Environmental Health Engineering and Management Journal 2022, 9(4), 419-424
http://ehemj.com
doi 10.34172/EHEM.2022.45

Environmental Health Engineering and Management Journal

Original Article

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Defluoridation potential of indigenous thirst-quenching herbal products commonly used in Kerala, India

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Abstract

Background: Drinking fluoride-contaminated water is a severe health hazard problem. Fluorosis -both skeletal and dental- is an important clinical and public health problem in about 24 countries including India. The best method to overcome the problem of excess fluoride in drinking water is defluoridation. Adsorption methods are simple, economical, and globally pursued techniques. Thirst-quenching herbal products locally called 'Dahashamini' are plant parts that are used during boiling drinking water. Possessing a defluoridation potential is an added benefit.

Methods: Two grams of each of dried and ground *Zingiber officinale*, *Elettaria cardamomum*, *Eugenia caryophyllus*, *Coriandrum sativum*, *Acacia catechu*, *Caesalpinia sapans*, *Vetiveria zizanioides*, *Cuminum cyminum*, and *Hemidesmus indicus*, were added to 100 millilitres of fluoridated water of baseline concentrations of 5 and 10 ppm, boiled till its boiling point, and cooled. The samples were then filtered and analyzed for fluoride content using fluoride ion specific electrode method. Statistical analysis was done using one-way ANOVA, followed by Tukey's post hoc test for pair-wise comparison.

Results: *Caesalpinia sapans*, *Vetiveria zizanioides*, *Acacia catechu*, *Eugenia caryophyllus*, and *Coriandrum sativum*, had a significant ability to adsorb fluoride from fluoridated water, with an efficiency ranging from 12% to 56% (at a baseline concentration of 10 ppm) and 19% to 82% (at a baseline concentration of 5 ppm).

Conclusion: The study indicates the possibility of the use of five ingredients in developing a cost-effective and acceptable method of defluoridation based on the adsorption method.

Keywords: Fluoride, *Vetiveria*, *Acacia*, *Eugenia*, *Elettaria*

Citation: Ramanarayanan S, Kuruvilla S, Methar S, Latti P. Defluoridation potential of indigenous thirst-quenching herbal products commonly used in Kerala, India Environmental Health Engineering and Management Journal 2022; 9(4): 419-424. doi: 10.34172/EHEM.2022.45.

Article History:
Received: 11 February 2022
Accepted: 10 May 2022
ePublished: 11 December 2022

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Introduction

Water is most abundant and is an essential component of our ecosystem. Water is considered as the most common and important resource on the Earth. Fluoride is naturally found in drinking water in various places, and hence, drinking water acts as the major source of fluoride. Both surface water and ground water contain fluoride (1).

Fluoride acts as a double-edged sword. At lower concentrations, it is therapeutic and has proven inhibitory effect on dental caries, while at higher concentrations, it poses a health hazard leading to fluorosis. Contamination of drinking water with fluoride is a severe health hazard (2). Symptoms affecting the soft tissues such as muscles and ligaments are also reported (3). Fluorosis - both skeletal fluorosis and dental fluorosis - is an important clinical and public health problem in about 24 countries including India (4).

According to the World Health Organization (WHO), the maximum acceptable fluoride concentration in drinking water is 1.5 mg/L. The Government of India reported that since April 2014, fluorosis has been prevalent in 230 districts across 19 Indian states. The population at risk is officially estimated to be around 11.7 million (5,6). In India, drinking water is the major source of fluoride (7).

The best method to overcome the problem of excess fluoride in drinking water is defluoridation. Among all available methods, adsorption is a simple, economical, and globally pursued technique (2).

Kerala is one of the states of India, which has a rich biodiversity of medicinal plants (2). One of the thirst-quenching herbal product locally called as 'Dahashamini', which is a commercially available crude mixture of 9 natural ingredients (3).

A customary practice in Kerala, is to add this mixture while boiling water for consumption, which also has additional medicinal benefits. Possessing a defluoridation potential is an added benefit for the herbal thirst quenchers, and thus, can be a cost-effective, natural, and a culturally acceptable alternative method of defluoridation. There are few studies assessing the defluoridation potential of these ingredients.

Therefore, the present study was conducted to evaluate the defluoridation potential of various indigenous herbal thirst-quenching products on boiling with water and to compare the defluoridation potential of a combination of these products locally known as 'Dahashamini'.

Materials and Methods

Fluoride stock solution was prepared by dissolving 2.21 g of anhydrous sodium fluoride (99.0% NaF, Sigma Aldrich) in 1000 mL distilled water in a volumetric flask (2). The fluoride solution of concentration of 5 parts per million (ppm) was prepared by diluting 500 ml stock solution with 1000 mL distilled water; and the fluoride solution of concentration of 10 ppm was prepared by diluting 1000 mL stock solution with 1000 mL distilled water.

'Dahasamini' is a commercially available crude mixture of 9 natural ingredients (3). The ingredients used are dry ginger (*Zingiber officinale*), cardamom pods (*Elettaria cardamomum*), Clove pods (*Eugenia caryophyllus*), Coriander seeds (*Coriandrum sativum*), Acacia catechu bark (*Acacia catechu*), Sappan wood bark (*Caesalpinia sappan*), Vetiver roots (*Vetiveria zizanioides*), Cumin seeds (*Cuminum cyminum*), and Indian Sarsaparilla root (*Hemidesmus indicus*) (3). Dahashamini powder and its constituents were purchased from the local market and authenticated by a botanist. The products were washed with tap water and double distilled water, dried, and ground in motor, and finally, sieved to get uniform particle size of 800 microns (2).

Two grams of 'Dahashamini' and its individual constituents were separately added to 100 mL of fluoridated water of concentrations of 5 and 10 ppm as two different batches, boiled till boiling point (100°C) and boiling was continued for 5 minutes, and then, the solution was cooled for 15 minutes. The samples were then filtered using a Whatman filter paper No. 4. The residues were kept aside and the filtered water was analyzed for fluoride content using fluoride ion-specific electrode (2). The experiment was triplicated to ensure consistency of the findings. The readings were recorded in parts per million (ppm). Water boiled without the addition of any ingredient was used as negative control.

Statistical analysis

The fluoride levels in water before and after boiling with the constituents were tabulated and summarized as mean ± standard deviation. The difference in the percentage of fluoride levels was summarized. Defluoridation potential between different constituents and 'Dahashamini' as a whole was compared using one-way ANOVA, followed by Tukey's post hoc test for pair-wise comparison. *P* value less that 0.05 was considered statistically significant. Data were analyzed using SPSS version 19.0 for Windows.

Results

Table 1 shows mean fluoride concentration after boiling, mean percentage change in fluoride concentration, and the results of comparative evaluation of mean fluoride concentration after boiling with the individual ingredients at a baseline concentration of 10 ppm. Five out of the nine constituents demonstrated a significant reduction in fluoride concentration after boiling with water. Inferential analysis revealed a statistically significant reduction in fluoride concentration from a baseline concentration of 10 ppm after boiling with the following constituents, *Caesalpinia sappan* (Sappan wood), *Acacia catechu* (Catechu), *Vetiveria zizanioides* (vetiver), Dahashamini mix, *Eugenia caryophyllus* (clove), and *Coriandrum sativum* (coriander seeds). A significant increase in fluoride concentration was observed with *Elettaria cardamomum* (cardamom), compared to all other ingredients.

Table 2 shows mean fluoride concentration after boiling, mean percentage change in fluoride concentration, and comparison of mean fluoride concentration after boiling with the individual ingredients at a baseline concentration of 5 ppm. Five out of the nine constituents demonstrated a significant reduction in fluoride concentration after boiling with water. Inferential analysis revealed a statistically significant reduction in fluoride concentration from a baseline concentration of 5 ppm after boiling with the following constituents, *Caesalpinia sapans* (Sappan wood), Dahashamini mix, *Acacia catechu* (Catechu), *Vetiveria zizanioides* (vetiver), *Eugenia caryophyllus* (clove), and *Coriandrum sativum* (coriander seeds). A significant increase was observed with *Elettaria cardamomum* (cardamom) and *Hemidesmus indicus* (Indian sarsaparilla) compared to all other ingredients.

Discussion

Water boiled with Dahashamini or few of its individual constituents has been consumed in Kerala since many years ago. These ingredients are readily available in the local market and have been extensively used in kitchens and traditional medicinal preparations (3). These ingredients have proven medicinal benefits, as cited in various ayurvedic texts and other scientific literature (7-12). Endemic fluorosis has been reported in 2 districts of Kerala, namely, Alappuzha and Palakkad, with a fluoride concentration ranging from 0.2 to 2.5 ppm (13).

Plant materials are reported to accumulate fluoride, and hence, considered as good defluoridation agents. Except studies on few individual constituents (2,14,15), namely, *Vetiveria zizanioides*, *Zingiber officinale*, and

Table 1. Mean fluoride concentration after boiling, mean percentage change in fluoride concentration and comparison of mean fluoride concentration after boiling with the individual ingredients at a baseline concentration of 10 ppm

Scientific name	Part Used	Fluoride after boiling	Mean % Change	ANOVA	Tukey's post hoc
1. *Zingiber officinale*	Root	10.10±0.26	1.00%	F=251.127 *P*≤0.001*	6 with 7 (*P*<0.001)* 7 with 5 (*P*=0.058) 5 with 3 (*P*<0.001)* 3 with 10 (P=0.705) 10 with 4 (*P*<0.001)* 4 with 8 (*P*<0.001)* 8 with 1 (*P*=0.992) 1 with 9 (*P*=0.951) 9 with 2 (*P*<0.001)* 11 with 1 (*P*=0.990) 11 with 2 (*P*<0.001)* 11 with 3 (*P*<0.001)* 11 with 4 (*P*<0.001)* 11 with 5 (*P*<0.001)* 11 with 6 (P<0.001)* 11 with 7 (*P*<0.001)* 11 with 8 (*P*=0.998) 11 with 9 (*P*=0.907) 11 with 10 (*P*=0.001)*
2. *Elettaria cardamomum*	Pod	11.60±0.26	16.00%		
3. *Eugenia caryophyllus*	Pod	7.76±0.06	-22.33%		
4. *Coriandrum sativum*	Seed	8.76±0.30	-12.33%		
5. *Acacia catechu*	Bark	6.40±0.26	-36.00%		
6. *Caesalpinia sappan*	Bark	4.33±0.32	-56.66%		
7. *Vetiveria zizanioides*	Root	5.66±0.35	-43.33%		
8. *Cuminum cyminum*	Seed	9.90±0.30	-1.00%		
9. *Hemidesmus indicus*	Root	10.36±0.15	3.66%		
10. *Dahashamini mixture*	-	7.66±0.35	-23.4%		
11. Boiled water (control)	-	10.06±0.15	0.66%		

*Significant.

Table 2. Mean fluoride concentration after boiling, mean percentage change in fluoride concentration and comparison of mean fluoride concentration after boiling with the individual ingredients at a baseline concentration of 5 ppm

Scientific Name	Part Used	Fluoride after Boiling	Mean % Change	ANOVA	Tukey's Post hoc
1. *Zingiber officinale*	Root	4.83±0.25	-3.33%	F=189.588 *P*≤0.001*	6 with 5 (*P*<0.039)* 5 with 10 (*P*<0.001)* 10 with 7 (*P*<0.001)* 7 with 3 (*P*=0.084) 3 with 4 (*P*=0.971) 4 with 8 (*P*=0.766) 8 with 1 (*P*=0.018)* 1 with 9 (*P*<0.001)* 9 with 2 (*P*=0.324) 11 with 1 (*P*=0.861) 11 with 2 (*P*<0.001)* 11 with 3 (*P*<0.001)* 11 with 4 (*P*<0.001)* 11 with 5 (*P*<0.001)* 11 with 6 (*P*<0.001)* 11 with 7 (*P*<0.001)* 11 with 8 (*P*=0.998) 11 with 9 (*P*=0.030)* 11 with 10 (*P*=0.001)*
2. *Elettaria cardamomum*	Pod	5.50±0.26	10.00%		
3. *Eugenia caryophyllus*	Pod	4.03±0.20	-19.33%		
4. *Coriandrum sativum*	Seed	3.83±0.35	-23.33%		
5. *Acacia catechu*	Bark	1.53±0.15	-69.33%		
6. *Caesalpinia sappan*	Bark	0.90±0.10	-82.00%		
7. *Vetiveria zizanioides*	Root	2.50±0.26	-50.00%		
8. *Cuminum cyminum*	Seed	4.13±0.06	-17.33%		
9. *Hemidesmus indicus*	Root	5.93±0.06	18.67%		
10. *Dahashamini mixture*	-	2.31±0.26	-53.8%		
11. Boiled water (control)	-	5.10±0.20	02.00%		

*Significant.

Eugenia caryophyllus, studies assessing the defluoridation potential of Dahashamini are not available in the electronic literature search.

Regarding all medicinal properties and benefits, if defluoridation potential exists, Dahashamini and its constituents can be used as a defluoridation agent, without the need for a behavioural change. Dahashamini can serve as a cost-effective, affordable, readily available, healthy, indigenous, culturally acceptable, and natural method of defluoridation. Although mostly used in Kerala, it can be used in other fluoride belts.

Among the conventional methods of fluoride removal that include precipitation, ion-exchange, reverse osmosis and adsorption, the former two are relatively expensive, therefore, adsorption is considered as a viable method (14). Adsorption technique has been quite popular due to its simplicity as well as availability of a wide range of adsorbents. Adsorption involves the passage of contaminated water through an adsorbent bed, where fluoride is removed by physical, ion-exchange or surface chemical reaction with adsorbent (15).

This study showed that five out of the nine constituents demonstrated a significant reduction in fluoride concentration after boiling with water.

The maximum reduction at a baseline concentration of 10 ppm was related to the use of *Caesalpinia sappan* (Sappan wood). There was a 56% reduction in fluoride levels (from 10 to 4.3 ppm). At a baseline concentration of 5 ppm, the reduction was 82%. The defluoridation potential of Sappan wood has already been demonstrated in another study conducted in Nagercoil by Anuja and Indrani (15). The study reported a reduction of 22-25% at a baseline concentration of 2 ppm, which is consistent with the results of the present study.

In this study, *Vetiveria zizanioides* (vetiver) demonstrated a fluoride removal efficiency of about

43.3% for 10 ppm solution and 50% for 5 ppm solution. Vetiver has been previously studied in an extensive manner assessing the effect of variables such as pH, agitation time, contact time, dose, etc. A previous study conducted by Harikumar et al (2) demonstrated up to 90% reduction following a surface treatment and vetiver activation. It was observed that the efficiency increases with a decreased particle size, increased agitation time, and decreased baseline concentration (80% at 2 ppm). Yet another study conducted by Anuja and Indrani (15) demonstrated an efficiency ranging between 24.29% at 3 g dose to 37.16% at 30°C temperature. Thus, the sorption process of fluoride ion on activated vetiver root was influenced by many experimental conditions.

Acacia catechu (Catechu) showed a mean percentage reduction of 36% when boiled with a concentration of 10 ppm and a higher percentage reduction of 69% when boiled with a concentration of 5 ppm. Except for a study conducted by Harikumar et al in Kozhikode, Kerala (2), which demonstrated 47% efficiency, at concentration of 2 ppm, there have been no other studies for comparison. The study analyzed the efficiency of catechu only in phase 1, and hence, was not subjected to further analysis like vetiver to assess the influence of the physical factors. This might probably be due to the fact that *Acacia catechu* might be more efficient at low concentrations of fluoride.

In this study, *Eugenia caryophyllus* (clove) demonstrated a percentage reduction of 22.3% at 10 ppm and 19.3% at 5 ppm concentration of fluoride. A study by Harikumar et al in Kozhikode, demonstrated a 70% reduction. The probable difference could be due to the baseline dose, which was 2 ppm against 10 ppm and 5 ppm, used in the study. This indicates the probable effect of clove at a lower fluoride concentration.

Coriandrum sativum (coriander seeds) demonstrated an efficiency of 12.3% at higher a concentration (10 ppm) and 23.3% at a lower concentration (5 ppm). Coriander seeds, as mentioned earlier, have been used to study the adsorption potential of other elements such as copper, zinc, and lead from water (16). This indicates the probable adsorption potential of coriander on fluorides. However, coriander leaves have been reported to have a fluoride concentration up to 5 ppm (17).

Zingiber officinale (dry ginger), hardly demonstrated any change (1%) in fluoride concentration after boiling at 10 ppm and a marginal 3.3% decrease at 5 ppm concentration, which is inconsistent with the results of other previous studies. The defluoridation potential of dry ginger, as assessed in the study conducted by Anuja and Indrani (15), demonstrated a significant reduction ranging from 9% to 49.2% after regulating various physical parameters such as pH, contact time, adsorbent dose, etc. The discrepancies in the results might be due to the low pH (49.2% at pH of 6) and increased contact time up to 60 minutes or heating at temperature of 30°C (25.57%) used in the previous study. Thus, it can be concluded that dry ginger probably requires a higher contact time, a lower pH and lower temperature to have an increased efficiency.

Cuminum cyminum (cumin), at a baseline concentration of 10 ppm, showed no effect on fluoride concentration after boiling (1%). However, at 5 ppm concentration, there was a significant increase in defluoridation potential (17.3%), indicating that cumin is effective at lower fluoride concentrations. In contrast to the observations of the present study, a study conducted by Santhi et al (18) demonstrated a reduction of 33% and 34% in fluoride concentration from baseline values of 6 and 8 ppm, respectively, after boiling with 2 g of *Cuminum cyminum.*

Hemidesmus indicus (Indian sarsaparilla) demonstrated a marginal rise (3.5%) in fluoride levels at 10 ppm baseline concentration and a highly significant fluoride increase of about 18% at 5 ppm baseline concentration. This suggests that the Indian sarsaparilla results in increasing fluoride concentration of water, more with lower concentrations.

Elettaria cardamomum (cardamom) demonstrated a mean increase of 16% and 10% at 10 and 5 ppm concentrations, respectively. However, the study reported by Santhi et al demonstrated a reduction of 65% and 33% in fluoride concentration from baseline values of 6 and 8 ppm, respectively, after boiling with 2 g of *Elettaria cardamomum* (18).

Dahashamini mix demonstrated a reduction of 23.4% and 53.8% at baseline concentrations of 10 and 5 ppm, respectively. However, locally available Dahashamini mix is a crude mixture of the available ingredients and uniformity of its composition need not be maintained in all packets. Therefore, an inference regarding its efficiency is only concerned with the specific packet used in this study.

This study was conducted to assess this defluoridation potential in a set of plant products, commonly available in various combinations of 9 ingredients referred to as Dahashamini. As about 5 of them proving to have a significant adsorption potential, they can be studied further in detail to have an in-depth understanding of the efficiency. Most of these ingredients are readily available and economical, and thus, can be used in all endemic fluoride areas. As these products are routinely consumed, they need not to be further assessed for safety.

Many of the studies have assessed the defluoridation potential at varying temperature ranges from 10 to 60°C. The temperature used in this study is the boiling temperature of water to simulate the customary practice of boiling the water with one or more of the ingredients before consumption. Fluoride analysis after boiling, allowing a contact time of 15 minutes, a duration after which the temperature is fit for consumption (lukewarm).

In the absence of several studies for comparison, this study can be considered only as a preliminary work in understanding the adsorption potential. Further research considering factors like particle size, temperature, contact time, etc. can help better understanding and analyzing the defluoridation properties of these biosorbents. Such detailed investigations could help in developi ng the accurate effective dose without compromising the water quality and standards.

Conclusion

It was observed that five of the nine ingredients namely *Caesalpinia sappan* (sappan wood), *Vetiveria zizanioides* (vetiver), *Acacia catechu* (catechu), *Eugenia caryophyllus* (clove), and *Coriandrum sativum* (coriander), had a significant ability to adsorb fluoride from fluoridated water. The study shows the possibility of the use of five ingredients in developing a cost-effective and acceptable method of defluoridation based on the adsorption method. The results indicate the potential and need for further and better comprehensive investigations. The study paves the way for further detailed research enabling the development of natural, cost-effective, and acceptable alternate methods of defluoridation.

Acknowledgments

This study received support from the Indian Council of Medical Research, as a part of Short-term Studentship Program (Institutional Ethics Committee ID: IEC/IGIDS/15/2019). The authors would like to acknowledge Dr. Venkitachalam for critically revising the manuscript.

Ethical issues

The study was approved by the Institutional Ethics Committee of Indira Gandhi Institute of Dental Sciences, Kothamangalam (Ethical code: IEC/IGIDS/15/2019).

Competing interests

None.

Authors' contributions

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