

# Evaluation of dietary exposure to fluoride from fruit juices and nectars in children and adolescents

## Abstract

Fluoride is present in a multitude of foods, mainly in water, but fruit juices and nectars are also relevant dietary sources, not only because of their high water content but also because of the high consumption figures, especially in children and adolescents. Fluoride is a beneficial hormetin to prevent the appearance of cavities but in high exposures it can pose a health risk. The European Food Safety Authority (EFSA) has established a maximum Tolerable Intake Level (UL) between 0.1 and 0.12 mg/kg weight/day depending on age. The objectives of this study were to determine the fluoride content in 90 samples of fruit juices and nectars representative of the Spanish market and to estimate and evaluate the dietary exposure to fluoride from this dietary source in order to characterize the risk in different consumption scenarios. Variables such as the production area, the flavor and the type of production (ecological vs. conventional) have been considered in the analysis. Determination by potentiometry using a selective ion electrode has shown that soft drinks from fruit juices are the ones that contain less fluoride (0.19 mg/L) compared to juices from concentrate (0.43 mg/L) and nectars (0.36 mg/L). Organic juices and nectars are richer in fluoride. For each of the three consumption scenarios (200, 400 and 600 ml/day) the Estimated Daily Intakes (EDI) of fluoride are higher for juices from concentrate followed by nectars and juices. The consumption of 200 ml/day supposes 0.086 mg/L (5.73% of the UL); 0.038 mg/L (2.53% of the UL) and 0.072 mg/L (4.8% of the UL) of fluoride in the case of juices from concentrates, nectars and juices, respectively. It is recommended to moderate the consumption of these drinks, especially in children, and to choose refreshing fruit juice drinks due to their lower levels of fluoride, and to promote an improvement in the labeling of these products, incorporating the content of this anion in order to improve consumer information.

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## Introduction

Diet is a source of exposure to multiple compounds of nutritional interest, but it is also a source of compounds of toxicological interest. The risks associated with exposure to these toxic compounds of dietary origin must be monitored and evaluated to ensure food safety. This risk analysis follows several phases such as hazard identification, hazard characterization, exposure estimation and risk characterization. In addition, the analysis of these risks must be accompanied by proper management and communication.<sup>1</sup>

Fluoride (F) is a highly electronegative ion naturally present in the environment and in food. F behaves like hormetin, that is, it is beneficial at low doses and toxic at high doses.<sup>2,3</sup> This anion has a beneficial role in the prevention of dental caries due to its great affinity for calcium, which facilitates its combination with hydroxyapatite, giving rise to fluorohydroxyapatite, a compound that is much more resistant to acid, allowing it to avoid demineralization of the teeth.<sup>4-6</sup> It also has a positive effect on the bones, preventing the development of osteoporosis.<sup>5</sup>

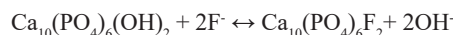
**Table 1** Tolerable Upper Intake Level (UL) for children and adolescents<sup>5</sup>

Age	7 to 11 months	1 to 3 years	4 to 6 years	7 to 8 years	9 to 14 years	15 to 17 years
UL (mg/day)	ND	1,5	2,5	2,5	5	7

\*ND: Not Available

**Table 2** Denomination of fruit juices and nectars<sup>10,11</sup>

Fruit juices from concentrate	The product obtained by reconstituting concentrated fruit juice as defined in point 3 with drinking water that complies with the criteria established in RD 140/2003 of 7 February 2003, which establishes the health criteria for the quality of water for human consumption.
Fruit nectar	The product capable of fermentation, but not fermented, which: (a) is obtained by the addition of water with or without the addition of sugars and/or honey to the products defined in points 1 to 5, to fruit purée, and/or to concentrated fruit purée, and/or to a mixture of these products; and (b) complies with Annex IV.



However, in regions of endemic fluorosis such as the Canary Islands or India, the population is chronically exposed to high levels of this hormetin, producing dental fluorosis characterized by loss of enamel shine and the appearance of white and opaque marks.<sup>7</sup>

In order to minimize the dietary risks derived from excessive intake, the European Food Safety Authority (EFSA) has established the Tolerable Upper Intake Level (UL) for the different age groups (Table 1) that is understood as the maximum amount of fluoride that can be ingested daily throughout life without posing a risk to the health of the consumer.<sup>5</sup>

Various food groups have been studied as dietary sources of fluoride, however there are few studies that assess the risk derived from the presence of fluoride in fruit juices and nectars.<sup>4,8,9</sup> Fruit juices and nectars can have different names depending on their ingredients or production procedures. In Spain, the specifications for each of them are included in Royal Decrees 781/2013 and 650/2011 (Table 2).<sup>10,11</sup>

Table Continued...

Refreshing fruit juice drink	<p>Refreshing drinks may contain any of the following ingredients, which shall comply with the relevant regulations: water for human consumption, prepared water, natural or spring mineral water, carbon dioxide, sugars, juices, purees, fruit or vegetable purees or mixtures thereof, compound syrup or basic preparation, fruit or vegetable extract or both, caffeine and quinine and authorised additives and flavourings.</p> <p>In particular, fruit juice drinks are characterised by the fact that they contain fruit juices, fruit purées, fruit or vegetable purees, fruit or vegetable separates or mixtures thereof.</p>
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The labeling of these products informs the consumer of the ingredients with which they have been made and also indicates their trade name, which provides a lot of information about the manufacturing process. However, the legislation does not include the obligation to indicate the fluoride content in these foods or the origin of the water used in its preparation.<sup>11</sup>

Regarding the profile of consumers, children and adolescents are identified as the population groups that consume the most fruit juices and nectars. According to the Report on Food Consumption in Spain, the Canary Islands is the Spanish region with the highest per capita consumption of these foods, being above the average (Figure 1).<sup>12-14</sup>

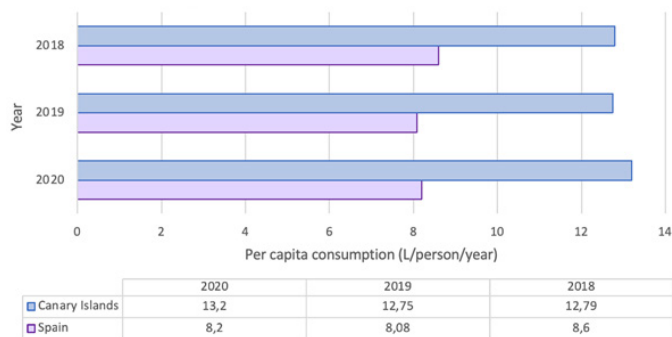


Figure 1 Data from the Food Consumption Report in Spain on juices and nectars.<sup>12-14</sup>

For all these reasons, the objectives of the study were to determine the levels of fluoride in different juices and nectars according to their flavor, trade name, and production (organic versus conventional); estimate the Estimated Dietary Intake (EDI) of F from them in different consumption scenarios; evaluate the risk based on the UL values established for the child and adolescent population and propose recommendations regarding consumption to minimize the risk.

## Material and methods

### Samples

90 samples of juices and nectars of three flavors have been analyzed between January and April 2020. The apple flavor (N=30) is marketed mainly in juice; peach (N=30) and pineapple (N=30) are marketed as nectar, juice and soft drink (Figure 2). Six commercial brands representative of the Spanish market were purchased, only one of which is locally produced (Tenerife). 15 of these 90 samples are organic juices made in the Spanish mainland, specifically in Murcia. Organic products produced in the Canary Islands were not analyzed because they were not marketed.

### Operative procedure

All the samples were shaken to homogenize their content before taking three 25 ml aliquots that were added to a plastic container with the help of a syringe made of the same material. Next, 5 ml of 0.75 M orthophosphoric acid (H<sub>3</sub>PO<sub>4</sub>) were added, which is the buffer solution responsible for adjusting the pH, ionic strength and eliminating interferences for subsequent analysis.<sup>15</sup> All the material

used was plastic and not glass because its composition can absorb fluoride and interfere with its measurable concentration.<sup>2</sup>

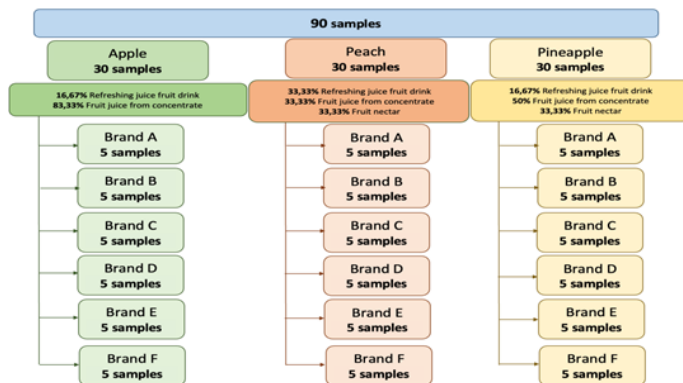


Figure 2 Analyzed samples classified according to flavor and brand.

### Fluoride determination

The analysis was performed using the potentiometric method with a selective fluoride ion (HACH, ISE F-9655C, Spain), whose quantification limits are 0.01-19000 mg/L, together with a potentiometer (CRISON, GLP22, Spain). To guarantee sample homogeneity, a magnetic stirrer (Selecta Agimatic-E, Spain) was used. First, the potential of the standard solutions whose concentrations ranged between 10<sup>-1</sup> and 10<sup>-5</sup> M of sodium fluoride (NaF) was determined, to obtain different calibration lines that convert the potentials of the samples into ion levels.<sup>16</sup> To prepare them, 0.428 g of NaF were weighed in the precision scale (METTLER TOLEDO, Spain) and dissolved in one liter of orthophosphoric, from which serial solutions were made.

### Statistic analysis

Statistical analysis was performed using the Graph Pad Prism 8.0.1 program to determine significant differences (p<0.05) based on flavor, trademark, trade name and type of production. As the data did not follow a normal distribution, a non-parametric independent variable test was applied, the Mann-Whitney test.

### Estimation and Evaluation of dietary exposure

The Estimated Daily Intake (EDI) has been calculated using Equation 1:

Equation 1 Calculation of the Estimated Daily Intake:

$$IDE = \text{Fluoride concentration (mg/L)} \cdot \text{Volume of juice/néctar consumed (L)}$$

For the evaluation of the EDI and the characterization of the risk, the contribution of the EDI to the UL of each population group under study has been estimated (Equation 2).

Equation 2 Calculation of the percentage of contribution to the UL

$$\%UL = \frac{IDE}{UL} \cdot 100$$

## Results and discussion

### Fluoride concentration

The results obtained for each flavour, trade name, origin and type of production are shown in Tables 3-6 and Figures 3-8. Pineapple juices and nectars have a higher concentration of fluoride (0.42 mg/L), although the maximum value was observed in apple juices (1.14 mg/L).

**Table 3** Fluoride concentration in juices and nectars of different flavors

Taste	N (90)	Fluoride (mg/L)		
		Media	Minimum	Maximum
Apple	30	0.36	0.11	1.14
Peach	30	0.31	0.06	0.72
Pineapple	30	0.42	0.2	1.1

**Table 4** Fluoride concentration by trade name

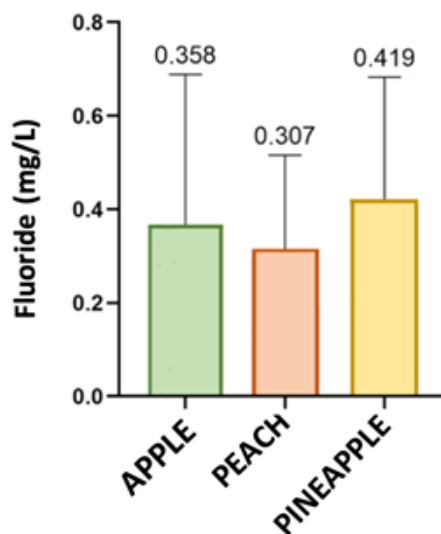
Trade name	N	Fluoride (mg/L)		
		Media	Minimum	Maximum
<b>Apple (N=30)</b>				
Fruit juice from concentrate	25	0.39	0.11	1.14
Refreshing fruit juice drink	5	0.18	0.17	0.2
Nectar	0	-	-	-
<b>Peach (N=30)</b>				
Fruit juice from concentrate	10	0.31	0.14	0.44
Refreshing fruit juice drink	10	0.19	0.06	0.33
Nectar	10	0.43	0.16	0.72
<b>Pineapple (N=30)</b>				
Fruit juice from concentrate	20	0.52	0.24	1.1
Refreshing fruit juice drink	5	0.21	0.2	0.22
Nectar	5	0.23	0.22	0.24
<b>Total samples (N=90)</b>				
Fruit juice from concentrate	55	0.43	0.11	1.14
Refreshing fruit juice drink	20	0.19	0.06	0.33
Nectar	15	0.36	0.16	0.72

**Table 5** Fluoride concentrations determined according to the place of manufacture of each flavour

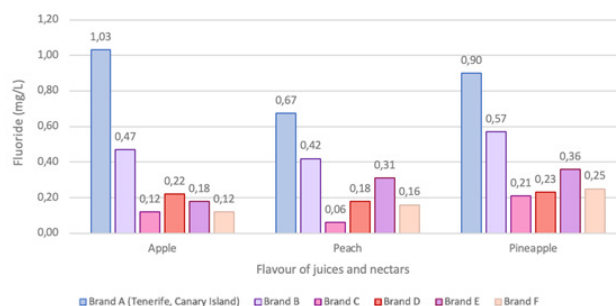
Location	N (90)	Fluoride (mg/L)		
		Media	Minimum	Maximum
<b>Apple</b>				
Tenerife, Canary Islands	5	1.03	0.99	1.14
Peninsula	25	0.22	0.11	0.49
<b>Peach</b>				
Tenerife, Canary Islands	5	0.67	0.62	0.72
Peninsula	25	0.23	0.06	0.44
<b>Pineapple</b>				
Tenerife, Canary Islands	5	0.9	0.77	1.1
Peninsula	25	0.32	0.2	0.79

**Table 6** Fluoride concentration in conventional and ecological fruit juices and nectars

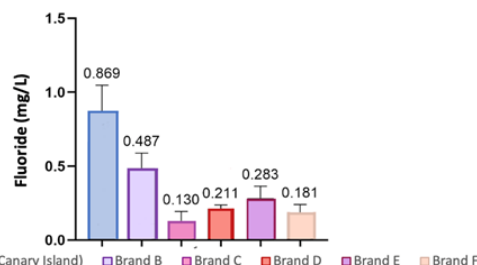
Type of production	N (60)	Fluoride (mg/L)		
		Media	Minimum	Maximum
<b>Apple</b>				
Ecological	5	0.47	0.44	0.49
Conventional	15	0.17	0.11	0.23
<b>Peach</b>				
Ecological	5	0.42	0.41	0.44
Conventional	15	0.22	0.14	0.33
<b>Pineapple</b>				
Ecological	5	0.57	0.43	0.79
Conventional	15	0.28	0.22	0.39



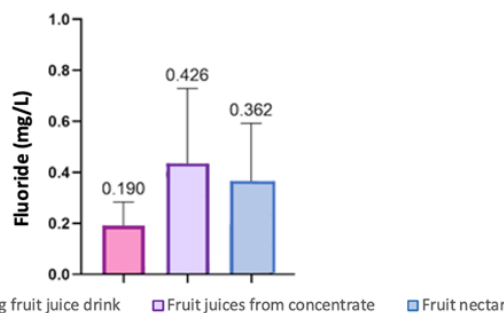
**Figure 3** Fluoride concentration in juices and nectars of different flavors.



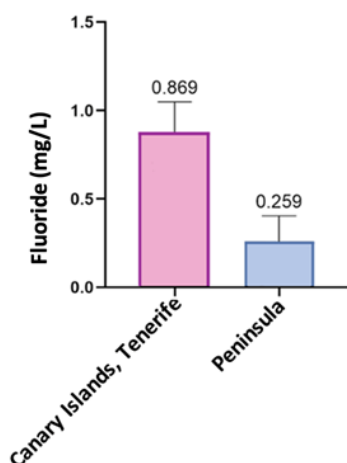
**Figure 4** Fluoride concentration in juices and nectars of three different flavors.



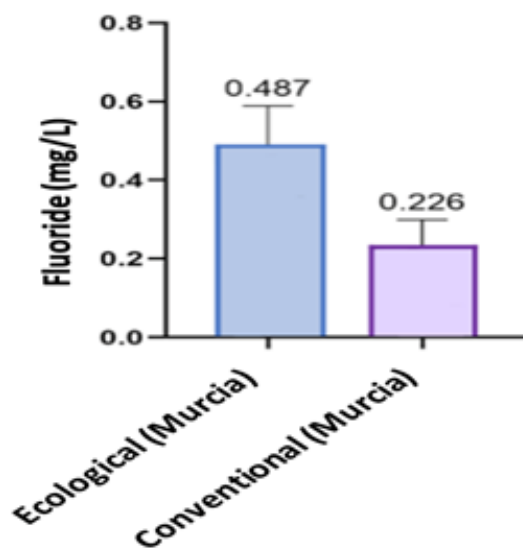
**Figure 5** Fluoride concentration in juices and nectars according to the commercial brand.



**Figure 6** Fluoride concentration determined in different types of fruit juices and nectars.



**Figure 7** Fluoride concentration determined in fruit juices and nectars depending on the place of manufacture.



**Figure 8** Fluoride concentration in juices and nectars according to the place of production.

In five of the six brands studied, the pineapple flavor has higher fluoride content (Figure 4). According to the statistical analysis it can be affirmed that there are significant differences between this flavor and the other two. Likewise, there are differences between the commercial brands and this may be due to the fact that they are not made in the same place, despite the fact that all except A and C are manufactured in Murcia (Spain), and the raw materials are not the same, as reflected in the list of ingredients (Figure 5).

The samples were also analyzed according to the commercial denomination of Royal Decrees 781/2010 and 650/2011 (Table 4, Figures 6). We have observed that the commercial denominations indicated on the products are not clear since there are occasions that give rise to errors because they incorporate terms that are not included in the legislation. Likewise, the fruit content only appears in nectars and soft drinks (50% of the samples) because it is required by law.

It is observed how the refreshing fruit juice drink, regardless of the flavor, is the one that contains the least fluoride, revealing significant differences with the others. This may be due to the fact that it contains

50% fruit juice or puree and other components mentioned in Table 2 that mean that the amount of water it contains is less than that of the other beverages because the fruit and the number of ingredients is less.

Table 5 and Figure 7 compare the levels of fluoride based on the origin of the samples and show the importance of the place of production. It is suspected that the water used in the production process determines the levels of fluoride in the final product. Those manufactured in the Canary Islands, specifically Tenerife, have higher levels of fluoride than those made in the Iberian Peninsula (mainland Spain). The Canary Islands is a volcanic region whose porous and permeable soil makes its water sources rich in this anion, whose levels range between 0.35-6.94 mg/L.<sup>17,18</sup> Four of the 4 samples originating from the Spanish peninsular territory are produced in Murcia, which is a region in which the waters have fluoride levels below 0.6 mg/L,<sup>19</sup> which would explain this relevant difference.

Mexico is another fluorosis-endemic region where research has been done on fluoride levels in different fruit juices manufactured by local companies. In the case of the apple flavor, the Canarian beverages (1.03 mg/L) present levels notably higher than the Mexican ones (0.34-0.87 mg/L) and with the pineapple flavor the levels double.<sup>4</sup> Therefore, despite both being regions known for their endemic fluorosis, the levels of fluoride in the juices and nectars produced on the island of Tenerife are higher. This can be derived from the high levels of fluoride in the waters of Tenerife (0.35-6.94 mg/L), levels slightly higher than those in some areas of Mexico (4.8 mg/L).<sup>18</sup>

The type of production was also taken into account. Regardless of flavour, organic juices and nectars are richer in fluoride (Table 6, Figure 8) which shows that organic products are not safer in terms of fluoride levels.

On the other hand, some authors have done research on juices and nectars specifically intended for children. In the case of peach nectar, the levels of fluoride previously observed (0.03 mg/L) are notably lower than those observed in the samples analyzed in our study (0.43 mg/L) and are characterized by not being specifically intended for this population. The fluoride content (0.11 mg/L and 0.04 mg/L) in different brands of apple juice is significantly lower than in beverages not intended for this specific public.<sup>20</sup>

### Exposure estimation and risk assessment

It has been estimated (Table 7) and evaluated (Table 8) the dietary exposure (EDI) and the risk of fluoride from the consumption of fruit juices and nectars in children and adolescents in three different consumption scenarios (200 ml/day, 400 ml/day and 600 ml/day). Regardless of the age of the individual, fruit juices from concentrate, followed by nectars, are the beverages that present a higher percentage of contribution to the UL because their fluoride levels are higher and this trend will be observed in the different consumption scenarios raised.

The consumption of 200 ml of fruit juice or nectar per day should not pose a risk for children and adolescents because the percentage of contribution in the different age ranges is similar to or less than 5%. This means that if the other foods in the diet that provide fluoride are considered, it is unlikely that the reference values will be exceeded.

Taking into account the recommendations of the Scientific Committee of "5 a day" that seeks to promote the consumption of five pieces of fruit and vegetables a day (one of these portions can be replaced by a glass (200-250 ml) of 100% juice or from concentrate, helping to maintain adequate hydration)<sup>21,22</sup> the consumer is not exposed to risk in terms of fluoride levels.

**Table 7** EDI by trade name for the different consumption scenarios

Trade name	EDI (mg/day)								
	200 ml/ day			400 ml/ day			600 ml/day		
	Media	Minimum	Maximum	Media	Minimum	Maximum	Media	Minimum	Maximum
Fruit juice from concentrate	0.086	0.022	0.228	0.172	0.022	0.228	0.258	0.066	0.684
Refreshing fruit juice drink	0.038	0.012	0.066	0.076	0.024	0.132	0.114	0.036	0.198
Nectar	0.072	0.032	0.144	0.144	0.064	0.288	0.216	0.096	0.432

**Table 8** Percentage of contribution to the UL according to the trade name for the different consumption scenarios

Age	% UL								
	Fruit juice from concentrate			Refreshing fruit juice drink			Nectar		
	200 ml/ day	400 ml/ day	600 ml/ day	200 ml/ day	400 ml/ day	600 ml/ day	200 ml/ day	400 ml/ day	600 ml/ day
7 to 11 months	-	-	-	-	-	-	-	-	-
1 to 3 years	5.73	11.46	17.2	2.53	5.06	7.6	4.8	9.6	14.4
4 to 6 years	3.44	6.88	10.32	1.52	3.04	4.56	2.88	5.76	8.64
7 to 8 years	3.44	6.88	10.32	1.52	3.04	4.56	2.88	5.76	8.64
9 to 14 years	1.72	3.44	5.16	0.76	1.52	2.28	1.44	2.88	4.32
15 to 17 years	1.23	2.46	3.69	0.54	1.08	1.63	1.03	2.06	3.09

If the daily consumption is 400 ml, there are situations where the contribution of fruit juices from concentrate and nectars to the UL exceeds or is close to 10% of the UL in children aged 1 to 3 years. In this age range, this intake could pose a risk if the rest of the diet is rich in this hormetin, especially in areas of endemic fluorosis such as the Canary Islands, where the consumption of water supplies can cause the amount of fluoride to which it is exposed. The individual is close to or above the tolerable upper level (UL). However, in the case of soft drinks made from fruit juices, the highest percentage of contribution is that observed in children aged 1 to 3 years and barely exceeds 5% contribution to the UL of F, practically half of what provided by the other two drinks.

In the case of a consumption of 600 ml/day, children between the ages of 1 and 3 years consume around 15% of the reference levels of F from these foods and those between 4 and 8 years old around 10%. With these contribution percentages, whether or not the child's health is affected will depend on the rest of the dietary sources of fluoride to which he or she is exposed.

In the case of adolescents in the three scenarios studied, the percentage contribution of these foods to the UL is significantly lower than that of the rest of the ages, since at no time does the contribution of F from these foods exceed 5% of the UL, except for the maximum values of juices from concentrate and fruit nectars, but does not exceed 10% of the UL.

Once the levels of fluorides in fruit juices and nectars have been determined, during risk management and communication the question may arise as to whether it is better to consume these beverages or water, either bottled or supplied. Water is the main source of fluoride, especially in the Canary Islands, where the levels detected in the water supply are so high that they are associated with toxic effects on the health of the consumer, since they exceed the parametric value of 1.5 mg. /L established by legislation.<sup>2,17</sup> It can be predicted that the consumption of fruit juices and nectars minimizes exposure to F because these foods, in any of the trade names, have lower levels of F than those of Tenerife's public water supply. If the consumption of bottled water is considered (depending on the brand, the F levels range between 0.24 and 0.62 mg/L) compared to the consumption of these juices and nectars, we can estimate that the consumption of bottled water provides fewer F concentrations that are lower than those of the juices and nectars analyzed.<sup>2</sup>

## Conclusion

Fruit juices and nectars should be considered an important dietary source of fluoride for children and adolescents, especially those under 8 years of age. Despite being made from fruit and nutritionally attractive, they can expose the child to risk if consumed in excess. Soft drinks made from fruit juices have lower levels of fluoride, so their choice should be prioritized over juices made from concentrate and nectars. In this sense, it is recommended that vending facilities located in schools follow this recommendation.<sup>23</sup>

The juices and nectars produced on the island of Tenerife (Canary Islands) have higher concentrations of fluoride than those produced on the mainland (continental Spain), so it is suspected that the geology of this volcanic archipelago and the richness of this anion in its waters condition F levels in these foods.

The Estimated Daily Intake (EDI) of fluoride from these foods in the different consumption scenarios proposed is lower than the value of the UL of F for all the age ranges studied. Only in children between the ages of 1 and 3 are higher percentages of contribution of the EDI to the UL detected, but in no case higher than 20%. However, the risk assessment based on the UL values for fluoride established for the child and adolescent population suggests reducing dietary exposure to fluoride by choosing refreshing fruit juice beverages over nectars and juices from concentrate. Likewise, it is recommended to limit the consumption of these packaged products to 200 ml/day in order not to reach high contribution percentages.

The labeling of these products could be improved because none of them indicate fluoride levels. We suggest regulating the obligation to indicate the fluoride content in these and other foods.

## Acknowledgments

None.

## Conflicts of interest

Authors declare that there is no conflict of interest.

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