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Evaluation of radionuclides in sugar samples from different regions of Brazil

Abstract

Sugar contains radionuclides and its consumption may result in an increase of the internal dose in humans. Thus, this work evaluates the profile and activity of radionuclides present in 4 VHP sugar samples obtained from producers from 4 cities in Brazil, located in the Southeast and Midwest regions: Pirajuba (State of Minas Gerais), Edeia (Goiás), Pitangueira (São Paulo) and Mendonça (São Paulo). The samples were collected between the years 2019 and 2020. The analyzes were performed by gamma spectrometry using a hyper pure germanium semiconductor detector (HPGe) counting the samples over a period of 24 hours. The same profiles of radionuclides ^{226}Ra, ^{212}Pb, ^{214}Bi, ^{40}K were observed in all samples, except for the sample from Mendonça, which presented a low counting rate for ^{214}Bi. Calculations of specific activity were carried out for the two most abundant radionuclides, ^{226}Ra and ^{40}K, obtaining the following values (in Bq kg^{-1}): for ^{226}Ra Pirajuba (22.43 Bq kg^{-1}), Edeia (22.38 Bq kg^{-1}), Pitangueira (18.25 Bq kg^{-1}) and Mendonça (36.16 Bq kg^{-1}) and for ^{40}K Pirajuba (0.66 Bq kg^{-1}), Edeia (0.99 Bq kg^{-1}), Pitangueira (0.66 Bq kg^{-1}) and Mendonça (0.33 Bq kg^{-1}). Additionally, the values of total absorbed dose rate were calculated: Pirajuba (43.61 nGy h^{-1}), Edeia (43.60 nGy h^{-1}), Pitangueira (41.82 nGy h^{-1}) and Mendonça (49.45 nGy h^{-1}). Therefore, according to the United Nations Scientific Committee on the Effects of Atomic Radiation, the results obtained did not show a significant transfer of absorbed dose due to radionuclides from sugar consumption under the conditions adopted in this work. No variation of radiation dose according to the region or year of production was observed.

Keywords: radionuclides, sugar, spectrometry, gamma spectroscopy

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Introduction

Considerable attention has been given worldwide to the study of high background radiation areas (HBRAs).[1–3] Some of these are found in China, India, Iran, United States, Canada, and Brazil.[3] A HBRA is defined as "an area or a complex of dwellings where the sum of exposures from cosmic radiation and natural radioactivity of soil, indoor and outdoor air, water and food intake result in an annual effective dose to the public above the level of the global average of 2.4 mSv y^{-1}" defined by the United National Scientific Committee on the Effects of Atomic Radiation (UNSCEAR).[4] HBRAs have been classified into four levels: low (<5 mSv y^{-1}); medium (5–20 mSv y^{-1}), high (20–50 mSv y^{-1}); and very high (>50 mSv y^{-1}).[5] Some of these areas have been under study for many years to determine the risks and effects of chronic low dose long term exposure for natural radiation.[6]

High backgrounds are normally due to high concentrations into the lithosphere of radionuclides from the three natural series starting with ^{238}U, ^{232}Th and ^{235}U. Plants grown in such areas are thus susceptible to bioconcentrate natural radioactivity. Human contribution to environmental contamination principally came from the military tests carried out during the 50ths and 60ths of last century or resulted from waste piles and effluents of uranium mining, nuclear wastes from nuclear power plants or nuclear accidents (e.g. Chernobyl, Fukushima,…) contributing to increase, locally or in large scale, backgrounds around the world.

Naturally occurring radionuclides are found in a variety of concentrations in every compartment of the earth: soil, water and atmosphere, and also in the tissue of all living beings (biosphere) subjecting human beings to a daily exposure.

Anthropic contamination is found more topically in areas affected by nuclear power plants explosions, nuclear bombs testing and more recently from the use of depleted uranium weapons. Such events affected mainly nearby soils, but also hydrographic basins and the atmosphere, mainly of the Northern hemisphere.

Consequently, people are exposed to ionizing radiation from naturally occurring and anthropic radionuclides that may be present in their food in unpredictable concentrations. Thus, besides anthropic ^{137}Cs *i.a.*, natural radionuclides belonging to the ^{232}Th and ^{238}U series as well as ^{40}K are the main contributors to radiation in foods.[5] As these radionuclides are not uniformly distributed in soils, the knowledge and measurement of their concentrations in foods play an important role in human radiation protection.

According to the International Food Safety Authorities Network (INFOSAN),[7] plants used as food commonly possess trace amounts of ^{40}K, ^{232}Th and ^{238}U and their progenies. And this is also the case of sugar cane, from which most of the sugar consumed over the world is produced.

Gamma-ray spectrometry using high-purity germanium detectors (HPGe) is a procedure widely used for determining the concentrations of natural and artificial radionuclides in environmental samples. As a nondestructive technique, this method possesses advantages in simultaneous multi-element analysis, simplified sample preparation (no chemical separation processes are required), and applicability for precise quantitative determination of the radioactive content in a sample. The most accurate and reliable method to determine the activity concentration of radionuclides is to use an adequate standard source with similar geometrical dimensions, density, and chemical compositions to the sample under study.[8]

This study aims to determine activities concentrations (AC) of the radionuclides ^{226}Ra, ^{212}Pb, ^{214}Bi and ^{40}K in sugar samples obtained from producers from 4 Brazilian cities located in the Southeast and Midwest regions: Pirajuba (State of Minas Gerais), Edeia (Goiás), Pitangueira (São Paulo) and Mendonça (São Paulo). The absorbed and effective doses due to sugar ingestion were also estimated.

Materials and methods

Sample collection and counting

The sugar samples analyzed were acquired from sugar producers from four cities situated the in the Southeast and Midwest regions of Brazil: Pirajuba (state of Minas Gerais), Edeia (Goiás), Pitangueira (São Paulo) and Mendonça (São Paulo).

The samples were obtained in the dry form submitting all of them to a drying process by treatment in an oven at 60°C for four hours or until they reached a constant weight. The dry mass value was used to determine the radionuclide activity concentration. The sample geometry recipient used for all samples in this work was a 2.1 L Marinelli beaker manufactured by Ga-Ma & Associates INC, made of polypropylene material. The Marinelli beakers were sealed and stored at the laboratory for a period of 45 days in order to let radionuclide activities reach the secular radioactive equilibrium conditions. Samples masses were measured on a digital balance, with a sensitivity of ± 0.01 g. Classical gamma spectrometric analytical techniques were applied to determine the AC of ^{226}Ra, ^{212}Pb, ^{214}Bi and ^{40}K. Initially measurements of ^{137}Cs were made in the sugar samples, but no such traces could be found. This step was executed at the Environmental Analysis of Nuclear Engineering Institute – IEN, Rio de Janeiro (RJ).

Gamma-ray detection system

The high-resolution gamma spectrometer included a Hyperpure Ge semiconductor detector (HPGe) with a Be window, manufactured by Canberra®, model GX2518 of cylindrical and coaxial geometries. The detector had a nominal efficiency of 30% and a resolution of 1.8 keV at the γ-ray energy of 1,332 keV of ^{60}Co. It was coupled to a multichannel MCA EAGLE PLUS equipped with 4,096 channels, whose pre-amplifier voltage was set at 3 kV, according to the manufacturer's specifications.[9] The Genie-2000® software provided by Canberra® was used to perform the manipulation and adjustments of the spectra.[10]

Activity concentration in the food samples

The AC and respective uncertainties were determined according to the statistical uncertainties of the peak areas provided by the Genie-2000 software. The specific AC of ^{40}K were measured at the gamma decay energy of 1,460.8 keV; those of ^{238}U, from the gamma emission line of ^{214}Bi (609,3 keV) and that of ^{226}Ra used the gamma emission line of 186,2 keV. The net counting area considered the total amount of counts under the peak energy in the area of interest discounted the Compton background, acquired per unit of time, and the background radiation (sample holder + shield). The count time was 28800 seconds. Activity concentrations in the sample were obtained from the net area using Equation 1[9]:

$$AC = \frac{N_L}{\varepsilon.m.k_g.\mathrm{t(s)}.\mathrm{P}_\gamma} \quad (1)$$

where AC is the activity concentration calculated for a given photopeak (Bq kg^{-1}), N_L the net area of photopeak energy E, ε is the counting efficiency at a specific energy, m the mass (kg) of analyzed sample, t is the counting time (s) and Pγ the emission probability of the measured gamma ray. For the study, the detection limits (DL) for quantification of the ^{238}U, ^{232}Th and ^{40}K were calculated using Equation 2[9]; several measurements of the laboratory background radiation were carried out:

$$DL = 4.66x\sigma \quad (2)$$

where σ represents the mean standard deviation associated with the background measurements and the factor 4.66 corresponds to a confidence level greater than 95%. The values of the detection limits obtained for the natural radionuclides assessed in the study can be seen in Table 1.

Table 1 Detection limit for HPGe-Be

Radionuclide	LD (Bq kg^{-1})
U-238	1.24
Ra-226	1.3
Th-232	2.03
K-40	2.47

The values determined for uranium and thorium took into account the average obtained as a function of the different limits for the energies used in the identification and quantification of each radionuclide, different for potassium that was estimated by its single gamma emission.

Dose calculation

The total air absorbed dose rate (nGy h^{-1}) for the sugar samples due to the specific activities of ^{226}Ra, ^{232}Th and ^{40}K (in Bq kg^{-1}) were calculated using Equation 3:

$$D_\gamma(\mathrm{nGy.h^{-1}}) = 0.427A_{Ra} + 0.662A_{Th} + 0.0432A_K + 0.03Ac_s + 34 \quad (3)$$

where D is the absorbed dose rate, A_{Ra}, A_{Th} and A_K are the AC for ^{226}Ra, ^{232}Th and ^{40}K, respectively.

To estimate the annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose (Sv Gy^{-1}) and outdoor occupancy factor proposed by Hendry J, et al.[5] were used. The effective dose rate in mSv y^{-1} was calculated by following equation 4:[10]

$$Def_{in}\left(Sv\,y^{-1}\right) = D\gamma.U.C_d \quad (4)$$

where Def_{in} is the calculated annual effective dose rate (Sv y^{-1}), U is the amount of food consumed in one year (kg y^{-1}), and C_d is the radionuclide specific concentration in the food (Bq kg^{-1}) and D_γ is the dose coefficient according to the International Commission on Radiological Protection (ICRP).[11]

Data available in the publication "Personal Food Consumption Analysis in Brazil - Brazilian Institute of Geography and Statistics/ IBGE"[12] were used to estimate the annual amount of food consumed. In addition, it was also considered that all radionuclides be transferred in the infusion process to the sugar. Such data facilitate understanding and application of the annual effective dose calculation.

Only the average values of the AC measured in this study were used to calculate the annual effective dose. Table 2 shows the dose coefficients[11] due to the intake of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K.

Table 2 Dose coefficients, D_γ (ICRP 119, 2012)

Radionuclides	Dose Coefficient (nSv Bq^{-1})
U-238	45
Ra-226	280
Th-232	230
K-40	6.2

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Results and discussion

The activity concentrations of radionuclides measured in sugar samples from Pirajuba (State of Minas Gerais), Edeia (Goiás), Pitangueira (São Paulo) and Mendonça (São Paulo) are shown in Table 3. The radionuclides ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K occurring naturally in sugar samples were present in low AC (in Bq kg^{-1}). No ^{137}C measurable AC could be found in the sugar samples; therefore, this anthropic radionuclide was not included in Table 3.

Sugar samples were analyzed by gamma spectrometry (Table 3). The AC of ^{238}U and ^{226}Ra are above the minimum detectable activity (MDA, see Table 1) whilst the values for ^{232}Th and ^{40}K are below. ^{40}K is below, possibly because being an essential element for plant development its concentration is physiologically regulated. The AC found for ^{238}U and ^{226}Ra were very close values, possibly because ^{238}U is in secular equilibrium with its decay product ^{226}Ra.

Table 3 Activity concentrations (mean ± SD, Bq kg^{-1}) of natural radioisotopes in sugar samples from four Brazilian regions compared to some foods reported.[5]

Radio-Nuclides	^{238}U	^{226}Ra	^{232}Th	^{40}K
Pirajuba	19.81 ± 0.41	22.43 ±7.12	0.03 ±0.56	0.66±0.41
Edeia	19.77 ± 0.51	22.38 ±7.46	0.03 ±0.59	0.99±0.41
Pitangueira	16.12 ± 0.43	18.25 ±6.71	0.01 ±0.56	0.66±0.34
Mendonça	31.93 ± 0.01	36.16 ±7.81	0.01 ±0.60	0.33±0.41
Fruits	11 ± 2.0	90.0 ± 40.0	10.0±3.0	47.0±10.0
Nuts	74 ± 5.0	2300±200	30.0±10.0	110.0±12.0
Vegetables	< 6	340.0±70.0	< 10	54.0±13.0
Cereals	< 6	150±5.0	< 10	134.0±15.0

According to Madruga, et al.,[13] soil can transfer ^{226}Ra to plants; as depending on the species, this isotope can be easily transferred from soil to vegetables. However, plant roots only absorb radionuclides when they are in solution in the soil.[14] In this case, radium may come into contact with the root surface. This is probably the case of Brazil nut trees known to bioaccumulate large amounts of radium isotopes in all areal parts.[15,16]

The intensity of radium absorption varies considerably from one plant to another, including between specimens of the same species, and is influenced by several factors, the main one being the radium concenration in the soil. Therefore, it can be stated that the presence of ^{238}U and ^{226}Ra in sugar depends mainly on the concentration of these radionuclides in the soil and the chemical form in which they are found.[13]

Most sugar samples have ^{232}Th values below 1.0 mBq.kg^{-1} (Table 3). Similar behavior is also observed for the ^{40}K isotope, the which does not belong to the thorium natural decay series. The difference in AC between the ^{238}U and ^{232}Th radionuclides in food products depends on their mobility in soils and their transfer to plants, which, in turn, depends on the type of soil and plant, climate, relief, season of vegetation and the radionuclide content in the soil.[17] Specifically in the case of Brazil, due to the indiscriminate use of fertilizers with phosphogypsum or gypsum by sugar cane producers,[18] the absorption of radionuclides such as ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K from sugar cane consumption may represent a public health problem, especially in by-products, such as brown sugar.[19] Control is therefore a need and is a legal must for sugar exportation.

Radionuclides such as ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K are of significant importance in radiological protection studies. Therefore, the concept of annual effective dose given by ICRP 119,[11] and occupational limits of radiological protection for the case of internal emissions were introduced based on the level of safety associated with the amount of radionuclides absorbed by the human body.[4] These safety limits can be used to compare with our results, since they are the maximum level allowed for the human body.[4] To better understand how contamination by radionuclides such as ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K works in the body, the International Commission on Radiological Protection[20] developed biokinetic models to promote a more reliable approach regarding the ingestion of radioactive substances. Such treatment seeks to understand both the movement of these substances in the body and the time they remain in each compartment, making it possible to calculate the internal risk associated to the various organs and tissues.[20]

The values of the radionuclides in sugar samples from the Brazilian cities here studied are lower than the span of values observed in the world.[4] Based on the conversion coefficients of specific activity to annual effective dose due to ingestion by members of the public (adults), tabulated in ICRP Publication 119,[11] the annual effective doses delivered by sugar from studied cities were calculated considering annual consumption of 1 kg of sugar. The total absorbed doses resulting from ingestion, by adults, for each radionuclide analyzed in this work and the annual effective dose are shown in Table 4.

Table 4 Total absorbed dose rate in air due to gamma radiation (D (nGy h^{-1})) and the outdoor annual effective dose rate (Def_{in} (mSv y^{-1})) assessment for sugar samples

Origin of Samples	Total absorbed dose D (nGy h^{-1})	Outdoor annual effective dose Def_{in} (mSv y^{-1})
Pirajuba	42.93	$4{,}43\times10^{-5}$
Edeia	43.17	$4{,}47\times10^{-5}$
Pitangueira	35.04	3.61×10^{-5}
Mendonça	68.43[a]	7.14×10^{-5}[b]

a. Values inside the world range (10-200 nGy h^{-1})

b. Values smaller than worldwide average (0.07 mSv y^{-1})

The effective doses for radionuclides ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K were calculated for dietary consumption by adult population groups[21] using Equation 4. Due to the biological half-life of potassium, the effective dose derived from ^{40}K is considered to be constant with a value of 0.66 mSv.y^{-1} for adults.[4]

The highest absorbed dose value (68.43 nGy h^{-1}) is observed in sugar from the city of Mendonça due to the highest AC of ^{226}Ra (36.16 Bq kg^{-1}) and ^{238}U (31.93 Bq kg^{-1}) (Table 3), different from the Pitangueiras region which presented the lowest total absorbed dose (35.04 nGy h^{-1}), values that put the absorbed dose in sugar samples within the range of international accepted values (10-200 nGy h^{-1}) for sugar intake.[4]

For all regions analyzed, the annual effective dose contribution for the four analyzed isotopes (Table 4) presents values in the same order of magnitude, with the highest annual effective dose contributions being found in the Mendonça region (7.14×10^{-5} mSv y^{-1}), considering the highest absorbed dose due to sugar ingestion as 68.43 nGy h^{-1} present in the analyzed samples, on the other hand, the lowest contribution to the annual effective dose was observed in sugar samples from the Pitangueira region (3.61×10^{-5} mSv y^{-1}); this value resulted from the absorbed dose by sugar intake equal to 35.04 nGy h^{-1}. The AC values for radionuclides such as ^{232}Th and ^{40}K, according to the results in Table 4, contribute very little to the calculation of total absorbed doses and annual effective doses.

Conclusion

The maximum concentrations of radioisotopes present in sugar were 36.16 Bq.kg^{-1} (^{226}Ra) and 31.93 Bq.kg^{-1} (^{238}U). These radiation levels are considered acceptable when compared with the literature and the recommendations of the International Commission on Radiological Protection (ICRP), being of no risk to human health. The sugar producing region with the lowest annual effective dose value for consumption of 1 kg y^{-1} was Pitangueira. The radionuclide that most contributes to the effective dose is ^{226}Ra, with more than 50% of the total. ^{40}K is evenly distributed in the body; its concentration is mainly under homeostatic control and does not pose a risk to human health; different from the long half-life ^{226}Ra isotope, whose biological behavior is similar to calcium. When ingested, radium is allocated into the individual's bone structures, increasing internal exposure, and consequently the effective dose. The results show that the radiation dose resulting from ingestion of the sugar samples does not represent any significant risk for the population's health from a radiological point of view.

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Conflicts of interest

The authors declare that there is no conflict of interest.

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