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ORIGINAL ARTICLE

Total and soluble fluoride concentration present in various commercial brands of children toothpastes available in Saudi Arabia – A pilot study

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Abstract *Objective:* The aim of this pilot study was to perform chemical analysis and investigate the total and soluble fluoride concentrations in various brands of children toothpastes.

Materials and methods: Three samples of five different commercial brands of children toothpastes were collected and divided into five groups; group A – Biorepair Oral Care toothpaste containing no fluoride (control), group B – Signal Kids Strawberry toothpaste having 500 ppm fluoride, group C – Aquafresh Milk Teeth toothpaste having 500 ppm fluoride, group D – Aquafresh Little Teeth toothpaste having 500 ppm fluoride, and group E – Siwak F Junior having 400 ppm F. The total fluoride (TF) and total soluble fluoride (TSF) concentration of the toothpastes was determined using fluoride ion selective electrode. Data were analysed using Paired sample *t*-test.

Results: The measured TF values were inconsistent with that of the declared concentrations by the manufacturers. Mean TF found in the toothpastes ranged between 2.37 and 515.74 ppm whereas, the mean TSF ranged between 2.00 and 503.4 ppm. For two groups, TF was more than the declared TF whereas for the other three groups, it was less than the declared concentration. All the differences between the declared and observed TF concentration were statistically significant ($p < .05$) except for one group. All the toothpastes demonstrated mean TSF slightly lower than their respective observed mean TF concentrations.

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Conclusion: The analysis of TF and TSF concentrations revealed variations from the labelled claims. Therefore, some of the toothpastes may have doubtful anti-caries effectiveness owing to deficiency of total and soluble fluoride.

1. Introduction

Dental caries involves loss of tooth structure as a result of acid attacks by cariogenic bacteria in the oral cavity after they ferment dietary carbohydrates (Featherstone, 2008). The use of fluoride based interventions has reduced the incidence of early childhood caries (Ammari et al., 2007). Fluoride (F^-) is an ion which replaces hydroxyl (OH^-) ion in the apatite structure of enamel and converts hydroxyapatite into fluorapatite, which is more resistant against dental caries (Mohammed et al., 2014). This F-OH exchange decreases the enamel's surface solubility thus making it stronger against acid attacks (Abou Neel et al., 2016).

Fluoride is commonly incorporated in toothpastes, gels, and mouthwashes for the prevention of caries (Ullah and Zafar, 2015). The relationship between fluoride and prevention of caries has been studied extensively in the literature previously (Somaraj et al., 2017; Clark and Slayton, 2014). In general, greater the concentration of fluoride incorporated in the toothpaste, higher will be the probability of caries prevention (Walsh et al., 2010). However, to be effectively anticarious, the formulation of toothpaste should be such that it provides sufficient soluble fluoride to enhance remineralization (Cury and Tenuta, 2008). The bioavailability of fluoride in toothpaste depends on the type of fluoride compound and abrasive present in the formulation (Cury et al., 2010). Three of the most common fluoride compounds added to a toothpaste are sodium fluoride (NaF), sodium monofluorophosphate (MFP), and strontium fluoride (SnF_2) whereas, most commonly used abrasives in toothpastes are silica, calcium carbonate, dicalcium phosphate dehydrate, calcium glycerophosphate, and tricalcium phosphate (Volpe, 1982).

Fewer researches are available worldwide which showed a difference between the declared and real concentration of fluoride present in children's toothpaste (Thakkar et al., 2015). Therefore, the aim of the study was to assess total fluoride (TF) and total soluble fluoride (TSF) existing in children toothpastes available in the markets of Saudi Arabia.

2. Materials and methods

The ethical approval (Ref: EA 2,016017) was obtained from the Scientific Research Unit of College of Dentistry, Imam AbdulRahman Bin Faisal University.

2.1. Toothpaste selection

Randomly selected, children toothpastes of different brands were divided in to five groups

Group A: Biorepair Oral Care toothpaste containing no fluoride.
Group B: Signal Kids Strawberry toothpaste having 500 ppm fluoride.
Group C: Aquafresh Milk Teeth toothpaste having 500 ppm fluoride.
Group D: Aquafresh Little Teeth toothpaste having 500 ppm fluoride.
Group E: Siwak F Junior having 400 ppm F.

2.2. Preparation of measurement apparatus

The details of active ingredients, expiry date, and abrasive in the toothpaste were noted. All the toothpastes were given codes to permit blind analysis of total fluoride (TF) and total soluble fluoride (TSF) concentration by direct potentiometry and the samples were run in triplicate. A multimeter (VWR® sympHony™, United States) was first prepared with an assembled fluoride ion selective electrode (Orion 9609BN, 710A meter, South Burlington, VT, USA) as an indicator electrode and a reference electrode (silver – silver chloride or double junction). A working solution of fluoride concentration varying from 10^{-1} to 10^{-7} M was prepared from a stock solution of 0.1 M sodium fluoride (NaF). Each standard solution with known volume of 0.1 M NaF was then mixed with 25.0 mL of total ionic strength adjustment buffer (TISAB) solution and diluted up to the mark into a 100 mL plastic volumetric flask. The solution was transferred to a plastic beaker for analysis. The whole process was repeated with the remaining concentration. A previously cleaned and dry indicator and reference electrode were inserted into the solution and swirled gently. The potential difference on the multimeter was recorded after the reading was stabilized. The electrodes were removed and gently swiped with a soft tissue, dried, and left in the air during analysis.

2.3. Analysis of total fluoride concentration (TF)

One gram of toothpaste was weighed in a clean and dry 100 mL beaker and the reading was recorded. After weighing the toothpaste, 25 mL of water and 25 mL of TISAB solution were poured into the beaker along with the toothpaste. The solution was boiled in a hot plate for 5 min to ensure thorough mixing of toothpaste in the solution. The solution was cooled and then stirred for 10 min to obtain a homogenous mixture. This mixed solution was then poured into a 100 mL plastic volumetric flask and diluted up to the mark. Afterwards, it was transferred to a plastic beaker for quantitative analysis.

2.4. Analysis of total soluble fluoride concentration (TSF)

The same steps for the preparation and calculation of TF were repeated except that before reading the potential difference, the sample solution was filtered to remove some of the sediment insoluble matters of the toothpaste.

2.5. Calculation formula

Using the collected standard calibration curve, the fluoride concentration in Molarity (M) of an unknown solution was calculated through an equation of the line. The graph was plotted between y-axis (potential difference, mV) and x-axis (logarithmic of concentration of F). The percentage of fluoride and ppm of fluoride were calculated by the following formulas

(a) $\%F = \frac{\text{Molarity of F (mol/L)} \times \text{Volume of sample (L)} \times 19 \text{ g/mol of F} \times 100}{\text{Mass/Weight of toothpaste (g)}}$

(b) ppm F = % F × 10,000

2.6. Statistical analysis

The SPSS software (version 19.0; SPSS Inc., Chicago, IL, USA) was used in the analysis. Paired sample *t*-test was applied to compare the difference in the declared and real TF. P-values of $< .05$ were considered statistically significant.

3. Results

The active ingredient is sodium fluoride in group B, C and D whereas group A and E contain hydroxyapatite and sodium monofluorophosphate respectively (Table 1).

All the toothpastes showed variations in TF concentration from their label claims. Mean TF found in the toothpastes ranged between 2.37 and 515.74 ppm whereas, the mean TSF ranged from 2.00 to 503.4 ppm (Table 2). For groups A and B, mean TF was more than the declared TF whereas for groups C, D, and E, it was less than the declared F concentration on the packaging (Table 2). All the differences between the declared and observed TF concentration were statistically significant ($p < .05$) except for group C. All the toothpastes demonstrated mean TSF slightly lower than their respective observed mean TF concentrations (Fig. 1).

4. Discussion

The TF concentration for all the analyzed toothpastes in this study was different from their respective declared TF concentrations. Previously carried out studies in different parts of the world also showed no harmony between the declared and observed concentrations of TF (Jordan et al., 2011, Kikwilu et al., 2008, and van Loveren et al., 2005). These worldwide differences existing between the declared and real TF concentrations in toothpastes could lead to under- or over-exposure of fluoride, both being detrimental to some extent for the consumer.

All the toothpastes in this study showed a lower concentration of TSF than their respective TF concentration. Previously, a similar study from Brazil also showed that around 50% of the dentifrices showed a TSF lower than that of their TF concentration (Cury et al., 2010). The probable reason for this is the incompatibility between the abrasive agent and the type of fluoride compound present in the composition of toothpaste, making the percentage of soluble fluoride less than TF concentration (Hattab, 1989). In general, NaF toothpastes have better compatibility with silica abrasive (Benzian et al., 2012), whereas, MFP toothpastes are more compatible with calcium containing abrasives (Volpe, 1982), although calcium carbonate abrasive can still inactivate fluoride in a MFP toothpaste (Sebastian and Siddanna, 2015).

In our study, three of five dentifrices had NaF as an active ingredient with silica as an abrasive agent, one toothpaste with MFP as an active ingredient had calcium carbonate as its abrasive agent, and one was labeled fluoride free. Although the three NaF toothpastes (groups B, C, and D) did show differences (two were statistically significant) between declared and real TF concentrations, these differences were not big (<30 ppm F) and this could be because of the better compatibility present between NaF compound and silica abrasive, previously proposed by Benzian et al., 2012. In a similar study carried out on toothpastes from various countries, it was reported that around 85% of the children's toothpastes had less TF concentration in comparison with their label claims (Benzian et al., 2012). These findings are similar to our study where two thirds of toothpastes showed less TF concentration and all groups showed discrepancies from their label claims.

The biggest difference (>140 ppm F) was observed for the MFP toothpaste (group E) having calcium carbonate abrasive and the reason for this difference could be the inactivation of MFP caused by calcium carbonate (Sebastian and Siddanna, 2015). In a MFP molecule, the fluoride is attached to phosphorus but the bond present between these two is unstable therefore, when the released fluoride reacts with the calcium of the abrasive (Conde et al., 2003), it precipitates as insoluble calcium fluoride (CaF_2), which becomes inactive against caries in the toothpaste (Ellwood et al., 2008). It has been suggested that if a combination of MFP and calcium carbonate has to be used, it should be determined that ample of free fluoride is available by adding extra or excess fluoride in the dentifrice (Thakkar et al., 2015). One of the toothpaste was labelled as "fluoride free" (group A) but on analysis, small quantity of TF and TSF were observed in it. This particular brand of toothpaste mentions its active ingredient as "microrepair® composed of hydroxyapatite" (http://www.biorepair.it/Products/Children). The possible reason for the appearance of fluoride in this toothpaste could be manufacturer's negligence or maybe the companies are using a combination of fluoride hydroxyapatite, as previously used in a study where fluoride ions were merged into the apatite lattice through precipitation and growth reactions (Oliveira and Mansur, 2007).

The findings from our study place a question mark on the anti-caries ability of the analysed toothpastes. Majority of

Table 1 Percentage of active ingredients and abrasives present in toothpaste samples according to the information present on the packaging.

Groups	Active ingredient	Percentage of active ingredient	Abrasive
A	Hydroxyapatite	Not mentioned	Silica
B	Sodium fluoride	500 ppm	Silica
C	Sodium fluoride	500 ppm	Silica
D	Sodium fluoride	500 ppm	Silica
E	Sodium monofluorophosphate	400 ppm	Calcium carbonate

Table 2 Evaluated toothpastes, coding for blind analysis, mean TF, declared TF, and mean TSF.

Name of the toothpaste	Coding	Mean TF (ppm)	Label claim (TF, ppm)	P-value (Significance)	Mean TSF (ppm)
Biorepair® oral care	A	2.37	0.0	.020*	2.00
Signal kids strawberry	B	515.74	500.0	.014*	503.43
Aquafresh milk teeth	C	489.85	500.0	.063	485.51
Aquafresh little teeth	D	472.92	500.0	.002*	469.42
Siwak F junior	E	259.64	400.0	.000*	257.38

* *T-test, significant at $p < .05$.*

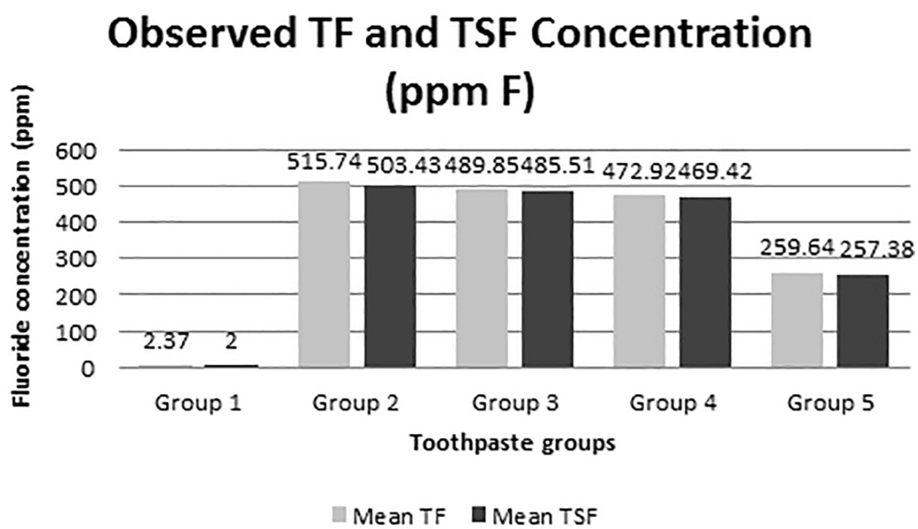

Fig. 1 Concentration of TF and TSF (ppm) in the toothpastes analyzed.

the toothpastes were not containing the declared TF concentration (except one which contained more than the expected TF concentration) which results in decreased presence of soluble fluoride which is essential for the remineralization of tooth structure. The reasons for the variations between declared and real concentrations of TF include carelessness during manufacturing of toothpastes, use of cheaper ingredients to increase profit margins, and presence of higher temperatures during storage (which can affect the stability of fluoride) (Sebastian and Siddanna, 2015).

Since it was a pilot study, one of the limitations of the present study is the small sample size. There is a less variety of children's toothpastes available in Saudi Arabian market and an attempt was made to include many (if not all) commercial brands in this pilot study. This should be kept in mind that these results are not absolute representative of the fluoride concentrations of all the other toothpaste samples of these commercial brands and the others can have different concentrations. Future studies which include more samples of children toothpastes are suggested by authors. The other limitation is the absence of a standardized methodology for analysis of fluoride concentrations. Different researchers use a variety of techniques and the methodology used in this study could be different from the others, yielding altered results.

It is recommended that manufacturers should try to avoid discrepancies between the mentioned TF and real TF concentration and should also declare the TSF concentration on their packaging. For the consumers, it is recommended that they should prefer toothpaste which has silica-NaF (abrasive-active ingredient) combination clearly mentioned on the packaging and the toothpaste is well within the period of its expiry date.

5. Conclusion

The analysis of total and soluble fluoride concentrations various brands of children's toothpastes available in Saudi Arabia showed variations from the labelled claims of the manufacturers in various brands. Therefore, some of the toothpastes may have doubtful anti-caries effectiveness owing to deficiency of total and soluble fluoride.

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Conflict of interest

None.

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