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^{210}Pb in Forage Consumed by Dairy Cows

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Abstract The aim of this research was to determine ^{210}Pb concentrations in forage from farms located in the municipalities of Pedra and Venturosa, in the Brazilian state of Pernambuco, where rolled blocks of mafic rock with a high percentage of U_3O_8 were found. The concentrations of ^{210}Pb varied from 2 to 503 Bq kg^{-1} in the forage dry matter samples evaluated. Due to the elevated levels of ^{210}Pb in the forage samples, it is concluded that this radionuclide is highly available to be ingested by dairy cows and this could represent a hazard to the human population due to possible milk contamination.

Keywords Radionuclides · Buffel grass · Milk · Prickly pear · Napier grass

^{210}Pb is one of the descendants of ^{238}U radioactive decay series and may be found in soils due to the natural fallout. The occurrence of ^{210}Pb in plants is due the root uptake (D'Souza and Mistry 1970) or as leaf uptake due to the natural fallout of ^{222}Rn (Athalye and Mistry 1972). In soils with high natural radioactivity, the ^{210}Pb is easily absorbed by forage plants that are consumed by dairy cows (Amaral et al. 1988). Depending on the area and due to the fallout of the ^{222}Rn, the ^{210}Pb is the main radionuclide that contaminates the pastures, especially if they are composed of leafy plant species (Hill 1960). This is very important from the radiological protection point of view because cow milk is considered as one of the greatest sources of radionuclide ingestion by humans, particularly where milk production is based on the ingestion of fresh fodder by dairy cows (Beresford and Howard 1999).

A recent study carried out on dairy farms in the counties of Pedra and Venturosa, located in the agreste region of Pernambuco state, Brazil, found high concentrations of ^{238}U in rock and soil samples (Santos Júnior et al. 2006). Within this region, the main crops cultivated and used as forage for dairy cow are prickly pear (*Opuntia ficus-indica*), buffel grass (*Cenchrus ciliares*) and Napier grass (*Pennisetum purpureum*). It is possible that the ingestion of these forage species by dairy cows could lead to high levels of ^{210}Pb concentrations in milk. However, studies on ^{210}Pb concentrations in these forage species are very scarce in literature. In this context, the aim of the present study was to quantify ^{210}Pb concentrations in samples of prickly pear, buffel grass and Napier grass cultivated in dairy farms located near the main uranium anomaly in the agreste region of the state of Pernambuco.

Materials and Methods

Twenty tree biomass samples of prickly pear, buffel grass and Napier grass were collected during November 2007 in nine dairy farms in the Pedra and Venturosa counties. Each sample was lead to a furnace at 80°C during 48 h. Then the temperature was gradually raised to 450°C, and left for 48 h at this temperate until white ashes were obtained.

The samples were analysed in triplicate. The ion exchange resin method was used (Godoy et al. 1998) to

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determine the ^{210}Pb concentrations in the samples. The proposed method starts with an overnight leaching of 2 g 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. The solution was then 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 was subsequently eluted with 50 mL of nitric acid (HNO_3) 1 M and heated until completely dry. Fifty millilitre of deionized water was added and the pH of the solution was corrected with ammonium acetate 40% to reach levels between 4.5 and 5.0 and after this it was heated again until boiling. Two millilitre of sodium chromate (Na_2CrO_4) 25% were added and the ^{210}Pb and stable Pb were precipitated as $PbCrO_4$. The solution was filtered with a quantitative paper, covering the precipitate. The ^{210}Pb was determined by beta counting with a Canberra Tennelec S5E detector of low background. The detector presented a mean background of 0.27 cpm on 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 the 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 = liquid counting obtained in the detector (cpm); t = ^{210}Bi ingrowth time (min); λ_{Bi} = ^{210}Bi decay constant (min^{-1}); η = detection efficiency for ^{210}Bi (cpm Bq^{-1}); y = chemical yield; and w = weight of dry sample (kg).

The S5E detector was calibrated using a standard solution of ^{210}Pb supplied by the Institute of Radioprotection and Dosimetry (IRD). The analyses of the samples were carried out in the Environmental Monitoring Laboratory (LMA) of the Department of Nuclear Energy from the Federal University of Pernambuco. The quality control of the ^{210}Pb analyses was performed using both internal and external parameters. The internal assessment was done by ^{210}Pb comparisons with a standard sample. The external control is done by the participation of the LMA on the National Intercomparison Program, coordinated by the IRD and whose objective is to evaluate the accuracy of the determination of concentrations of ^{210}Pb by the method used on the present study. Standard samples were prepared using certified material provided by IRD. In this case, 2 g of ashes from four samples were taken with known concentration values of ^{210}Pb, adding 3 Bq of standard solution activity of ^{210}Pb with the objective of determining the efficiency of the standard concentration during the normal analysis procedures. The results obtained showed an average efficiency of 90 ± 7% of standard concentration in the method. This technique has a detection limit of about 0.081 Bq.

Results and Discussion

The mean concentrations of ^{210}Pb found in this study are presented in Table 1. For the calculation of the standard deviation of the mean concentrations, a 5% error was adopted for the radiochemical analysis of the samples, in accordance with the National Intercomparison Program of IRD (1983). The ^{210}Pb values found in the forage species (Table 1) were very elevated in relation to those found in literature. Amaral et al. (1988) determined ^{210}Pb concentrations varying from 9.4 to 83 Bq kg^{-1}DM in forage samples from dairy farms in the anomalous region of Poços de Caldas in Minas Gerais state, Brazil. In another study, Pereira and Júnior (2002) found mean ^{210}Pb concentrations of 0.56 and 1.99 Bq kg^{-1} DM, respectively, in prickly pear and grass samples from milk producing farms in the vicinity of the uranium concentration unit in Caetité, Bahia State, Brazil.

As seen in Table 1 there is an elevated variability in the results obtained. Any numeric value with a position in a box-plot above inter quarterly intervals of 1.5 can constitute an anomalous value (S-Plus 1997). According to the

Table 1 Mean concentration of ^{210}Pb in vegetables samples

Farm code	Vegetables	Concentration (Bq kg^{-1} DM)
F-1	Bufell grass	237 ± 20[a]
F-1	Elephant grass	260 ± 25
F-1	Forage palm	3.5 ± 1
F-1	Forage palm	23 ± 8
F-2	Bufell grass	191 ± 12
F-2	Forage palm	10 ± 2
F-3	Bufell grass	314 ± 35
F-3	Bufell grass	326 ± 34
F-3	Forage palm	23 ± 4
F-3	Forage palm	96 ± 15
F-4	Forage palm	4.5 ± 1
F-5	Bufell grass	42 ± 11
F-5	Forage palm	6 ± 1
F-5	Forage palm	27.5 ± 5
F-6	Forage palm	15 ± 2
F-7	Bufell grass	503 ± 50
F-7	Forage palm	127 ± 14
F-7	Forage palm	66 ± 7
F-7	Forage palm	2 ± 1
F-8	Bufell grass	361 ± 38
F-8	Bufell grass	30 ± 4
F-8	Forage palm	160 ± 19
F-9	Bufell grass	41 ± 5

DM dry matter

[a] Standard deviation (95% of confidence)

S-Plus (1997), any concentration that does not belong to the range m_e-1.5$A_4 < X < m_e$-1.5A_4, where m_e and A_4 are respectively the median and inter quartile range, is an anomalous value. The inter quarterly parameters obtained from the results of ^{210}Pb concentrations in the forage samples are presented in Table 2.

Figure 1 shows a normal qq-plot of ^{210}Pb concentrations in the prickly pear samples, which indicates the existence of anomalous values, very distant from the others, i.e., they deviate from the distribution of the other values. Comparing Tables 1 and 2, it is observed that in the data set of ^{210}Pb concentrations in the samples of buffel grass presented no anomalous values. Moreover, in the data set of prickly pear samples there was the occurrence of two anomalous values (127 and 160 Bq kg^{-1} DM). This suggests that high amounts of ^{210}Pb occur more often in buffel grass than in prickly pear.

Based on the study conducted by Hill (1960) and Athalye and Mistry (1972), possibly because as it is a leafy plant, the bufell grass accumulated more ^{210}Pb than the prickly pear which has no leaves, but only cladodes, which have a lower leaf area index and, therefore, lower probability to catch ^{210}Pb natural fallout. As shown in Table 1, the same trend of accumulation of ^{210}Pb in the bufell grass was also observed in Napier grass, because in these two plant species the radionuclide concentrations were higher. Despite the fact the prickly pear presents no leaves, the absorption of ^{210}Pb by this plant species was very significant. This can be explained taking into account the study done by D'Souza and Mistry (1970). According to these authors, ^{210}Pb is also transferred from soil to plants by root uptake.

According to a study of IAEA (1994), the ^{210}Pb ingested by dairy cows is transferred to the milk. This is a serious concern to radiological protection because cow's milk is considered as one of the major sources of radionuclide ingestion by humans, particularly where milk production is based on the ingestion of fresh fodder by dairy cows (Beresford and Howard 1999). The ^{210}Pb, a beta emitter, has an affinity for hard tissue, with potential hazards to human health. Therefore, the study of the transport of this radionuclide through the human food chain, as well as the determination of the parameters involved, is of interest to the long-term environmental impact assessment of regions with high levels of ^{210}Pb. The results presented in the this study showed the extreme need for a complete study that investigates the transference of ^{210}Pb to milk, with the aim of evaluating the risk of radionuclide ingestion due to milk consumption by the population in the agreste region of the state of Pernambuco.

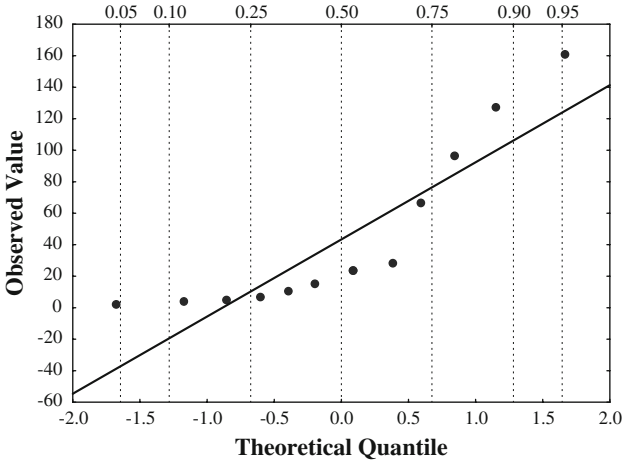

Fig. 1 Normal qq-plot of ^{210}Pb concentrations in forage palm samples (*Opuntia* spp.)

Table 2 Inter quarterly parameters obtained from the results of ^{210}Pb concentrations in the forage samples

Parameters	Forage palm (Bq kg^{-1} DM)	Bufell grass (Bq kg^{-1} DM)
First quantile ($q_{1/4}$)	42	6
Median (m_e)	237	23
Third quantile ($q_{3/4}$)	326	66
Interquarterly amplitude	284	60
Interquarterly interval	[−189; 663]	[−67; 113]

DM dry matter

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