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Intake of uranium and radium-226 due to food crops consumption in the phosphate region of Pernambuco – Brazil

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Abstract

The phosphate region located in the Northeast of Brazil covers an area of approximately 150 km long with an average width of 4 km, along the coast of the states of Pernambuco and Paraíba. The inhabitants of this area are exposed to natural radioactivity levels higher than the background values recorded in the literature, mainly due to the presence of uranium and its decay products in the phosphatic sediments. The main aim of this study was to determine the activity concentration of uranium and ^{226}Ra in foodstuffs cultivated in this area, where the phosphate mineral has been extracted. The activity concentrations found for uranium and

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^{226}Ra in the foodstuffs analyzed varied from 13 to 186 mBq kg^{-1} (wet weight), with a mean value of 46 mBq kg^{-1} and from 43 to 2209 mBq kg^{-1} (wet weight), with a mean value of 358 mBq kg^{-1}, respectively. The annual intake of these radionuclides, for rural residents, was 7.45 Bq for uranium and 69.3 Bq for ^{226}Ra.

Keywords: Phosphate; Uranium; ^{226}Ra; Food; Crops; Intake

1. Introduction

According to Gonzales and Anderer (1989), natural radioactivity is responsible for approximately 70% of the dose received by the population due to all sources (natural and artificial). The most important terrestrial sources of natural background radiation are ^{40}K and the ^{238}U and ^{232}Th decay series. These radionuclides are mainly responsible for internal exposure, through ingestion of food and water, and through inhalation of air particulates. Of special concern are the long-lived alpha emitters of the uranium decay chain.

Knowledge of radioactivity levels in human diet is of particular concern for the estimation of possible radiological hazards to human health. However, very few surveys of radioactivity in foods have been conducted in Brazil. The few data available were mainly carried out in regions of high levels of natural radioactivity. One of these regions, located in the Northeast of Brazil, is known for its urano-phosphate deposits. The mineralized area covers an extension of approximately 150 km in length with an average width of 4 km in the coastal region of the states of Pernambuco and Paraíba, with natural uranium (U_{nat}) contents ranging from 10 to 530 ppm (Amaral, 1994). The area is occupied by houses, industries, and small farms, being fairly populated. Foodstuffs cultivated in the area include fruits (bananas, mangoes and cashews), tubers and root vegetables (sweet potatoes, yam and cassava), and grains (brown beans and corn).

This sedimentary phosphate deposit was first observed in 1949, in the search of calcareous layers used for the analysis of mineral water. In 1952, a group of the "Departamento Nacional de Produção Mineral – DNPM" started a project aiming to study the area, in order to have more information about the extension of the phosphate deposit and its natural uranium content (Almeida, 1978). It is known that phosphatic rocks of sedimentary origin contain small amounts of uranium, and their content is usually dependent upon the phosphate concentration in the rock (Saad, 1974). Thus, characterizing the zones mineralized with phosphorus, automatically characterizes the zones where uranium occurs. According to Almeida (1978), the phosphatic sedimentary basin along the coast of the State of Pernambuco and State of Paraíba, originated from organic fossil remains accumulated in the basal zone, with a long period of sedimentation. The thickness and lithologic composition of these basal layers are directly dependent upon its calcareous and clay content.

Fisenne and Keller (1969) measured the ^{226}Ra contents in the main components of the United States diet, which varied from 3.7 to 220 mBq kg^{-1} (0.1–0.6 pCi kg^{-1}) of wet weight. Wrenn (1977) also studied the ingestion of ^{226}Ra in the population of New York, Chicago, San Francisco, San Juan, United Kingdom, Bombay and Kerala. The ingestion average rate at those locations varied from 25.9 mBq day^{-1} in San Juan (Puerto Rico) to 103.6 mBq day^{-1} in Kerala (India). In most of the areas studied, radium ingestion was attributed to consumption of vegetables, grains and fruits, with the highest ingestion rate in Kerala and lower ingestion rate in the United Kingdom.

Cothern and Lappenbusch (1983) determined the uranium concentration in public water supplies of the United States. They measured approximately 22 000 samples, and concluded that the water is a major contributor for the internal doses due to the ingestion of uranium. Uranium is also found in foodstuff, since this radionuclide is present in all soils. The daily ingestion of uranium through foodstuff in the United States was found to range from 1.0 to 2.0 μg day^{-1} (Fisenne and Keller, 1969; Wrenn, 1977; Hamilton, 1972).

This study aims to determine the concentration of natural uranium and ^{226}Ra in food crops, which constitute the main diet of the people who live in the phosphate region of Pernambuco, and to evaluate the annual levels of ingestion of these radionuclides.

2. Material and methods

Due to the extension of the phosphate deposit in the Northeast region of Brazil (Fig. 1), it was decided to select an area where the phosphate mineral had been extracted, to start with, near the coast where the populational density is higher. The region selected, covering an area of 40 km^2, includes Igarassu city in the North and Paulista city in the South, and is delimited by the Atlantic Ocean to the East and the county of Abreu and Lima to the West (Fig. 2).

The foodstuffs were selected according to the local production and use, in specific seasons, from January 1991 up to September 1993 except the fruits, which are produced only once per year. The following samples were collected:

Grains – corn (*Zea mays* Hinm) and bean (*Phaselus vulgar* Hinm).
Tubers and roots – sweet potato (*Ipomoea batatas* Poir), cassava (*Manihot dulcis* Pax) and yam (*Dioscorea* sp).
Fruits – banana (*Musa sapientum* Linm), cashew (*Ancailiumaccidentale* Linm) and mango (*Mangifera indica* Linm).

The production of these foodstuffs is confined to the rainy season since the local producers lack financial support to build an artificial irrigation system. The foodstuffs were selected in accordance with the diet of the local population. A total of 40 samples were analyzed. For calculation of the annual "per capita" alimentary consumption, for different foodstuffs, the harvest time of each species in the delimited region was observed.

Fig. 1. Location of the phosphatic region of the Northeast of Brazil.

Also, soil samples were collected, from 28 different points inside of the delimited area, in layers of 0–40, 40–80 and 80–120 cm, to verify a possible variation in radionuclide concentrations of interest. The 120 cm depth is equivalent to the maximum root depth. Before being analyzed, the soil samples were dried outdoors, for 72 h, and sieved in 2 mm mesh, for thick and fine fractions separation. The uranium and ^{226}Ra determinations were realized from the fine fractions, using fluorimetry and gamma spectrometry methods, respectively.

The foodstuff samples were washed with water, as for human consumption, weighed and divided into small parts, dried in a stove at a temperature of 80 °C for 24 h and subsequently, calcinated at 500 °C, for a period of 96 h, to destroy the organic fraction. Once ashes were obtained, a 10 g aliquot was removed and this was dissolved in concentrated nitric acid (Godoy, 1983; IRD, 1983). The solution was used for the determination of ^{226}Ra concentration by the radon emanation technique (Rushing et al., 1964). Radium was first concentrated by co-precipitation with barium sulphate. The precipitate was then dissolved with EDTA ($C_{10}H_{14}N_2Na_2O_8 \cdot 2H_2O$) and the solution was transferred to a radon bubbler, de-emanated by passing compressed air through the bubbler, and stored to allow ingrowth of ^{222}Rn. The ^{222}Rn was then measured by using a scintillation cell coupled to the bubbler.

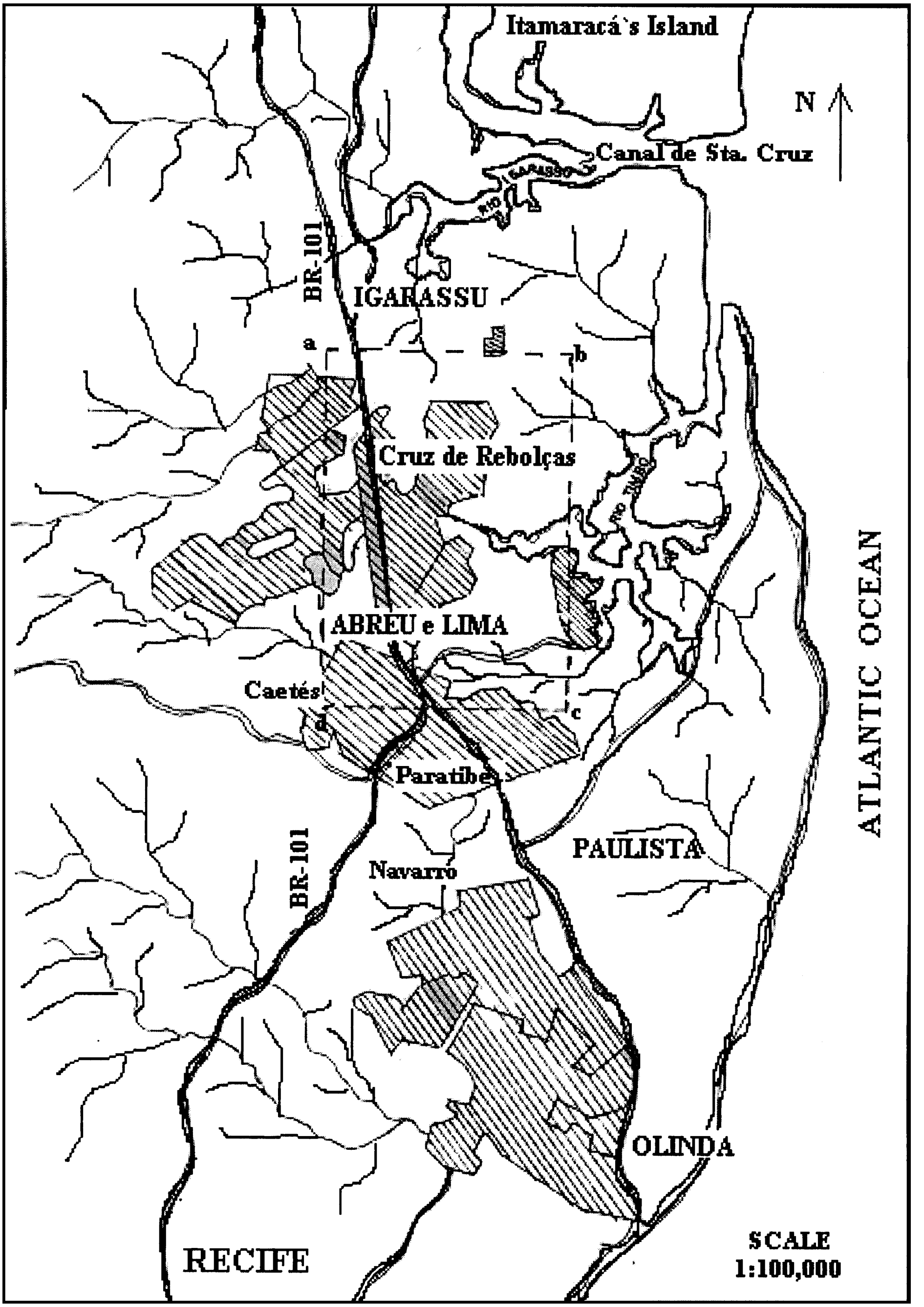

Fig. 2. Map of area selected for the study.

For the uranium determination, 2 g of ashes was dissolved in a combination $HNO_3/HclO_4/HF$ (*nitric-percloric-fluoridric acid*) (IRD, 1983). Then the solution was diluted up to total of 50 ml with distilled water. Aliquots of 20 ml of all solutions together with the standards, were then placed in extraction tubes.

The uranium content was determined by the fluorophotometric technique. This consists of the extraction of uranium with an organic extractor TOPO (tri-*n*-octyl-phosphine), followed by deposition on NaF/LiF disks, in 2% ratio of lithium fluoride and 98% of sodium-fluoride, weighing approximately 300 mg, previously prepared in platinum crucibles. This methodology leads to better results in terms of sensitivity and precision (around 6 times compared to the use of pure NaF) and also better compared to other existing methods (Centanni et al., 1956). The uranium-spiked disks were then fused at 1000 °C and cooled according to the procedure described by Price et al. (1953). An empty platinum crucible was used to determine the beginning of the scale (white) while another crucible containing LiF/NaF and 100 μl of a uraniun solution of 1 ppm was used for defining the high end of the scale (standard).

The uranium fluorescence was evaluated using fluorimeter (Jarrel-Ash Mod. 27-000) and related to the uranium content of the sample, by using calibration curves.

3. Results and discussion

In order to precisely evaluate the annual food consumption rate, the data published by the Instituto Brasileiro de Geografia e Estatística (IBGE, 1988) for the metropolitan area of Recife (this area includes the studied region) were compared with the information obtained directly from the rural population living in the area selected for the present study. This obtained information is presented in Table 1.

The activity concentrations of uranium and ^{226}Ra found in the analyzed foodstuffs are presented in Table 2. The results show that fruits present the highest uranium concentration, followed by grains, and roots and tubers. The highest concentration for ^{226}Ra, on the other hand, was found in roots and tubers, followed by grains and fruits. Among the species included in the group of fruits, cashew samples showed an unusually high uranium concentration, shifting the average concentration to a higher value.

The daily dietary intake of uranium and ^{226}Ra by the rural population of the phosphatic region of Pernambuco was obtained by taking into account the activity concentrations presented in Table 2 and the average annual consumption presented in the last column of Table 1. The considered foodstuff was produced and consumed by the local rural population. The daily ingestion found for the rural residents was 20.4 mBq for uranium and 190 mBq for ^{226}Ra. It should be stressed that these values do not correspond to the total dietary intake.

The values found for the annual ingestion of uranium and ^{226}Ra due to the consumption of foodstuffs cultivated in the phosphatic region of Pernambuco were 7.45 and 69.3 Bq year^{-1}, respectively.

Table 1
"Per capita" average and standard deviation annual consumption

Foodstuff groups	Foodstuffs	Metropolitan region[a] (kg $year^{-1}$)	Rural area[b] (kg $year^{-1}$)
Grains	Bean	12.8 ± 4.1	28.3 ± 5.1
	Corn	1.7 ± 0.8	34.0 ± 8.3
		27.8[d] ± 9.6	
Fruits	Mango	0.40 ± 0.03	17.4 ± 7.6
	Banana	14.5 ± 4.4	15.6 ± 6.5
	Cashew[c]	0.10 ± 0.06	1.6 ± 0.3
		47.2[d] ± 7.6	
Tubers and roots	Sweet potato	2.3 ± 0.7	21.2 ± 9.7
	Yam	5.3 ± 2.1	28.1 ± 9.4
	Cassava	1.1 ± 0.4	38.3 ± 10.1
		26.3[d] ± 6.3	

[a] IBGE, (1988).
[b] This work.
[c] In the evaluation of the amount of tuber and roots consumed per year the cassava flour was not taken into account.
[d] Total amount consumed per year for each group of foodstuff according to IBGE.

The uranium concentration in soil samples ranged from 15 to 300 Bq kg^{-1} of dry soil, with range factor of 20 between the different 28 points. The ^{226}Ra concentration ranged from 14 to 240 Bq kg^{-1} of dry soil, with range factor of 17. These results are according to those found in Planalto de Poços de Caldas (Amaral et al., 1988; Amaral, 1992).

Analyzing the results presented in Table 2 and the uranium and ^{226}Ra concentration values for soil samples, although the uranium concentrations were approximate to the ^{226}Ra concentrations, the ^{226}Ra absorption for the foodstuffs was

Table 2
Arithmetic mean and standard deviation and maximum activity concentration of uranium and ^{226}Ra

Food group	Concentration (mBq kg^{-1} of wet weight)			
	Arithmetic mean		Maximum concentration	
	Uranium	^{226}Ra	Uranium	^{226}Ra
Grains (7)	54 ± 10	483 ± 62	137	784
Bean (4)	81 ± 8	748 ± 61	137	784
Corn (3)	17 ± 6	130 ± 9	19	179
Fruits (13)	48 ± 10	129 ± 31	186	300
Mango (3)	41 ± 5	114 ± 7	53	149
Banana (7)	36 ± 6	157 ± 29	62	300
Cashew (3)	81 ± 7	76 ± 6	186	131
Tubers and roots (20)	36 ± 12	461 ± 62	88	2209
Sweet potato (6)	51 ± 8	672 ± 31	88	2209
Yam (5)	29 ± 5	367 ± 24	38	875
Cassava (9)	34 ± 7	409 ± 61	57	1607

Values in the parenthesis indicate the number of samples used for these results.

Table 3
Values reported in the literature for natural uranium and ^{226}Ra concentrations in different samples

Source	Concentration range (mBq kg^{-1} of wet weight)	
	Natural uranium	Radium-226
This work		
Bean	30.9–137	709–784
Corn	15.1–19.2	92.1–179
Tubers	12.9–87.7	90.8–2209
Fisenne et al., 1987	–	3.7–185
Vasconcellos et al., 1986		
Bean	–	163–840
Corn	–	70–229
Fisenne and Keller, 1969		
Bean	–	26–178
Fisenne and Keller, 1969[a]	60	56.8
Tracy et al., 1983		
Tubers[a]	15	181
Bean[a]	45	248
Corn[a]	12	70

[a] Average values.

greater. This could be due to small amount of uranium found in the soil which can be absorbed by the plants.

Table 3 shows, for comparison, other results obtained for the activity concentration of uranium and ^{226}Ra in foodstuff obtained throughout the world. These data show that ^{226}Ra concentrations in beans and corn found in the phosphatic region of Pernambuco are of the same order of magnitude as those found by Vasconcellos et al. (1986) in Poços de Caldas, Minas Gerais. This region is an alkaline igneous intrusion in which several radioactive anomalies exist. However, our results are higher than those measured by Fisenne and Keller (1969) and Fisenne et al. (1987). The uranium concentration in beans was a little higher than the value reported by Fisenne et al. (1987). The levels of ^{226}Ra found in a vegetable-garden contaminated with waste from a uranium plant in Ontario, Canada (Tracy et al., 1983), presented a mean geometric in tubers (potatoes) and grains (bean and corn) lower than those found in the phosphatic area of the Northeast of Brazil (Amaral, 1994).

The results obtained for the daily ingestion of U in the phosphate region of Pernambuco are in good agreement with results found in literature for other countries, as can be seen in Table 4. The results obtained for the daily ingestion of ^{226}Ra in the present work were 2- to 6-fold higher than those presented in literature, except for the values of India.

The results obtained for uranium and ^{226}Ra activity concentrations presented a high variation, mainly for sweet potato and cassava. The lowest relative standard deviation was obtained for corn samples. The ^{226}Ra activity concentration for all foodstuffs analyzed (fruits included) ranged from 43 to 2209 mBq kg^{-1}, whereas uranium concentration varied from 13 to 186 mBq kg^{-1}. Bean samples presented

Table 4
Mean values reported in the literature and this work for daily ingestion of uranium and ^{226}Ra in several locations

Location	Average ingestion (mBq day^{-1})	
	Uranium	^{226}Ra
This work	20.4	190
Hamilton, 1972		
Europe	0.82	–
San Juan diet	–	25.9
United Kingdom	20.3	–
Kerala diet	–	103.6
Fisenne and Welford, 1986		
New York diet	32.2	–
Japan diet	37.2	–
United Kingdom diet	24.8	–
Watson et al., 1984		
India diet	–	140.6
Florida diet	–	55.5
Morse and Welford, 1971		
New York diet	–	59.2

higher mean activity concentrations for ^{226}Ra and uranium; cashew samples also presented a higher mean activity concentration only for uranium.

The radionuclides of the uranium natural series are in secular equilibrium in the host rock. However, this equilibrium may be disrupted due to weathering conditions (UNSCEAR, 1988). Due to its chemical similarity with calcium, the radionuclide ^{226}Ra initially present in the rock, when dissolved, can be easily absorbed and/or adsorbed and enter in the food chain (Grzybwska, 1974; Burkart et al., 1983). When humans ingest radium, about 20% is absorbed into the body's circulatory system. Absorbed radium is initially distributed to soft tissues and bone, but its retention is mainly in growing bone (ICRP 67, 1993). According to Lima and Penna-Franca (1988), food constitutes the main source of human daily ingestion of radium.

For the fruits analyzed, higher ^{226}Ra concentrations were found for bananas, whereas, cashew samples presented higher concentrations of uranium. Among tubers and roots, higher levels of ^{226}Ra and uranium were observed for sweet potato samples, whereas among grains, bean samples presented higher concentrations of uranium and ^{226}Ra. In all cases uranium concentrations were always lower than ^{226}Ra concentrations.

The high variation of the experimental results obtained for the foodstuffs analyzed is mainly due to differences in the uptake of radium and uranium (Burr, 1984; Mays et al., 1985; Simon and Ibraim, 1990) differences in the soil ^{226}Ra and uranium concentrations, as well as possible differences in the soil composition in the studied region. It was observed that higher concentrations of ^{226}Ra and U were found in samples collected in areas where the phosphate mineral has been extracted. The results obtained could also be used to monitor possible alterations in the natural background due to human activities related to the phosphate exploitation.

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