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Total and soluble fluoride content in commercial dentifrices in Chile

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Abstract

Objective. Regardless of the total amount of fluoride (F) in a dentifrice, only the soluble fraction presents anti-caries activity and its concentration must be above 1000 ppm. No information regarding the quality of F in Chilean toothpastes is available. The aim of this study, therefore, was to determine the F concentration present in commercially-available toothpastes in Chile. ***Materials and methods:*** Eighteen toothpastes of five different brands were tested, each purchased from three different chain drugstores in Chile. For each toothpaste, total, total soluble and ionic F concentrations were assessed and expressed as ppm of F (μg F/g). ***Results.*** Among the dentifrices evaluated, 77.8% contained NaF (sodium fluoride), 16.7% MFP (monofluorphosphate) and 5.6% both types of F salts; 77.2% of the dentifrices showed similar total F content to that declared by the manufacturer in the labeling with only five with lower concentration than expected, beyond a 5% disagreement tolerance range. Total soluble F ranged from 958.8–1496.3 ppm (mean ± SD: 1270.5 ± 183.7). Five toothpastes showed lower total soluble F than total F. Only one toothpaste showed soluble salt levels below the recommended dose for this type of dentifrice. ***Conclusion:*** Most Chilean dentifrices evaluated contained soluble F within the range of 1000–1500 ppm and, therefore, may have anti-caries activity.

Key Words: *dentifrice, toothpaste, fluoride, soluble fluoride, anti-cariogenic, dental caries, Chile*

Introduction

Dental caries remain an important Public Health concern in many countries [1]. Despite a decline in most developed countries [2], caries is still the most prevalent oral disease in several developing countries [3]. Topical application of fluoride (F) has proven to be an effective method to prevent dental caries [4]. Among topical F, dentifrice-containing F has become the most widespread way to provide the ion [4,5]. Indeed, incorporating F in toothpastes explains the reduction in caries experienced during the latest years in developed [4] and developing [6] countries. Many investigations have clearly shown the beneficial effect of fluoridated toothpastes in caries prevention [7,8]. Furthermore, toothpaste is the delivery vehicle of F most accessible for the majority of the population, regardless of their socioeconomic status [9].

Recently, two Cochrane systematic reviews were published [8] showing that toothpastes containing 1000 or more ppm of F, as sodium fluoride (NaF) or monofluorphosphate (MFP), are effective to prevent caries, when compared to placebo and 250 ppm F toothpastes. Likewise, it was reported that a higher concentration of F associates with caries decline in children and adolescents. The beneficial effect only becomes significant when a threshold concentration of 1000 ppm F is present in the toothpaste [8]. According to the US Food and Drug Administration regulation, dentifrices must contain at least 60% of its total F content as soluble ion F (F-) and soluble ions (F- and FPO_3^{2-}) for NaF- and MFP-containing, respectively. The content of total soluble F (F^- + MFP ion) is dependent on the presence of calcium contained in the abrasive to form insoluble salts that are ineffective against caries. Thus, manufacturers must provide total F concentrations that ensure F bioavailability within those ranges. F declared in the labeling of commercial dentifrices, however, do not provide information on the active soluble F and only inform about the total F

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concentration. Since only total soluble F should be considered potentially active against caries, knowing the content of this fraction when consumers choose a dentifrice is important. Information available from various countries shows that, in several cases, the declared F content does not match the actual concentration [10].

In Chile, a governmental regulation establishes that the dentifrices with F between 1000–1500 ppm should not be used by children under 6 years old [11]. The regulation also states that toothpastes with more than 1500 ppm F must only be prescribed by professionals [12]. Although the recommendation establishes that dentifrices intended for children older than 6 years old should not contain less than 1000 ppm and no more than 1500 ppm of total F [11], the regulation does not refer to the minimum total soluble F dentifrices must have. Manufacturers, therefore, are not constrained to label total soluble F concentration and no information regarding soluble F is available. The aim of this study was to evaluate the concentration of total F and its soluble fraction in commercial dentifrices available in Chile.

Materials and methods

Dentifrices tested

Eighteen dentifrices belonging to five brands commercialized in Chile were evaluated. Only dentifrices sold in three different chain drugstores during the time of the study were chosen for the analysis. One tube was purchased from each of the three stores. Each dentifrice was coded with a letter to allow blind analysis (Table I). All the samples were purchased and analyzed before their expiration date and the analyses were performed in duplicate.

Fluoride preparation

To determine F concentration, three different F fractions were assessed: (1) ionic F (IF), (2) total chemically soluble F (TSF or free F), which represents IF + F as MFP ion, and (3) total F (TF). TF corresponds to TSF + insoluble F (InF). InF is F bound to the abrasive. All the analyses were carried out in duplicates according to a protocol which has been used since 1980 at the Laboratory of Oral Biochemistry at Piracicaba Dental School, UNICAMP, Brazil [13]. In brief, 90–110 mg of each toothpaste was weighed (±0.01 mg) and homogenized in 10.0 mL of deionized water to create a suspension from which the different F fractions will be obtained and measured by a selective F electrode, as follows:

(1) TF: 0.25 mL in duplicate of the suspension was transferred to test tubes and 0.25 mL of 2 M HCl was added to break MFP ion into F ion and to dissolve InF bound to the abrasive. Acidified suspension was maintained 1 h at 45°C and neutralized afterwards using 0.5 mL 1 M NaOH followed by 1 mL TISAB II (1 M acetate buffer, pH 5.0, containing 1 M NaCl and 0.4% CDTA).
(2) TSF: dentifrice suspension was centrifuged (10 min at 3,000 *g*) at room temperature to remove InF bound to the abrasive. To 0.25 mL of the supernatant, 0.25 mL of 2 M HCl was added to break MFP into IF and the tube maintained for 1 h at 45°C. Acidified solution was then buffered with 0.5 mL 1 M NaOH followed by 1 mL of TISAB II.
(3) IF: To obtain the ionic F fraction, 1 mL TISAB II followed by 0.5 mL 1 M NaOH and then 0.25 mL 2M HCl was added to 0.25 mL of the supernatant.

Fluoride assessment

To determine F concentration in each of the three suspensions analyses, a F electrode (Orion model 96-09, Orion Research, Cambridge, MA) coupled to an ion analyzer (Orion EA-740) was used, which was calibrated with F standards containing 0.06–8.0 ppm F made with the same reagents used to prepare the samples. The concentrations of IF, TF and TSF were expressed in ppm (μg F/g) and the concentration of F as ion MFP was calculated by subtracting TSF minus IF. The concentration of InF was obtained by subtracting TF minus TSF concentrations and the percentage of InF was calculated by the formula % InF = 100[InF]/[TF].

Results

Among the 18 dentifrices evaluated, 77.8% contained NaF, 16.7% MFP and 5.6% both types of F (Figure 1). TF found in the F-toothpastes ranged from 1008.2–1505.9 ppm F (mean 1295.8 ± 165.2) (Table II). Although all the studied dentifrices had TF between 1000–1500 ppm, only 77.2% of them showed similar TF content to that declared by the manufacturer (Figure 2). On the other hand, some of the dentifrices (E, J, N, O and P) showed TF that exceeded the labeled amount. Toothpastes A, D, G, K and L had lower TF content than expected (Figure 2). To consider a dentifrice containing less than expected, a disagreement of ≤5% had to be found between the concentration labeled in the tube and the measured values [13].

F concentration in the TSF fraction ranged from 958.8–1496.3 ppm (mean 1270.5 ± 183.7) (Table II). When measured TF was compared with TSF, the active fraction, some of the dentifrices (C, D, N, O and P) showed lower TSF than TF (Figure 3). The missing part of TSF with respect to

Table I. Toothpastes under analysis. Codes used and composition declared by the manufacturers.

Commercial brand	Code	Labeled content of F (ppm) as NaF or MFP	Abrasive	Lot
Sensodyne Proteccion Total	A	NaF. 1450 ppm	Silica	LL0270 LL0269 LL0270
Yucral Fumadores	B	NaF 1450 ppm F	Hydrated Silica	C0904 C0904 C007
Colgate Total 12 Professional Clean	C	NaF 1450 ppm F	Hydrated Silica	9325BR 9343BR 9054BR
Pepsodent Xtra Whitening	D	MFP 1.11%, 1450 ppm F	Calcium Carbonate	11023 30413 30812
Aquafresh Triple Protection Kids Bubblemint	E	MFP 0.15% w/v F	Calcium Carbonate, Hydrated Silica	(L)9H10B (L)9H10B (L)9C16A
Sensodyne Blanqueador Extra Fresh	F	NaF 1384 ppm F	Silica	ND 0105 ND0105 ND0105
Pepsodente Doble Frescura	G	NaF 1450 ppm F or 0.32%w/w F	Silica	10152 10293
Pepsodent 3generation	H	NaF 0.32% w/w 1450 ppm F	Silica	10162 10091 10132
Aquafresh White & Shine	I	NaF 0.15% w/v F	Hydrated Silica	(L)9C23B (L)9C23A (L)9C23B
Aquafresh Triple Protection Toothpaste	J	MFP 1100 ppm F	Calcium Carbonate	(L)9F29A (L)9I07E (L)9I07F
Aquafresh Triple Protection Advanced 25% stronger	K	NaF 0.14% w/v F	Silica	(L) 8J06F (L)8I29B (L)8I29E
Aquafresh Extreme Clean	L	NaF 0.15% w/v F	Hydrated Silica	(L)9C09C (L)8H25B (L)8H25B
Aquafresh Ultimate White	M	NaF 0.15% w/v F	Hydrated Silica	(L)9G20E (L)9G20C (L)9G13B
Colgate MaxWhite	N	NaF 0.32% 1450 ppm F	Hydrated Silica	9229MX 9229MX 9229MX
Colgate Total 12 professional Clean Mint	O	NaF 0.32% 1450 ppm F	Hydrated Silica	9296BR 9083BR 9254BR
Colgate Máxima Protección Anticaries	P	MFP0.76% /NaF 0.1%: 1450 ppm F	Dicalcium Phophate	9271MX 9069MX 9272MX
Pepsodent White Now	Q	NaF 0.32% w/w 1450 ppm F	Silica	10253 NA 06/12C10
Pepsodent Doble Frescura Acquamint	R	NaF 0.32% w/w 1450 ppm F	Silica	10061 10071 10281

TF of those toothpastes corresponds to InF bound to the abrasive. Only one toothpaste (D) showed TSF values below the recommended dose for this type of paste (Figure 3). Taken together, almost all the Chilean dentifrices studied contained TSF within the range of 1000–1500 ppm, regardless of the TF contained.

Discussion

Fluoridated products have an indisputable role as anti-caries agents and are widely used [14]. Despite the relevance in knowing the F content in 'over the counter' toothpastes [10], no study has reported F concentration in the toothpastes commercialized in

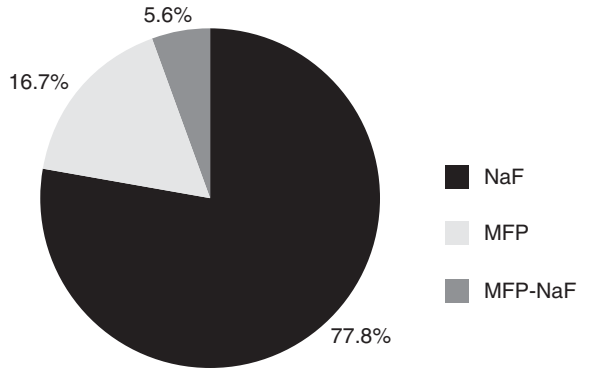

Figure 1. Fluoride formulation contained in Chilean toothpastes. Circular plot shows the percentage of the tested dentifrices containing; sodium fluoride (NaF), monofluophosphate (MFP) or both compounds.

Chile. From the results of this study, Chilean toothpastes commercially available seem to meet the regulation of F content within 1000–1500 ppm [15]. This requirement does not specify whether F must be soluble or not. In order to be active, F contained in a dentifrice must be soluble [8]. Even if TF content of toothpastes is high, important portions of the available F may have reacted with the abrasive, affecting the purpose of preventing caries or enhancing remineralization [16]. Previous studies have shown that when $CaCO_3$ or DCPD is used as an abrasive for MFP-containing toothpastes, a fraction of F precipitates as CaF_2 [17], which decreases the amount of soluble F. Decreases occur during storage of the products [18]. This is the reason to increase the amount of TF to ~ 1500 ppm when this combination is used [13]. With very few exceptions, most of the Chilean dentifrices tested here showed TSF close to TF (Figure 3), which indicates that most of the F present in the product is in an active form. Although five dentifrices showed lower TF than labeled, most of the products had concentrations close to the expected TF amount and within a 5% tolerance margin [13]. Although samples E, J and P present low IF concentrations (Table II), their total soluble F concentration (F^- + MFP ion) is appropriate to control caries. These toothpastes are formulated with MFP, which undergoes hydrolysis during storage releasing F ion. When toothpastes E, J and P are used to toothbrush, the anti-caries properties would be due to the immediate effect of F^- already present in the toothpaste and also to the ionic F released from MFP hydrolysis [19].

Perhaps the most important finding was that most of the dentifrice analyzed showed more than 1000 ppm of TSF, which, according to systematic reviews, is the minimum F concentration to achieve anti-caries efficacy [7,8]. The only toothpaste with less than 1000 ppm of TSF, nonetheless, had 958.8 ppm, which may not be clinically relevant as it is close to 1000 ppm TSF.

In general, toothpastes contain F either as ionic fluoride derived from NaF or from MFP. To interfere with the caries process, dentifrices must provide

Table II. Dentifrice codes, fluoride type, expected total fluoride (TF), measured ionic fluoride (IF), measured total soluble fluoride (TSF) and measured total fluoride of the tested dentifrices. Each measured value corresponds to the mean of three different tubes made in duplicate ± SD.

Dentifrices brand	Expected TF (ppm F)	Measured TF (ppm F)	SD	Measured IF (ppm F)	SD	Measured TSF (ppm F)	SD
Sensodyne Proteccion Total (A)	1400	1329.3	30.7	1330.8	38.9	1347.1	38
Yucral Fumadores (B)	1450	1397.4	31.7	1434.2	38.5	1443.5	34.5
Colgate Total 12 professional Clean (C)	1450	1386	77.8	1374.6	21.3	1361.9	10.6
Pepsodent Xtra Whitening (D)	1450	1359	119.9	455.3	69.6	958.8	82
Aquafresh Triple Protection Kids Bubblemint (E)	1100	1124.5	73.5	169.8	20.9	1125	71.2
Sensodyne Blanqueador Extra Fresh (F)	1384	1346	58.5	1395.1	87.4	1408.5	87.5
Pepsodent Doble Frescura (G)	1450	1334.8	94.1	1351	50.2	1356.7	59.2
Pepsodent 3generation (H)	1450	1387.2	57.7	1407.1	30.3	1395.9	32.1
Aquafresh White & Shine (I)	1100	1095.6	55.3	1107.2	54.7	1116.4	38
Aquafresh Triple Protection Toothpaste (J)	1100	1150	71.3	176.5	6.3	1149.8	84.6
Aquafesh Triple Protection Advanced (K)	1100	1028.1	26.8	1054.9	24.6	1064	25.6
Aquafresh Extreme Clean (L)	1125	1008.2	5.2	1051.9	8.1	1029.5	26.2
Aquafresh Ultimate White (M)	1100	1087.5	63.2	1072.4	60.5	1108.6	34.5
Colgate MaxWhite (N)	1450	1474.3	43.6	1457.8	51.5	1471.8	18.9
Colgate Total 12 professional Clean Mint (O)	1450	1505.9	26.6	1508.8	29	1496.3	55
Colgate Máxima Protección Anticaries (P)	1450	1484.8	10.2	333	18.6	1092.9	29.9
Pepsodent White Now (Q)	1450	1425.7	40.9	1522.1	37.6	1486.4	39.7
Pepsodent Doble Frescura Acquamint (R)	1450	1399.7	7.8	1472.7	13.9	1455.3	11

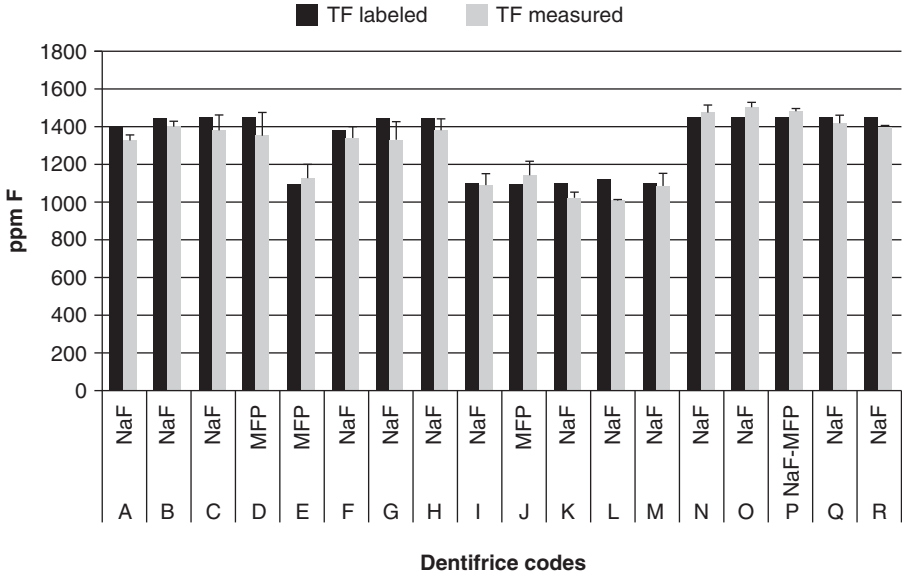

Figure 2. Labeled and actual total fluoride in toothpastes. Plot shows total fluoride content expected, from the manufacterers labeling (dark bars) and measured by the analyses (light bars) in ppm F. Light bars correspond to the mean total fluoride of three dentifrices per each commercial brand in duplicate ± SD.

soluble F in their formula, ensuring F bioavailability in the oral cavity during brushing [20]. Soluble F, either as fluoride ion or MFP ion, is capable to prevent demineralization and enhance remineralization of the hard dental tissues [21].

F bioavailability in the mouth depends on the chemical compatibility between the fluoridated agent, NaF or MFP, and the abrasive used. Most MFP-based dentifrices analyzed contained dicalcium phosphate dihydrate (DCPD) as an abrasive. Importantly, a fraction of MPF undergoes hydrolysis into the toothpaste tube and releases ionic F, which in turn binds to the abrasive. In the bound form, F becomes insoluble and inactive against caries [16,22]. Since most of the dentifrices analyzed had SiO_2 as an abrasive, NaF was used as a source of soluble F (Table II). Only three products contained MFP/$CaCO_3$ (D, E and J) in the formula, which represents 16.7% of the samples. Considering that in some fresh dentifrices the TSF is slightly lower than TF, further studies on aged dentifrices are desirable to verify F stability over time. Especially important, MFP-containing dentifrices should be monitored over time as the amount of InF in dentifrices containing $CaCO_3$ or DCPD as abrasives increases during storage [17,18]. Conversely, previous studies in different parts of the world

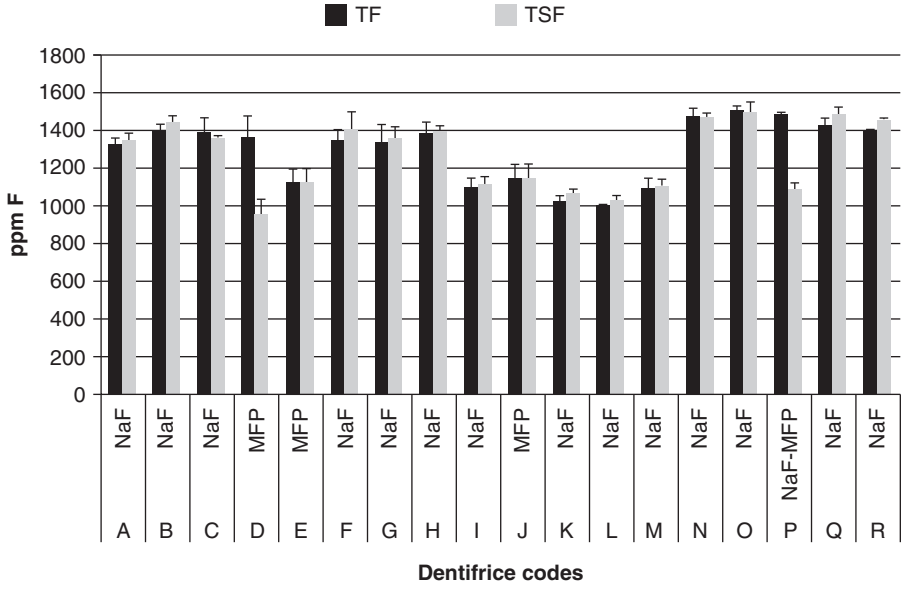

Figure 3. Total fluoride and total soluble fluoride. Plot shows measured total fluoride in the assays (dark bars) and measured total soluble fluoride by the analyses (light bars) in ppm F. Bars depict mean values of three dentifrices per commercial brand in duplicate ± SD.

have shown a marked trend to use MFP/$CaCO_3$ instead of NaF/SiO_2 [13,23], due to their lower price. As a matter of fact, in Brazil, at least 90% of the available toothpastes use MFP as the fluoridated compound and $CaCO_3$ as the abrasive agent. Since the majority of fluoridated dentifrices in Brazil contain MFP and its introduction coincides with a decline in caries incidence in the country [6,24,25], the efficacy of MFP as an anti-cariogenic agent is widely acknowledged.

In conclusion, toothpastes available in Chile match the declared content of TF. Dentifrices contain TSF close to TF, ensuring bioavailability, and meet the internationally acceptable soluble F concentration of 1000 and 1500 ppm to attain an anti-caries effect.

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