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Fluoride content of bottled waters available in Northern Greece

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Summary. *Objective.* The aim of this study was to evaluate the fluoride content of bottled drinking waters commercially available in northern Greece and to report on the accuracy of the labelling of fluoride concentration.
Materials and methods. Twenty-two randomly selected commercial brands of bottled water were obtained from three supermarkets in Thessaloniki, Greece. Three bottles of each brand were purchased. Following calibration, six tests were conducted on each bottle using a combination fluoride-ion selective electrode (Orion, 96-09-00, MA, USA). The average reading for each brand was estimated and also compared with the fluoride content printed on the label.
Results. The mean (± SD) fluoride content of the bottled water samples was 0·35 (± 1·00) mg F/L with a range from 0·05 to 4·8 mg F/L. Only 18% ($N = 4$) of brands tested mention the fluoride concentration on the label, and 90% ($N = 22$) had a tested fluoride between 0·05 and 0·21 mg F/L. Of the remaining two brands, one was found to contain 0·3 mg F/L without having the fluoride concentration indicated on the label, and the other was labelled at 6 mg F/L, whereas the concentration was estimated as 4·8 mg F/L.
Conclusions. The use of bottled water may be a significant source of systemic fluoride and therefore be considered as a risk factor for dental fluorosis in young children. This article shows that bottled drinking waters contain differing concentrations of fluoride. The manufacturers' labelling of fluoride concentrations may be inaccurate. When prescribing fluoride supplements, dentists should be aware of the fluoride content of bottled waters used by paediatric patients, especially brands with a concentration higher than 0·3 mg F/L. In view of the wide variation of fluoride concentration in the tested bottled waters, regulatory guidelines for controlling concentration in order to prevent dental fluorosis are recommended.

Introduction

One of the most common trends among consumers in recent years in various countries, including Greece, is the replacement of their daily water intake with bottled water. According to a commercial report, bottled water consumption in Greece increased during 1996–1999 by 11·05% compared to the 1994–1996 period. In 1999, 477 million litres of bottled water or 45 L per person were sold in Greece, whereas in 1989 it was only 13 L per person [1]. Since 1999, the consumption has increased further, with an annual increase of 12% and such that, in 2002, bottled water consumption was 870 million litres. The rate of increase remained the same during the years 2003–2004 [2]. The replacement of tap water by bottled water happens mainly for two reasons: the questionable quality of the public water and the strong chlorine taste, resulting from generalized water chlorination in most areas. Unlike chlorine, however, in Greece, the fluoride content of public water supplies is not widely monitored.

In Thessaloniki, the second largest city in the country, with more than half of the population of northern Greece (1·3 million people), the mean fluoride concentration in the public water is less than 0·1 mg F/L. Fluoride toothpaste is seen as the cornerstone of fluoride use in Greece, and the use of fluoride tablets is limited. Recently, paediatric dentists recommended the use of fluoride tablets only for high-risk children. The proposed dosage is 0·25 mg from the 6th month until the third year of age and twice this amount for children aged 3–6 years old [3]. Dentists and paediatricians, however, often prescribe fluoride supplements without considering other fluoride sources.

As far as bottled water is concerned, the fluoride content may be highly variable among different brands, and this may have oral health implications for individuals, and especially infants and children, who use bottled water as their primary source of

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drinking water. Flaitz *et al*. [4] found that 9·32% of 1126 randomly selected paediatric dental patients, aged 2 weeks to 13 years old, were using bottled water as primary water source. When the fluoride content of the bottled water was correlated with the patient's age, the authors noted that although 16·9% of their patients were receiving less than an optimal level of fluoride in their drinking water, a significantly greater percentage (72·4%) were receiving greater than the optimal amount recommended for their age group. Only 10·7% of the patients were receiving the recommended dosage of fluoride in their drinking water. In addition, paediatricians had prescribed systemic fluoride supplementation for approximately 10% of the patients. Patients receiving these supplements were also estimated to be drinking bottled water with fluoride contents that ranged from 1·00 to 1·40 mg F/L.

The concentration of some elements such as calcium, sodium, and aluminium in bottled water is regulated in some countries [5]; however, there is no regulation regarding the fluoride content of bottled water in Greece or other European countries. Manufacturers are encouraged to list the nutritional contents of their products, but labelling of the fluoride levels on bottled water is not legally required. Furthermore, when fluoride concentrations appear on the bottle labels, they may not always be accurate. Wienberger [6] found the accuracy of the printed concentrations of fluoride to be doubtful in 16 of 17 bottles tested in a Canadian study. Toumba *et al*. [7] reported that only three of the seven tested labelled brands were in agreement with the results of their study. Finally, in a further UK study, only 6 of 26 bottles tested had the fluoride concentrations printed on their labels [8].

It is important to know precisely the concentration of fluoride in the water that a patient is drinking in order to propose an effective preventive program with minimum risk of fluorosis. In cases where fluoride concentration is too high, particularly in children between 22 and 26 months of age, dental fluorosis may appear on the maxillary permanent central incisors [9]. In contrast, if the concentration is too low, supplementation may be necessary.

In view of the expanding range of bottled waters available for purchase in Greece, the first objective of this study was to provide information on the fluoride concentrations of commercially available bottled waters. The second aim was to report the accuracy of the labelling of fluoride concentration on the bottles.

Methods

Bottles of 22 commercial brands of water were obtained from three Thessaloniki (GR) supermarkets (Table 1). Of these, 68% (15 brands) had their source or production site in Greece. Three bottles of each brand, each with a different batch number and date of bottling, were purchased. All bottles were stored in a dark place and in their original closed plastic containers at room temperature until the fluoride analysis was made.

After shaking the bottle of water, a 5-mL sample was taken and mixed with 5 mL of Total Ionic Strength Adjusting Buffer II (TISAB II, Orion, MA, USA). Following calibration, six tests were conducted on each bottle using a combination fluoride-ion selective electrode (Orion, Combination Fluoride Ionalyzer, 96-09-00, MA, USA) in conjunction with an ionanalyser (Crison MicropH 2002, Barcelona, Spain) by using an acetate buffer system (TISAB). The pH of each sample was measured by using a combination pH electrode (Glass – Ag, AgCl, Mettler Toledo, 405-S7/120, Switzerland) before and after the addition of TISAB in order to ensure that the pH of the solution was between 5 and 7. Fluoride standards containing 0·05, 0·10, 0·50, 1·00, and 5·00 mg/L fluoride prepared and after the addition of TISAB solution were measured and used to construct standard curves. Final millivolt readings were converted to fluoride ion concentration using the standard correlation curve.

One batch number (of three) for each of the 22 bottled water samples was randomly selected and the samples re-analysed to assess the reliability of the method. SPSS (Statistical Package for Social Sciences, version 11) was used to derive descriptive data.

Results

The fluoride content, when given on the labels, the measured mean fluoride content and pH values obtained before and after the addition of TISAB are shown on Table 1. For each water sample, the concentration is shown in mg/L fluoride as given on the label (or NL if not labelled). The reliability of the method of fluoride analysis was determined to be 99%. The mean (± SD) fluoride content of the 22 bottled water samples was 0·35 (± 1·00) mg/L with a range from 0·05 to 4·80 mg/L. The highest mean concentration was found in bottled water from Vichy, France, which had a measured mean fluoride

Table 1. Fluoride concentrations of 22 bottled waters (mean ± SD values for 18 samples) and pH values with and without TISAB solution. Values in parenthesis concern the labelled pH.

Bottled water brand	Country of origin	Fluoride concentration (mg F/L)	Labelled content* (mg F/L)	pH	
				Without TISAB	With TISAB
Athos	Greece	0·20 ± 0·01	0·10	7·40 (7·50)	5·61
Ayra	Greece	0·19 ± 0·01	NL	7·35 (7·60)	5·68
Bikos	Greece	0·17 ± 0·02	NL	7·09 (7·62)	5·58
Contrex	France	0·30 ± 0·02	NL	7·06 (NL)	5·50
Dorna	Romania	0·07 ± 0·01	NL	5·85 (6·00)	5·61
Evian	France	0·12 ± 0·01	NL	6·99 (7·20)	5·62
Ioli	Greece	0·14 ± 0·01	NL	7·27 (7·50)	5·61
Iris Loutraki	Greece	0·16 ± 0·02	NL	7·93 (7·60)	5·75
Ivi-Loutraki	Greece	0·12 ± 0·01	NL	7·82 (8·20)	5·62
Kimi	Greece	0·06 ± 0·01	NL	7·25 (NL)	5·63
Korpi	Greece	0·14 ± 0·01	NL	7·21 (7·30)	5·64
Louttraki-Karadanis	Greece	0·07 ± 0·01	NL	8·10 (8·20)	5·75
Mitsikeli, Epirus	Greece	0·05 ± 0·01	NL	7·40 (7·50)	5·61
Pigi Xonaiou Thetis	Greece	0·20 ± 0·01	NL	7·55 (7·70)	5·63
Ranis	Greece	0·12 ± 0·01	0·15	7·32 (7·50)	5·65
Samaria	Greece	0·09 ± 0·01	NL	7·58 (7·80)	5·61
San Benedetto	Italy	0·11 ± 0·02	NL	6·55 (NL)	5·55
Spa	Belgium	0·12 ± 0·01	NL	6·30 (NL)	5·63
Vichy	France	4·80 ± 0·13	6	6·21 (6·80)	5·84
Vittel	France	0·18 ± 0·01	NL	6·82 (NL)	5·61
Ydor, Sourotis	Greece	0·11 ± 0·01	NL	7·00 (7·20)	5·60
Zagori	Greece	0·14 ± 0·01	0·39	7·28 (7·52)	5·60

*NL, not labelled.

content of 4·8 (± 0·13) mg/L. With the exception of this brand, no substantial difference was found between samples from different countries of origin. The mean fluoride concentrations of samples from Greece, Italy, Romania, Belgium, and France were 0·12 (± 0·05), 0·11 (± 0·02), 0·07 (± 0·01), 0·12 (± 0·01), and 1·35 (± 2·30) mg/L, respectively.

Regarding the quality of the labelling of bottled waters, only 4 of the 22 brands (18%) of bottled waters state the fluoride concentration on the labels. Two of these were in agreement with our measures (Athos and Ranis). For the other two brands (Zagori and Vichy), the displayed fluoride concentration was above the level measured in this study – it was labelled 0·39 and 6 mg/L compared with a measured fluoride content of 0·14 and 4·8 mg/L, respectively.

Twenty brands (90%) were found with a fluoride concentration between 0·05 and 0·21 mg/L, one (4·5%) was found 0·3 mg/L, and one (4·5%) was measured 4·8 mg/L. There were no significant differences among the three batch numbers for each brand (Table 2).

Discussion

The dental health of a child who utilizes bottled water as the primary source of drinking water may be significantly affected in one of three ways: the child may receive an appropriate level of fluoride from bottled water, allowing for optimal caries prevention, or receive suboptimal levels of fluoride, with a resultant increase in dental caries. Finally, an elevated level of fluoride in a child's drinking water could result in fluorosis. Although in Greece there is no official regulation defining the limits of fluoride in bottled water, in other countries the highest limit has been set at 4 mg/L [10].

In this study, the concentration of fluoride in bottled drinking waters purchased from local stores was found to vary widely between 0·1 and 4·8 mg F/L. This variation was generally consistent with previous studies [5–7,11]. The fluoride content of one brand (Evian), however, showed a difference between this study (0·12 mg F/L) and that reported by Zohouri *et al*. [5] (0·06 mg F/L). Our result was similar to the findings of Toumba *et al*. [7], where the recorded value was 0·15 mg F/L. In this study, fluoride concentration in another water brand (Vittel, 0·18 mg F/L) was also similar to that recorded by Zohouri *et al*. [5] (0·12 mg F/L). In addition, another analysis conducted in Belgium [5] reported the mean fluoride content of Evian and Vittel at 0·08 and 0·17 mg F/L, respectively, and both results are close to our findings.

Table 2. Fluoride concentrations (mg F/L) of three different batch numbers of each bottled water, measured six times each.

	Fluoride concentration (mg F/L)		
Bottled water brand	Batch 1	Batch 2	Batch 3
Athos	0·20	0·20	0·20
Ayra	0·20	0·19	0·19
Bikos	0·18	0·17	0·16
Contrex	0·30	0·29	0·30
Dorna	0·07	0·07	0·06
Evian	0·12	0·12	0·12
Ioli	0·14	0·15	0·15
Iris Loutraki	0·17	0·16	0·17
Ivi-Loutraki	0·12	0·13	0·12
Kimi	0·05	0·06	0·06
Korpi	0·13	0·15	0·15
Louttraki-Karadanis	0·07	0·07	0·07
Mitsikeli, Epirus	0·06	0·05	0·04
Pigi Xonaiou Thetis	0·21	0·20	0·20
Ranis	0·12	0·12	0·12
Samaria	0·10	0·10	0·08
San Benedetto	0·10	0·12	0·11
Spa	0·12	0·11	0·13
Vichy	4·73	4·87	4·80
Vittel	0·18	0·18	0·19
Ydor, Sourotis	0·11	0·10	0·12
Zagori	0·15	0·14	0·15

It has been shown that the fluoride content of water depends on a significant degree on weather changes, i.e. heavy rains [12]. Both Evian and Vittel are extracted from French mountains, and so one explanation for the fluoride concentration differences among the studies could be the seasonal fluctuation.

In an epidemiological study conducted in an agricultural prefecture of Greece, it has been shown that the dmft and DMFT values in 6- to 11-year-old children vary from 1·94 to 8·08 and from 0·97 to 4·12, respectively, whereas fluoride concentrations in drinking water vary from 0·01 to 3·2 mg/L [13]. Further data of the same study showed that lower values of both indices were derived from the village with the highest concentration of fluoride in the water.

Greece is not considered an endemic area for dental fluorosis although previous studies have shown that children may present with low–mild and mild mottled enamel in some areas of the country [14,15]. Naturally, fluoridated water is found in Greece only in a few areas, mainly small towns with a population of less than 3000. Adamidi and Apostolopoulos [13] reported that mild to severe fluorosis lesions were present in 44% of the 6- to 11-year-old examined children in such areas where fluoride concentration in the water was measured 3·2 mg F/L. A recent study [16], however, reported that from a randomly selected group of 287 8-year-old children living in urban areas (Athens, Greece), only 5% showed fluorosis-like opacities in one of their maxillary central incisor. It should be noted that fluoride concentration of tap water in Athens was measured to be less than 0·01 ppm. That study compared the prevalence of fluorosis in seven European areas derived from different countries, and the authors reported that the highest values (30%) were recorded in Cork (Ireland), where water supplies were artificially fluoridated to a concentration between 0·8 and 0·11 mg F/L.

The level of fluoride in the bottled drinking water has implications for the prescription of fluoride supplements as well as for the estimation of the risk of fluorosis. A first step towards evaluating how bottled water consumption might affect fluoride exposure is to determine the fluoride content of bottled waters. It is equally important to evaluate daily water intake among children living in different areas, with respect to their age, gender, and during seasonal changes of temperature. Based on this information, the total fluoride intake from different sources, and consequently the risk for fluorosis, can be estimated. According to the data from the National Diet and Nutrition Survey for young people aged 4–18 years in UK [17], the mean total daily bottled and tap water intake is 108 and 155 mL, respectively. The mean total daily fluoride intake, when the source of water intake is solely tap water and when it is a combination of tap and bottled water, is estimated to be 0·264 and 0·164 mg/L, respectively. Unfortunately, evaluation of the same parameters for Greece is not yet possible as there is simply a lack of available data.

It has been suggested that if the level of fluoride in drinking water is below 0·7 mg/L, supplements may be required in some cases, whereas if the level is greater than 0·7 mg/L, supplementation is not necessary [4,18]. Furthermore, if a high concentration of fluoride in drinking water is used to make up a baby formula that have been shown to contain variable amounts of fluoride themselves [19,20], then potentially very high intakes of fluoride are possible in young children.

This study showed that in all but one tested brands of bottled water, the fluoride concentration was between 0·05 and 0·30 mg/L. The extremely high fluoride concentration in one brand (Vichy) could cause fluorosis if the water is consumed by young children alone or in addition with other fluoride supplements.

Only 18% of the tested samples displayed the fluoride content of the water on the labels. Dentists should be aware that values on labels might not be reliable; as our findings indicate that in 50% of the bottled water samples, there was a significant difference between the measured fluoride content and that marked on the label.

As the sales of bottled waters have more than tripled in recent years and with the vast plethora of different brands available, it is important for dentists to be aware of the fluoride concentration of any bottled water used by child patients, especially when prescribing fluoride supplements. The American Academy of Paediatrics [21] stated that for optimal dental health benefits the total daily intake should be 0·05–0·07 mg fluoride per kilogram of body weight, and to avoid the risk of dental fluorosis, fluoride intake should not exceed a daily level of 0·10 mg fluoride per kilogram of body weight.

What this paper adds

- This is first study of fluoride content of bottled waters in the Greek market.
- Most of the bottled waters tested contained less than 0·3 ppm fluoride, but was as high as 4·8 ppm in one brand of bottled water.
- Most brands do not mark fluoride content on their labels.

Why this paper is important for paediatric dentists

- Water consumed by children in northern Greece may contain widely varying fluoride levels, independent of tap water concentrations.

Conclusions

This study shows that bottled drinking water in Greece may contain various concentrations of fluoride. The manufacturers seldom display the fluoride concentration on the labels and where depicted, they are not always accurate. Dentists should be aware of the fluoride concentrations of the drinking water of their child patients when prescribing fluoride supplements. This is of particular importance as there is a potential risk of dental fluorosis as a result of high levels of fluoride.

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