applied sciences

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Fluoride Risk Assessment from Consumption of Different Foods Commercialized in a European Region

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Received: 29 August 2020; Accepted: 15 September 2020; Published: 21 September 2020

Abstract: Fluoride is a halogen found in soil and water from natural and anthropogenic sources. Foods, such as cereals, fruits, and vegetables, among others, absorb and accumulate fluoride. High intakes of this element produce toxic effects such as dental or skeletal fluorosis. Fluoride content was determined in a total of 144 samples from different food groups (cereals and derivatives, fruits, tree nuts, dry fruits, mushrooms, vegetables, and legumes) using selective fluoride ion potentiometry. The fluoride concentration stood out in almonds (3.70 ± 0.96 mg/kg), walnuts (3.53 ± 0.62 mg/kg), bread (2.54 ± 0.85 mg/kg), and rice (2.28 ± 0.93 mg/kg). Consumption of 236 g/day of bread or 263 g/day of rice represents 100% of the recommended daily intake (DRI) set at 0.6 mg/day for children aged 1–3 years. In the case of rice consumption by children of these ages, it is recommended to use bottled water for rice preparation. The consumption of the analyzed foods by teenagers and adults does not pose a health risk.

Keywords: fluoride; human exposure; risk assessment; food

1. Introduction

Fluorine is a highly electronegative chemical element necessary for the human organism. It is widely distributed in nature as part of minerals, as well as in water, where it is found in its ionized form [1]. The main sources of fluoride in the environment are natural, such as volcanic emissions and marine aerosols [2]. Volcanic regions or regions with geothermal activity present high levels of fluoride in soils and water [3,4]. However, anthropogenic activities such as the use of pesticides, sewage, or industrial discharges cause an increase in fluoride concentration in the environment [5,6].

Fluoride is absorbed by the gastrointestinal route [7,8]. Thus, diet is the main fluoride source. Plant foods, such as cereals, fruits, and vegetables, among others, absorb fluoride mainly from the soil and irrigated water. Kudo and Garrec [9] showed that after adding 22 mg/L of fluoride in an experimental tank to aquatic plants, the concentration of fluoride in these plants increased up to 35 times in 24 h. The absorption and accumulation capacity of fluoride in plant organisms depends on factors such as soil type and pH. The absorption process is favored in soils with acidic pH due to fluoride's ionic form [10]. Food fluoride levels vary considerably from country to country [11].

Consumption of plant-based foods, such as cereals, fruits, and vegetables, can lead to high intakes of fluoride [12,13]. Fluoride is an essential element that is necessary for enzymatic reactions in the body, as well as in the reduction of dental caries topically [14–16]. The European Food Safety Authority (EFSA) established recommended daily intake (DRI) values for fluoride according to age and sex [17]. The recommended daily intake is defined as the minimum required amount of a nutrient or essential element for the proper functioning of the organism [18]. However, a prolonged and high intake of fluoride over time can trigger a series of toxic effects such as myopathy, neurological damage, dental fluorosis, or skeletal fluorosis [2,19]. Currently, Europe does not have an acceptable daily intake limit (ADI) for this ion. However, the Food and Nutrition Board of the American Organization Institute of Medicine established ADI values according to age and sex [20]. The acceptable daily intake is defined as the estimate of the total amount of a chemical substance or element contained in food and/or drinking water that can be ingested daily throughout the life of a person, without appreciable risk to health [18].

2. Material and Methods

The material used was washed with laboratory detergent (Alconox, Alconox Inc., NY, USA) and Mili-Q quality distilled water obtained from a filtration system (Millipore, Merck KGaA, Darmstadt, Germany). The use of glass material was avoided to prevent any interferences with fluoride.

2.1. Samples

A total of 144 samples of different food types were analyzed. The samples were acquired in large commercial areas and supermarkets on the island of Tenerife (Canary Islands, Spain). The samples were processed immediately after their acquisition. Table 1 shows the number of different samples analyzed.

Table 1. The number of analyzed samples according to the type of food.

Group	Food Type	No. Samples
Cereals and derivatives	Rice	8
	Gofio	8
	Corn	8
	Bread	8
Fruits	Citrus	8
	Apples	8
	Bananas	8
Tree nuts	Almonds	8
	Walnuts	8
Mushrooms	Mushrooms	8
Vegetables	Onions	8
	Lettuce	8
	Potato	8
	Tomatoes	8
	Carrots	8
Legumes	Bean	8
	Lentils	8
	Chickpeas	8

2.2. Solutions

The different solutions used in the treatment of the samples were as follows: 10–1 M fluoride solution was obtained by dissolving 4.199 mg of NaF (Sigma Aldrich, Steinheim, Germany) in 1 L of distilled water. Buffer solution TISAB (Total Ionic Strength Adjustment Buffer) with CDTA (Cyclohexanediaminetetraacetic Acid Monohydrate) was obtained by dissolving 58 g of NaCl (Sigma Aldrich, Steinheim, Germany), 57 mL of CH_3-COOH (Sigma Aldrich, Steinheim, Germany), and 4 g of 1,2-diamine-cyclohexane tetraacetic acid (DCTA) in 500 mL of deionized water. The pH was adjusted to 5–5.5. We obtained 8 M NaOH solution (Sigma Aldrich, Steinheim, Germany) by dissolving 320 g of NaOH (Sigma Aldrich, Steinheim, Germany) in distilled water to a total volume of 1 L, and 12 M HCl solution (Sigma Aldrich, Steinheim, Germany) was obtained by diluting 493.2 mL of HCl (Sigma Aldrich, Steinheim, Germany) in 500 mL of distilled water.

2.3. Treatment of the Samples

Previously homogenized samples of 0.50 g each were weighed in nickel crucibles (0.6 mm thick and 50 mL capacity). The samples were subjected to digestion in basic medium [21–23], adding 8 mL of 8 M NaOH and were left in an oven (Nabertherm, Lilienthal, Germany) for 24 h at 80 °C. Subsequently, they were placed in a muffle furnace (Nabertherm, Lilienthal, Germany) with two temperature-time programs, the first being 16 h at 200 °C, and the second being 3 h at 525 °C. The ashes obtained were dissolved in distilled water and their pH was neutralized with a 12 M HCl solution. The content was filtered and transferred to a 25 mL plastic volumetric flask. Filtering was necessary to remove insoluble cation oxides such as Si, Fe, Al, Ca, and Mg, which could disrupt the measurement [21].

2.4. Analysis Method

Potential measurements were obtained using a digital potentiometer with a fluoride selective ion electrode (Crison, Bercelona, Spain), coupled with a magnetic stirrer.

The standard addition method was used to determine the fluoride concentration in the solution, measuring the fluoride concentration before and after the addition of a known amount of fluoride. Furthermore, 6 mL of TISAB-DCTA buffer solution was added to the solution in order to adjust the ionic strength [23] and the potential difference was measured. The fluoride molar concentration was obtained as follows:

$$\text{Fluoride concentration (M)} = \text{Cs} \cdot [\text{Vs}/(\text{Vp} + \text{Vs})]/[10^{\Delta E/S} - ((\text{Vp} + \text{Vs})/\text{Vp})] \quad (1)$$

where Cs is the fluoride concentration of the added solution (M), Vp is the volume of the analyzed solution (mL), Vs is the volume of the added solution (mL), ΔE is the the potential difference of the measured (mV), and S is the slope of the electrode (mV).

With the molecular weight of the ion, the volume of the solution, and the quantity of food initially used, the concentration of fluoride (mg/kg) was obtained for each analyzed food.

2.5. Statistical Analysis

The statistical analyses were performed using IBM Statistic SPSS 23.0 software (Statistical Package for the Social Sciences) (IBM, NY, USA) for Windows™.

The Kolmogorov–Smirnov and Shapiro–Wilk tests were applied to check if the analyzed data followed a normal distribution [24]. The Levene statistic was used to check the homogeneity of the variances for each of the analyzed parameters [25]. When the data followed a normal distribution, parametric tests were used through the one-way ANOVA (analysis of variance) test. For the data that did not follow a normal distribution, a non-parametric study was performed using the Kruskal–Wallis test and the Mann–Whitney U test.

These statistical analyses were performed to confirm the existence of significant differences between the different food groups in this study ($p < 0.05$) [26].

2.6. Dietary Intake Assessment

The dietary intake was assessed by calculating the estimated daily intake (EDI), calculated as:

$$\text{EDI (mg/day)} = \text{Fluoride concentration (mg/kg)} \times \text{Consumption} \quad (2)$$

Once the EDI was calculated, the contribution percentage was obtained considering the recommended and maximum fluoride values established by EFSA [17] and IOM (Institute of Medicine) [20], respectively.

$$\text{Contribution (\%)} = [\text{EDI/Guideline value}] \times 100 \quad (3)$$

3. Results and Discussion

3.1. Fluoride Levels in the Different Food Groups Analyzed

Table 2 shows the mean concentrations, standard deviations, and maximum and minimum values of the analyzed samples.

Table 2. Fluoride concentration, standard deviation, and maximum and minimum values found in the analyzed samples.

Group	Food Type	Concentration ± SD (mg/kg)	Max–Min
Cereals and derivatives	Rice	2.28 ± 0.93	3.62–0.53
	Gofio	1.44 ± 0.66	2.32–0.53
	Corn	1.10 ± 0.91	3.24–0.50
	Bread	2.54 ± 0.85	3.62–1.23
Fruits	Citrus	Not detected	
	Apples		
	Bananas	1.72 ± 0.73	1.98–0.86
Tree nuts	Almonds	3.70 ± 0.96	4.80–2.32
	Walnuts	3.53 ± 0.62	4.15–2.79
Mushrooms	Mushrooms	1.28 ± 0.29	1.59–0.86
Vegetables	Onions	0.01 ± 0.02	0.06–0
	Lettuce	0.08 ± 0.05	0.16–0.08
	Potato	0.99 ± 1.90	3.53–0
	Tomatoes	0.96 ± 0.55	2.14–0.40
	Carrots	0.04 ± 0.01	0.04–0
Legumes	Bean	1.05 ± 1.79	3.86–0
	Lentils	0.32 ± 0.66	1.84–0
	Chickpeas	0.68 ± 1.41	1.36–0

Walnuts were the food group in which the highest concentrations of fluoride were registered among all the groups studied. Almonds (3.70 ± 0.96 mg/kg) and walnuts (3.53 ± 0.62 mg/kg) registered the highest mean levels of fluoride. The highest concentration recorded in these foods may be due to the concentration of elements that occurs after desiccating tree nuts [27]. The New Zealand Total Diet Study (NZTDS), carried out in 2016 by the Ministry for Primary Industries (MPI), registered an average concentration of fluoride in almonds of 5.66 mg/kg, higher than that found in this study [28,29]. The differences in the origin of the sample have a considerable influence on the level of fluoride registered in the walnuts.

The studied cereals and derivatives presented the following descending concentration sequence: bread > rice > gofio > corn. The level of fluoride registered in bread (2.54 ± 0.85 mg/kg) and rice (2.28 ± 0.93 mg/kg) stood out considerably. Levels found by Liteplo et al. [10] in baked goods and cereals were between 0.04 and 1.85 mg/kg, lower than those registered in the present study. Battacharya et al. [30], in rice cultivated in two regions in western India, registered concentrations of 0.56 ± 0.14 mg/kg (Bankura) and 0.83 ± 0.19 mg/kg (Purulia); in both cases, these values are lower than those registered in the present study. Jha et al. [31] recorded higher levels in rice cultivated in other regions of India ranging from 6.7 ± 0.6 to 12.0 ± 0.9 mg/kg. The differences in the fluoride content recorded in the rice samples depend on the crop area.

In the fruit group, fluoride was detected only in bananas, registering an average level of 1.72 ± 0.73 mg/kg. Notably, banana is a fruit with a high percentage of water.

Statistical analysis revealed the existence of significant differences ($p < 0.05$) in the fluoride content between almonds and walnuts compared with the rest of the analyzed foods. Statistical differences were found between bread and rice compared with the rest of the foods.

3.2. Assessment of the Fluoride Intake

Table 3 shows the estimated daily intake of fluoride by type of food, as well as the percentages of contribution to the recommended daily intake (DRI).

The consumption of 100 g per day of the analyzed foods ensures contribution percentages of 0.17–61.7% of the DRI set at 0.6 mg/day for boys and girls aged 1–3 years [17]. The percentage of contribution from the consumption of almonds (61.7%), walnuts (58.3%), bread (42.3%), and rice (38.0%) stood out considerably. At these ages, boys and girls consume greater amounts of food such as bread and rice, with which the established DRI may be exceeded. Thus, with a consumption of 236 g/day of bread or 263 g/day of rice, 100% of the DRI set for this age group would be reached. Considering the maximum intake value was established by IOM [20] at 1.3 mg/day for children of 1–3 years, it would be necessary to consume 511 g/day of bread or 570 g/day of rice. In the case of rice, it is necessary to consider that its preparation needs water. Normally, tap water is used for cooking; therefore, the regions with high concentrations of fluoride in water, such as the Canary Islands, will have high levels of fluoride in cooked rice. It is recommended, in the case of children, that bottled water be used to prepare this type of food or to reduce the consumption of rice.

Adolescents (15–17 years old) and adults are the groups that record the lowest percentages of contribution to the DRI (under 13.2%). Therefore, these groups are not at risk of exceeding the fixed values of DRI and ADI.

Table 3. Values of estimated daily intake (EDI) and contribution percentages (%) to the dietary recommended intake (DRI).

Group	Food	Conc ± SD (mg/kg)	EDI * (mg/day)	Contribution (%) to DRI **											
				Children								Teens		Adults	
				1–3 years		4–6 years		7–10 years		11–14 years		15–17 years			
				Boy	Girl	Boy	Girl	Boy	Girl	Boy	Girl	Boy	Girl	Men	Women
Cereals and derivatives	Rice	2.28 ± 0.93	0.228	38.0		22.8	25.3	15.2	16.3	10.4	9.91	7.13	8.14	6.71	7.86
	Gofio	1.44 ± 0.66	0.144	24.0		14.4	16.0	9.60	10.3	6.55	6.26	4.50	5.14	4.24	4.97
	Corn	1.10 ± 0.91	0.110	18.3		11.0	12.2	7.33	7.86	5.00	4.78	3.44	3.93	3.24	3.79
	Bread	2.54 ± 0.85	0.254	42.3		25.4	28.2	16.9	18.1	11.5	11.0	7.94	9.07	7.47	8.76
Fruits	Banana	1.72 ± 0.73	0.172	28.7		17.2	19.1	11.5	12.3	7.82	7.48	5.38	6.14	5.06	5.93
Tree nuts	Almonds	3.70 ± 0.96	0.370	61.7		37.0	41.1	24.6	26.4	16.8	16.1	11.6	13.2	10.9	12.8
	Walnuts	3.53 ± 0.62	0.353	58.3		35.3	39.2	23.5	25.2	16.1	15.3	11.0	12.6	10.4	12.2
Mushrooms	Mushrooms	1.28 ± 0.29	0.128	21.3		12.8	14.2	8.53	9.14	5.82	5.57	4.00	4.57	3.76	4.41
Vegetables	Onions	0.01 ± 0.02	0.001	0.17		0.10	18.9	0.07	0.07	0.05	0.04	0.03	0.04	0.03	0.03
	Lettuce	0.08 ± 0.05	0.008	1.33		0.80	0.89	0.53	0.57	0.36	0.35	0.25	0.30	0.24	0.28
	Potato	0.99 ± 1.90	0.099	16.5		9.90	11.0	6.60	7.07	4.50	4.30	3.10	3.54	2.91	3.41
	Tomatoes	0.96 ± 0.55	0.096	16.0		9.60	10.7	6.40	6.86	4.36	4.17	3.00	3.43	2.82	3.31
	Carrots	0.04 ± 0.01	0.004	0.67		0.40	0.44	0.27	0.30	0.18	0.17	0.13	0.14	0.12	0.14
Legumes	Alubias	1.05 ± 1.79	0.105	17.5		10.5	11.7	7.00	7.50	4.77	4.57	3.28	3.75	3.09	3.62
	Lentils	0.32 ± 0.66	0.032	5.33		3.20	3.56	2.13	2.30	1.45	1.39	1.00	1.14	0.94	1.10
	Chickpeas	0.68 ± 1.41	0.068	11.3		6.80	7.56	4.53	4.86	3.10	2.96	2.13	2.43	2.00	2.34

* Considering a consumption of 100 g/day. ** DRI values set at 0.6 mg/day (1–3 years), 1 mg/day (boy, 4–6 years), 0.9 mg/day (girl, 4–6 years), 1.5 mg/day (boy, 7–10 years), 1.4 mg/day (girl, 7–10 years), 2.2 mg/day (boy, 11–14 years), 2.3 mg/day (girl, 11–14 years), 3.2 mg/day (boy, 15–17 years), 2.8 mg/day (girl, 15–17 years); 3.4 mg/day (men) and 2.9 mg/day (women) [17].

4. Conclusions

The foods with the highest levels of fluoride were almonds, walnuts, bread, and rice. Significant differences ($p < 0.05$) were detected in the fluoride content between almonds and walnuts compared with the rest of the foods, as well as differences between rice and bread compared with the rest of the analyzed foods.

Children between 1 and 3 years of age are more exposed to fluoride because they need lower amounts of this element and the consumption of some foods in this population group may be higher. Thus, the consumption of rice can cause a possible risk for the health of children (1–3 years) if tap water is used in the preparation of this food. This water can contain high levels of fluoride, which may lead to the excess of this element in the daily diet. It is recommended to reduce the consumption of rice in children from 1 to 3 years old or prepare rice with bottled water. The adolescent and adult population is not at risk because the consumption of 100 g/day of the analyzed food does not entail exceeding the DRI or ADI values.

Author Contributions: A.H. and J.R.J. conceived and designed the experiments; J.R.J. performed the experiments; Á.J.G. analyzed the data; Á.J.G. and A.H. contributed reagents/materials/analysis tools; S.P. and C.R. wrote the paper. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Conflicts of Interest: The authors declare no conflict of interest.

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