

Fluorine content of several brands of chocolate bars and chocolate cookies found in Brazil

Conteúdo de flúor em diversas marcas de chocolate e bolachas de chocolate encontradas no Brasil

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ABSTRACT: Chocolate bars and chocolate cookies are foodstuffs highly appreciated by children. The possibility of having fluorine (F) among their components, associated with an excessive consumption, may make them decisive contributors to the total daily F intake. Thus, they could participate in the establishment of dental fluorosis. The aim of this study was to analyze the fluorine concentration [F] of the chocolates bars (CB) Baton, Confeti, Garoto Ball, Kinder Ovo, M&M's, Milkybar, Nescau, Nescau Ball, Surpresa, Surpresa Bichos, Tortuguita; and of the chocolate cookies (CC) Danyt's, Hipopó, Nescau, Passatempo, Pokémon, Sítio do Pica-Pau Amarelo and Trakinas. Samples were purchased in Bauru, São Paulo, Brazil. Three grams of each product were previously ashed at 525°C (CB and cookies fillings) and at 550°C (cookies dough), during 4 hours. Fluorine was separated from the ash by hexamethyldisiloxane (HMDS)-facilitated diffusion. Fluorine analysis was carried out with the specific electrode. Mean [F]s \pm SD and amplitude (unit $\mu\text{g/g}$) were: CB = 0.30 ± 0.45 (0.07 – 1.60, n = 12) and CC = 1.08 ± 2.64 (0.04 – 7.10, n = 7). It was concluded that some of the analyzed foods may be important contributors to the total daily F intake. As for the product that had the highest [F] (Danyt's), when only 3 units are consumed just once a day, they may supply up to 40% of the maximum recommended daily F intake (0.07 mg/kg body weight) for a 2-year-old child (12 kg). The [F] in these products should be informed on their labels.

DESCRIPTORS: Chocolate; Fluorine; Fluorosis, dental; Child.

RESUMO: Chocolates em barra e bolachas de chocolate são guloseimas altamente apreciadas pelas crianças. A possibilidade de conter flúor (F) em seus componentes, associada a seu excessivo consumo podem torná-los contribuintes decisivos para a ingestão diária total de F. Assim, eles poderiam participar no estabelecimento da fluorose dental. O objetivo deste estudo foi analisar a concentração de flúor [F] dos chocolates em barra (CB) Baton, Confeti, Garoto Ball, Kinder Ovo, M&M's, Milkybar, Nescau, Nescau Ball, Surpresa, Surpresa Bichos, Tortuguita; e das bolachas de chocolate (CC) Danyt's, Hipopó, Nescau, Passatempo, Pokémon, Sítio do Pica-Pau Amarelo e Trakinas. Os produtos foram adquiridos em Bauru, São Paulo, Brasil. Três gramas de cada produto foram previamente calcinadas a uma temperatura de 525°C (CB e recheio das bolachas), e de 550°C (massas das bolachas), durante 4 horas. O F foi separado das cinzas por difusão facilitada por hexametildisiloxano (HMDS). As análises de F foram feitas com o eletrodo específico. As [F]s médias \pm DP e amplitude ($\mu\text{g/g}$) foram: CB = $0,30 \pm 0,45$ (0,07 – 1,60, n = 12) e CC = $1,08 \pm 2,64$ (0,04 – 7,10, n = 7). Concluiu-se que alguns dos alimentos analisados podem ser importantes contribuintes para a ingestão diária total de F. No caso do produto que apresentou a maior [F] (Danyt's), quando apenas 3 unidades são consumidas uma única vez ao dia, elas podem fornecer mais de 40% da ingestão diária máxima de F recomendada (0,07 mg/kg peso corporal) para uma criança de 2 anos de idade (12 kg). A [F] presente nesses produtos deveria ser informada em seus rótulos.

DESCRIPTORES: Chocolate; Flúor; Fluorose dentária; Criança.

INTRODUCTION

It has been suggested that the incidence and severity of dental fluorosis has become greater in the last decade in both optimally fluoridated and non-

fluoridated areas in many countries, as well as in Brazil^{5,11,18,23}. This has been attributed to an increase in the fluoride level of foods and beverages through processing with fluoridated water, to the

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inadvertent ingestion of fluoride toothpaste, and to the inappropriate use of dietary supplements¹⁹.

During infancy and childhood, the main sources of fluoride are considered to be commercially available beverages and foods. This period coincides with the calcification of different stages of the developing permanent teeth crowns. This is also a critical time for ensuring that the optimal levels of ingested fluoride are not exceeded. Several previous studies have determined the fluoride content of children foods, such as milk^{1,3,14,15,26,30}, dinners and desserts^{6,12,21,22,25,31} and beverages^{1,13,16}. However, these studies have been limited mainly to products manufactured in North America.

Little information is available on products from Brazil^{4,10,24}. The available studies were limited to a very small number of infant foods.

Chocolate bars and chocolate cookies are foodstuffs highly appreciated by children. Thus, the aim of the present study was to analyze the fluoride concentration [F] of chocolate bars and chocolate cookies commercially available in Brazil.

MATERIALS AND METHODS

Twelve product samples of chocolate bars (CB) and seven of chocolate cookies (CC) from different brands (Table 1) were purchased in Bauru, São Paulo, Brazil. The products were chosen because they are the most consumed by infants and young children. Foodstuff type, brand name, manufacturer's name and production site were recorded for each studied product.

Preparation and fluoride analysis

The products' packages were opened on the day of the analysis. CBs were frozen and then grated. Their fillings, when present, were separated. As for the CCs, the baked dough was separated from the fillings and ground. After this preparation, 3 g of each product was ashed for four hours, at 525°C for the CBs and fillings, and at 550°C for the dough of the CCs.

The ashes were dissolved in 3 ml of deionized water. Fluoride determinations were carried out after overnight hexamethyldisiloxane (HMDS)-facilitated diffusion using the ion-specific electrode (model 9609, Orion Research, Cambridge, MA, USA)²⁷. A set of standards (ranging between 0.200-6.400 µg F/ml) was prepared, using serial dilution from a 100 ppm NaF stock solution (Orion #940907) and diffused in triplicate in the same way as the samples. The millivoltage poten-

TABLE 1 - Chocolate bars and chocolate cookies analyzed.

Foodstuff type	Product name	Manufacturer	Production site
Chocolate bars	Baton	Garoto	Vila Velha
	Confeti	Kraft Foods	Curitiba
	Garoto Ball	Garoto	São Paulo
	Milkybar	Nestlé	Caçapava
	Nescau	Nestlé	Caçapava
	Nescau Ball	Nestlé	Caçapava
	Surpresa	Nestlé	Caçapava
	Surpresa Bichos	Nestlé	Caçapava
	M&M's Minis	EFFEM-Brasil, Mars, Inc.	Abreu e Lima
	Kinder Ovo	Ferrero do Brasil	Italy
	Tortugueta Caramel	Arcor	Bragança Paulista
	Tortugueta Strawberry	Arcor	Bragança Paulista
Chocolate cookies	Danyt's	Danone	Campinas
	Hipopó	Cory	Ribeirão Preto
	Pokemón	Cory	Ribeirão Preto
	Sítio do Pica-Pau Amarelo	Cory	Ribeirão Preto
	Nescau	Nestlé	Marília
	Passatempo Carinhas	Nestlé	Marília
	Trakinas	Nabisco	Piracicaba or Argentine

tials were converted to µg F using a standard curve with a coefficient correlation of $r \geq 0.99$. All samples were analyzed in duplicate. The mean reproducibility of the readings, based on the duplicate samples, was 94 percent.

RESULTS

Table 1 shows all the products analyzed, as well as the manufacturer. Table 2 shows the [F] determined for all the products. Fluorine concentration is expressed as µg/g. Mean [F] ± SD (amplitude, n) were 0.30 ± 0.45 (0.07 - 1.60, n = 12) and 1.08 ± 2.64 (0.04 - 7.06, n = 7) for the CBs and CCs, respectively. Most of the products had low [F],

TABLE 2 - Chocolate bars (CB) and chocolate cookies (CC) fluorine concentration ($\mu\text{g F/g}$).

Foodstuffs	CB/CC (mass)	CB/CC (filling)	Total [F]*
Baton			0.17
Confeti			0.07
Garoto Ball			0.21
Milkybar	0.12	0.09	0.11
Nescau			0.09
Nescau Ball			0.71
Surpresa			0.16
Surpresa Bichos	0.17	0.12	0.11
M&M's Minis			1.60
Kinder Ovo			0.23
Tortuguita Caramel	0.34	0.04	0.21
Tortuguita Strawberry	0.12	0.08	0.11
Danyt's	7.16	1.15	7.06
Hipopó	0	0.12	0.04
Pokemón	0.01	0.35	0.09
Sítio do Pica-Pau Amarelo	0.07	0.60	0.21
Nescau	0.03	0.13	0.05
Passatempo Carinhas	0.03	0.12	0.05
Trakinas	0.06	0.09	0.07

*When the product had filling, harmonic mean was calculated.

except one brand of CB (M&M's Minis) and one brand of CC (Danyt's) that had mean [F] of 1.60 and 7.06 $\mu\text{g/g}$, respectively.

DISCUSSION

Due to the increase in the prevalence and severity of dental fluorosis in both optimally fluoridated and non-fluoridated areas in many countries, as well as in Brazil, the increase in the consumption of fluoride from many sources, including foods and beverages, causes preoccupation. Many studies have demonstrated that it is necessary to know the fluorine concentration of infant foods, foodstuffs and beverages to estimate the total fluorine ingestion by children^{12,13,17}. Although it is difficult to precisely determine the total fluorine intake from the diet, it is clear that there is substantial variation in the intake of different foods, foodstuffs and beverages, and in the fluorine content of these products. Since products are not required to have their fluo-

rine content displayed, only through a fluorine assay it is possible to determine dietary fluorine intake.

It is considered that the optimal range of fluorine intake is 0.05 – 0.07 mg F/kg body weight/day^{2,11}. Most of the foodstuffs analyzed revealed no concentrations in excess of 0.40 $\mu\text{g F/g}$ so that these sources of fluorine appear to be acceptable, at least at present (Table 2). This is in reasonable agreement with the findings of Vlachou et al.³¹ who found fluorine concentrations between 0.01 and 0.31 $\mu\text{g F/g}$ for dairy products. However, high fluorine concentrations were found in CB M&M's Minis (1.60 $\mu\text{g F/g}$) and in CC Danyt's (7.06 $\mu\text{g F/g}$). We cannot rule out the possibility that the high fluorine concentrations found in these products may make them considerable contributors to the total daily fluorine intake. When 215 g of M&M's or 3 units of Danyt's, which contain approximately 0.210 mg of F, are consumed just once a day, they may provide up to 40% of the upper limit of the ranges believed to be associated with increased risk of enamel fluorosis (0.07 mg F/kg body weight/day) for a 2 year-old child, weighing around 12 kg^{2,11}.

Another possibility that must be considered is the concomitant ingestion of these foodstuffs with other fluorine-rich products. This is the case of a ready-to-drink chocolate milk (Toddyinho, Quaker, Guarulhos, SP, Brazil), which contains fluorine levels between 1.08 and 1.28 ppm⁴. Soymilk or even powdered or liquid concentrate infant formulas appear to have the same problem. In this situation, the total fluorine intake will be higher.

Buzalaf et al.⁴ tested two commercial brands of ready-to-drink chocolate milk. In one of them (Nescau, Nestlé, Caçapava, SP, Brazil), fluorine concentrations ranging between 0.150 and 0.170 ppm were found while in the other (Toddyinho, Quaker, Guarulhos, SP, Brazil), fluorine levels between 1.08 and 1.28 ppm were observed. Both products were produced in optimally fluoridated areas. The authors had previously detected high fluorine concentrations in Toddyinho²⁴. However, they were not able to identify the possible source of the fluorine. They found low fluorine concentrations (ranging between 0.05 and 0.09 $\mu\text{g F/g}$) in powdered chocolate. This may support the exclusion of cocoa as the possible fluorine source in foodstuffs that have chocolate among their components.

The bioavailability of fluorine from fluorine-rich products is an important issue to be considered. Trautner, Siebert²⁹ compared the bioavailability of fluorine from dietary sources and observed low bioavailability for most solid foods, such as fish and algal flour and for mammalian fish bones. The authors attributed this to the high calcium content of these products. The CC that showed the highest fluorine concentration in this study is calcium-rich. Thus, it is possible that this product may contribute little to dietary fluorine intake. This should be tested in bioavailability experiments in humans, ideally, or *in vitro*, simulating the gastric conditions.

Another factor that should be taken into account is that the presence of food bolus when fluorine is ingested may increase the degree of fluorine absorption by the stomach by increasing the residence time²⁸. There is also some evidence that high dietary fat levels may increase the absorption of ingested fluorine²⁰. High level of fat in the duodenum reduces the rate of gastric emptying, increasing fluorine absorption. Chocolate foodstuffs contain fat among their components and may, therefore, increase the degree of fluorine absorption. Hence, the degree of absorption of fluorine may be reduced if it is ingested simultaneously with foods containing appreciable amounts of divalent or trivalent cations, because insoluble complexes or precipitates are formed.

Another crucial factor when the association between fluorosis and infant foods is considered is the critical period of fluorine exposure for the development of fluorosis. Enamel fluorosis can occur following either an acute or chronic exposure to fluorine during tooth formation⁷. Fluorosed enamel is characterized by retention of amelogenins in the early-maturation stage of development and by the formation of a more porous enamel with sub-surface hypomineralization. Secretory enamel is believed to be more susceptible to acute fluorine exposure, and the transition/early maturation

stage of enamel formation is the most susceptible to chronic fluorine ingestion above the threshold levels. Evans, Stamm⁹ determined the occurrence of fluorosis in Hong Kong when the fluorine level in the drinking water was reduced from 1.0 to 0.7 ppm. The susceptibility of developing enamel in the various birth cohorts to the change in water fluorine concentration indicated that the permanent maxillary central incisors were most susceptible to fluorosis during a critical period of twenty-two to twenty-six months of age. For the permanent incisors, fluorine exposure during the months before this period carried less risk than did continued exposure for up to thirty-six months beyond this critical time that corresponds to the maturation phase of permanent incisor development. In a subsequent report concerning the same population, Evans, Darvell⁸ refined the estimate of the critical period for susceptibility to enamel fluorosis in permanent maxillary central incisors. The authors concluded that the maxillary central incisor, as a whole, appears mostly at risk of fluorosis from dietary fluorine between the ages of fifteen and twenty-four months for males, and between the ages of twenty-one and thirty months for females. Hence, the critical period for the anterior permanent teeth would be after the first twelve months of life. At this time, there is a high fluorine intake from infant foods and beverages.

CONCLUSION

This work showed that some chocolate bars and chocolate cookies may be important contributors to the total daily fluorine intake, especially when combined with other fluorine sources. Thus, their consumption by children who are in the period of tooth formation, and consequently are in risk of developing dental fluorosis, should be controlled. The product labels should provide information on their fluorine content in order to permit pediatric dentists and doctors to advise about their consumption by patients.

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