



Activity levels of gamma-emitters in Brazil nuts

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ABSTRACT

Activity concentrations of the radionuclides ¹³⁷Cs, ⁴⁰K, ²²⁶Ra and ²²⁸Ra were determined in Brazil nuts acquired at points of sale between 2010 and 2013. Results indicated that the estimated annual effective radioactive dose due to ingestion of Brazil nuts is 27% of the annual dose limit of 1 mSv y⁻¹ for public exposure, according to the International Commission on Radiological Protection (ICRP). To estimate this dose the highest activity concentration obtained for each radionuclide was considered, assuming an annual consumption of 1.5 kg y⁻¹ per individual.

Keywords: Brazil nuts; Gamma-ray spectrometry, Ingestion dose.

1. INTRODUCTION

In recent years, nutritionists have recommended controlled daily intake of Brazil nuts due to its high selenium content. Some studies have shown that the daily consumption of one unit of Brazil nuts is sufficient to restore the status of selenium in obese women and the elderly [1-2]. Brazil nuts (*Bertholletia excelsa*, family Lecythidaceae) grow on wild trees, some reaching up to 50

meters in height and scattered throughout the Amazon region [3]. Selenium is an important part of antioxidant enzymes that act in the prevention of age-related illnesses such as cancer and cardiovascular diseases [4-6]. In addition to selenium, it is known that Brazil nuts also accumulate barium and radium [7]. Due to the presence of radium in these nuts, the level of radioactivity is 2-4 times higher than that of other vegetable foodstuffs, of the same region. Smith [8] also reported that the Brazil nut tree has a unique capacity of accumulating barium and radium in its fruit. This accumulation of Ba and Ra has been explained as a possible formation of organic complexes that increases the mobility of alkaline earth ions, particularly for these elements, favoring their redistribution during fruit development.

Several radioactive substances are considered carcinogens (Group 1 agents), according to the International Agency for Research on Cancer [9]. The carcinogenic activity is attributed to radiation, such as the radiation emitted by ^{224}Ra or ^{226}Ra or ^{228}Ra and their decay products. For this reason, monitoring of certain foods is a form of health protection. Radium isotopes are bone accumulating radionuclides with a significant residence time and high contribution to the internal dose [10]. They are among the most radiotoxic natural occurring radionuclides as can be seen by their high effective dose coefficients [11]. Besides natural radiation, artificial radionuclides, such as ^{137}Cs , also contribute to the total dose mainly in locations subjected to the radioactive fallout. Despite their capacity of accumulating earth alkaline elements (including Ra-isotopes) has been known for some decades, which few papers reporting the effective dose due to Brazil nuts ingestion [12]. Besides, continuous dose assessment is worthwhile due to the great appeal for consumption of this food today.

The aim of this study was to evaluate the concentrations of natural (^{226}Ra , ^{228}Ra , and ^{40}K) and artificial (^{137}Cs) radionuclides in samples of Brazil nuts acquired at final points of sale, in various Brazilian regions, between 2010 and 2013. Gamma-ray spectrometry measurements were used to determine the radionuclide activity concentrations.

2. MATERIALS AND METHODS

2.1. Brazil nut sampling and treatment for the analysis

Samples of about 200 g of shelled Brazil nuts, the edible portions, were purchased in stores located in various regions of Brazil. Five collections were performed each year over the period of 2010 to 2013 resulting in 20 samples. This approach was used in such a way that samples from different production locations over time could be obtained. The weight of a whole nut ranged from 2.8 to 5.1 g.

In order to obtain homogeneous material for the measurements, nuts were ground in mills with a titanium knife, without previous treatment. Besides grinding no other treatment was performed on the samples as the nuts have high oil content.

2.2. Gamma-ray spectrometry of ^{226}Ra , ^{228}Ra , ^{40}K and ^{137}Cs radionuclides

Sub samples of the homogeneous materials were placed in cylindrical plastic pots (53 mm x 20 mm), which were sealed (to prevent radon release). These sealed pots were kept for at least 30 days to reach an equilibrium condition with progeny nuclides of ^{226}Ra . Three important data: date of sealing, name and weight of the sample were recorded for every pot.

To obtain the activity concentration of radionuclides in nuts, the comparative method of analysis was used. In this case, the net peak area of the element of interest in the sample is directly compared with the net peak area, in the same energy, of certified reference materials (CRM). The CRMs: IAEA-375, IAEA-RGK-1, IAEA-RGU-1 and IAEA-RGTh-1 were used as comparative standards for determining respectively, the activity concentration of ^{137}Cs , ^{40}K , ^{226}Ra and ^{228}Ra . All CRM samples were prepared and measured in an identical geometry as the nut samples. To check the validity of this procedure, the IAEA-327 material was analyzed as a control sample.

The measurements were carried out in a high resolution gamma spectrometer: Canberra model GX2020 high purity Ge detector, coupled to a model 1510 Integrated Signal Processor and MCA System 100, both from Canberra. The detector used had a resolution (FWHM) of 0.9 keV for 122 keV gamma rays of ^{57}Co and 1.9 keV for 1332 keV gamma-ray of ^{60}Co . The counting time

for samples and background was 28 h. All measurements were carried out with the samples placed in contact with the detector.

The determination of ^{226}Ra , ^{228}Ra , ^{40}K and ^{137}Cs activity concentrations in nut samples was based on detection of 609 keV gamma rays of ^{214}Bi , 911 keV gamma rays of ^{228}Ac , 1,460 keV gamma rays of ^{40}K and 661 keV gamma rays of ^{137}Cs , respectively. In all cases the background radiation was considered and subtracted from the photopeak measured in the samples. No auto-absorption correction was applied since all the gamma transition energies considered in the calculations was above 200 keV [13]. The combined standard uncertainty was estimated through the propagation of errors due to counting statistics.

2.3. Calculation of the annual effective dose

For the assessment of the annual effective dose it was admitted an approximate consumption of 1.5 kg y^{-1} per individual, assuming an average weight of 4 g per nut, and considering the recommended daily intake of one unit of Brazil nut [1-2]. The conversion factors used for annual effective dose due to radionuclide ingestion are shown in Table 1.

Table 1: Conversion factors of the effective dose by ingestion of radionuclide [14]

Conversion factor of the effective dose, $\mu\text{Sv Bq}^{-1}$			
^{137}Cs	^{40}K	^{226}Ra	^{228}Ra
1.3×10^{-2}	6.2×10^{-3}	0.28	0.69

3. RESULTS AND DISCUSSION

3.1. Gamma spectral analysis

The gamma-ray spectra obtained for all Brazil nut samples were similar. Gamma-ray peaks belonging to decay products of ^{226}Ra such as ^{214}Pb (295.2 and 351.9 keV) and ^{214}Bi (609.3 and 1120 keV), the peaks from ^{228}Ac (338, 911 and 968.9 keV) decay product of ^{228}Ra , peak from ^{40}K (1460 keV) and in some cases, peak from ^{137}Cs (661 keV) were well defined. Since the gamma rays of ^{210}Pb (46 keV, progeny of ^{214}Bi) and ^{234}Th (63 keV, progeny of ^{238}U) in the samples were of the same order of magnitude of background radiation, they were not considered.

3.2. Quality control

In order to validate the obtained results, ^{137}Cs , ^{40}K , ^{226}Ra and ^{228}Ra activity concentrations were determined in CRM IAEA-327. The results showed good agreement with the certified values, as shown in Table 2.

Table 2: ^{137}Cs , ^{40}K , ^{226}Ra and ^{228}Ra activity concentrations obtained for the IAEA-327, in Bq kg^{-1}

Radionuclide	Recommended value (Confidence Interval)	Obtained value (Uncertainty) ^a
^{137}Cs	24.9 (24.6 – 25.2)	28 (7)
^{40}K	621 (612 – 630)	636 (17)
^{226}Ra	34.1 (32.7 – 35.5)	32 (5)
^{228}Ra	38.7 (37.8 – 39.6)	40 (4)

^aUncertainty resulting from the propagation of errors due to counting statistics.

3.3. Radionuclide concentrations in Brazil nuts

Table 3 shows the mean values and ranges of activity concentrations obtained for ^{137}Cs , ^{40}K , ^{226}Ra and ^{228}Ra in Brazil nuts, using high resolution gamma spectrometry.

Table 3: Mean and range of activity concentrations in Brazil nuts.

Year	Mean activity concentration \pm SD ^a – (Range of activity) Bq kg^{-1}			
	^{137}Cs	^{40}K	^{226}Ra	^{228}Ra
2010	7 \pm 11 (0.5 – 26)	244 \pm 95 (167 – 379)	52 \pm 10 (35 – 60)	235 \pm 59 (150 – 317)
2011	6 \pm 8 (1.4 – 20)	192 \pm 35 (150 – 246)	26 \pm 7 (21 – 38)	186 \pm 35 (143 – 237)
2012	13 \pm 9 (0 – 21)	189 \pm 56 (113 – 251)	34 \pm 11 (26 – 45)	135 \pm 96 (17 – 226)
2013	1 \pm 3 (0 – 7)	275 \pm 50 (193 – 324)	65 \pm 28 (31 – 104)	57 \pm 17 (34 – 75)

^aStandard deviation, n=5

In some samples, in the years 2012 and 2013, the peak at 660 keV of ^{137}Cs was not detected. On the other hand, the activity concentration observed for ^{137}Cs in some Brazil nut samples are higher than that found in edible mushroom (also known to concentrate this radioelement) consumed in São Paulo city, Brazil [15]. The highest activity concentrations obtained for ^{40}K can be

expected because potassium is an essential microelement for living organisms, and it is present in all samples from the environment. In about 75% of the samples analyzed (Table 3), the activity concentration ratios $^{228}\text{Ra} / ^{226}\text{Ra}$ are higher than one, indicating that the levels of ^{228}Ra are higher than those of ^{226}Ra . This has been observed in tropical soils, due to a tendency of these soils to have higher levels of Th than U [16].

For the calculation of annual effective dose the highest activity concentration obtained for each radionuclide (Table 3), consumption rate of Brazil nuts of 1.5 kg y^{-1} per individual and the effective dose conversion factor (Table 1) by ingestion of nuclide were considered. Thus the estimated annual effective dose of ingestion of Brazil nuts was of 0.27 mSv. The contribution of ^{228}Ra to that dose is 0.24 mSv. The total dose due to the analyzed radionuclides is 27% of the annual dose limit of 1 mSv y^{-1} for public exposure, according to the ICRP [17]. Therefore, there is no apparent health risk involved in the consumption of Brazil nuts according to results obtained for analyzed samples.

4. CONCLUSIONS

Among the radionuclides determined in samples of Brazil nuts ^{137}Cs , ^{40}K , ^{226}Ra and ^{228}Ra , the ^{228}Ra was the main contributor to the dose estimated annual intake of 0.27 mSv, with a contribution of 0.24 mSv. Contribution from radioactive fallout, fission product ^{137}Cs , was negligible. Based on the data obtained in this study, it can be said that the radioactivity present in Brazil nuts does not offer health risks for a daily ingestion of one unit.

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