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Original Article

Dentistry Section

Total and Free Fluoride Concentration in Various Brands of Toothpaste Marketed in India

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ABSTRACT

Background: For fluoridated toothpaste to be effective in controlling dental caries, an adequate concentration of soluble fluoride must be available in the oral cavity.

Aim: To determine the total and free fluoride concentration in various brands of toothpaste marketed in India.

Materials and Methods: Three samples of 12 different toothpastes were purchased from supermarkets in Mysore city, Karnataka, India. Toothpastes were analysed in duplicate using a fluoride ion-specific electrode. The concentration of total fluoride (TF) and total soluble fluoride (TSF) were determined.

Results: Measured TF was consistent with that declared by the manufacturer in five products. Four toothpastes showed lower TF and two higher TF than declared. Most toothpastes exhibited TSF concentrations similar to the TF content except four samples that displayed considerably lower TSF than TF.

Conclusion: The measurement of total and free fluoride concentrations of toothpastes available in India showed inhomogenities. Therefore there is a need for stringent regulatory control measures for the determination of fluoride content in toothpastes in developing country like India.

Keywords: Dental caries, Dentrifice, Fluoride content, Fluoride ion electrode

INTRODUCTION

Recent reports in dental literature demonstrate a decline in the dental caries trend in most developed nations and this is mainly attributable to the use of fluorides in different forms [1]. However, the problem of dental caries is still existing as a disease of high propensity in many under developed and developing countries of Africa and Asia including India due to lack of public awareness and motivation, inadequate resources for dental treatments and changing dietary habits [2]. Indeed, the use of fluorides in different forms is still recognized as one of the most successful measures for caries prevention [3]. A panel of experts convened in Geneva for the "Global consultation on Oral Health through Fluoride" in the year 2006. They stated that the only realistic way of reducing caries burden is by using fluoride [4]. Subsequently a joint meeting of World Health Organization, FDI, World Dental Federation and International Association for Dental Research was conducted at Beijing in the year 2007. During the meeting there was a call to action stating that:

- Fluoride toothpaste is the most significant form of fluoride used globally and the most rigorously evaluated vehicle for fluoride use [5].
- Since 1940, over 100 clinical trials have assessed the effectiveness of fluoride toothpaste and these trials have confirmed the anti-caries efficacy of fluoride toothpaste [5].
- Irrespective of low, normal or high fluoride exposure from other sources, fluoride toothpaste is safe to use [5].

This call to action recommends the promotion of fluoride toothpaste for the mass prevention of dental caries [6]. However, fluoride must be chemically free in the toothpaste formulation [7], so as to ensure adequate fluoride bioavailability in the oral cavity during tooth brushing [8]. This bioavailability depends on the chemical compatibility between the type of fluoride added and the abrasive used in the toothpaste formulation [9]. The fluoride compounds present in toothpastes such as NaF, NH_4F, SnF_2 are not compatible with abrasives that contain calcium (eg: calcium carbonate, dicalcium phosphate dehydrate, calcium glycerophosphate, tricalcium phosphate). By contrast, toothpaste which contains sodium mono fluorophosphates, the PO_3F_2- component has greater compatibility with calcium containing abrasives [10]. This is because the fluoride which is present in PO_3F_2- is firmly bound to the phosphate and cannot bind to soluble calcium to form insoluble calcium fluoride [6].

In most developing countries, the majority of the toothpastes contain calcium carbonate as abrasive, which can inactivate fluoride present in the toothpaste, even if they are formulated with sodium monofluorophosphate. A study in Brazil reported that 13 out of 14 MFP toothpastes with calcium containing abrasives used by children contained 15-50% less free available fluoride than the total fluoride content [9]. By contrast in all the NaF toothpastes which contain silica as abrasive, almost all the fluoride was available in the free ionized form and these findings raise concerns regarding the compatibility of calcium containing abrasive with PO_3F_2- component [6].

A recent systematic review of literature evidenced that only toothpastes with 1000 ppm F or more would have anti-caries effect. This has increased the relevance of the requirement that toothpaste should have soluble fluoride in their formulations [11]. It is therefore necessary to determine the concentration of total and free fluoride in toothpastes in order to evaluate the quality and stability of toothpastes [12]. The aim of this study, therefore, was to analyse total and free fluoride concentrations of commercially available toothpastes in India.

MATERIALS AND METHODS

Fluoride Toothpaste Samples

The study was conducted in the Department of Environmental Engineering, Sri Jayachamarajendra College of Engineering, Mysore during the month of October 2013 after obtaining prior permission from the respected authorities. Twelve tooth pastes, commercially intended for adults, belonging to eight different companies were purchased from super markets in Mysore city, India. The 12 tooth pastes were bought in triplicate, each of them from one store (total=36) verifying that they were from different lots. Only tooth pastes sold in three different stores were purchased. All the tooth paste samples were checked for information provided on the packaging. The information on the package was checked for

descriptive names of the fluoride component and its concentration in parts per million (ppm). Descriptive names of abrasives on the packaging were recorded. In tooth pastes in which both silica and calcium containing abrasives were declared, the calcium containing abrasive was recorded. The expiry date was recorded for every sample and in case if the date of production was declared, then the expiry date was recorded as three years later.

Determination of Total and Free Fluoride Concentration

All tooth paste samples were coded with letters to allow blind analysis and F determination was made in duplicate. As to the determination of F concentration, 2 forms of F available in the toothpastes were determined: total soluble F (TSF) and total F (TF) (the sum of total soluble fluoride (TSF) plus insoluble F (IF) that is the F bound to the abrasive).

Analyses were carried out in duplicates according to a protocol modified from Pearce [13], which has been used since 1980 at the Laboratory of Oral Biochemistry at Piracicaba Dental School, as described: 90 to 110 mg of toothpaste were weighed (± 0.01 mg), homogenized in 10.0 mL of deionized water and duplicates of 0.25 mL of the suspension were transferred to test tubes for TF analysis. The remaining of the suspension was centrifuged (3,000 g, 10 min, r.t.) to remove IF bound to the abrasive. Duplicates of 0.25 mL of the supernatant were transferred to assay tubes to determine TSF concentrations. For the TF and TSF tubes, 0.25 mL of 2.0 M HCl was added, and after 1 h at 45^{0}C, the samples were neutralized with 0.5 mL 1.0 M NaOH and buffered with 1.0 mL of TISAB II (1.0 M acetate buffer, pH 5.0, containing 1.0 M NaCl and 0.4% CDTA).

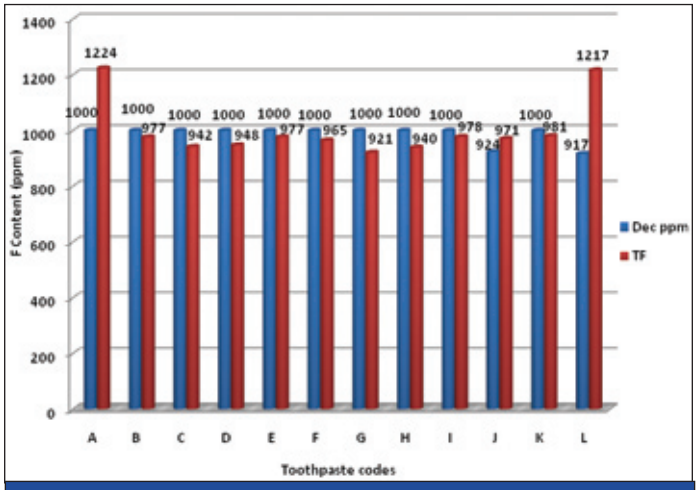
To determine fluoride concentration, fluoride electrode (Orion model 96-09, Orion Research, Cambridge, MA) coupled to anion analyser (Orion EA- 740) was used, which was previously calibrated with F standards containing 0.06-8.0 ppm fluoride, prepared with the same reagents used to prepare the samples [9].

STATISTICAL ANALYSIS

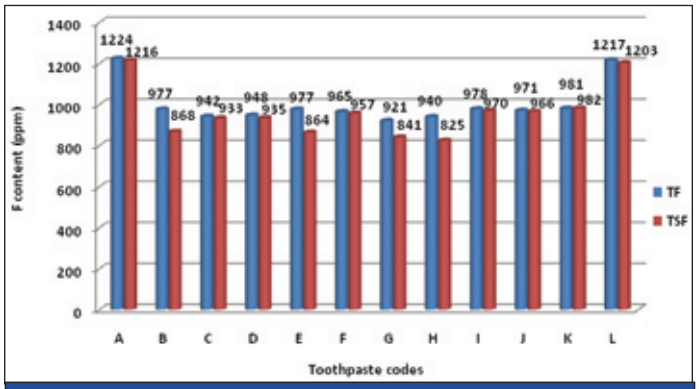
To consider a toothpaste containing less or more than expected value, a disagreement of <5% or >5% had to be found between the concentration labeled in the toothpaste tube and the measured value [14]. A linear regression between F concentration in standards and mV was constructed using the software Microsoft Excel, which was used to determine fluoride concentration in each toothpaste expressed as ppm. During repeated analysis (duplicate), the average variation coefficients were less than 2%. The average and standard deviation values of fluoride concentration of each toothpaste brand were calculated using Statistical Package for Social Sciences (SPSS version 19.0, Chicago IL, USA) [14]. All the analyses were conducted in duplicate from each toothpaste tube.

RESULTS

Twelve tooth pastes intended for adults were purchased from Indian market [Table/Fig-1]. On the basis of their declared fluoride content, dentrifices can be arbitrarily divided in those containing 1000 ppm F (dentifrices A,B,C,D,E,F,G,H,I and K) and those containing less than 1000 ppm F (dentifrices J and L). When the F salt was considered, 33.3% of the dentifrices contained NaF as the active ingredient and 66.6% were formulated with mono fluoro phosphate (MFP). Regarding the abrasive, MFP-based tooth pastes contained calcium carbonate or silica and tooth pastes formulated with NaF, on the other hand, had silica.



[Table/Fig-2]: Concentration (ppm F) of Total Fluoride declared by the manufacturer (Expected F) and found in the toothpaste analysed (TF)
A-Colgate Total 12; B-Colgate Sensitive; C-Close Up Deep Action; D-Colgate Gel; E-PepsodentGermi Check; F-Pepsodent Gum Care; G-Cibaca; H-Anchor; I-Sensodyne; J-Sensodent- KF; K-Parodontax; L-Senquel-F



[Table/Fig-3]: Concentration (ppm F) of Total Fluoride (TF) and Total Soluble (TSF) found in the toothpaste analysed
A-Colgate Total 12; B-Colgate Sensitive; C-Close Up Deep Action; D-Colgate Gel; E-Pepsodent Germi Check; F-Pepsodent GumCare; G-Cibaca; H-Anchor; I-Sensodyne; J-Sensodent- KF; K-Parodontax; L-Senquel-F

Commercial Brand	Code	Fluoridated Agent	Fluoride expected	Abrasive agent	Mean TF Analysed	Mean TSF Analysed
Colgate Total 12	A	NaF	1000 ppm	Silica	1224 ppm	1216 ppm
Colgate Sensitive	B	NaMFP	1000 ppm	$CaCO_3$	977 ppm	868 ppm
Close up Deep action	C	NaF	1000 ppm	Silica	942 ppm	933 ppm
Colgate Gel	D	NaMFP	1000 ppm	Silica	948 ppm	935 ppm
Pepsodent Germicheck	E	NaMFP	1000 ppm	$CaCO_3$	977 ppm	864 ppm
Pepsodent Gumcare	F	NaF	1000 ppm	Silica	965 ppm	957 ppm
Cibaca	G	NaMFP	1000 ppm	$CaCO_3$	921 ppm	841 ppm
Anchor	H	NaMFP	1000 ppm	$CaCO_3$	940 ppm	825 ppm
Sensodyne	I	NaMFP	1000 ppm	Silica	978 ppm	970 ppm
Sensodent- KF	J	NaMFP	924 ppm	Not mentioned	971 ppm	966 ppm
Parodontax	K	NaF	1000 ppm	Not mentioned	981 ppm	982 ppm
Senquel-F	L	NaMFP	917 ppm	Not mentioned	1217 ppm	1203 ppm

[Table/Fig-1]: Toothpastes analysed. Codes for blind analysis and information provided by the manufacturers

Mean TF found in the low F tooth pastes (dentifrices J and L) was 1094 ppm (971-1217 ppm F). For high F tooth pastes (dentifrices A,B,C,D,E,F,G,H,I and K) mean TF content was 985.3 ppm (921-1224 ppm F). Five products (41.6%) showed measured TF content in consistency with that declared by the manufacturer i.e. disagreement < 5% [Table/Fig-1]. Dentifrices C, D, G and H showed lower TF concentration than the content labeled by the manufacturer with a disagreement > 5%, whereas dentifrices A, J and L showed higher concentration than expected i.e. disagreement > 5% [Table/Fig-2].

When TSF content was considered, high F tooth pastes (dentifrices A,B,C,D,E,F,G,H,I and K) had a mean value of 939.1 ppm (825-1216 ppm F), whereas low F dentifrices (dentifrices J & L) showed 1084.5 ppm F (966-1203 ppm F). Most of the evaluated tooth pastes showed similar measured TF to measured TSF [Table/Fig-2] which indicates good bioavailability of F. Only tooth pastes B, E, G and H showed lower TSF than TF [Table/Fig-3].

DISCUSSION

There have been substantial decline in dental caries in industrialized countries during the last few decades which have been attributed to the widespread use of fluorides in both systemic and topical forms with. Among the topical delivery vehicles, fluoride toothpaste is being the most widespread form of topical usage [15]. Tooth brushing with fluoride toothpaste has become an important public health measure in preventing dental caries [16]. Around 500 million people of the world's population use fluoride toothpaste, thereby making it the most important topical fluoride delivery system [17].

Fluoride is incorporated into various products such as toothpastes, mouth rinses due to its ability to inhibit dental caries [12]. The regular application of fluoride from toothpaste provides protection by exerting a topical effect on the tooth surfaces. Fluoride interacts with the plaque/ tooth system mainly in four ways: enhancement of remineralization of incipient lesions, reduction of enamel solubility, prevention of mineral dissolution and inhibition of acid production by plaque bacteria [18,19]. For fluoridated toothpaste to be effective in controlling dental caries, it is essential that an adequate concentration of fluoride must be soluble. However, the total fluoride contained in the toothpaste is not completely available as some forms of fluoride may link to the abrasive contained in the toothpaste formulation [20]. As there can be a discrepancy between manufacturer reported and biologically active fluoride for caries prevention, assessment of fluoride in toothpastes become relevant [13].

Previous studies from different parts of the world have shown questionable anti-caries efficacy of fluoridated toothpaste as a result of lack of free available fluoride. A study conducted by Jordan et al., reported inhomogenities in the measurement of total and free fluoride concentrations of toothpaste available in Gambia (West Africa) [21]. Similar study conducted by Kikwilu et al., reported that toothpaste manufactured in Tanzania had free fluoride concentrations below the optimum level for dental caries prevention [22]. Lack of consistency between manufacturer reported and actual fluoride concentration will compromise the anti-caries efficacy. Hence monitoring and controlling products such as toothpaste should be routinely performed [13].

Toothpaste A and L had measured TF higher than the content labeled by the manufacturer, with a disagreement of 22.4% and 32.7% respectively. When TSF concentration of the toothpastes is analysed considering 1000 ppm F as the minimum effective anticaries concentration, only products A, I, J, K and L would have full preventive effect. The other toothpastes contain lower TSF concentration, particularly MFP toothpastes. Reasons for inconsistency in free fluoride concentrations include, but are not limited to:

1. Global manufacturers produce toothpastes locally through sub-contracted companies. These products may not be produced correctly or may be compromised for quality.
2. Toothpaste which is of low price may use cheaper ingredients especially abrasives so as to increase profit margins.
3. Dishonest distributors in our country may sell large batches of toothpaste which is nearing its expiry date.
4. Fake products that initiate recognized brands may be of poor quality.
5. Depending on the shop, location, season etc, toothpastes may have a slow turnover rate. This will result in lengthy shelf life and sales near or beyond the expiration date.
6. High temperature during storage will have a negative influence on the stability of toothpaste [6].

Labels of commercial toothpaste declare only information regarding the total fluoride and not total soluble fluoride concentration. Total soluble fluoride should be considered as the only source of fluoride responsible for anti-caries effect during tooth brushing [23]. A Cochrane review by Wong et al., found evidence on the caries protective action of using toothpaste of 1000 ppm or more when compared to those containing 250 ppm fluoride [15]. Hence it is essential that fluoride toothpastes contain sufficient amount of free available fluoride so as to ensure their effectiveness [6]. Our study has shown that some of the toothpaste samples have insufficient levels of fluoride concentrations and this can have implication in their efficacy in reducing dental caries. This will predispose our people to a public health affliction particularly in those areas where the fluoride intake is not balanced through drinking water.

Strong national drug and consumer product regulatory authorities should be established in our country so as to tackle the weak quality control and lack of compliance on the part of toothpaste manufacturers. These bodies can also be responsible for ensuring that fluoride products on the market comply with minimum labelling and packaging requirements [6].

LIMITATIONS

This study has several limitations and therefore its results should be interpreted with caution. These limitations include:

- **Sampling:** The toothpaste samples included in this study were not obtained in a systematic manner. Because of small sample size, the results cannot be considered representative of fluoride content or labelling quality of any particular brand although we list the respective brand names in the result tables, we do not imply that other samples of these brands, or the brand overall would show the same results in further analysis.
- **Analysis methodology:** Currently there is no standardized methodology to measure free available fluoride concentrations in toothpaste. The methodology used in this study may differ from methods used by other laboratories or the manufacturer themselves.
- **Genuine and fake products:** It has become increasingly complicated, if not possible, to distinguish between a genuine and a counterfeit product. Labelling and packaging are all too often copied so accurately that even expert has difficulty in clearly identifying fake products. It is thus possible that some of the samples included in the study are in fact fake products. Although this would be an important finding with far reaching consequences for consumers, national regulatory bodies and manufacturers, we can neither exclude nor confirm this assumption [6].

In spite of these limitations, our results highlight problem areas related to fluoride toothpaste, particularly in countries in which quality control bodies for consumer products are weak.

RECOMMENDATIONS

1. Check the expiry date of toothpaste before purchasing.
2. Do not purchase toothpaste that does not show an expiry date.
3. Silica-based fluoride toothpaste without any calcium containing abrasives is likely to have more anti-caries efficacy [6].

CONCLUSION

The measurement of total and free fluoride concentration of toothpastes available in India showed inhomogenities. Some of the toothpaste samples in our study may have questionable anti-caries efficacy due to lack of free available fluoride. This point towards the need for regulatory control measures for the analysis of fluoride concentrations in dental products in developing countries like India.

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