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THORIUM AND RARE EARTH ELEMENTS IN CRYSTAL AND BROWN SUGAR CONSUMED IN BRAZIL AND ARGENTINA

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ABSTRACT

Human exposure to contaminants in foods is a matter of general health concern. There is a growing interest in determine and quantify contaminants in food chain including natural radionuclides and rare earth elements (REE). Irradiation effects of radioactive nuclides and REE may cause lesions from their interaction with the human body. This study aimed to identify the presence of thorium and rare earth elements in crystal and brown sugar samples available for consumption in Brazil and Argentina. To determine the chemical elements, the 5g-sample methodology established at CDTN/CNEN, in Belo Horizonte, using the neutron activation technique, k_0-method, was applied. The element Sm was determined in crystal sugar samples analyzed that were available to consumption in both countries. Similarly to the brown sugar samples which presented La, Sc and Sm. The elements Ce and Th were found in brown sugar sample available to consumption in Brazil. Thus, the detection of these elements in sugar samples is important insofar as the increasing consumption of sugar around the world. The presence of impurities and its concentration may contribute to health issues to consumers.

1. INTRODUCTION

About fifty elements have been identified and quantified as part of biological components in living organisms. Among them it is possible to identify the trace elements that constitute a group of elements considered essential for transportation and maintenance of molecular interactions in the human organism [1]. According to Peereboom [2], trace elements are those elements which their concentrations in the human body are lesser than 100 mg. kg^{-1}.

The essential elements play an important physiological role as part of maintenance and fulfillment of molecular interactions. They are involved in proteins links, forming metalloproteins compounds or being part of the enzymatic processes [1]. Studies demonstrate that the deficiency, or even excess, of essential elements may be able to contribute to the

occurrence of diseases in humans [3]. Therefore, high concentrations of certain elements may be toxic, favoring the onset of diseases and injuries in human tissues [1, 4-6].

Such toxicity also can occur with other elements, considered non-essential, especially those whose properties can simulate the interactions with some substances as trace elements [7]. In this context are the rare earth elements (REE) that are not considered essential to the maintenance functions in the organism, or even highly toxic to the environment [8]. Studies have shown several interactions between REE and biological systems that comprising the human body [8-12].

The main source of essential and non-essential elements in human organism occurs by dietary intake. The food chain contamination is important entrance rote to elements in the body. This type of contamination is present due to vegetables growing in soils with such elements in its composition. The irrigation water used, the application of sewage sludge and the proximity of industrial and urban centers contributes to the deposition of these elements on soils [13].

The REE are, generally, found in a large variety of minerals such as phosphates, carbonates, fluorides and silicates [8]. Phosphate fertilizers are commonly contaminated by radioactive elements, mostly uranium, thorium and their daughters [14, 15] and depending on the concentration of radioactive elements in the fertilizer products, they may be able to lead toxicity to individuals that manipulate it, causing skin lesions, such as cancer [16, 17].

Human tissues such as blood, bone and brain are able to accumulate REE demonstrating their presence in human body [18-20]. Studies have showed that exposures to high concentrations of REE may cause health issues [21-23].

Studies have shown the presence of REE in soils and plants in regions close to places with high concentrations of REE [24, 25]. Zhao *et al*. [26] found that REE can be absorbed by sugarcane through the leaves in case of atmospheric exposure, and also through the roots due to the application of contaminated fertilizers in the soil. In both situations it is possible that the REE are redistributed through different parts of the vegetal.

The per capita availability of sugar increased, worldwide, in 32% between the years 1962 and 2000 [27]. The optimal sugar consumption should be about 10 kg/person/year [28]. In this sense, this study aims at identifying the presence of Th and REE in samples of crystal and brown sugar available to consumption in Brazil and Argentina applying the 5-g sample methodology by neutron activation analysis, k_0 method.

2. METHOD

2.1. Sample Collection

For the present study, samples of crystal and brown sugar were obtained by using bags of 3 to 5 g available in restaurants, snack shops and pubs randomly in Brazil and Argentina. However, it is not known the place where the sugar was produced or even the cultivation raw material. Analyzing the sugar composition, there is no difference between the sugars obtained from beet or sugarcane, because there is one hundred percent of saccharose on both. For this

reason, the producers are not required to inform on the labels of their products the origin of sugars, if it is from beet or sugarcane [29].

2.2. Analysis of the Samples

The samples were analysed by k_0-Instrumental Neutron Activation method [30], applying the larger sample methodology of analysis, recently established at CDTN [30], using the TRIGA MARK I IPR-R1 reactor, located at CDTN/CNEN, in Belo Horizonte [31].. For quality control, certified reference materials were analyzed, one of geological material, IAEA/Soil 7, and another of biological material GBW 0805, Tea leaves.

For analysis, 3-5g samples were weighed into suitable irradiation tubes. The irradiation time was 8 hours, enough to activate the isotopes. After the irradiation and suitable decay time, the samples were counted in a gamma spectrometry system, consisted of a gamma detector HPGe, relative efficiency 50%, with electronics associated and the program of spectra acquisition Genie 2000, CANBERRA. The gamma spectra were acquired by a necessary time so that they could reach a good counting statistics. The gamma spectra evaluation was carried out with the program HyperLab [33, 34] and the elemental concentrations were calculated by the program Kayzero for Windows® (*Kayzero for Windows for reactor neutron activation analysis (NAA) using the k_0 standardization method Version 2, User's Manual*, 2005).

3. RESULTS AND DISCUSSION

The results of the samples of certified reference IAEA/Soil 7 and GBW 0805, Tea Leaves, are presented in Table 1. It is possible to observe a good agreement between the experimental and the recommended values.

Table 1: Reference Materials– experimental results and recommended values for REE and thorium

	GBW 0805 (Tea leaves)		**IAEA/Soil7**	
Elements	**Recommended Values (mg kg^{-1})**	**Experimental Values (mg kg^{-1})**	**Recommended Values (mg kg^{-1})**	**Experimental Values (mg kg^{-1})**
Ce	0.686 ± 0.09604	0.74 ± 0.04	61 ± 6.50	59 ± 2
La	0.458 ± 0.0229	0.44 ± 0.02	28 ± 1.00	28 ± 1
Nd	NR	< 2	30 ± 6	28 ± 1
Sc	NR	< 0.01	8.3 ± 1.05	8.7 ± 0.3
Sm	NR	< 1	5.1 ± 0.35	4.90± 0.03
Tb	NR	< 0.2	0.6 ± 0.2	0.63± 0.03
Th	0.105 ± 0.0126	0.114 ± 0.004	8.2 ± 1.05	7.8 ± 0.3
Yb	NR	< 0.1	2.4 ± 0.35	2.3 ± 0.1

NR. Not reported.

Table 2 shows the range of results for REE and Th only for those elements that presented concentrations higher than detection limits.

Table 2. Elements determined in crystal and brown sugar samples.

Elements	Samples collected in Argentina		Samples collected in Brazil	
	Crystal Sugar (mg kg^{-1})	Brown Sugar (mg kg^{-1})	Crystal Sugar (mg kg^{-1})	Brown Sugar (mg kg^{-1})
Ce	< 0.05	< 0.05	< 0.05	0.005 – 0.048
La	< 0.002	0.0003 – 0.003	< 0.02	0.001 – 0.019
Sc	0.0001 – 0.0003	0.0002 – 0.003	0.0001 – 0.0002	0.0001 – 0.0036
Sm	< 0.0002	0.00004 – 0.0005	< 0.0002	0.3 – 3.2
Th	< 0.004	< 0.004	< 0.004	0.0004 – 0.0039

As a result, the presence of the La, Sc and Sm were observed in the samples of crystal sugar available to consumption in both countries. Brown sugar acquired in Brazil had higher concentrations of contaminants, Ce, and Th which were not detected in samples available in Argentina.

The brown sugar does not undergo the bleaching, crystallization or refining process [35, 36] which justifies the higher concentrations levels and the presence of different elements in brown sugar samples.

The element Th was only determined in brown sugar samples available in Brazil. Thorium is a tetravalent metal, as the U, its ionic size causes similar behavior to bivalent calcium [37]. Presenting similar behavior to Ca in the body, the tissue which suffers greater damage caused by natural Th is the bone [38]. Just small concentrations of Th are able to be stuck to the bone tissue through the bloodstream and can remain in the bones for years [39]. Intake of ^{228}Ra, which is absorbed from the gastrointestinal tract, has its deposition in bones where decays to ^{228}Th and ^{232}Th [40] and may cause bone cancer.

About the REE elements, Sm was determined in crystal sugar samples analyzed that were available to consumption in both countries. Similarly, the brown sugar samples presented La, Sc and Sm. The element Ce was detected in brown sugar sample available to consumption in Brazil.

Although REE concentrations in the earth's surface are small, like the Ce, the element with the highest concentration about, 66 $\mu g.g^{-1}$ [8, 25]. Some elements are found largely dispersed in a variety of forms, mostly as accessory minerals [8]. The element Ce is used in catalytic converters in automotive vehicles [41]. Cerium can act similar to calcium in human organism, so it may accumulate in the bone tissue [42] and also in the lungs and lymph nodes [43]. Cerium oxides are capable to cause lesions in the organism as bronchitis, pneumonitis, and granulomatous lesions [44, 45]. The elements Ce and La may cause pneumoconiosis [46].

Lanthanum is used extensively catalyzing the splitting of long chain hydrocarbon and carbon lighting applications [41]. The La is used therapeutically as lanthanum carbonate for treat hyperphosphatemia in patients with chronic kidney disease [47-48]. The kidney does not

suffer the toxic effects of the La, because its main route of excretion is through the liver [49, 50]. Lanthanum can be found in lung workers using this element as raw material, leading to pneumoconiosis [51]. The toxicity effects of lanthanum in nervous system are unclear. According to Zhu *et al*. [52] the long-term intake of lanthanides would cause changes in the conduction of nerve impulses.

The element Sc is a light material with a higher melting point. Therefore, its use is applied in different industrial sectors [41]. The Sc toxicity is observed mainly in developing tissues, when compared to mature tissues which are also susceptible to trivalent cations [53]. In rats, the scandium chloride was able to reduce the breathing pattern, causing tremors of extremities and fainting [54].

Like other REE, Sm has many applications in different areas. The Sm is used as a catalyst in plastics decomposition [55], and to dehydration and dehydrogenation of ethanol [56]. The interactions of samarium compounds in the human body are still controversial. Insoluble salts of Sm are non-toxic, while the soluble ones are only slightly toxic. From its entry into the gastrointestinal tract, about 0.05% of samarium salts is absorbed into the bloodstream. Then, about 45% goes to the liver, and 45% is deposited on the bones [41].

4. CONCLUSIONS

The presence of La, Sc and Sm was observed in the samples of crystal sugar available to consumption in both countries. Brown sugar acquired in Brazil had higher amounts of contaminants, containing the elements Ce and Th which were not detected in samples available in Argentina.

Many times, brown sugar is thought to be a natural product, with less quantity of contaminants. However, the fact that it doesn't undergo to industrial processes causes it to hold greater variety of impurities in its composition. Thus, it was the sugar that presented a higher variability of detectable chemical elements.

Anthropogenic actions have increased the presence of certain elements such as radionuclides and REE which may be transferred to air, water and soil where plants grow. Food intake contaminated with these elements is the main cause of its concentrations in the human organism. High concentrations of these elements in sugars, over time, can be detrimental to the health of people consuming products that have sugar in the composition.

This study [57, 58] constitutes a preliminary research. Thereby more samples need to be analyzed. Nevertheless it was possible to identify the presence of REE and Th as impurities in sugar samples available to consumption in Brazil and Argentina.

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