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Fluoride and aluminum in teas and tea-based beverages

Flúor e alumínio em chás e bebidas à base de chás

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Keywords
Fluorine, analysis. Aluminum, analysis. Beverages, analysis. Fluorosis, dental. Tea-based beverages.

Abstract

Objective
To evaluate fluoride and aluminum concentration in herbal, black, ready-to-drink, and imported teas available in Brazil considering the risks fluoride and aluminum pose to oral and general health, respectively.
Methods
One-hundred and seventy-seven samples of herbal and black tea, 11 types of imported tea and 21 samples of ready-to-drink tea were divided into four groups: I-herbal tea; II-Brazilian black tea (*Camellia sinensis)*; III-imported tea (*Camellia sinensis)*; IV-ready-to-drink tea-based beverages. Fluoride and aluminum were analyzed using ion-selective electrode and atomic absorption, respectively.
Results
Fluoride and aluminum levels in herbal teas were very low, but high amounts were found in black and ready-to-drink teas. Aluminum found in all samples analyzed can be considered safe to general health. However, considering 0.07 mg F/kg/day as the upper limit of fluoride intake with regard to undesirable dental fluorosis, some teas exceed the daily intake limit for children.
Conclusions
Brazilian and imported teas made from *Camellia sinensis* as well as some tea-based beverages are sources of significant amounts of fluoride, and their intake may increase the risk of developing dental fluorosis.

Descritores
Flúor, análise. Alumínio, análise. Bebidas, análise. Fluorose dentária. Bebidas à base de chá.

Resumo

Objetivo
Avaliar as concentrações de flúor e alumínio em chás brasileiros de ervas, preto e bebidas a base de chá, como em chás preto importados considerando seus riscos para, respectivamente, a saúde oral e geral.
Métodos
Foram analisadas 177 amostras de chá de ervas e chá preto, 11 tipos de chá preto importados e 21 amostras de bebidas à base de chá. Flúor e alumínio foram determinados após infusão dos chás de ervas e pretos. As determinações de flúor e alumínio foram feitas por eletrodo específico e absorção atômica, respectivamente.
Resultados
A quantidade de flúor e alumínio encontrada nos chás de erva foi muito baixa, mas

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foi alta nos chás preto e nas bebidas à base de chá. A quantidade de alumínio em todas amostras analisadas pode ser considerada segura para a saúde geral. Entretanto, considerando 0,07 mg F/kg/dia como o limite superior de risco de fluorose esteticamente comprometedora, alguns chás preto e algumas bebidas à base de chá podem por si só submeter crianças a uma dose diária superior ao limite.

Conclusões

Chás preto brasileiros e importados, assim como bebidas à base de chá são fontes de quantidades significantes de flúor e sua ingestão diária pode aumentar o risco de fluorose dental

INTRODUCTION

Fluoride is a recognized substance used worldwide to control dental caries. However, the ingestion of high fluoride amounts during tooth formation and mineralization is also responsible for dental fluorosis. The upper limit for clinically acceptable dental fluorosis development is not well known, but the value of 0.05-0.07 mg F/kg body weight/day[2] is generally accepted as a reference. Aluminum has been associated with some neurological disorders[16] and the limit dose of 1 mg Al/kg body weight/day is considered an acceptable daily intake.[7]

Tea (*Camellia sinensis)* is a naturally rich source of fluoride[10] and other components including aluminum.[8] The tea plant absorbs fluoride and aluminum from acid soil by passive diffusion, which are accumulated in the leaves during the plant's life span.[18] In countries where regular tea consumption is culturally determined, tea plays an important role in triggering fluoride undesirable effects on tooth formation. In some Chinese villages, where dental fluorosis is endemic, fluoride intake from tea has been strongly correlated to this condition.[3,10]

Tea consumption has also spread all over the world through the industrialization of tea-based beverages. Thus, the concern about daily intake of fluoride and aluminum should not be limited to consuming tea infusions prepared at home but include the wide variety of ready-to-drink flavored teas now available to the public, and so attractive to children and youngsters. Moreover, recent dietary studies indicated that water intake among children has declined, whereas consumption of soft drinks and juices has increased,[13] which would inevitably include tea-based beverages. However, information on fluoride and aluminum concentration in these products is scarce.

In the past, tea used to be grown in natural soil but nowadays fertilizers are used to boost production. It is known fluoride and aluminum concentration in tea depends on the soil,[12] and plants can take up more fluoride when the soil is fertilized.[20]

Studies have focused primarily on black tea.[4,9] However, there is a wide variety of tea and herbal tea now available with no data on fluoride and aluminum concentration. In addition, recent publication has showed that the daily fluoride intake by Brazilian children from diet and dentifrice (0.09 mg F/kg) is 30% higher than the safe threshold for fluoride exposure to avoid dental fluorosis.[14] Data on daily intake of Al in Brazil were not found in the literature indexed.

The purpose of this study was to determine fluoride and aluminum concentration in herbal, black, ready-to-drink and imported teas available in Brazil considering the risks fluoride and aluminum pose to oral and general health, respectively.

METHODS

Sampling

One-hundred and five samples of herbal tea (bags), 72 samples of Brazilian black tea (bags), 11 different types and brands of imported tea (bags) and 21 samples of ready-to-drink tea (200-350 mL cans) were purchased in Brazilian supermarkets and divided into 4 groups: I – Herbal tea; II – Brazilian black tea (*Camellia sinensis)*; III – Imported tea from USA, England, Canada, China, and Japan (Black, green and Oolong tea, all extracted from *Camellia sinensis*); IV – Ready-to-drink tea-based beverages (black and mate tea). Batch numbers of all types of tea were recorded.

Preparation of tea infusions

Tea infusions were prepared in the laboratory using three tea bags from each box. This number was considered representative because the coefficient variation between analyses was lower than 5%. Thus, simulating home tea preparation, 90 mL of distilled deionized water was brought to boil in a beaker, the

Table 1 – Estimated amount (mean SD; n=105) of fluoride and aluminum in one bag of herbal tea. Piracicaba, Brazil, 1999.

Herbal tea (Types)	mg/bag Fluoride	Aluminum
Chamomile	b.q.l.	b.q.l.*
Balm-mint	b.q.l.	b.q.l.
Anise	b.q.l.	b.q.l.
Other herbs	b.q.l.	b.q.l.
Flowers and fruits	b.q.l.	b.q.l.
Mate herb	0.0025 0.0010	b.q.l.

*b.q.l. – below quantification limit

heater was turned off and one tea bag was added to the beaker. After three minutes of infusion, the tea bag was taken out. When the solution reached room temperature, water was added in a volumetric flask to make up a 100 mL volume. Ready-to-drink teas were analyzed by taking samples directly from cans.

Fluoride and aluminum determinations

Fluoride was analyzed in all samples in duplicate using an ion specific electrode (Orion - 96-09) after buffering with the same volume of TISAB II (acetate buffer 1.0 M, pH 5.0 containing NaCl 1.0 M and 1,2 cyclohexanediaminetetracetic 0.4%). The electrode was calibrated with five standard solutions ranging from 0.03125 to 0.500 µg F/mL. The calibration curve (r=0.996) was tested against fluoride standard solutions (Orion) and blanks. When sample concentration was lower than the lowest limit of the curve, the value found was only considered as an approximation. When the concentration was higher than the highest value of the curve, dilutions were made and the analysis repeated. The sensitivity limit for fluoride detection was 0.02 µg /mL using the mV value of the blank solution as reference.

Aluminum was determined in duplicate by atomic absorption using nitrous oxide, acetylene flame and a hollow cathode lamp at 309.3 nm. The spectrophotometer (VARIAN – AA-50) was calibrated with five standard solutions ranging from 0.5 to 25.0 µg Al/mL. All samples were analyzed without any pre-treatment procedure. The sensitivity limit was 0.1 µg Al/mL.

Fluoride and aluminum concentrations in the samples were determined and converted into amount per bag or can of tea (mg F/bag and mg Al/bag). The results (mean ± SD) represent an estimated amount of fluoride and aluminum intake every time a cup (prepared with one bag) or one can of tea is consumed. Pearson's correlation between fluoride and aluminum in each sample group was also determined. In addition, the number of cups or cans of tea that would expose a 1 to 3-year-old child to the upper limit of F (0.05-0.07 mg F/kg body weight/day) and to the acceptable daily intake of aluminum (1.0 mg /kg body weight/day) was estimated.

RESULTS

Tables 1 to 5 summarize the results of all analyses in terms of mg fluoride and aluminum intake when one cup (prepared with one bag) or one can of tea is ingested. Table 1 shows that herbal teas showed very low amounts of fluoride and, in fact, most results were below the quantification limit (b.q.l.) for the analysis. The amount of aluminum in all herbal teas was below the quantification limit for the method used.

Brazilian black teas (Table 2), however, showed higher values of fluoride and aluminum ranging from 0.08 to 0.40 mg F/bag and 0.30 to 0.98 mg Al/bag, respectively. A broad range could be seen among different samples of black tea. The correlation between F and Al was statistically significant but low (r=0.36, p=0.002).

Table 3 also shows that fluoride and aluminum were found in detectable amounts in the imported tea analyzed. The correlation between fluoride and aluminum in this group was statistically significant high (r=0.85; p<0.001).

The amount (mg) of fluoride and aluminum per can of ready-to-drink teas is showed in Table 4. Mean fluoride was 0.54 mg but a broad range (0.01–1.91) was observed, which may be attributed to the low and high amounts in Matte D and Iced Tea E, respectively. Table 4 also shows a statistically significant high correlation (r=0.98; p<0.001) between fluoride and

Table 2 – Estimated amount (mean SD; n=72) of fluoride and aluminum in one bag of Brazilian black tea. Piracicaba, Brazil, 1999.

Brazilian black tea (Brands)	mg/ bag Fluoride	Aluminum
Brand A	0.34 0.032	0.53 0.089
Brand B	0.14 0.027	0.56 0.061
Brand C	0.13 0.026	0.51 0.069
Brand D	0.23 0.075	0.71 0.206
Brand E	0.13 0.034	0.49 0.136
Brand F	0.24 0.050	0.56 0.122
Mean	0.20 0.091	0.57 0.142
(Range)	(0.08-0.40)	(0.30-0.98)

Table 3 – Estimated amount (mean SD; n=16) of fluoride and aluminum in one bag of imported tea. Piracicaba, Brazil, 1999.

Imported tea (Types)	mg/ bag Fluoride	Aluminum
Black tea	0.33 0.146	0.88 0.223
Green tea	0.08 0.006	0.32 0.058
Oolong tea	0.33 0.01	1.03 0.024
Mean	0.30 0.147	0.84 0.282
(Range)	(0.07-0.57)	(0.28-1.25)

Table 4 – Estimated amount (mean SD; n=21) of fluoride and aluminum in one unit of ready-to-drink tea-based beverage. Piracicaba, Brazil, 1999.

Ready-to-drink tea (Brands)	mg/unit	
	Fluoride	Aluminum
Iced tea Brand A – 350 mL	0.15 0.015	0.57 0.057
Iced tea Brand B – 200 mL	0.12 0.019	0.29 0.069
Mate Brand C – 200 mL	0.11	0.26
Mate Brand D 340 mL	0.013 0.004	0.56 0.001
Iced Tea Brand E (Peach) – 340 mL	1.27 0.504	1.68 0.229
Iced Tea Brand E (Lemon) – 340 mL	1.58 0.152	1.80 0.256
Mean	0.54 0.663	1.04 0.695
(Range)	(0.01-1.91)	(0.26-1.96)

aluminum in ready-to-drink teas.

Table 5 shows the estimated minimum number of cups or cans of tea consumed that would expose a 1 to 3-year-old child weighing 13 kg to the upper limit of daily fluoride and to the maximum acceptable daily aluminum intake.

DISCUSSION

Tea, a traditional beverage originally from China, is prepared from the infusion of *Camellia sinensis* leaves. There are three different kinds of tea produced from the plant *Camellia sinensis* in the process of oxidation: green tea is non-oxidized, Oolong tea is semi-fermented and black tea is fully oxidized. Other plants infusions are not considered to be tea, but are called herbal teas. For example, "mate" is a very popular herbal tea in the south of Brazil and is prepared from leaves of the plant *Ilex paraguaensis*, which is a native of Paraguay.

In the study, the fluoride amount released from most herbal teas in the infusion process was below the quantification limit of the analytical method used (Table 1). This reflects these teas' composition, which consists of fruits, flowers, herbs, spices, and plants other than regular tea. The very low amount of fluoride in "mate" tea is in agreement with these authors' previous data.[5] Aluminum amount found in herbal teas was below the quantification limit of the analytical method used. Thus, the intake of fluoride and aluminum from herbal teas can be considered safe in terms of unwanted effects.

The amount of fluoride found in one bag of Brazilian black tea ranged from 0.08 to 0.40 mg F/cup (Table 2). Fluoride concentrations in the infusions (0.8 to 4.0 ppm F) are comparable with those reported in Brazil[5] and other countries.[4,9] The concentration range can be explained by factors such as leaf age, plant genetics, rainfall, altitude, fertilizer, type of soil, and even sample preparation.[9,18] In addition, commercially available teas are a blend from a variety of sources, which is reflected by the wide fluoride range in the infusions. The aluminum results, ranging from 0.30 to 0.99 mg Al/bag, are in agreement with those published in the literature[12] and such variation could be explained as for fluoride.

Regarding fluoride and aluminum in imported tea (Table 3), all types showed detectable amounts as they were extracted from the same plant family (*Camellia sinensis*). Green tea, which is most similar to the natural plant, showed lower fluoride content, probably because it merely consists of the tealeaf after a drying process. In the present study, Oolong and black tea showed similar and higher fluoride and aluminum content than green tea, respectively. It is likely that the oxidation process has a role in that, although no data were found about Oolong tea in the literature. When comparing Brazilian and imported black teas, the Brazilian showed a lower fluoride and aluminum content, and this agrees with previous studies published in Brazil[5] and may reflect soil conditions or plant origin.

Relevant results were found for ready-to-drink teas (Table 4) mostly in one brand. The consumption of one can/day of Iced Tea-E would expose a 1 to 3-year-old child to a higher amount of fluoride (mean of 1.45 mg) than the daily fluoride intake from water plus dentifrice, or even from fluoride supplements. The first explanation for such high amount of fluoride in this brand is that it could have been prepared with water containing a high concentration of natural fluoride. However, the fluoride amount in Matte D, which is produced by the same manufacturer, was low (Table 4) and also this beverage is made of mate leaves that do not accumulate fluoride.[5] Therefore, this would suggest that the high fluoride content

Table 5 – Estimated daily intake* (range) of cups or cans of tea to reach the upper limit dose for F and the tolerable daily intake for Al. Piracicaba, Brazil, 1999.

Tea	Fluoride	Aluminum
Brazilian black tea	2–13	14–43
Ready-to-drink tea	1–90	7–50
Imported tea	2–13	11–47

*For a 1 to 3-year-old child

could have originated from the tea extract. In fact, after analyzing the tea source from the manufacturing company, it was found 1,384 µg F/g dry weigh. These data show that guidelines should be established to control fluoride concentration in dietary products in order to avoid dental fluorosis.

With regard to the correlation coefficient between fluoride and aluminum, it was found a value of 0.855 when all samples were considered (from groups II to IV), which is in agreement with other reports.[19] This can be explained by the fact that both elements form complexes in tea leaves.[11] The study data showed that there was a low correlation (0.368) only for Brazilian black teas (Table 2) and soil conditions where it is grown may explain that. There was another unexpected finding in the herbal tea "mate," which showed a low fluoride (Table 1) and unknown aluminum concentration. The study results show (Table 1) that Al was not detected in the infusion. However in ready-to-drink tea, it was found a high amount of aluminum (Table 4). These data suggest that either more aluminum is extracted from mate leaves in the industrialization process than by infusion at home or there is contamination from the can or during the industrialization process. Can contamination is a probable explanation because it has been reported in the literature.[1]

As for fluoride and aluminum intake and health risks, herbal teas are completely safe. The results for Brazilian and imported black tea are similar (Table 5), showing that aluminum intake does not represent a concern, even when taking into account heavy tea-drinkers that ingest about 2-6 cups of tea daily[9]. Furthermore, bioavailability of aluminum is very low,[6] probably because not much aluminum is potentially available for absorption.[17] With respect to fluoride intake from Brazilian black teas, this would not be a concern for children, because in Brazil only the adult population consumes this kind of tea. However, our data show that the ingestion of one cup of tea represents 50% of the upper limit dose of fluoride for a 1-3 year-old child, which would be a valid concern in countries where tea is consumed daily irrespective of age, because the bioavailability of fluoride from tea is high.[21]

With respect to ready-to-drink teas and health risks, the amount of aluminum intake is not of concern (Table 5). However, fluoride intake from these beverages is so high that daily consumption of one unit exceeds the upper limit in terms of risk to aesthetically acceptable dental fluorosis.

In conclusion, the amount of aluminum from tea consumption does not represent a hazard to general health according to the acceptable daily intake suggested by FAO/WHO.[7] However, Brazilian and imported teas as well as some ready-to-drink teas can effectively contribute to an intake of significant amount of fluoride, which is a concern with regard to dental fluorosis. These data are relevant because Brazilian children are exposed to fluoride from water and dentifrice at a higher dose than the upper risk limit of dental fluorosis.[14] However, the present findings should not be considered a local problem limited to Brazil because recent communication[15] showed that iced-tea in the US could provide 2-3 mg of fluoride per day if moderately consumed.

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