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Fluoride Levels in Publicly Supplied and Bottled Drinking Water in the Island of Tenerife, Spain

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The interest of fluoride content in public water supply lies in its ability to prevent *dental caries* (Martín *et al.* 1991; Martín *et al* 1990). In fact, a proven correlation exists between communities that consume waters with moderate levels of fluoride (around 1.5 mg/L.) and lower rates of dental caries. Nevertheless, a relationship has been confirmed between consumption of water with high levels of ion fluoride and onset of fluorosis, a disease characterized by "mottled teeth". A correlation also seems likely between high fluoride intake and the onset of renal disorders, accompanied by bone decalcification - due to the sequestration by the fluoride of quantities of Ca^{2+} that are essential to the correct development of bones (Martín *et al.* 1991; Martín 1989; Sanz and Sanz 1998; Hardisson 1996).

There are settlements in Tenerife, mainly in the north of the island (La Guancha, Icod de los Vinos and San Juan de la Rambla), with cases of fluorosis. Their water supply shows an excess of this ion (Hardisson 1996; Rodríguez *et al.* 1995), the water coming from springs at the feet of high mountains or in areas next to geological deposits of marine origin (Martín 1989). In particular, the high fluorine content in the waters of Icod de Los Vinos and San Juan de La Rambla is due to their being supplied by the Vergara ravine gallery, in La Guancha borough.

On the other hand, the continuous growth of the island population, together with the remarkable reduction of rainfall in our islands, increases the water needs of the province. This creates technical and health problems, and forces one to pay special attention to the water issue, in general (Hardisson *et al.* 1998), and to the water fluoride concentration, in particular.

The reason for this is that drinking water is the main intake path for that ion, as well as the cheapest and most efficient way to provide optimal levels of fluoride to most of the population (Martín 1989; Mella *et al.* 1992; Mella *et al.* 1994; Jiménez and Bernal 1986). Apart from the public water supply, bottled water is an increasingly popular source. It is thus necessary to monitor the fluorine reaching the population from this source (Jiménez and Bernal 1986; Martín *et al.* 1992; Martín et al. 1993).

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The study carried out by Martín et al. (Martín *et al.* 1990) showed that the mean fluorine content in the most consumed waters of the Canary Islands was 1.47 mg/L. The same authors found, in subsequent studies, that the mean fluorine content in beer and soft drinks was 0.49 mg/L and 0.40 mg/L, respectively (Martín et al. 1992).

We present a study that shows the high concentrations of fluorine in the waters of the district health of Icod de Los Vinos (Tenerife Island), zone that presents an endemic fluorosis. Also, we have analyzed the contents of fluorine in the bottled waters of great consumption in the island.

MATERIALS AND METHODS

Forty-two water samples were analyzed, 21 of which came from the public water supply of three boroughs located in the north of the island of Tenerife. The three boroughs are dental fluorosis areas where efficient reduction of the high concentration of this anion in the public water supply has not been achieved. We used polyethylene bottles to take and preserve the samples for fluoride analyses, not using in any case an excess of bleaching agent (sodium thiosulfate). The remaining 21 samples came from bottled drinking water, of different origin and characteristics: 14 classified as bottled mineral water, 3 classified as bottled spring water, 3 as prepared bottled drinking water, and 1 as medicinal-mineral water. They were acquired in food stores in Tenerife, selecting the more popular brands.

The solutions used for the determination of fluoride in public supply and bottled waters were: 10^{-2}M fluoride standard solution; TISAB-CDTA solution, of the following composition: 58 g of NaCl, 4 g of CDTA (cyclohexylene dinitrilo tetraacetic acid) and 57 ml of glacial acetic acid per liter. The solution pH was adjusted to 5-5.5 using 50% NaOH. (All products used were analytical-grade reagents.)

The analytical technique used for the determination of fluoride in water is based on the official method for analysis of this anion in the public water supply. This consists of the determination of fluoride content in mg/L through a comparison with the previously drawn calibration curve, and it is based on direct potentiometry by means of a fluoride ion specific electrode. From the two buffering solutions proposed in the official method, we chose the TISAB-CDTA solution (Martín 1989; Martín *et al.* 1991; Martín *et al.* 1990). The sample volume (mL): buffer solution volume (mL) was 20:20.

In a sample of water, a known quantity of fluoride was added. The measure of fluoride was realized eleven times. The recovery percentage was 99.6% and the standard deviation 1.74%. the limit of detection is 0.2 mg/L and range of the standard curve was between 0.1 mg/L and 10 mg/L of fluoride.

RESULTS AND DISCUSSION

Table 1 shows the results of the analysis carried out on the public water supply, giving the area from which the sample was drawn.

Table 1. Fluoride concentration (mg/L) in public supply waters of Icod de Los Vinos health district

Sample	Origin	Borough	Concentration F^- (mg/L)
1	Public supply	S. J. de La Rambla	2.33
2	Public supply	S. J. de La Rambla	2.08
3	Public supply	S. J. de La Rambla	2.48
4	Public supply	S. J. de La Rambla	2.49
5	Public supply	S. J. de La Rambla	2.77
6	Public supply	S. J. de La Rambla	3.12
7	Public supply	S. J. de La Rambla	2.25
8	Public supply	I. de los Vinos	0.97
9	Public supply	I. de los Vinos	6.91
10	Public supply	I. de los Vinos	5.60
11	Public supply	I. de los Vinos	6.94
12	Public supply	I. de los Vinos	5.77
13	Public supply	I. de los Vinos	5.65
14	Public supply	I. de los Vinos	0.35
15	Public supply	La Guancha	5.98
16	Public supply	La Guancha	6.05
17	Public supply	La Guancha	5.81
18	Public supply	La Guancha	2.88
19	Public supply	La Guancha	6.20
20	Public supply	La Guancha	5.91
21	Public supply	La Guancha	6.03

Table 2 shows the mean, maximum and minimum values of fluoride concentration, as well as the standard deviation, in the public water supplies from the boroughs of Icod de los Vinos, San Juan de la Rambla and La Guancha. All

waters exceed the maximum permisable value of 1.5 mg/L stated in spanish legislation (B.O.E. 1990).

Table 2. Statistical parameters of results obtained

Borough	Mean (mg/L)	σ	Maximum (mg/L)	Minimum (mg/L)
La Guancha	5.55	1.18	6.20	2.88
San Juan de La Rambla	2.50	0.35	3.12	2.08
Icod de los Vinos	4.59	2.75	6.94	0.35

σ = Standard deviation.

If we consider the range of mean concentrations (2.5 to 5.55 mg/L) and set the mean water consumption to 2 l per day, the fluoride intake would be between 5 and 11.1 mg/L of fluoride/person/day. These values far exceed recommended dietary allowances (RDA_S 1991), which are: 1.5 to 4 mg/person/day, for adults; 0.1 to 1 mg/person/day, for children under one; 0.5 to 1.5 mg/person/day, for children between one and three; and up to 2.5 mg/person/day for children under 12. This accounts for this area having a high incidence of dental fluorosis . Our results are slightly higher than those found in 1988 by Hardisson and Reyes (1986) in the public water supply. In that year, they found a value of 3.90 mg/l, and in the present paper, we found an average value of 4.22 mg/L.

Table 3 shows the change with time of the fluoride content in the galleries of La Guancha borough. This Tenerife borough has very high concentrations of fluoride ion and is considered an area where dental fluorosis is endemic, together with two other boroughs of the same health district, San Juan de la Rambla and Icod de los Vinos. This is explained by the fact that the water comes from the La Guancha area and, in particular, from the Vergara ravine gallery, because the rest of the galleries cannot be harnessed due to water losses. The table shows there has been a clear increase in fluorine concentration with time (1976-1997) because in 1986 the combination of mains water with waters from different galleries with less fluorine content was discontinued. However, during the 90's, when the only water source is the Vergara ravine gallery, the fluoride concentration in water from the mains is below the one of the gallery itself. This could be explained by rainfall, by water supply system losses due to chemical reactions with Ca^{2+} and other ions, and by possible adsorption phenomena in the materials of the water mains.

Table 4 shows a comparison of fluoride concentrations in drinking waters from different areas. Thus, and although the contents vary, as is the case in our island, we can see that there are some waters in India (Sarma and Rao 1997) with high fluoride contents, similar to those of our waters in the health district of Icod de los Vinos.

Table 3. Change with time of fluoride content (mg/L) in the waters of La Guancha borough for the period 1976-1997

Gallery	Year					
	1976	1981	1984	1986	1995	1997
Vergara ravine	6.41	7.80	8.00	9.40	10.2	10
El Frontón	4.72	5.10		4.40		
El Pinalete	4.18			4.30		
Sta. Teresa	3.96	5.00		3.90		
Los Palomos	3.25	5.98		4.30		
El Porvenir	2.58	3.10		3.10		
El Derriscadero	2.54	2.00		3.30		
La Esperanza	0.57					
Fuente Pedro		5.20	5.00			
Bilbao		4.10	3.50			
El Partido		2.90	2.75			
Public supply (mixture)				3.90	5.55	5.80

Table 4. Fluoride concentration in drinking waters of other areas

Geographical area	Method	F (mg/L)	Reference
Portugal (Riberáo Prieto)	(1)	0.1 – 1	(Spadaro *et al.* 1990)
India (Visakhapatnam)	(1)	0.31 - 8.35	(Sarma and Rao 1997)
Spain (País Vasco)	(1)	Álava < 0.05- 1.12 Guipúzcoa:< 0.05 - 0.26 Vizcaya: < 0.05 - 0.45	(Arevalo et al. 1984)
Spain (La Rioja)	(1)	0.22 – 1.1	(Jimenez and Bernal 1986)
Spain (Tenerife)	(1), (2)	6% >1.5 64% ≤ .5 - 1.5; 20% < 0.5	(Our current data)

(1) Ion selective; (2) SPADNS colorimetry

Table 5 shows the concentrations of ion F^- in the 21 samples of bottled drinking waters analyzed, together with their origin and characteristics. We have obtained minimum and maximum values of 0.10 and 13.77 mg/L, with an mean value of 1.09 mg/L and a standard deviation of 2.89.

We can see that 85.7% of the samples fall within the range 0.1 to 0.8 mg/L of F^-; 4.8% are between 0.8 and 2.0 mg/L, and only 9.5% exceed 2.0 mg/L. If we group

Table 5. Origin, water type and fluoride concentrations (mg/L) of the 21 samples of bottled drinking waters analyzed

Sample	Origin	Type of water	F^- concentration (mg/L)
1	France	Natural mineral	0.15
2	Italy	Natural mineral	0.64
3	Tenerife	Spring	0.54
4	Tenerife	Spring	2.77
5	Germany	Natural mineral	0.19
6	Germany	Natural mineral	0.14
7	Germany	Natural mineral	0.17
8	Germany	Natural mineral	0.67
9	Germany	Natural mineral	0.90
10	Cataluña	Medicinal mineral	13.77
11	Tenerife	Prepared drinking	0.36
12	Gran Canaria	Spring	0.24
13	Barcelona	Natural mineral	0.19
14	Italy	Natural mineral	0.66
15	Gran Canaria	Prepared drinking	0.16
16	Gran Canaria	Natural mineral	0.26
17	Cantabria	Natural mineral	0.11
18	France	Natural mineral	0.10
19	Cuenca	Sparkling mineral	0.11
20	Gran Canaria	Prepared drinking	0.12
21	Barcelona	Natural mineral	0.58

the samples according to their origin, we find that 11 samples come from Spain (except sample 10, low consumption medicinal-mineral water), including those samples of branded waters bottled in Canary Islands. The mean concentration is 0.49 mg/L F^-, with a standard deviation of 0.74, and minimum and maximum values of 0.11 and 2.77 mg/L of F^-, respectively. If we consider the samples bottled in the Canary Islands alone, the mean concentration is 0.63 mg/L, with standard deviation 0.88 and minimum and maximum values 0.12 and 2.77 mg/L,

respectively. In the same way, if we take into account their legal classification (B.O.E. 1990; B.O.E. 1991), the highest mean concentration is that of the bottled spring waters (1.18 mg/L), followed by the mineral waters (0.35 mg/L). The lowest mean concentration is that of the prepared drinking waters (0.21 mg/L).

Comparing our results with those of Martín et al. (1990), we find that our mean (1.09 mg/L) is below their 1.47 mg/L. The same is found when we compare our results with those of Armijo et al. (1976), who obtain an mean concentration of 1.77 mg/L with respect to our 1.09 mg/L. In addition, we agree with them in considering that there are bottled drinking waters exceeding the admissible values stated by law for bottled drinking waters, and that they should be declared as "fluoridated" waters, according to the Spanish Technical Health Rules (B.O.E. 1990).

Various techniques for the elimination of fluoride from natural water could be applies: precipitation and adsorption methods. The precipitation methods, however, recuiry the addition of a substance to the water (brushite, calcium hydroxide), whereas the ion exchange methods are more expensive.

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