Evaluation of the fluoride concentration and consumption of mineral water

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Keywords

Abstract
Objective
Considering that water is an importance source of fluoride intake, and that the consumption of mineral water and prevalence of dental fluorosis have been increasing, the aim of this study was to evaluate the consumption of mineral water and its fluoride concentration.

Methods
The study was performed in residential districts of the municipality of Bauru, State of São Paulo, by means of stratified sampling via clusters. Each cluster corresponded to one residential block. For randomization purposes, the residential blocks were numbered within the 17 districts established by the city plan. One thousand homes were thus visited. Mineral water samples were collected using previously labeled 50 ml plastic flasks. Fluoride analysis was done using an ion-sensitive electrode (Orion 9609), after buffering using TISAB II. Information on the consumption of mineral water was obtained by means of applying a questionnaire.

Results
Around 29.72% of the city’s population was consuming mineral water. In the 260 samples analyzed from 29 different brands of water, the fluoride concentration ranged from 0.045 to 1.515 mg/l. For one brand, the label stated that the fluoride concentration was 0.220 mg/l, but analysis revealed a concentration of 1.515 mg/l. Moreover, some brands did not specify the fluoride concentration on the label and, for these, the analysis showed concentrations ranging from 0.049 to 0.924 mg/l.

Conclusions
The results demonstrated wide variation in fluoride concentrations and reinforce the importance of the control of such waters by the sanitary surveillance agency.

INTRODUCTION

The exploitation of mineral water or bottled water in Brazil is regulated by the National Mineral Production Department of the Ministry of Mines and Energy. The definition and control of drinkability are under the control of the National Sanitary Surveillance Agency of the Ministry of Health.4 Because of the need to standardize the utilization of mineral waters that are commercialized by means of bottling, and for other purposes, Decree Law no. 7841, known as the “Mineral Water Code”, was published in the Federal Official Gazette (DOU) on August 20th, 1945. This law remains in force today, with some modifications.3

For a long time, it was believed that mineral waters differed in origin from underground water. However, it is now known that they have the same origin, i.e. they are surface waters that penetrate the subsoil, and that their salt content is directly related to heat, since the ability to dissolve minerals and incorporate solutes increases with temperature. Mineral waters are

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regarded as such when they have reached greater depths, thereby achieving enrichment in salts and acquiring new physicochemical characteristics.

According to Regulation no. 54 of June 15th, 2000, mineral water is understood to be any water obtained directly from natural springs or artificially extracted from the subsoil that is characterized by a defined and constant content of mineral salts (ion composition) and by the presence of trace elements and other components. Collection of these waters should be conducted under conditions that ensure the maintenance of the original features found at their wells or springs, which should be kept within the variation limits and have no direct influence from surface waters. In Brazil, most occurrences of mineralized waters are in the form of natural springs.

Water fluoridation is a recognized method for preventing dental caries when it is undertaken at the ideal concentration for each region, according to the local mean temperature. However, it is also a risk factor for dental fluorosis when the fluoride (F-) concentration is above the recommended levels. The impact of water fluoridation is mainly indirect, through the use of water in food and beverage manufacturing processes and also in food and beverage reconstitution at home (Burt, 1992; Winkle et al., 1995; Villena et al., 1996; Buzalaf et al., 2001; Buzalaf et al., 2003).

The production and consumption of mineral water in Brazil has increased by about 20% per year over the last five years, while the increase was 9% worldwide and 10% in the United States. The Brazilian per-capita consumption increased from 15.13 liters per year in 1995 to 23.8 liters in 2001. The southeastern region accounts for the largest production in the country, namely 53%, of which 36.9% is in the State of Sao Paulo. It should be highlighted that the production of the northeastern region has increased by 40% and now accounts for 25.10% of national production.

Considering the variety of the sources of mineral water and the lack of data from Brazil regarding such consumption, it becomes essential to know the fluoride content of the water consumed, so as to be assured of its benefits in the prevention of caries without running the risk of dental fluorosis (Featherstone, 1999). Thus, the present study had the aim of evaluating fluoride concentration and mineral water consumption.

**METHODS**

The data were collected in residential areas of the city of Bauru, State of Sao Paulo, Brazil, by means of stratified sampling by area, achieved via clusters. Each cluster corresponded to one residential block. The percentage of the population living in each of the 17 districts established by the Bauru city plan was calculated (Table 1). Within each district, the clusters were numbered and randomly selected until the proportional number of homes in relation to the population of each district was obtained. The sample size was set at 1,000 homes, based on a confidence interval of 3% for a confidence level of 95%. Thus, 1,000 homes were visited, in which a total of 3,586 people were living (mean of 3.6 inhabitants per home), since the study aimed at evaluating the overall consumption of mineral water in the city and not the percentages of one or other mineral water brand consumed.

The consumption profile for bottled mineral water among the population was evaluated by means of a questionnaire applied in each of these homes. The following were investigated: how many people lived there and what their ages were; whether mineral water was consumed or not; what brand of mineral water was used; how long they had been consuming mineral water; what fluoride concentration was specified on the label; whether some type of water purifier was used; and what awareness there was of the presence of fluoride in the mineral water and its action in caries prevention.

For the analysis of the fluoride concentration in the mineral water, the samples were collected in previously labeled 50-ml plastic flasks and frozen at -20°C until analysis. Sample analysis was carried out at the Biochemistry Laboratory of Bauru Dental School (FOB/USP).

**Fluoride analysis**

The fluoride (F-) concentration of the water sam-
samples was analyzed in duplicate using an ion-sensitive electrode (Orion 9609) connected to a potentiometer (Procyon, model 720). A sample size of 1.0 ml was utilized, to which 1.0 ml of TISAB II buffer (Orion) was added. Prior to the analysis, calibration was carried out in triplicate, using standard fluoride solutions of concentrations ranging from 0.02 to 1.6 µgF-/ml. These were obtained through serial dilution from a standard stock solution containing 100 µgF-/ml (Orion). Tests with known fluoride concentrations were performed every hour to check the calibration. The readings in mV from the standards of known fluoride concentrations were entered into the Microsoft Excel spreadsheet and converted to µgF-. The calibration curves were only accepted when the variation was no more than 5%. The mean repeatability of the readings, based on the duplicate analyses, was 91.5%. In addition, new analyses were performed on 32% of the sample, thereby revealing a reproducibility of 92.2%.

Analysis of the questionnaire

Analysis of the data obtained via the questionnaire was done by means of descriptive statistics, with the utilization of absolute and relative frequencies that were described in tables and graphs.

Although mineral water consumption was reported in 312 homes out of the 1,000 visited, no water was available for sample collection in 26 homes. A further 26 water samples were excluded during the tabulation of the data from the questionnaire because, even though the consumer knew the brand, there was no label on the water receptacle to enable confirmation of the brand and fluoride concentration. Thus, 260 samples were taken into account in preparing the tables and graphs.

RESULTS

Around 29.72% of the population was consuming mineral water, of whom 14% had had this habit for five years or more and 15.6% had acquired it over the last five years (1998 to 2003). In the 260 samples analyzed from 29 different brands of mineral water, the fluoride concentration ranged from a minimum of 0.045 to a maximum of 1.515 mg/l (Table 2). Among the water samples collected from nine known sources, seven of them (77.8%) presented an average fluoride concentration of between 0.115 and 0.522 mg/l that was greater than what was stipulated in the label. Among the mineral waters from unknown sources, the range was from a minimum of 0.049 mg/l to a maximum of 1.180 mg/l (Table 3). Overall, for the 260 samples analyzed, it was seen that 62.3% of the mineral waters presented fluoride concentrations ranging from 0.045 to 0.100 mg/l (non-fluoridated), 24.6% from 0.100 to 0.600 mg/l (under-fluoridated) and just 3.9% from 0.600 to 0.800 mg/l. It was further observed that 9.2% of the samples showed fluoride concentrations of 0.800 to 1.515 mg/l (over-fluoridated) (Figure). For ten brands, only one sample of each could be obtained, and these were therefore not included in the tables. For these brands, the fluoride concentrations ranged from 0.045 to 0.778 mg/l.

Table 2 - Mineral water consumed in Bauru in 2003, according to fluoride concentration shown on the label, minimum and maximum values found and sample mean in mg/l.

<table>
<thead>
<tr>
<th>Brand of mineral water</th>
<th>N of samples</th>
<th>mg F⁻/l on the label</th>
<th>Minimum mg F⁻/l</th>
<th>Maximum mg F⁻/l</th>
<th>Mean mg F⁻/l</th>
</tr>
</thead>
<tbody>
<tr>
<td>Source 1</td>
<td>7</td>
<td>0.010</td>
<td>0.061</td>
<td>0.083</td>
<td>0.071</td>
</tr>
<tr>
<td>Source 2</td>
<td>33</td>
<td>-</td>
<td>0.085</td>
<td>0.924</td>
<td>0.174</td>
</tr>
<tr>
<td>Source 3</td>
<td>4</td>
<td>0.059</td>
<td>0.099</td>
<td>0.140</td>
<td>0.115</td>
</tr>
<tr>
<td>Source 4</td>
<td>3</td>
<td>0.255</td>
<td>0.460</td>
<td>0.640</td>
<td>0.522</td>
</tr>
<tr>
<td>Source 5</td>
<td>10</td>
<td>0.025</td>
<td>0.049</td>
<td>1.008</td>
<td>0.155</td>
</tr>
<tr>
<td>Source 6</td>
<td>9</td>
<td>0.035</td>
<td>0.079</td>
<td>0.517</td>
<td>0.158</td>
</tr>
<tr>
<td>Source 7</td>
<td>44</td>
<td>0.220</td>
<td>0.075</td>
<td>1.215</td>
<td>0.183</td>
</tr>
<tr>
<td>Source 8</td>
<td>3</td>
<td>0.034</td>
<td>0.092</td>
<td>0.092</td>
<td>0.086</td>
</tr>
<tr>
<td>Source 9</td>
<td>5</td>
<td>0.020</td>
<td>0.051</td>
<td>1.021</td>
<td>0.376</td>
</tr>
</tbody>
</table>

F⁻ - fluoride
*fluoride data obtained from the label provided by some participants in the study.

Figure 1 - Variation in the fluoride concentration (mg/l) found in the mineral waters consumed in Bauru, 2003.
DISCUSSION

Increased consumption of mineral water in substitution or addition to the public water supply may have implications for safe fluoride supplementation (Brandão et al., 1998; Lindemeyer, 1996; Tate & Chan, 1994; Winkle et al., 1995). Similarly, as has been seen for mineral water, the consumption of infant formulae (supplemented milk) has increased greatly over recent years. These formulae are commercially available in three presentations: ready to use, concentrated liquids, or powdered. The latter is the type most frequently found in Brazil. The fluoride concentrations in formulae ready for consumption and concentrated liquids are relatively low and are usually around 0.1 to 0.3 mg/l. However, liquid formulae require the addition of an equal volume of water before consumption. Thus, the final fluoride concentration will be directly related to the water utilized in the dilution. In powdered formulae, the mean F concentration is around 0.69 mg/l for milk-derived products and slightly higher for soybean-derived products. The reconstitution of powdered products requires the addition of an average of 145 g of powder to 880 ml of water. Thus, the final fluoride concentration also depends on the fluoride concentration of the water utilized for dilution, which usually ranges from 0.276 to 0.980 mg/l (Formon et al., 2000; Silva & Reynolds, 1996). For children aged more than four months who consume an average of 120 ml/kg/day of diluted formula, their fluoride intake is around 0.072 to 0.118 mg/kg/day for the concentrated and powdered formulae, respectively. However, it should be borne in mind that children at this age also ingest fluoride from sources other than milk (Formon et al., 2000).

Buzalaf et al. (2001) evaluated the fluoride concentration of ten brands of powdered milk, prepared according to the manufacturers’ instructions using deionized water, the public water supply (0.9 mgF/l) and seven different brands of mineral water. The fluoride concentration ranged from 0.01 to 0.75 mg/l in the milks prepared with deionized water, 0.91 to 1.65 mg/l in those prepared with the public water supply and 0.02 to 1.37 mg/l in those prepared with mineral water. Considering that the optimal daily fluoride intake recommended is 0.05 to 0.07 mg/kg of body weight (Burt, 1992), some types of milk exceeded the limit when prepared using water from the public water supply, considering the mean daily intake of milk recommended for each child. Since consumption above these levels may cause dental fluorosis, utilization of fluoridated water with fluoride concentrations close to 1.0 mg/l as diluents for powdered milk for infants and young children should be avoided. Non-fluoridated mineral water is recommended for this purpose (Buzalaf et al., 2001).

Milk is the first and main source of nourishment for children during their first months of life, when the front teeth are being formed. Human breast milk has a low fluoride concentration, usually around 0.005 to 0.010 mg/l, because the fluoride transportation from serum to milk is poor (Ekstrand et al., 1984). The fluoride concentration in the milk of other mammals is also low (Dias et al., 2000; Formon et al.). Thus, both the type of milk and the water consumed by children should be regarded as important factors in evaluating the risk of fluorosis and prescribing fluoride supplements (Lindemeyer et al., 1996; Tate & Chan, 1994; Winkle et al., 1995). It can therefore be seen that it is extremely important to evaluate the F concentration of mineral water and its consumption by children (Stannard et al., 1990; Toumba et al., 1994).

Ordinance no. 14 of January 1st, 1977, from the Ministry of Health (Resolution 25/76) lays down that the label should state the composition of the product per liter as indicated in the respective analysis certificate. Resolution no. 54 of June 15th, 2000, from the National Sanitary Surveillance Agency establishes the obligation for the label to clearly and precisely state the following information, among other matters:

- "Contains fluoride", when the product has more than 1 mg/l of fluoride;

Table 3 - Brands of mineral water without indication of source that were consumed in Bauru in 2003, according to fluoride concentration shown on the label, minimum and maximum values found and sample mean in mg/l.

<table>
<thead>
<tr>
<th>Mineral water brand</th>
<th>N of samples</th>
<th>mgF⁻/l on the label</th>
<th>Minimum mg F⁻/l</th>
<th>Maximum mg F⁻/l</th>
<th>Mean mg F⁻/l</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>4</td>
<td>-</td>
<td>0.070</td>
<td>0.770</td>
<td>0.311</td>
</tr>
<tr>
<td>2</td>
<td>4</td>
<td>-</td>
<td>0.083</td>
<td>0.931</td>
<td>0.507</td>
</tr>
<tr>
<td>3</td>
<td>16</td>
<td>-</td>
<td>0.049</td>
<td>0.914</td>
<td>0.148</td>
</tr>
<tr>
<td>4</td>
<td>11</td>
<td>0.050 ± 0.100</td>
<td>0.070</td>
<td>1.180</td>
<td>0.370</td>
</tr>
<tr>
<td>5</td>
<td>3</td>
<td>0</td>
<td>0.097</td>
<td>0.982</td>
<td>0.674</td>
</tr>
<tr>
<td>6</td>
<td>5</td>
<td>0</td>
<td>0.861</td>
<td>0.882</td>
<td>0.871</td>
</tr>
<tr>
<td>7</td>
<td>2</td>
<td>0</td>
<td>0.110</td>
<td>0.130</td>
<td>0.120</td>
</tr>
<tr>
<td>8</td>
<td>49</td>
<td>0.036 ± 0.070</td>
<td>0.073</td>
<td>1.105</td>
<td>0.264</td>
</tr>
<tr>
<td>9</td>
<td>5</td>
<td>0.038 ± 0.013</td>
<td>0.091</td>
<td>0.227</td>
<td>0.160</td>
</tr>
<tr>
<td>10</td>
<td>3</td>
<td>0.072</td>
<td>0.080</td>
<td>0.960</td>
<td>0.569</td>
</tr>
</tbody>
</table>
• “This product is not suitable for breastfeeding mothers or children aged under seven years” when the product has more than 2 mg/l of fluoride;
• “Fluoride above 2 mg/l is not recommended for daily consumption” when the product has more than 2 mg/l of fluoride.

Furthermore, according to the mineral water legislation, water is regarded as fluoridated when the fluoride concentration is greater than or equal to 0.1 mg/l, and this should be made clear on the label. However, Table 2 shows that the analysis of seven samples from this source revealed fluoride concentration ranging from 0.061 to 0.083 mg/l, with a mean of 0.071 mg/l. The opposite was found for 33 samples from Source B, which did not describe its fluoride concentration on the label, even though the analysis found concentrations ranging from 0.065 to 0.924 mg/l, with a mean of 0.174 mg/l. This source should therefore be regarded as fluoridated. Water from Source G, which had a considerable consumption rate, considering that 44 samples were obtained, declared a fluoride concentration of 0.220 mg/l on its label, yet the concentration revealed by the analysis ranged from 0.075 mg/l to 1.515 mg/l, a maximum that is much greater. The difference between the concentration stated on the label and what was actually found was even larger for water from Source I, which stated a concentration of 0.020 mg/l on the label and in fact ranged from 0.051 to 1.021 mg/l in the analysis of its five samples, with a mean of 0.376 mg/l. Of the nine brands of mineral water of known origin collected (Table 2), seven brands (77.77%) displayed a mean fluoride concentration of 0.115 to 0.522 mg/l that was greater than the level stated on the label. In the same way as in the investigations by Franco & Maltz11 (1991) and Villena et al24 (1996), which were also conducted on mineral waters found on the Brazilian market, the present study found high fluoride concentrations (above the recommended level) that were not specified on the labels, and also low concentrations (below the minimum level considered to be fluoridated) for waters that their producers described as fluoridated. This reveals that, even 12 years after the first study and 7 years after the second, the problem still persists.

Table 3 presents the mineral waters for which the sources were not identified but the brands were. It reveals large differences between the fluoride concentrations stated on the label and what was found in the analyses. The mean values observed in the samples of these ten different brands ranged from 0.045 to 0.871 mg/l. Eighty percent of them were within the minimum and maximum limits, thus being considered as safe fluoridated waters, whereas the remainder had low concentrations of between 0.045 and 0.096 mg/l.

Even though there were large differences between the fluoride concentrations stated on the labels and what was revealed by analysis of the 260 samples of the 29 brands of mineral water, the maximum values of the means, namely 0.871 mg/l, were within the acceptable levels for water for daily consumption (Tables 2 and 3). Of these, five brands (20%) had a mean that was less than the minimum level regarded as fluoridated (<0.1 mg/l). Considering all the samples analyzed (260), it was observed that 62.3% of the brands showed fluoride concentrations ranging from 0.045 to 1.00 mg/l (non-fluoridated), 24.6% from 1.00 to 0.600 mg/l (under-fluoridated) and just 3.9% from 0.600 to 0.800 mg/l. It was further observed that 9.2% of the samples revealed fluoride concentrations of 0.800 to 1.515 mg/l (over-fluoridated). These results are similar to those found by Winkle et al25 (1995), who evaluated 78 brands of mineral water in Iowa and found fluoride concentrations ranging from 0.02 to 1.36 mg/l, of which 83% were from 0.02 to 0.16 mg/l, 7% from 0.34 to 0.56 mg/l and 9% over 1 mg/l.

There has been a continuous increase in the consumption of mineral water in Brazil and the world, both for drinking and for preparation of foods, ice cubes and beverages, especially for children. Studies have indicated a concern among populations for seeking water of higher quality, which has been sought through the consumption of mineral water (Ayo-Yusuf et al1 2001; Brandão & Valsecki,2 1998; Stannard et al21 1990; Villena et al23 1996)

The present study found that around 29.7% of the population of Bauru was consuming mineral water: 14% had been using it for five years or more, while 15.6% had started consuming it during the last five years (1998-2003). That is to say, there was an increase in consumption of more than 50% over this period. It was further observed that 82.5% of the population that did not consume mineral water had some type of domestic water purifier. Moreover, 17.5% of the population that consumed mineral water also had water purifiers. Around 47.5% of all the homes made use of some type of water purifier: 7.7% tap-mounted purifiers, 9.8% wall-mounted purifiers and 30% bottle purifiers. These numbers are much higher than those found in Iowa, where 11% of the homes have water purifiers (Winkle et al23 1995). However, it is known that activated carbon filters, which are the type most widely employed in Brazil, do not remove fluoride from the water (Buzalaf et al8 2003). With
regard to the presence of fluoride in mineral water, around 45% of the population studied was aware of this fact, and 50% knew about its action in caries prevention.

According to Federal Law no. 8080 of September 19th, 1990, sanitary surveillance is defined as “a set of actions that can eliminate, reduce or prevent health risks and intervene in sanitary problems that result from the environment, the production and distribution of goods and the provision of healthcare-related services” (Brasil,1990). According to Narvai17 (2001), sanitary surveillance is one of the bases of public health and sanitary surveillance actions in the field of public health dentistry should encompass three fields: “dental care services, oral hygiene products, and foods and beverages” (Narvai,16 1998). The Brazilian sanitary surveillance system was reorganized by means of Law no. 9782 of October 25th, 1999, when the National Sanitary Surveillance Agency (ANVISA) was created. Its purpose is “to promote and protect public health by means of sanitary control over the production and commercialization of products and services that are subject to sanitary surveillance, including the environments, processes, supplies and technologies that are related to such products and services” (ANVISA, 2001). Since fluoride is a chemical element that may be present in various products, such as the public water supply, mineral water, beverages, juices, soft drinks, foods, and some drugs and nutritional supplements, the control and maintenance of its proper concentration are of interest to sanitary surveillance (Murray,15 1992; Narvai,17 2001; OMS,16 1972; Santoro,19 1997).

REFERENCES


