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Brown, Crystal, Refined and Organic Sugar Samples from Several Countries: Evaluation of Chemical Impurities

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1. Introduction

Sugar is among the foods that are most consumed by the world population. According to the International Sugar Organization – ISO [1], the sugar from either cane or beet is produced today by about 110 countries; among them, eight countries are producing sugar from both cane and beet. The great production of sugar comes from sugar-cane, about 80% of global production. India, Brazil, Thailand, China, USA, Mexico, Russia, Pakistan, France and Australia are the top ten producing countries. The world trade in 2016 - 2018 pointed out that the top five exporters were Brazil, Thailand, EU, Australia and India, responsible on average for nearly 70%. Brazil, as the largest producing and exporting country in the world, accounting for about 45% of global export, is dominating the world trade. On the other hand, Indonesia, China and the United States were world's largest importing nations in 2018. The global increase in consumption is associated with the increase in world urbanization, population growth, per capita incomes, the price of sugar and better agricultural technology [1]. Besides the sugar, several products came from sugar crops such as livestock feed, fiber and energy, particularly biofuels (sugar-based ethanol) and co-generation of electricity (cane bagasse). The industry has been using sugar in sugar-sweetened beverages, sweets and desserts, condiments, prepared foods to improve palatability and increase viscosity, texture, color and durability. Added sugars refer to refined sugar, brown sugar, corn and glucose syrups, fructose-based sweeteners, honey and molasses [2]. There is a wide variety of sugars derived from a variety of vegetables and obtained from different methods, however, usually the term "sugar" refers to sucrose, whose chemical formula is $C_{12}H_{22}O_{11}$ and which is a sweet carbohydrate, white when pure. Sugar originated from sugarcane and popularly known in the trade as white crystal sugar is obtained through processes of milling and grinding the stems. From the extracted juice that is treated to remove impurities, it is concentrated by boiling, forming a thick syrup. The crystallized sugar is separated from the syrup through rotation in a centrifuge, obtaining raw sugar, which after going through the refining process, has the final product [3]. In general industry it is used in production of pharmaceuticals mainly antibiotics, cosmetics, and other products for human consumption, production of medication and other uses [4,5]. At the same time, there is the health concern debate. There are many evidences that a high-sugar diet can come with damaging health risks. For instance, sugar has been linked to increased risk of high blood pressure, high cholesterol, inflammation, insulin resistance, obesity, type 2 diabetes, and heart disease [6]. Even though it is classified as a safe food ingredient, high and prolonged consumption can lead to exposure to inorganic elements - which may be present as impurities - that represent a health risk [7,8].

Therefore, it is important to identify and quantify chemical elements considered impurities in sugar that reach consumers, since the Brazilian legislation ANVISA (National Health Surveillance Agency) [9] and ICUMSA (International Commission for Uniform Methods of Sugar Analysis) [10,11] establish maximum values of sugar concentrations for some elements. Inorganic elements in sugar can come, for example, from the planting of the vegetable, the geographic location, the type of soil, the quality of drainage water and the type of plantation cultivated in the vicinity. The elemental concentrations in different samples can be determined by neutron activation technique [12]. This technique is usually applied to samples with a mass of approximately 200 mg, considering the as small or punctual size. Thus, several simplifications are made in relation to irradiation, as it is assumed that there is not neutron self-shielding or flux gradient in the sample. Also, it is assumed that there is not gamma self-attenuation. However, when analyzing a punctual sample, there is not guarantee of representativeness or homogeneity. In order to overcome these difficulties, a methodology was established to analyze samples with masses from 500 mg to 5 grams [8,12] using the existing infrastructure at the Nuclear Technology Development Center, CDTN. Thus, the established methodology that consists of analyzing samples with cylindrical geometry, which is the format of the sample holder used, applying the k_0 method of neutron activation, was applied in this study to sugar samples of Brazilian and international origins.
The objectives of this study were to verify the presence of inorganic impurities in sugar samples available in sachets to consumers in places such as restaurants, coffee shops and others, in different countries applying the large sample analysis methodology, k_0 method, and verify if the concentrations of impurities found are in accordance with Brazilian and international legislations.

2. Methodology

Brown, crystal and organic sugar sachets were collected in different places (restaurants, coffee shops), from different brands and countries (Germany, Argentina, Austria, Brazil, Canada, Chile, Slovenia, United States, England, Japan, Montenegro, Portugal, Sweden). The samples were weighed from around 3 g to 5 g in a polyethylene sample holder according to large sample methodology [12]. Neutron monitors, Al-Au alloy (0.1%), IRMM-530R, from the IRMM, Institute for Reference Materials and Measurements, Belgium, were prepared as disks (0.1mm thick and 6mm in diameter) and weighed. For irradiation, each cylindrical sample was inserted into a larger vial for irradiation, with a monitor placed under it and another in the cap of this sample holder. The samples and the neutron monitors were irradiated for 8 hours in the TRIGA Mark I reactor IPR-R1 of the CDTN in the irradiation positions of the carousel. At a power of 100 kW, the average thermal neutron flux is 6.30×10^{11} neutron cm^{-1} s^{-1} and the average spectral parameters f (ratio between thermal and epithermal neutron fluxes) and α (parameter that measures the distance of the epithermal neutron distribution from ideality in the irradiation position) were 21.67 and 0.0026, respectively [13]. After 3 days decay time, the acquisition of gamma spectra was performed in an HPGe detector, 50% of nominal efficiency, associated electronics and Genie 2000 software for gamma spectra acquisition. Each sample was measured three times: the first measurement was to detect radionuclides with half-lives lesser than 15 hours, the second to detect half-lives lesser than 72 hours, and the third, radionuclides with longer half-lives. For the deconvolution of the spectra, the program HyperLab 2002 was used. The Kayzero for Windows® ver. 2.42 was applied to calculate the elemental concentrations.

3. Results and Discussion

Twenty-eight elements were detected (As, Au, Ba, Br, Ca, Cd, Ce, Co, Cr, Cs, Cu, Eu, Fe, Hf, Hg, K, La, Na, Rb, Sb, Sc, Sm , Sr, Ta, Th, U, W and Zn) in sugar samples, and 92.9% of these elements were determined in brown sugar (As, Au, Ba, Br, Ca, Ce, Co, Cr, Cs, Eu, Fe, Hf, K, La, Na, Rb, Sb, Sc, Sm, Sr, Ta, Th, U and Zn), 60.7% in crystal/refined (Au, Br, Ca, Co, Cr, Cs, Eu, Fe, Hg, K, La, Na, Rb, Sb, Sc, W and Zn) and 46.4% in organic (Au, Br, Ca, Co, Cr, Fe, K, La, Na, Rb, Sc, Sm and Zn).
The results show that brown sugar had a greater number of elements. This can be explained by the fact that it does not go through the refining process like crystal/refined sugar, thus, it conserves more calcium,

iron and mineral salts that are important for the human body, making people choose to use it for this type. However, as it does not undergo these industrial refining processes, it has shown that it also contains a greater variety of chemical elements in its composition than the other types. The presence of elements such as Au, Ce, Cs, Hf, La, Sc, Sm, Th, U, W can also be observed, mainly in brown sugar, which are not classified as essential for humans and are potentially toxic. The accumulation of potentially toxic elements due to long-term consumption of sugar can pose health risks. Through the Technical Regulation on Maximum Limits of Inorganic Contaminants in Food, RDC nº 42, 8/29/2013, ANVISA [9] establishes maximum normative values in sugar only for As and Pb, however without specifying which type of sugar. International legislation, ICUMSA [10,11], establishes values for As and Pb in refined/crystal sugar and As, Cd, Cu, Hg, Pb and Zn, in brown. Pb was not determined by neutron activation as it did not present adequate characteristics for the technique used. Tables 1 shows the mass fraction range of elements and the maximum concentration limits established by legislations.

Table 1. Mass fraction range (mg kg^{-1}) of the elements detected in sugar samples and the maximum values established by Brazilian and international legislations.

Elem.	Sugar (mg kg^{-1})					
				International Legislation		Brazilian Legislation
	Brown	Crystal/ Refined	Organic	Brown [10] ICUMSA 800/1200	Crystal [11] ICUMSA 45	All types, ANVISA [9]
As	0.08 – 0.29	< 0.003	< 0.002	1	1	0.1
Au	0.0006	0.00009	0.00005	VNF	VNF	VNF
Ba	1.2 – 1.7	< 0.9	< 0.4	VNF	VNF	VNF
Br	2.1 – 2.7	0.005 - 0.3	0.02	VNF	VNF	VNF
Ca	198 - 1726	186	111	VNF	VNF	VNF
Cd	< 1	< 0.03	< 0.03	0.05	VNF	VNF
Ce	0.05 – 0.07	< 0.01	< 0,01	VNF	VNF	VNF
Co	0.03 – 0.04	0.001 – 0.02	0.002	VNF	VNF	VNF
Cr	0.2 – 0.7	0.07 – 0.2	0.1	VNF	VNF	VNF
Cs	0.007	0.008	< 0.0002	VNF	VNF	VNF
Cu	< 20	< 0.01	< 0.01	3	3	VNF
Eu	0.0006	0.0004	< 0.0003	VNF	VNF	VNF
Fe	3.1 – 62	2.1 – 2.4	2.3 – 7.2	VNF	VNF	VNF
Hf	0.008	< 0.004	< 0.002	VNF	VNF	VNF
Hg	< 0.01	0.01	< 0.01	VNF	NF	NF
K	781 – 5044	9.0 – 433	126 – 151	VNF	VNF	VNF
La	0.02 – 0.04	0.002	0.003	VNF	VNF	VNF
Na	3.3 – 34	0.02 – 13.3	0.62 – 1.7	VNF	VNF	VNF
Rb	1.0	0.3	0.2	VNF	VNF	VNF
Sb	0.006 – 0.04	0.004 – 0.01	< 0.001	VNF	VNF	VNF
Sc	0.002	0.0007	0.002	VNF	VNF	VNF
Sm	0.01 – 0.002	< 0.0002	0.001	VNF	VNF	VNF
Sr	1.5 – 10.5	< 0.9	< 0.8	VNF	VNF	VNF
Ta	0.002	< 0.001	< 0.001	VNF	VNF	VNF
Th	0.007	< 0.002	< 0.001	VNF	VNF	VNF
U	0.02	< 0.003	< 0.001	VNF	VNF	VNF
W	< 0.01	0.02	< 0.002	VNF	VNF	VNF
Zn	16 – 37	19 – 40	20 – 31	VNF	VNF	VNF

Elem., Element; <, lesser than Detection Limit; VNF, Value Not Foreseen

4. Conclusions

Several elements were determined that are not considered essential for humans and potentially toxic, which can pose a health risk, such as rare earths, thorium and uranium. The elements Au, Br, Ca, Co, Cr, Cs, Fe, K, La, Na, Rb, Sc, Zn were determined in the three types of sugar analyzed. Brown sugar had arsenic in a concentration that exceeded the limit established by Brazilian legislation. Related to the analyzed samples, brown sugar, which presented 92.9% of the determined elements, is the one that potentially poses more health risk when consumed. White sugar presented 60.7% and organic 46.4%.

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