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Inorganic elements in sugar samples consumed in several countries

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Abstract Sugar is considered safe food ingredient, however, it can present inorganic elements as impurities uptake during cultivation and production process. Therefore, this study aimed at identifies the presence of these elements in granulated and brown sugar samples available for consumption in public places in several countries. The neutron activation technique applying the methodology to analyse larger samples, 5 g-sample, established at CDTN/CNEN based on k_0-method was used to determine the elemental concentrations. Several essential and non-essential elements were determined in a large range of concentrations. The results are discussed comparing to maximum values foreseen in the international and Brazilian legislations.

Keywords k_0-method · Larger sample · Sugar · Neutron activation analysis

Introduction

The most popular sugar, white sugar, is composed of crystallized sucrose extracted from either sugarcane [1] or sugar beets [2], although it can be extracted from many plants. World sugar production is based on 70 % of sugarcane and on 30 % of sugar beet [3–5]. Several kinds of sugar are usually available for the end consumer in public places, but the most common ones are the white and brown sugars. Granulated sugar from white refined sugar is made from raw sugar that has undergone a refined process to remove the molasses. It is pure white crystalline ground sucrose [6]. Partly unrefined sugar is brown sugar, also called muscovado, dark muscovado or moist sugar. This kind of sugar is a specialty raw sugar, moist with high molasses content and strong molasses flavor [7]. Several kinds of brown sugars have been widely accepted mainly by organic low-processed food products consumers [3].

Although white sugar, produced from sugarcane and beet, is a very pure food product (>99.9 %) [5, 8], it may contain some soluble and/or insoluble organic and inorganic impurities that affect its quality and market value [6, 8, 9]. During sugar production, salts of organic and inorganic acids are added to sugarcane juice, about 3–4.5 % of the juice. Intermediate or final products of sugars transformations contain organic and inorganic salts [10]. Brown sugar is usually handmade. Purification, separation or sugar drying procedures are not used. The size of grains and

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crystals is irregular. Brown sugar grains which contain many soluble sugarcane juice components are a lighter color [11].

Inorganic elements present in sugar may come from plant cultivation place, raw material production, manufacturing process and technology applied for this purpose [8, 12, 13].

Knowledge of inorganic impurities in sugar is important not only due to the commitment with the quality of the final product [8, 11] but also because of dietary nutritional role. When ingested, those inorganic impurities will be transferred to humans and may be toxic [14].

The highest concern is usually about the presence of As, Cd, Hg, Pb in sugar, due to their toxicity [14–19]. Other elements, namely Cu, Fe, Mn and Zn, which may contaminate sugar during production by the corrosion of tubing and machinery, are also worrisome for the sugar industry and market [9, 12, 13, 20]. Besides contamination during sugar production [13], it is well known that the chemical composition of plants usually reflects the elemental composition of the place on which they are cultivated. Higher plants take up chemical elements from the air (or water) through their leaves [21–23] Absorption from soil through roots can be the main pathway of elements to plants [12, 24, 25]. The concentration of several elements in soils is associated with biological and geochemical cycles and is influenced by anthropogenic activities, such as agricultural practices, transport, industrial activities, waste treatment and disposal [26–30].

It is important to determine the presence of inorganic elements in the final product of granulated and brown sugars mainly because food safety [8, 13]. Flame absorption spectrometry (FAAS) was applied to determine traces of Ca, Cu, Fe, K, Mg, Na and Zn in white sugar samples [8, 9, 31]. The anodic stripping voltammetry (ASV) were used to determine As, Cd, Cu, Pb and Zn in white refined beet sugar [32, 33]. Impurities as Ag, Al, As, Au, Ba, Be, Br, Ca, Cd, Ce, Cl, Co, Cr, Cs, Cu Dy Eu, Fe, Ga, Hf, Hg, K, La, Li, Lu, Mg, Mn, Na, Nb, Ni, P, Pb, Sb, Sc, Se, Sm, Sn, Sr, Ta, Th, Ti, U, V, W and Zn in crystal and brown sugars were determined by Instrumental neutron activation analysis (INAA) [14, 34], atomic absorption spectrometry (AAS) [14, 34] and inductively coupled plasma-atomic emission spectrometry (ICP-AES) [34, 35]. Oxygen plasma ashing flameless atomic absorption was the technique applied to determinate Cr in white and brown sugars [36].

Therefore, this paper is about the determination of chemical elements in those types of sugar available for consumption in public places from several countries. The objective was to determine the quality of the sugars offered to customers. However the product packaging does not give consumers any information related to the place where the sugar was produced or to the origin of the sugarcane. The values of the elemental concentrations were compared to those foreseen in the current international and Brazilian legislations.

Experimental

Sampling

Samples of crystal (white) and brown sugar usually available in bags of 3 to 5 g were randomly collected from public places as restaurants, snack shops and pubs located in Argentina, Brazil, Croatia, France, Germany, India, Montenegro, Portugal, Slovenia and The Netherlands.

Analysis of the samples

One sample from each kind of sugar was analyzed by neutron activation technique, applying the k_0-standardization method, k_0-INAA [37], using the TRIGA MARK I IPR-R1 research reactor located at CDTN/CNEN, in Belo Horizonte. The analysis procedure consisted of steps like weighing samples, irradiating in the research reactor, executing the gamma spectrometry and determining the elemental concentrations.

In this study, 3 to 5 g-samples, considered for neutron activation analysis larger samples, were analyzed applying the new methodology established at CDTN/CNEN [38] and also with 200 mg, small sample—usual procedure, in order to validate the procedure to analyze larger sample. The samples were weighed in polyethylene vials (6.6 mm inner radius, 40 mm inner height) appropriated to irradiation. The samples were irradiated with Al-(0.1 %)Au discs (6 mm in diameter and 0.2 mm thick), alloy IRMM-530, from Central Bureau for Nuclear Measurements, Geel, Belgium, as monitors. The irradiation time was 8 h, period enough to activate the isotopes with nuclear characteristics adequate to determine the elements whose radionuclides have medium half-lives (As, Au, Br, Ga, K, La, Na and Sm), and long half-lives (Fe, Sb, Sc, Sr and Zn). The irradiation was in the carrousel of the TRIGA MARK I IPR-R1 reactor, irradiation channels IC-6, IC-7 and IC-8, with 6.35×10^{11} cm^{-2} s^{-1} average thermal neutron flux at 100 kW, and parameters f and α, 22.32 and -0.0022, respectively [37].

After irradiation, the cooling time was enough to decay the radionuclides with shorter half-lives so that they could interfere in the gamma spectrometry, about 3 days. The gamma detection system was composed by a gamma detector HPGe, 40 % relative efficiency. The spectra were acquired by Genie 2000 program CANBERRA for, at least, 4 h, and applied the HyperLab software ver. 2009 [39, 40] for net peak area evaluation. The elemental concentrations

Table 1 Materials of reference: experimental results and recommended values

El.	GBW 0805 (Tea leaves)			IAEA-SOIL-7 (Trace elements in soil)				
	Recommended values [37][a] (mg kg^{-1})	Small sample		Recommended values [36][a] (mg kg^{-1})	Small sample		Larger sample	
		Experimental values[b] (mg kg^{-1})	E_n number		Experimental values[b] (mg kg^{-1})	E_n number	Experimental values[b] (mg kg^{-1})	E_n number
As	0.191 ± 0.03	0.199 ± 0.017	0.18	13.4 ± 0.85	13.2 ± 0.5	0.15	13.1 ± 0.5	0.23
Ba	15.7 ± 2.04	14.8 ± 1.2	0.29	159[c]	149 ± 9	–	150 ± 11	–
Ca	2840 ± 227.20	3134 ± 195	0.65	163,000[c]	165,900 ± 6400	–	166,000 ± 6066	–
Ce	0.686 ± 0.09604	0.74 ± 0.04	0.43	61 ± 6.5	59 ± 2	0.26	55 ± 2	0.79
Co	NR	<1	–	8.9 ± 0.85	9.0 ± 0.5	0.08	7.9 ± 0.3	0.96
Cr	NR	<0.1	–	60 ± 12.5	62 ± 3	0.14	64 ± 2	0.30
Cs	NR	<0.1	–	5.4 ± 0.75	5.3 ± 0.2	0.12	5.0 ± 0.2	0.47
Fe	373.0 ± 26.11	399 ± 14	0.68	25,700*	26,750 ± 930	–	27,070 ± 950	–
Hf	NR	<1	–	5.1 ± 0.35	4.8 ± 0.2	0.56	4.6 ± 0.2	0.94
K	19,700 ± 1379.00	20,860 ± 733	0.58	12,100[c]	12,190 ± 480	–	12,490 ± 950	–
La	0.458 ± 0.0229	0.44 ± 0.02	0.39	28 ± 1.0	28 ± 1	0.00	27 ± 1	0.44
Na	142 ± 14.20	159 ± 6	0.91	2400[c]	2422 ± 22	–	2442 ± 85	–
Nd	NR	<2	–	30 ± 6	28 ± 1	0.32	24 ± 1	0.92
Rb	36.9 ± 1.476	39 ± 1	0.84	51 ± 4.5	49 ± 4	0.22	47 ± 2	0.66
Sb	0.037 ± 0.00,333	0.042 ± 0.002	0.96	1.7 ± 0.2	1.6 ± 0.1	0.35	1.6 ± 0.1	0.42
Sc	NR	<0.01	–	8.3 ± 1.05	8.7 ± 0.3	0.32	7.9 ± 0.3	0.33
Sm	NR	<1	–	5.1 ± 0.35	4.90 ± 0.03	0.56	4.6 ± 0.2	0.99
Sr	10.8 ± 1.84	12.4 ± 0.9	0.62	108 ± 5.5	<50,500	–	<50,500	–
Ta	NR	<0.1	–	0.8 ± 0.2	0.8 ± 0.1	0.00	0.67 ± 0.02	0.64
Tb	NR	<0.2	–	0.6 ± 0.2	0.63 ± 0.03	0.14	0.60 ± 0.02	0.00
Th	0.105 ± 0.0126	0.114 ± 0.004	0.60	8.2 ± 1.1	7.8 ± 0.3	0.32	7.3 ± 0.3	0.72
U	NR	<0.1	–	2.6 ± 0.55	2.1 ± 0.2	0.74	2.3 ± 0.1	0.51
Yb	NR	<0.1	–	2.4 ± 0.35	2.3 ± 0.1	0.25	2.0 ± 0.1	0.99
Zn	38.7 ± 3.87	38 ± 3	0.10	104 ± 6	108 ± 5	0.34	107 ± 4	0.30

NR not reported

[a] Uncertainty with a coverage factor $k = 2$ (95 % CI)

[b] U_{Lab_Comb} ($k = 1$)

[c] Information value

were calculated by the program Kayzero for Windows® [41]. For quality control, 200 mg and 5 g-samples of IAEA-SOIL-7 [42] and 200 mg sample of GBW 0805 [43], Tea leaves, reference materials, were analyzed.

Results and discussion

Statistical evaluation

The E_n number [44] was calculated for IAEA-SOIL-7, analyzed as small sample (200 mg) and large sample (5 g), and for GBW 0805 analyzed as small sample. The E_n number takes into account the expanded uncertainty of experimental and assigned values with a coverage factor $k = 2$ (95 % confidence interval). The following equations were used in the calculations:

E_n number

$$E_n = \frac{Value_{Experimental} - Value_{Assigned}}{\sqrt{U^2_{Experimental} + U^2_{Assigned}}} \quad (1)$$

where $U_{Experimental}$ and $U_{Assigned}$ are the expanded uncertainties ($k = 2$) of the experimental results and the assigned result, respectively, and

$$U_{Experimental} = 2 \cdot U_{Lab\_Comb} \quad (2)$$

$$U_{Lab\_Comb} = \sqrt{u^2_{AREA} + u^2_{method}} \quad (3)$$

Table 2 Elemental concentrations in Brazilian granulated sugar determined in small and larger samples

El.	Brazilian crystal white sugar	
	Small sample (mg kg^{-1})	Larger sample (mg kg^{-1})
As	<0.02	<0.01
Au	<0.0002	0.00055 ± 0.00003
Ba	<4	<1
Br	0.020 ± 0.002	0.017 ± 0.001
Ca	<200	<150
Cd	<1	<1
Ce	<0.2	<0.05
Co	<0.01	0.009 ± 0.001
Cr	<0.3	0.06 ± 0.01
Cs	<0.02	<0.005
Cu	<20	<20
Fe	<15	3.4 ± 0.4
Hf	<0.02	0.011 ± 0.001
Hg	<0.01	<0.01
K	8 ± 1	9 ± 1
La	<0.01	<0.002
Na	3.9 ± 0.2	3.8 ± 0.1
Nd	<0.2	<0.1
Rb	<0.9	<0.2
Sb	<0.004	0.009 ± 0.001
Sc	<0.001	0.0002 ± 0.0001
Se	<0.1	<0.02
Sm	<0.001	<0.0002
Sn	<100	<100
Sr	<5	<2
Th	<0.01	<0.004
Zn	18 ± 1	26 ± 1

where u_{AREA} is the uncertainty of the net peak area and u_{method} is the overall uncertainty of k_0-NAA established at the CDTN/CNEN as 3.5 % with a coverage factor $k = 1$. The uncertainty of the method is an estimate of the systematic uncertainty due to all standard uncertainties of parameters involved in k_0-standardization (k_0-factors, Q_0, f, α, ε_p, and true coincidence effects) [45, 46].

To compare the results of the two geometries with reference data, the criterion $|E_n| \leq 1$ was applied meaning that the evaluation of the performance of the method was satisfactory and if $|E_n| > 1$, the performance was unsatisfactory. Table 1 shows the experimental results for GBW0805 (small sample) and for IAEA-SOIL-7 (small and large samples) and the E_n number related to recommended values. It is observed that all results of E_n number are ≤ 1 pointing out that the performance of the method was satisfactory.

One sample collected in Brazil, granulated sugar, was analyzed as small and large samples [37]. The objective was to confirm whether the results would be consistent analyzing only 200 mg or 5 g, since, in this study, samples would be analyzed with a mass higher than the usual in neutron activation mass, approximately 5 g. The results are presented in Table 2 and are in good agreement. Additionally, in the case of values lower than detection limits, the large sample presented lower detection limits.

Tables 3 and 4 display the elemental concentrations determined in granulated and brown sugars, respectively. From some places, only one sample was collected, from others, more than one sample. In this case, a range of results is presented.

Observing Table 3, it is verified that granulated sugar sample from India presented the highest elemental concentration when compared to samples from other countries. Related to brown sugar samples, Table 4, Brazil presented the highest results.

Table 5 summarizes the results for all samples—granulated and brown sugars—presented in ranges (lower and higher values). Brazilian samples were separated from other countries in order to be easier the comparison with the values reported elsewhere, determined applying several techniques as neutron activation analysis, atomic absorption spectrophotometry and inductively coupled plasma mass spectrometry. It can be seen that the concentration of elemental impurities are varied.

Several inorganic elements were determined in sugar samples in a large range of concentration. In order to verify whether the values were in a safe range to be consumed by population, the values were compared to the legislation. The International Commission for Uniform Methods of Sugar Analysis (ICUMSA) [48] specifies for white sugarcane maximum of As and Pb values in refined/crystal sugar and the elements As, Cd, Cu, Hg, Pd and Zn in brown sugar. For beet sugar values to As, Cu and Pb are established. ICUMSA is an international organization that covers the activities of National Committees for Sugar Analysis in more than thirty member countries.

The Brazilian legislation, ANVISA, Agência Nacional de Vigilância Sanitária (*National Health Surveillance Agency*) decree number 55,871 May 26, 1965 [49] establishes maximum values to additives and inorganic contaminants As, Cd, Cr, Cu, Ni, Pb, Sb, Se and Zn in foods. The ordinance 685 August 27, 1998 [50] establishes normative values to the elements As, Cd, Cu, Hg, Pb and Sn. Both determine the amount of As and Pb in sugar, but without specifying the type of sugar [51]. In this paper, it was decided to present the values published in 1965 decree [49] as 1998 ordinance [50], assuming the most recent values.

The highest elemental concentration determined for each type of sugar and the maximum elemental values

Table 3 Results of elemental concentrations in crystal white sugar samples from several countries, analyzed as larger samples

El.	Granulated crystal white samples from several countries (mg kg^{-1})									
	Argentina $n = 3$	Brazil $n = 5$	Croatia $n = 1$	France $n = 1$	Germany $n = 1$	India $n = 1$	Montenegro $n = 1$	Portugal $n = 1$	Slovenia $n = 5$	The Netherlands $n = 1$
As	0.019 –0.021	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Au	0.00014–0.00044	0.00018–0.00055	0.00012 ± 0.00001	0.00019 ± 0.00001	0.00052 ± 0.00002	<0.0001	0.00040 ± 0.00002	<0.0001	<0.0001–0.00059	0.00032 ± 0.00002
Ba	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Br	0.032–0.053	0.009–0.018	0.013 ± 0.001	0.016 ± 0.001	0.008 ± 0.001	6.2 ± 0.2	0.023 ± 0.001	<0.005	0.010 –8.8	0.010 ± 0.001
Ca	<150	<150	<150	<150	<150	5761 ± 667	<150	<150	<150	<150
Cd	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Ce	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Co	<0.005–0.007	0.007–0.009	0.008 ± 0.001	0.008 ± 0.001	<0.005	0.111 ± 0.005	<0.005	<0.005	0.008–0.15	<0.005
Cr	<0.07	<0.07–0.06	<0.07	<0.07	<0.07	0.41 ± 0.05	<0.07	<0.07	<0.07–0.05	<0.07
Cs	<0.005	<0.005	<0.005	<0.005	<0.005	0.017 ± 0.002	<0.005	<0.005	<0.005–0.15	0.008 ± 0.001
Cu	<20	<20	<20	<20	<20	<20	<20	<20	<20	<20
Fe	<4	<4–3.4	<4	<4	<4	88 ± 4	<4	<4	<4	<4
Hf	<0.005–0.012	0.011–0.014	0.014 ± 0.001	0.012 ± 0.001	<0.005	<0.005	<0.005	<0.005	<0.005–0.15	<0.005
Hg	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
K	9–114	4.7–9	<3	27 ± 2	20 ± 1	5675 ± 485	50 ± 2	0.007 ± 0.001	25–10610	29 ± 2
La	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002
Na	3.5–3.6	3.0–5.9	<0.02	4.4 ± 0.2	5.9 ± 0.2	3888 ± 137	1.7 ± 0.1	0.006 ± 0.001	7.1–5610	4.7 ± 0.2
Nd	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Rb	<0.2	<0.2	<0.2	<0.2	<0.2	8.4 ± 0.4	<0.2	<0.2	<0.2	<0.2
Sb	0.001–0.008	<0.001–0.009	0.011 ± 0.001	0.011 ± 0.001	0.003 ± 0.0002	<0.001	<0.001	<0.001	0.011–0.015	0.009 ± 0.001
Sc	<0.0001–0.0003	<0.0001–0.0005	<0.0001	<0.0001	<0.0001	0.018 ± 0.001	<0.0001	<0.0001	<0.0001–0.004	<0.0001
Se	<0.02	<0.02	<0.02	<0.02	<0.02	0.18 ± 0.01	<0.02	<0.02	<0.02	<0.02
Sm	<0.0002	<0.0002	<0.0002	<0.0002	<0.0002	<0.0002	<0.0002	<0.0002	<0.0002	<0.0002
Sn	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100
Sr	<2	<2	<2	<2	<2	38 ± 2	<2	<2	<2	<2
Th	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004
Zn	25 ± 1	26–30	33 ± 1	31 ± 1	26 ± 1	61 ± 2	26 ± 1	0.021 ± 0.001	29–38	38 ± 1

n number of different samples analysed

Table 4 Results of elemental concentrations in brown sugar samples from several countries, analyzed as larger samples

El.	Brown sugar samples from several countries (mg kg^{-1})			
	Argentina $n = 2$	Brazil $n = 1$	Croatia $n = 1$	Slovenia $n = 4$
As	<0.01–0.021	0.29 ± 0.01	0.006 ± 0.001	<0.01
Au	0.00019–0.00022	0.0006 ± 0.0001	0.00023 ± 0.00002	0.00023–0.0010
Ba	<2	1.2 ± 0.1	<2	<2
Br	0.051–0.19	2.7 ± 0.1	0.073 ± 0.003	0.138–0.208
Ca	<150–146	1726 ± 283	825 ± 72	<150
Cd	<1	<1	<1	<1
Ce	<0.05	0.048 ± 0.005	<0.05	<0.05
Co	0.006–0.017	0.037 ± 0.001	0.017 ± 0.001	<0.005–0.023
Cr	<0.07	0.70 ± 0.03	<0.07	<0.07–0.09
Cs	<0.005	0.0067 ± 0.0004	0.007 ± 0.001	<0.005
Cu	<20	<20	<20	<20
Fe	<4	43 ± 2	9 ± 1	<4–7
Hf	<0.005–0.011	0.0080 ± 0.0005	0.015 ± 0.001	<0.005–0.014
Hg	<0.01	<0.01	<0.01	<0.01
K	119–195	5044 ± 177	730 ± 26	163–214
La	<0.002–0.0025	0.019 ± 0.001	0.0029 ± 0.0003	0.0022 ± 0.0004[a]
Na	4.5 ± 0.2*	10.5 ± 0.4	9.5 ± 0.3	3.4–4.8
Nd	<0.1	<0.1	<0.1	<0.1
Rb	<0.2–0.16	4.9 ± 0.2	1.5 ± 0.1	<0.2
Sb	0.0045–0.008	0.0083 ± 0.0005	0.010 ± 0.001	0.0059–0.019
Sc	<0.0001–0.0030	0.0036 ± 0.0001	0.0015 ± 0.0001	0.0016–0.0025
Se	<0.02	<0.02	0.058 ± 0.004	<0.02
Sm	<0.0002–0.0005	0.0022 ± 0.0002	0.00061 ± 0.001	0.0005 ± 0.0001[a]
Sn	<100	<100	<100	<100
Sr	<2	3.2 ± 0.3	3.2 ± 0.3	<2
Th	<0.004	0.0039 ± 0.0004	<0.004	<0.004
Zn	25 ± 1*	29 ± 1	31 ± 1	28–51

El. element

[a] Only one sample presented the element

Table 5 Summary of results of crystal white and brown sugar samples analyzed in this work and values reported in the literature

El.	Crystal white sugar samples			Brown sugar samples		
	Brazilian samples (mg kg^{-1})	Other countries samples (mg kg^{-1})	Literature [35, 47] (mg kg^{-1})	Brazilian samples (mg kg^{-1})	Other countries samples (mg kg^{-1})	Literature [12, 14] (mg kg^{-1})
As	<0.01	0.001–0.019	NR	0.01–0.29	0.001–0.006	0.018 ± 0.178
Au	0.00003–0.00018	0.00003–0.00008	NR	0.0001–0.0006	0.0001–0.00022	NR
Ba	<1	<1	NR	0.1–1.2	<1	NR
Br	0.001–0.009	0.300–0.008	NR	0.1–2.7	0.01–0.051	0.41–5.88
Ca	<150	667–5761	143.30	283–1726	72–146	14.36–5432.6
Cd	<0.1	<0.1	0.013–0.016	<0.1	<0.1	<0.1
Ce	<0.05	<0.05	NR	0.005–0.048	<0.05	NR
Co	0.001–0.007	0.0008–0.007	0.007–0.131	0.001–0.037	0.002–0.006	NR
Cr	0.01–0.03	0.05–0.05	NR	0.03–0.70	0.01–0.05	NR
Cs	<0.005	0.001–0.008	NR	0.0004–0.0067	0.001–0.007	NR
Cu	<20	<20	0.01–0.43	<20	<20	0.07–7.09
Fe	0.4–2.6	2.6–4.0	3.55	2–43	1–4.24	0.42–360.4

Table 5 continued

El.	Crystal white sugar samples			Brown sugar samples		
	Brazilian samples (mg kg^{-1})	Other countries samples (mg kg^{-1})	Literature [35, 47] (mg kg^{-1})	Brazilian samples (mg kg^{-1})	Other countries samples (mg kg^{-1})	Literature [12, 14] (mg kg^{-1})
Hf	0.001–0.011	0.001–0.012	NR	0.0005–0.0080	0.001–0.011	NR
Hg	<0.01	<0.01	NR	<0.01	<0.01	9 ± 0.9
K	0.2–5.9	0.007–774	91.16	177–5044	26–119	18.11–37510
La	<0.02	<0.002	NR	0.001–0.019	0.0003–0.0020	NR
Na	1–3	0.0055–198	NR	0.4–10.5	0.3–3.4	NR
Nd	<0.1	<0.1	NR	<0.1	<0.1	NR
Rb	<0.2	8.4–1.0	NR	<0.02	0.03–0.11	NR
Sb	0.001–0.008	0.001–0.009	NR	0.0005–0.0083	0.001–0.0045	12 ± 1.6
Sc	0.0001–0.0002	0.001–0.0002	NR	0.0001–0.0036	0.0003–0.0015	NR
Se	<0.02	0.01–0.18	NR	<0.02	0.004–0.058	153 ± 16
Sm	<0.0002	<0.0002	NR	0.3–3.2	0.001–0.00043	NR
Sn	<2	<100	NR	<100	<100	NR
Sr	<100	2–38	NR	0.3–3.2	0.4–2.3	NR
Th	<0.004	<0.004	NR	0.0004–0.0039	<0.004	NR
Zn	1–26	0.001–0.021	1.25	1–29	2–25	0.01–10.85

NR not reported; < limit of detection

Table 6 Maximum elemental concentrations determined in sugar samples consumed in several countries and the maximum elemental values foreseen in the legislation

El.	Brown sugar (mg kg^{-1})	Crystal white sugar (mg kg^{-1})	International legislation [48]			Brazilian legislation [49, 50] ANVISA (mg kg^{-1})
			Brown sugar ICUMSA 800/1200 (mg kg^{-1})	Cane sugar ICUMSA 45 (mg kg^{-1})	Beet sugar ICUMSA 45 (mg kg^{-1})	
As	0.29 ± 0.01	0.021 ± 0.001	0.5	0.5	1.0	1.0[b]
Cd	<1	<1	0.05	NF	NF	1.0[b]
Cr	0.70 ± 0.03	0.41 ± 0.05	NF	NF	NF	0.1[a]
Cu	<20	<20	1.0	NF	3.0	30.0[b]
Hg	<0.01	<0.01	1.0	NF	NF	0.01[b]
Sb	0.0083 ± 0.0005	0.015 ± 0.001	NF	NF	NF	2.0[b]
Se	0.058 ± 0.004	0.18 ± 0.01	NF	NF	NF	0.3[b]
Sn	<100	<100	NF	NF	NF	150.0[b]
Zn	51 ± 2	61 ± 2	3.0	NF	NF	50.0[a]

El. element; *NF* not found

[a] 1965

[b] 1

foreseen for each type of sugar in the international and Brazilian legislations are shown in Table 6 for As, Cd, Cr, Cu Hg, Sb, Se, Sn and Zn. The elements Au, Br, Ca, Co, Cs, Fe, Hf, K, La, Na, Rb, Sc, Sm, Sr and Th had their concentration determined in the samples, however there are no limits foreseen for them in the legislation.

Conclusions

The technique neutron activation analysis was used in this study applying the k_0 method and the new methodology established at CDTN/CNEN, LS-NAA, making the analysis more efficient. Granulated sugar was analyzed as small and larger samples. More elements were determined in the

larger sample while in small ones the results reached only the detection limit.

The satisfactory performance of the method applied was pointed out by the results of E_n number calculated for IAEA-SOIL-7, analyzed as small sample (200 mg) and larger sample (5 g), and for GBW 0805 analyzed as small sample.

Concerning the sugar samples collected in Brazil and in other countries, they presented several elements—As, Au, Br, Ca, Co, Cr, Cs, Fe, Hf, K, Na, Rb, Sb, Sc, Se, Sr and Zn in the samples of granulated sugar and in brown sugar, the same elements besides Ba, Ce, La, Sm and Th. However, the international and Brazilian legislations present limits foreseen only for As, Cd, Cr, Cu, Hg, Sb, Se, Sn and Zn, other elements as Au, Ba, Ce, La, Sm, Sr and Th are not in the list of essential or probably essential elements for humans, meaning that they are potentially toxic [52, 53]. Related to the legislation, only Cr and Zn presented results higher than the values foreseen. More samples should be analyzed in order to have a better assessment of the quality of the sugar available for consumers in public places.

The consumption of products obtained naturally is a trend among consumers who care about the nutrition. Many people prefer to use brown sugar instead of white sugar because of the idea that this sugar is more natural. In fact, this kind of sugar is not submitted to industrial processes that crystal and refined sugar undergo. Due to this, brown sugar presents a wider variety of elements not considered essentials for human being, like rare-earth elements and thorium, in its composition. It was confirmed in this study because it presented higher elemental concentration and higher diversification of chemical elements. These results suggest that in spite of not being submitted to industrial processes, suggesting a natural and healthier product, the presence of chemical elements not considered essentials for human beings, as impurities make the brown sugar potentially unhealthy.

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