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Evaluation of Effect of Brushite-Calcite and Two Indigenous Herbs in Removal of Fluoride from Water

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ABSTRACT

Introduction: The acceptable concentration of fluoride in drinking water is 1.5mg/l. Excess fluoride in drinking water causes fluorosis. Fluorosis is an important public health problem in India. Several treatment technologies suggested in the past for removing excess fluoride generated and causes various chemical byproductswhich are hazardous to public. In recent years, there has been a resurgence of interest to use natural materials due to cost and associated health and environmental concerns of synthetic organic polymers and inorganic chemicals.

Aim: The aim of this study was to evaluate and compare the defluoridating capability of the brushite-calcite with that of two indigenous herbs, tulsi and wheat grass.

Materials and Methods: One gram of brushite-calcite combination, tulsi and wheat grass were separately added to 10 containers, each containing 1.0 l of prepared distilled water with a fluoride concentration of 5ppm and naturally fluoridated water at 2ppm. Half of the samples were boiled for one minute in a domestic electric kettle for one minute and allowed to cool. The remaining half of the samples was left un-boiled. Fluoride concentration in all the samples was assessed at the end of 30 minutes and 24 hours using fluoride ion selective electrode method. Data was analyzed using unpaired t-test and one-way ANOVA.

Results: For water with 2ppm and 5ppm fluoride, brushite-calcite had shown highest de-fluoridation capacity (p=0.001) at the end of both 30 minutes and 24 hours in boiled samples whereas tulsi (p=0.001) was most effective in un-boiled samples.

Conclusion: The results of the study suggest that tulsi can be used for domestic water defluoridation as it is economic, safe and effective.

Keywords: Dental, Defluoridation, Fluorosis, Tulsi, Wheat grass

INTRODUCTION

Water is life for people and for the planet. It is essential to the well-being of humankind, a vital input to economic development, and a basic requirement for the healthy functioning of all the world's ecosystems. Ironically, more than 125 million children under five years of age live in households without access to an improved drinking-water source [1].

The importance of water was stressed in the Millennium Development Goals adopted by the General Assembly of the United Nations (UN) in 2000. The UN General Assembly also declared the period from 2005 to 2015 as the International Decade for Action, "Water for Life" to achieve the Millennium Development Goal (MDGS) to reduce by half the proportion of the world's population without sustainable access to safe drinking water and sanitation by 2015 [2]. Fluoride is one of the very few chemicals that have been shown to cause significant effects in people through drinking water. Fluoride is often described as a 'double-edged sword' as inadequate ingestion is associated with dental caries, where as excessive intake leads to dental, skeletal and soft tissue fluorosis which has no cure [3].

According to the World Health Organization (WHO), the maximum acceptable concentration of fluoride is 1.5mg/L [4]. The acceptable and permissible limits in Indian context are 1mg/l and 1.5mg/l respectively [5].

It is estimated that around 260 million people worldwide are drinking water with fluoride content more than 1.0 mg/l. In India alone, endemic fluorosis is thought to affect around one million people and is a major problem in 17 states, especially Rajasthan, Andhra Pradesh, Tamil Nadu, Gujarat and Uttar Pradesh [6]. As fluorosis is an irreversible condition that has no cure, prevention is the only solution for this menace. Hence, provision of water with optimal fluoride concentration is the only way to protect future generations against this disease [7].

Various technologies were developed to remove fluoride from drinking water by ion exchange, adsorption, precipitation, electro chemical defluoridation and reverse osmosis, etc. But due to high cost involved and by-products generated, such technology is not reasonable for a developing country like India. Therefore, there is a great need for environmental friendly and low cost technology for domestic usage [8].

Natural materials have been used in water treatment since ancient times. In recent years there has been a resurgence of interest to use natural materials and usage of algal biomass and plant products for defluoridation has been reported. Thus, a study was attempted to evaluate water defluoridating capability of untreated plant materials like tulsi leaves and wheatgrass and in turn compare it with that of brushite-calcite combination reported by Larsen and Pearce [9].

MATERIALS AND METHODS

This in-vitro experimental study was conducted at Bapuji Institute of Engineering Technology, Davangere, India, in April 2015 for a period of one month. The experimental procedure followed could be explained under the following steps.

Procurement of test products: Tulsi leaves (*Ocimum sanctum*) and wheatgrass (*Triticum aestivum*) were obtained from the local market. All plant specimens were identified by a botanist for their authenticity. After washing with distilled water, the plants were sun dried for a week and the leaves were grinded in a flour mill to a fine powder. Brushite ($CaHPO_4.2H_2O$; Analytical grade; Oxford Laboratory, Mumbai, India), Calcite ($CaCO_3$; Analytical grade; Nice Chemicals, Kochi, India) and Sodium fluoride (NaF; Analytical grade; Fischer scientific, Mumbai, India) were purchased from a scientific store. TISAB (Total Ionic Strength Adjustment Buffer) solution required for fluoride analysis in water samples was obtained from Bapuji Institute of Engineering Technology, Davangere, India.

Preparation of stock solution: The fluoride stock solution of 1000ppm was prepared by diluting 2.21g of sodium fluoride salt in one litre of double deionised distilled water at room temperature. The solution was diluted as required to obtain working test solution of 5ppm.

Procurement of natural fluoridated water: To check the practical feasibility of the procedure in day to day life, ground water with a natural fluoride concentration of 2.03 ppm was obtained from Bharamasamudra village near Davangere city, India.

Experimental procedure: One litre each of 5ppm water solution was taken in 30 plastic containers, 10 designated to each of the three test products. One gram of tulsi powder was added to each of 10 containers in Group I, one gram of wheatgrass powder to each of 10 containers in Group II and one gram of a combination of brushite and calcite (0.5g each) to 10 containers in Group III. All the containers were shaken vigorously for one minute to allow uniform dispersion of the test products. Five containers in each group were left undisturbed. Water in the remaining five containers was boiled separately in a domestic electric kettle for one minute and transferred back. The electric kettle was cleaned between each run with 0.1 mol/L of acetic acid followed by five rinses with deionised water. Samples were drawn from the supernatant After 30 minutes and 24 hours from all the thirty containers. Entire procedure was repeated again using 2ppm natural fluoridated water. Fluoride concentration in each sample was determined by using an ion electrode (Hach-Sension MM 374, Hach Company, USA) after addition of TISAB solution and denoted as fluoride in parts per million (mg/ml). Each sample was analysed in duplicate and average reading was taken.

STATISTICAL ANALYSIS

All the data were entered in a Microsoft excel sheet 2010 and subjected to statistical analysis using SPSS version 20. For intragroup comparisons between boiled and non-boiled samples, unpaired t-test was used and for intergroup comparisons between three test products, One-way ANOVA followed by Tukey's post Hoc analysis was done.

RESULTS

[Table/Fig-1,2] show fluoride concentration in 5ppm and 2ppm water samples following the addition of the test products followed by subsequent treatment at the end of 30 minutes and 24 hours respectively.

Addition of test products followed by boiling for one minute: As evident from [Table/Fig-1], addition of test products to artificially fluoridated water at 5ppm followed by boiling for one minute resulted in reduction of fluoride concentration to 3.28 ppm, 4.57 ppm and 0.86 ppm in Groups I, II and III respectively at the end of 30 minutes. Higher reduction of 82.8 % was seen with brushite-calcite when compared to tulsi (34.4%) and wheatgrass (8.6%) and this difference was statistically highly significant (p=0.001). [Table/Fig-2] shows that similar reduction in fluoride concentration was also observed in naturally fluoridated water of 2ppm after boiling. Here, the fluoride ion concentration fell to 1.38ppm, 1.81ppm and 0.49ppm in Groups I, II and III respectively at the end of 30 minutes. Higher reduction of 75.5% was seen with brushite-calcite when compared to tulsi (31%) and wheatgrass (9.5%) and this difference was statistically highly significant (p=0.001). Even at the end of 24 hours, brushite-calcite combination had shown superior defluoridating potential than tulsi and wheatgrass (p=0.001).

Addition of test products and without boiling: From [Table/Fig-1], it is apparent that 5ppm artificial fluoridated water when not boiled following addition of test products, a reduction in fluoride concentration to 3.78ppm, 4.68ppm and 4.90ppm was seen in Groups I, II and III respectively at the end of 30 minutes and to 3.59ppm, 4.60ppm and 4.88ppm at the end of 24 hours. Tulsi had shown higher reduction rates of about 24.4% at the end of 30 minutes and 28.2% at the end of 24 hours when compared with other test products and the difference was statistically highly significant (p=0.001). As seen in [Table/Fig-2], similar results were also observed in 2ppm fluoridated natural water samples after addition of test products at various time intervals (p=0.001).

Test Product	Fluoride concentration at the end of 30 minutes		t-test	Fluoride concentration at the end of 24 hours		t-test
	On boiling for 1 minute	Without boiling		On boiling for 1 minute	Without boiling	
Tulsi (T)	3.28 ± 0.04 (34.4%)	3.78 ± 0.03 (24.4%)	p= 0.001	3.10 ± 0.05 (38%)	3.59 ± 0.04 (28.2%)	p= 0.001
Wheat Grass (W)	4.57 ± 0.05 (8.6%)	4.68 ± 0.05 (6.4%)	p= 0.005	4.46 ± 0.04 (10.8%)	4.60 ± 0.03 (8%)	p= 0.001
Brushite-Calcite (BC)	0.86 ± 0.03 (82.8%)	4.90 ± 0.01 (2%)	p= 0.001	0.67 ± 0.01 (86.6%)	4.88 ± 0.45 (2.4%)	p= 0.001
ANOVA	p=0.001	p=0.001	-	p=0.001	p=0.001	-
Tukey's Post-Hoc Analysis	T vs W p=0.001	T vs W p=0.001		T vs W p=0.001	T vs W p=0.001	
	T vs BC p=0.001	T vs BC p=0.001		T vs BC p=0.001	T vs BC p=0.001	
	W vs BC p=0.001	W vs BC p=0.001		W vs BC p=0.001	W vs BC p=0.001	

[Table/Fig-1]: Fluoride concentration after addition of different test products in 1litre of 5ppm artificially fluoridated water.

Test Product	Fluoride concentration at the end of 30 minutes		t-test	Fluoride concentration at the end of 24 hours		t-test
	On boiling for 1 minute	Without boiling		On boiling for 1 minute	Without boiling	
Tulsi (T)	1.38 ± 0.03 (31%)	1.47 ± 0.04 (26.5%)	p= 0.003	1.16 ± 0.06 (42%)	1.44 ± 0.05 (28%)	p= 0.001
Wheat Grass (W)	1.81 ± 0.03 (9.5%)	1.96 ± 0.01 (2%)	p= 0.001	1.82 ± 0.03 (9%)	1.94 ± 0.02 (3%)	p= 0.001
Brushite-Calcite (BC)	0.49 ± 0.01 (75.5%)	1.99 ± 0.01 (0.5%)	p= 0.001	0.37 ± 0.02 (81.5%)	1.97 ± 0.01 (1.5%)	p= 0.001
ANOVA	p=0.001	p=0.001	-	p=0.001	p=0.001	-
Tukey's Post-Hoc Analysis	T vs W p=0.001	T vs W p=0.001		T vs W p=0.001	T vs W p=0.001	
	T vs BC p=0.001	T vs BC p=0.001		T vs BC p=0.001	T vs BC p=0.001	
	W vs BC p=0.001	W vs BC p=0.001		W vs BC p=0.001	W vs BC p=0.001	

[Table/Fig-2]: Fluoride concentration after addition of different test products in 1litre of 2.03ppm natural fluoridated water obtained from Bharamasamudra village.

DISCUSSION

If alternate source of water is not available, defluoridation of drinking water becomes the only option to overcome the problem of excessive fluoride in drinking water. Desirable characteristics of any defluoridation process include: cost-effectiveness, ease to handle, no alteration in taste of water, no addition of other undesirable substances and be independent of input fluoride concentration, alkalinity, pH and temperature [10].

Adsorption of fluoride ions onto the surface of an active agent is a popular method for defluoridation. Among a wide array of materials suggested for adsorption, a combination of brushite and calcite developed by Larsen and Pearce had shown superior defluoridating ability [11].

Plants have been used for water treatment since centuries. Plant materials like seeds, leaves, bark, roots and fruits are known to possess defluoridating capabilities [12]. In the present study, locally available plants like tulsi and wheatgrass were selected on the basis of local availability and edibility in addition to medicinal value.

Available literature on usage of these herbs for defluoridation is sparse. However, an attempt was made to compare selected results wherever possible maintaining the validity of comparisons.

Initially, the study was planned with artificially fluoridated distilled water at 5ppm. However, the reduction in fluoride concentration seen prompted us to replicate the study on naturally fluoridated water at around 2ppm to check the practical applicability of the study results.

In the present study, when water was boiled following addition of test products, brushite-calcite combination had shown a superior defluoridating capacity (ranging from 75.5% to 86.6%) than the plant products. The defluoridating capability of brushite-calcite combination in the present study was similar to that shown in studies by Larsen MJ et al., and Lakshminarayan L et al., [9,13]. However, no such reduction was seen in un-boiled samples. Boiling converts the two salts rapidly into apatite crystals which in turn adsorb the fluoride ions present in the water causing a pronounced decrease in the fluoride concentration. When left undisturbed, sedimentation of adsorbed fluoride in the form of fluoroapatite, hydroxyapatite and unutilized calcite occurs. Generation of such harmful chemical substances necessatizes prior separation before consumption which itself is a tedious process. Also boiling drinking water before consumption alters the taste of the water [9]. Considering the domestic application of this technique, these factors are not desirable.

Among the three products, tulsi is the only material that had shown a consistent defluoridation capacity both with boiling (31% to 42%) and without boiling (24.4% to 28.2%). This reduction could be attributed majorly to the coagulant proteins in plant leaves [12]. The reduction in fluoride level after addition of tulsi was consistent with two Indian studies reported. Mahehwari R et al. and Patni M et al., reported a reduction of fluoride concentration by 95% and 85.3% respectively after addition of tulsi leaves [14,15]. In these studies, water was pre-treated and pH adjusted with chemicals to eliminate interference with ions other than fluoride which resulted in higher defluoridation potential. But in the present study, no prior chemical adjustments were done. Though the amount of reduction observed in the present study was lower, the methodology used was simple and easy to replicate at the domestic household level. Addition of tulsi altered the taste of water. However, the taste is culturally acceptable in Indian scenario considering the importance the plant has in daily life. In addition, the medicinal properties of tulsi enrich the water making it safe and healthy for consumption [16].

Wheatgrass has shown a lower defluoridating capability on boiling among the three test materials. However, it was better than brushite-calcite combination when water was not boiled following addition. A major alteration in taste was also reported. All these factors limit its applicability as a defluoridating agent.

Alkalinity, pH, temperature and presence of other chemicals also influence the fluoride concentration of drinking water [15]. The results obtained in the study were encouraging. Though the effect was modest, tulsi can be tried for domestic defluoridation considering its ease of availability, ease to handle, acceptable taste alteration and non-generation of any harmful chemical wastes. These factors make tulsi a desirable defluoridating agent.

LIMITATION

A high level of chemical technological expertise beyond the scope of dentistry was required to check the interferences of other factors on defluoridating capacity. This was a major limitation of the study.

CONCLUSION

The present study investigated defluoridating capacity of brushite-calcite chemical combination in comparison with two herbs – tulsi and wheatgrass. Among the three materials used, brushite-calcite combination though found to be superior, suffers from harmful chemical wastes and wheatgrass had almost no defluoridating capacity. Based on the results obtained, it can be concluded that addition of tulsi leaves appears to be an economical, effective and natural method for domestic defluoridation of water on a small scale. However, as the fluoride removal efficiency may vary according to many site-specific chemical, geographical and economic conditions, further research is warranted with varying fluoride concentrations using different concentrations of tulsi and at different time intervals to evaluate its application in day to day life.

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