 3.3×10^{-8}/year (Mays et al. 1985). In this case, based on the accumulated risk value, the number of estimated bone sarcoma cases was 0.03/year per one million people. In contrast, the expected normal risk estimate is 750 cases of sarcoma per 10^6 persons per year (FUSAM 2002). Therefore, it is concluded that the concentrations of ^{226}Ra in the samples of sugar cane juice consumed by the population of the metropolitan region Recife, correspond to unsubstantial risk levels in terms of appearance of head carcinoma and bone sarcoma.

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References

Amaral ECS, Carvalho ZL, Godoy JM (1988) Transfer of 226Ra and 210Pb to forage and milk in brazilian high natural radioactivity region. Radiat Prot Dosim 24:119–121

Bettencourt AO, Teixeira MMGR, Elias MDT, Faisca MC (1988) Soil to plant transfer of radium-226. J Environ Radioact 6:49–60. doi: 10.1016/0265-931X(88)90067-7

FUSAM (2002) Boletim Informativo das Doenças-Crônico-Degenerativas. Recife-PE

Godoy JM (1990) The environmental behaviour of radium. Technical Report Series 310. IAEA, Vienna

Goldin AS (1961) Determination of dissolved radium. Anal Chem 33:406

IBGE (2003) Estatísticas do Século XX. Comunicação Social

IRD (1983) Manual de procedimentos técnicos do Departamento de Proteção Radiológica Ambiental. CNEN, Rio de Janeiro

Lima VT, Penna-Franca E (1988) Uptake of endogenous and exogenous ^{226}Ra by vegetables from soils of a highly radioactive region. Radiat Prot Dosim 24:123–126

Mays CW, Rowland RE, Stehney AF (1985) Cancer risk from the lifetime intake of Ra and U isotopes. Health Phys 48:635–647. doi:10.1097/00004032-198505000-00005

Rowland RE, Stehney AF, Brues AM, Littman MS, Keane AT, Patten BC, Shanahan MM (1978) Current status of the study of ^{226}Ra and ^{228}Ra in human at the center for human radiobiology. Health Phys 35:159–166