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Estimation of Dose Due to Ingestion of ^{210}Pb in Milk from Dairy Cattle in the Semi-Arid Region of Pernambuco, Brazil

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Abstract ^{210}Pb is widely distributed in the environment. In this context, the aim of this research has been: (1) to determine ^{210}Pb concentrations in milk and dairy products from farms located in the cities of Pedra and Venturosa in Pernambuco, Brazil; and (2) to calculate the dose due to the ingestion of ^{210}Pb in these products. The ion exchange resin method was used and the concentration of ^{210}Pb varied from 62 to 650 mBq l^{-1} in fresh milk, from 202 to 1,494 mBq kg^{-1} in curdled cheese and from 11 to 253 mBq l^{-1} in milk whey. The estimated dose due to ingestion of milk and dairy products in this region was 0.2 Sv, which is at least two orders of magnitude above the maximum recommended levels.

Keywords Radioactivity · Lead · Risk · Man

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 some areas of the counties of Pedra and Venturosa, in the semiarid region of the state of Pernambuco, in northeastern Brazil. In these areas, uranium levels reached concentrations up to 22,000 mg kg^{-1} in rock samples (Costa et al. 1977). Milk production is one of the main activities in the counties of Pedra and Venturosa and also in the adjacent counties, an area known as the "Dairy Basin" of Pernambuco. The combined production of milk in the counties of Pedra and Venturosa represents about 10% of the total milk production in the state of Pernambuco. The milk and derived products from these counties are destined predominantly to the state of Pernambuco itself, where they are commercialized mostly in supermarkets and bakeries (SEBRAE 2002).

Radioactive anomalies were detected in a farm located about 9 km from Venturosa, near the Ipanema river. In this region, mafic rocks with a high concentration of uranium were found (Costa et al. 1977). The uranium mineral is located in rolled blocks, found near the porfiric granite contact with the migmatites, which are two regional emergent units (Costa et al. 1977). The analysis of these rocks revealed concentrations of up to 2.2% of natural uranium (Costa et al. 1977). Among the radioisotopes, ^{210}Pb, a descendant of ^{238}U, is the most abundant in the environment (Jaworowski 1969).

A more recent study, carried out in the milk producing farms in the counties of Pedra and Venturosa, located in the semi-arid region of Pernambuco, found elevated concentrations of ^{210}Pb in rock and soil samples, ranging from 3 to 201 kBq kg^{-1} and 195 to 86,400 Bq kg^{-1}, respectively (Silva et al. 2009a). The ^{210}Pb weathered from the rock is transferred to the soil and water and may be taken up by plants. If these plants are ingested by animals the ^{210}Pb may enter the human food chain through the consumption of meat or dairy products (Jaworowski 1969). The main plant species used as feed for dairy cows in farms in Pedra and Venturosa are prickly pear (*Opuntia* spp.), buffel grass (*Cenchrus ciliares*) and napier grass (*Pennisetum purpureum*). Analyses of plant samples from dairy farms in Pedra

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and Venturosa revealed high concentrations of ^{210}Pb in plant biomass, with values ranging from 2 to 503 Bq kg^{-1} in dry matter (Silva et al. 2009b). Part of the lead ingested by a dairy cow through the consumption of contaminated forage is transferred to the milk (Potter et al. 1971; Stanley et al. 1971). This is a very important pathway, from the radiological protection point of view, because milk is considered as one of the greatest sources of radionuclide ingestion by humans, particularly when dairy cows are fed with fresh fodder (Beresford and Howard 1999).

^{210}Pb is a beta emitter radionuclide with chemical affinity for hard tissue and with potential health hazards to humans. Salmon et al. (1999), stated that this radionuclide is mainly deposited in the trabecular and cortical bones of humans. These authors also stated that 22% of the total amount of ^{210}Pb present in human plasma eventually ends up deposited in bones. This accumulation occurs through the ion exchange between Pb^{2+} and Ca^{2+}, due to the metabolic similarity of lead with calcium in the human body, although the mechanisms of metabolism are not necessarily identical for these two elements (Pounds and Mittelstaedt 1983). Therefore, the study of the transport of this radionuclide through the human food chain, as well as the determination of the parameters involved on it, is of great interest for the long-term environmental impact assessment of regions with high levels of ^{210}Pb.

Materials and Methods

Twelve 5-L samples of fresh milk were collected from nine farms (namely F-1 to F-9) located near the area with anomalous levels of uranium. The samples were collected in June 2002 (rainy period), December 2003 (dry period) and May 2004 (rainy period). The selection criteria for the sampling locations were the distance of the farms to the occurrence of uranium and the quantity of milk produced in each farm. The farms chosen in the present study are the major producers of milk in the region, with a daily production of approximately 6,000 L. Since a significant fraction of the milk produced in the region is used to manufacture cheese, three samples of curdled cheese and milk whey were also collected from the three main dairy manufacturers in the counties of Pedra and Venturosa, which are located near the area with high levels of uranium. Several farms nearby, including the nine farms evaluated in the present study, supply milk to the three manufacturers of dairy products investigated in this research.

The possibility of milk contamination from sources other then the uranium anomalies is minimal, given the location of the farms that were studied. First of all, this region is characterized mainly by subsistence agriculture, therefore there are very few trucks that pass through the dirt roads that connect the farms. In addition, there are no industries or paved roads in the region where the farms is located, besides the three dairy manufacturers included in the study. Finally, the shortest distance between any of the farms included in this study to a paved road is approximately 10 km, and the nearest industry to the area of the study is about 30 km.

The selection criteria for choosing the cheese manufacturers were primarily the quantity of curdled cheese and milk whey produced and their proximity to the occurrence of uranium. The possibility of ^{210}Pb contamination during the milking process in the farms is minimal, since practically all farmers use polyethylene containers to store milk before sending it to the dairy manufacturers. A few of the larger farms use mechanical milking systems according to the standard procedures (Brasil 1980), therefore without materials that could contaminate the milk with ^{210}Pb. Besides, the facilities of the dairy manufacturers do not have processes that could expose the milk to materials that could allow for lead contamination.

The analysis of the milk, cheese and milk whey samples were carried out in the Environmental Monitoring Laboratory of the Departamento de Energia Nuclear of the Universidade Federal de Pernambuco (UFPE), in Recife, Pernambuco, Brazil.

The fresh milk samples were collected directly from the storage tank of each dairy farm, with a 5-L polyethylene container. Immediately after the collection of the milk sample, 5 mL of acetic acid and 10 mL of 37% formaldehyde were added to the container. After that, dry material was obtained by heating the sample to 80°C during 48 h. The material was taken to the furnace and the temperature was gradually raised up to 450°C, and left for 48 h at this temperate until ashes were obtained (IRD 1983).

Twenty litters of milk whey were collected in each dairy manufacturer by filling four 5-L polyethylene containers directly from the storage tanks. Immediately after collection, 10 mL of acetic acid and 20 mL of 37% formaldehyde were added to the containers (IRD 1983). After evaporation, the milk whey samples were submitted to the same treatment given to the milk samples.

Three-kilogram samples of curdled cheese were collected in three dairy manufacturers of the region. After that, each cheese sample was placed to dry at 80°C, until constant weight was obtained. After this procedure, the other steps of sample preparation were the same as described for the milk and whey samples (IRD 1983). It is important to point that, during the study, it was found that not all the cheese collected from the dairy manufacturers was necessarily produced with the milk coming from farms F-1 to F-9.

The ion exchange resin method (Godoy et al. 1998) was used to determine the concentrations of ^{210}Pb in milk,

curdled cheese and milk whey. The proposed method initiates with an overnight leaching of a 2 g sample of ashes with 100 mL 0.5 M HBr and 1 g hydroxylamine hydrochloride, for a period of 12 h. The solution was filtered and 1 mL of lead carrier (20 mg Pb^{2+} ml^{-1}), that was previously dissolved with nitric acid, was added. After this, the solution was percolated into a column that contained ion exchange resin type BIO-RAD DOWEX 1-X8 50–100 mesh chloride form. During this stage, the ^{210}Pb was retained in the resin and subsequently eluted with 50 mL of nitric acid (HNO_3) 1 M. The solution obtained was heated until it was totally dry. 50 mL of deionized water was added and the pH corrected with ammonium acetate 40% to reach levels between 4.5 and 5.0. The solution was heated until boiling, when 2 mL of sodium chromate (Na_2CrO_4) 25% were added, and then filtered on quantitative paper, covering the precipitate (lead was precipitated as $PbCrO_4$) and taken to the detector to determine the beta counting. For this, a Canberra Tennelec S5E detector of low background was used, which presented a mean background of 0.27 cpm at the beta plateau and a mean counting efficiency of ^{210}Bi of 20%. The concentration of ^{210}Pb ($C_{Pb\text{-}210}$) in Bq kg^{-1} was determined using Eq. 1 (Jia and Torri 2007).

$$C_{Pb-210} = \frac{A_L}{(1 - e^{-\lambda_{Bi}t})\eta y w} \quad (1)$$

where A_L = the liquid counting obtained in the detector (cpm); t = the ^{210}Bi ingrowth time (min); λ_{Bi} = the ^{210}Bi decay constant (min^{-1}); η = the detection efficiency for ^{210}Bi (cpm/Bq); y = the chemical yield; w = the sample dry weight (kg).

The S5E detector was calibrated using a standard solution of ^{210}Pb supplied by the Instituto de Radioproteção e Dosimetria (IRD/CNEN). The Laboratório de Monitoração Ambiental (Environmental Monitoring Laboratory) of the Departamento de Energia Nuclear (Nuclear Energy Department) of the Universidade Federal de Pernambuco (Federal University of Pernambuco) is part of the National Intercomparison Program, whose objective is to evaluate the accuracy of the determination of concentrations of ^{210}Pb by the method used in the present work.

The reliability of the analytical method used was assessed by internal intercomparisons, prior to the determination of ^{210}Pb concentrations in the samples. For this, standard samples were prepared, using certified material provided by IRD. In this case, a standard ^{210}Pb solution with 3 Bq of activity was added to 2 g of ashes from four samples with known concentrations of ^{210}Pb, with the objective of determining the chemical yield of the standard concentration during the normal analysis procedures.

Since ^{210}Pb, when ingested by humans, is accumulated mainly in bones (Salmon et al. 1999), it was considered that the systematic ingestion of cow milk by the inhabitants of the studied area generate a uniform distribution of this radionuclide throughout the skeletal structure, as suggested in ICRP-67 (1993). The estimation of dose is a very important step to assess the risk of incorporation of ^{210}Pb in the skeleton of humans. Some considerations must be examined on the dosimetry of ^{210}Pb, given the energy released during the decay of this radionuclide in the skeleton: (1) the ^{210}Pb decays to ^{210}Bi by emitting beta particles with average energy of 4 keV; (2) ^{210}Bi produces ^{210}Po, when other beta particles are emitted with average energy of 0.4 MeV, (3) ^{210}Po (last element generated by the radioactive decay chain of ^{238}U) emits alpha particles of energy 5.3 MeV. However, for dosimetric standards, the energy of 4 keV can be neglected (Salmon et al. 1999). In the case of incorporation of radionuclides into bones, the dosimetry of alpha particles is the most important from the radiological protection point of view (Rowland et al. 1978). In general, the alpha particle absorbed dose $D_{a,T}$ (in Gy) to a soft tissue (T) from internal ^{210}Pb is calculated by Eq. 2 (Salmon et al. 1999).

$$D_{\alpha,T} = \frac{(A_T)(5,297,000)(t)(1.6 \times 10^{-19})}{W_T} \quad (2)$$

where A_T = activity of the nuclide in the tissue T (Bq); 5,297,000 J $\cong$ 5.3 MeV; t = incorporation time (s); 1.6×10^{-19} = the energy of 1 eV (joules); W_T = tissue weight (kg). For committed dose equivalent in Sv, alpha absorbed dose is multiplied by 20 Sv Gy^{-1}.

Results and Discussion

The fresh milk, milk whey and curdled cheese samples were analyzed in triplicate and the mean concentrations of ^{210}Pb 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 IRD (1983).

The accuracy of the results was checked by running three replicates of the certified reference material. The results obtained showed an average chemical yield of 90 ± 7% of standard concentration in the method. In the calculation of the detection limit the Brazilian Agency of Sanitary Agency (ANVISA) from the Brazilian Ministry of Healthy definitions were used (ANVISA 2003). This technique has a detection limit of 81 mBq (0.081 Bq).

The concentrations of ^{210}Pb in the milk samples are coherent with the occurrence of the main anomalous ^{238}U rock outcrop in the region, which is located in F-7 (Santos Jr. et al. 2006). High ^{210}Pb concentrations in milk were also observed in F-8, which may be explained by the fact that

Table 1 Mean concentration of ^{210}Pb in fresh milk, milk whey and curdled cheese samples

Farm	Latitude	Longitude	Year	Concentration of ^{210}Pb		
				Milk (mBq L^{-1})	Whey (mBq L^{-1})	Cheese (mBq kg^{-1})
F-1	8°49′16″	37°00′24″	2002	120 ± 3[a]		
F-1	8°49′16″	37°00′24″	2003	97 ± 12		
F-2	8°48′30″	37°00′51″	2003	575 ± 13		
F-2	8°48′30″	37°00′51″	2004	89 ± 9		
F-3	8°55′9″	37°00′59″	2003	628 ± 16		
F-4	8°49′9″	37°00′42″	2004	91 ± 8		
F-5	8°44′16″	37°00′41″	2003	857 ± 8		
F-6	8°2′14″	37°00′32″	2003	297 ± 10		
F-6	8°2′14″	37°00′32″	2004	722 ± 14		
F-7	8°39′10″	37°00′19″	2004	1,557 ± 22		
F-8	8°39′20″	37°00′18″	2004	1,471 ± 18		
F-9	8°53′39″	37°00′44″	2004	567 ± 12		
DM-1	8°47′18″	37°00′54″	2004		87 ± 3[a]	
DM-2	8°48′25″	37°00′38″	2004		161 ± 7	
DM-3	8°52′19″	37°00′54″	2004		253 ± 12	
DM-1	8°47′18″	37°00′54″	2004			1,403 ± 14[a]
DM-1	8°47′18″	37°00′54″	2004			1,262 ± 12
DM-1	8°47′18″	37°00′54″	2004			1,208 ± 10
DM-2	8°48′25″	37°00′38″	2004			2,219 ± 10
DM-2	8°48′25″	37°00′38″	2004			2,347 ± 13
DM-3	8°52′19″	37°00′54″	2004			1,356 ± 16
DM-3	8°52′19″	37°00′54″	2004			1,202 ± 12
DM-3	8°52′19″	37°00′54″	2004			1,388 ± 17

[a] Standard deviation (95% confidence)

the soils in F-8 have high levels of ^{238}U originated from the weathering of the existing anomalous rock out crops on the neighbour F-7.

As mentioned previously, in the area of the present study no anthropogenic alterations were found to justify the presence of ^{210}Pb in the environment. It was verified that the sample collection procedure and storage of milk and milk whey carried out in all 9 farms were in accordance with the Ministry of Agriculture Legislation (Brasil 1980) that defines: every recipient used for milk sampling or milk storage should be made of stainless steel, aluminium or tinned iron, of perfect finishing, without faults. This procedure supports the hypothesis that the lead found in the fresh milk, milk whey and curdled cheese samples (Table 1) came only from the natural occurrence of this element in the soils of the farms. Thus, the decay of ^{238}U can be considered as the main source of soil ^{210}Pb in the counties of Pedra and Venturosa. Parkpian et al. (2003) stated that the ingestion of lead contaminated soil adhered to the forage is the main process responsible for the presence of lead in cow's milk.

Studies on ^{210}Pb concentrations in fresh milk samples are very scarce in literature. Compared to the available studies, the values presented in Table 1 are considered high. Amaral et al. (1988) determined ^{210}Pb concentrations varying from 5.0 to 60 mBq l^{-1} in fresh milk of dairy farms in the anomalous region of Poços de Caldas in Minas Gerais State, Brazil. Another study conducted by Pereira and Júnior (2002) found a mean ^{210}Pb concentration of 270 mBq l^{-1} in fresh milk samples from milk producing farms in the vicinity of the uranium concentration unit in Caetité, Bahia, Brazil. On the other hand, in the literature there are no data about concentrations of ^{210}Pb in samples of curdled cheese and milk whey.

The average daily milk ingestion by the residents of the Pedra and Venturosa cities is about 0.3 L, taking in account the consumption of all food products that contain milk (SEBRAE 2002). If we take into account the milk sample with the highest ^{210}Pb concentration (1,557 mBq l^{-1}), then the daily ingested activity could be as high as 467 mBq. According to Salmon et al. (1999) the concentration of ^{210}Pb in cortical and trabecular bone increases with age with a particularly rapid increase between the ages of 10–20 years, associated with the pubertal growth spurt. Considering a life expectancy of 68.6 years for the population of the studied region (IBGE 2003), the transference of

^{210}Pb during only 58.6 years is considered for committed dose equivalent. In this case, it was considered that that a 10-year old person consumes the same amount of milk as an adult.

The daily ingestion of 467 mBq during 58.6 years results in 1.0×10^7 mBq, which is the ^{210}Pb activity ingested during the average lifetime of a person in the study region. According to Salmon et al. (1999), 22% of the total ^{210}Pb ingested by man is transferred to the bones. Therefore, 22% of 1.0×10^7 mBq is equal to 2.2×10^6 mBq. Transforming this value to Bq, we obtain 2.2×10^3 Bq. The alpha absorbed dose was estimated with the Eq. 2, considering a uniform distribution in the skeleton as suggested by ICRP-67 (1993). A skeleton mass (W_T) equal to 7 kg was considered, according to the reference man (ICRP 1977). The activity accumulated in the skeleton (A_T) during the lifetime was 2.2×10^3 Bq. The time of incorporation was 58.6 years which is approximately equal to 1.85×10^9 s. In this case, the cumulative dose absorbed was approximately 0.50 Gy, while the equivalent cumulative dose was 10 Sv. If this cumulative dose of 10 Sv is divided by 58.6 Sv year, the result correspond to an annual dose of approximately 0.2 Sv.

Knowledge of dose levels in radiation protection is an important step for risk assessment. Thus, to restrict the exposure to ionizing radiation, international authorities on radiation safety recommend dose limits to workers and members of the general public (ICRP 1993). The ICRP-67 (1993) recommends an annual effective dose limit of 0.001 Sv to members of the general public. If necessary in some cases a higher dose may be accepted, provided that the average over 5 years does not exceed 0.001 Sv. This dose limit is very low when compared with the world mean effective dose, i.e. 0.002.4 Sv, from natural background radiation (ICRP 1993).

Several studies have failed to correlate natural background radiation levels and cancer incidence, even in regions with experimentally high natural background such as areas of India and China (Nambi and Soman 1987; Luxin et al. 1990). However, the annual dose equivalent of approximately 0.2 Sv estimated in the present study is two orders of magnitude greater than 0.001 and 0.0024 Sv. This is very important considering the effects of ionizing radiation. According to ICRP-67 (1993), there is no safe dose of exposure to radiation, in relation to genetic effects, therefore, any exposure to radiation may involve some risk of induction of somatic and hereditary effects.

Information obtained from the Center of Hematology of Pernambuco (HEMOPE), demonstrates that there are cases of acute promyelocytic leukemia diagnosed in residents of the cities of Pedra and Venturosa. According to Pounds and Mittelstaedt (1983), ^{210}Pb causes changes in blood cells and, more specifically, Salmon et al. (1999), state that ^{210}Pb may cause leukemia in humans. Currently, cytogenetic dosimetry has proven to be an important tool to evaluate the effects of the ionizing radiation in humans (Amaral 2005).

Regulatory actions should be considered in order to protect the residents of the counties of Venturosa and Pedra against the possible effects caused by the ingestion of ^{210}Pb through the consumption of contaminated milk. To get a more complete assessment of the radiation dose to residents of the municipality of Pedra and Venturosa, the authors suggest the conduction of a cytogenetic study.

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