International Journal of Community Medicine and Public Health
Havale R et al. Int J Community Med Public Health. 2024 Nov;11(11):4354-4360
http://www.ijcmph.com
pISSN 2394-6032 | eISSN 2394-6040

DOI: https://dx.doi.org/10.18203/2394-6040.ijcmph20243298

Original Research Article

Natural solution for fluoride removal: a comparative analysis of three adsorbents

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Received: 24 August 2024
Revised: 10 September 2024
Accepted: 17 October 2024

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ABSTRACT

Background: Defluoridation of groundwater is a major global health concern, and research has been conducted worldwide to determine the optimal level of fluoride in drinking water. Adsorption is a common method for defluoridation. The present research planned to use natural adsorbents for defluoridation, they are effective and economical. This study aimed to evaluate and compare the efficacy of powders of Pineapple peel, Banana peel and Ginger as adsorbents to defluoridate the fluoridated drinking water in chosen demographic area.
Methods: Water samples from a highly fluoridated area were collected and their initial fluoride levels were determined. Peels from pineapple, banana and ginger were collected, washed, dried in the shade, powdered and sieved. The powdered peels were then added to the fluoridated water samples, were shaken for 24 hour on a rotary shaker and subsequently filtered. The filtered samples were analyzed for fluoride content using an Ion selective electrode.
Results: The study found that powders of pineapple peel, banana peel and ginger were effectively reduced fluoride levels in drinking water. Pineapple peel and ginger powder showed the highest efficiency, removing 17.65% of fluoride ions regardless of dosage. Banana peel showed a dose dependent increase in fluoride removal with higher dosages.
Conclusions: Pineapple peel, ginger powder, and banana peel powder effectively remove excess fluoride from drinking water, offering a cost-effective and sustainable solution to mitigate harmful health effects.

Keywords: Adsorption, Banana peel, Defluoridation, Ginger, Pineapple peel

INTRODUCTION

High fluoride content in potable drinking water has become a serious problem across the world.[1] The World Health Organization (WHO) (1984) and Bureau of Indian standards (1991) set the acceptable limit of drinking water at 1.0 ppm and the maximum permissible limit at 1.5 ppm.[2] India Fluorosis Research and Rural Development Foundation (2001) has reported that 15 states of India has dental fluorosis, including Karnataka, due to high fluoride levels in drinking water.[3] Excessive fluoride consumption can lead to fluorosis, causing a range of harmful effects on bones, teeth, and brain function.[2] In response to this nationwide issue , the government launched the National Programme for Prevention and Control of Fluorosis (NPPCF) in the 11th Five-Year Plan . Karnataka is one of the states severely affected by fluoride contamination, with several districts.[4]

According to NPPCF data, Raichur is one among the other districts having significant levels of fluoride in their groundwater. Raichur, located in northeastern part of Karnataka relies heavily on groundwater as its primary source of drinking water.[5] The prevalence of dental fluorosis among the school children aged between 5 to 10 years was reported to be 32.6% in six villages of Raichur district.[5] Wadloor village in Raichur district has high levels of fluoride in its groundwater, which is used for drinking. Although the water is treated using the reverse osmosis process, many families do not fully utilize this facility due to lack of awareness, negligence, and the distance between their homes and the treatment source.

The WHO states that the amount of fluoride absorbed by plants depends primarily on the plant species, soil characteristics, and the existing levels and forms of fluoride in the soil, which influence the uptake and accumulation of fluoride by plants.[6] The research was carried out to estimate fluoride levels in staple crops (rice, redgram, jowar) and soils were compared in fluoridated and non-fluoridated regions. Fluoridated areas had higher levels (0.79-8.8 ppm), while non-fluoridated areas had lower levels (0.07-0.81 ppm). The correlation between crop and soil fluoride levels was strong, indicating that crops in fluoridated areas absorb more fluoride.[7] Another research was carried out in Raichur district showed that the ability of jowar crops to retain fluoride was linked to the severity of dental fluorosis in the local population.[8]

Fluoride-containing water must be treated using appropriate, practical, and affordable techniques. There are various techniques used for water defluoridation which include membrane filtration, precipitation, nanofiltration, ion exchange, electrocoagulation flotation, reverse osmosis, and adsorption.[9-11] Among these, adsorption method has proven to be successful at low concentrations and less expensive[9,10] having least disadvantages when compared with other above mentioned techniques. Use of natural adsorbents have led a novel way in water defluoridation .It includes activated alumina-coated silica gel, activated sawdust, activated coconut shell carbon, coffee husk, bone charcoal, multani mitti, red mud, calcined clay, concrete, pineapple peel powder, chalk powder, orange peel powder, and rice husk.[12] Among these, the commonly, easily and economically available natural adsorbents are pineapple peel, banana peel and ginger, hence research was aimed to investigate and compare the efficacy of pineapple peel, banana peel and ginger powder as natural adsorbents in removing fluoride from high fluoridated drinking water.

METHODS

Before initiating the research, the ethical clearance was obtained from the Institutional Ethical Committee of AME'S Dental College and Hospital, Raichur. (Ref.No.AME/DC/67/22-23). A cross sectional research design was used to study the defluoridating capacity of the drinking water in Wadloor, Raichur district. The study period was 3 months (September 2023 to November 2023). The study was carried out in the two steps:

Estimation of Fluoride level and pH: Ground water was collected from the study village. It was subjected to ion selective electrode for fluoride testing, which demonstrated 8.5 ppm. The pH of water sample was calculated using pH meter which displayed the pH of 9.5. To substantiate this, dental fluorosis was assessed in the children of the study village.

Assessment of dental fluorosis: Of the two government schools in the village, one school was randomly selected and permission from head master were obtained for conducting the study. A convenient sampling method was employed in the present study; the study was conducted in 320 children aged between 5-12 years to assess the status of dental fluorosis. The selection of children's were carried out on the basis of the inclusion and exclusion criteria.

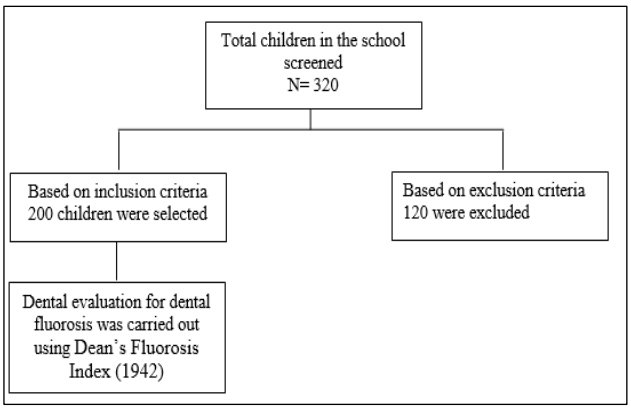

Figure 1: Study design for evaluation of dental fluorosis.

Inclusion criteria

The children who were continuously present in the village since the birth, consumed the ground water for drinking were included in the study.

Exclusion criteria

The children who consumed filtered treated water for drinking were excluded in the study.

Clinical examination

The dental examination was performed in the 200 children in the school settings under natural day lighting. The Dean's fluorosis index [1942] was used to assess the dental fluorosis in the children of aged 5-12 as this is the period of mixed dentition where the permanent tooth development undergoes several changes due to increase in the fluoride levels than normal where the permanent teeth were examined using mouth mirror and probe to score fluorosis.

Table 1: Dean's fluorosis index (1942).

Score	Criteria
Normal (0)	The enamel represents the usual translucent semivitriform type of structure. The surface is smooth, glossy and usually of a pale, creamy white color.
Questionable (0.5)	The enamel discloses slight aberrations from the translucency of normal enamel, ranging from a few white flecks to occasional white spots. This classification is used in those instances where a definite diagnosis of the mildest form of fluorosis is not warranted and a classification of "normal" not justified.
Very mild (1)	Small, opaque, paper white areas scattered irregularly over the tooth, but not involving as much as approximately 25% of the tooth surface. Frequently included in this classification are teeth showing no more than about 1-2 mm of white opacity at the tip of the summit of the cusps of bicuspids or second molars.
Mild (2)	The white opaque areas in the enamel of teeth are more extensive, but do not involve as much as 50% of tooth.
Moderate (3)	All enamel surfaces of the teeth are affected and surfaces subject to attrition show wear. Brown stain is frequently a disfiguring feature.
Severe (4)	All enamel surfaces of the tooth are affected and hypoplasia is so marked that the general form of the tooth may be affected. The major diagnostic sign of this classification is discrete or confluent pitting. Brown stains are widespread and teeth often present a corroded-like appearance.

Source: Dean 1942, American Association for the Advancement of Science.

Preparation of natural adsorbents

The three natural adsorbents namely pineapple peel, banana peel from juice center and ginger from vegetable vendor were collected, washed thoroughly under water, chopped, shade dried for 3-4 days followed by which they were grinded into a fine powder and sieved to the size of 100 microns.

Procedure

100 ml of each water sample was added to each 4 conical flasks, for this 0.5, 1, 1.5 and 2 grams each of natural adsorbents were added respectively. These flasks were placed on the rotary shaker for 24hours and after which the samples were filtered and underwent fluoride testing using ion selective electrode.[1] pH of these water samples were also calculated using pH meter.

Statistical description

The present study consists of providing intervention by three natural adsorbents for defluoridation of water. The results comprise of a single value of percentage of fluoride removal for each adsorbent, thus in the present study no statistical analysis was able to perform to compare the percentage of fluoride removal between three natural adsorbents in defluoridation.

RESULTS

The demographic profile of the 200 subjects revealed that the mean age of the participants was 8.5 years. Of that, 111 were boys, and 89 were girls. 66% of the children had fluorosis, where 11% of the children had severe form of fluorosis, 12.5-14% had mild to moderate form of fluorosis depicted in Table 2.

Table 2: Showing the status of Dean's Fluorosis Index in the 200 children of the study village.

Status	Percentage
Normal (0)	34
Questionable (0.5)	21
Very mild (1)	7.5
Mild (2)	12.5
Moderate (3)	14
Severe (4)	11

Effect of natural adsorbents on fluoride removal

The three adsorbents studied showed a relative reduction in fluoride levels in fluoridated drinking water samples. Pineapple peel and ginger powder consistently demonstrated the highest defluoridation capacity, removing 17.65% of fluoride regardless of the adsorbent dosage [0.5-2 grams]. In contrast, banana peel showed a dose-dependent increase in defluoridation, with increasing adsorbent dosages [0.5, 1, 1.5, 2 grams] resulting in corresponding increases in fluoride removal [5.88%, 9.41%, 11.70%, and 14.11% respectively] (Table 3, Figure 5).

pH of the water samples

The pH levels of the water samples (Table 4) were analyzed to determine the acceptability of the fluoridated water after undergoing the adsorption procedure for defluoridation.

Table 3: Describes the defluoridating efficacy of pineapple peel, ginger and banana peel water samples.

Adsorbent dosage (grams)	Initial fluoride conc. (ppm)	Defluoridation of water					
		Pineapple peel		Ginger		Banana peel	
		Final conc. (ppm)	% of fluoride removal	Final conc. (ppm)	% of fluoride removal	Final conc. (ppm)	% of fluoride removal
0.5	8.5	7.0	17.65	7.0	17.65	8.0	5.88
1.0	8.5	7.0	17.65	7.0	17.65	7.7	9.41
1.5	8.5	7.0	17.65	7.0	17.65	7.5	11.70
2.0	8.5	7.0	17.65	7.0	17.65	7.3	14.11

Ppm =Parts per million, conc =Concentration; % =Percentage.

Table 4: Effect of natural adsorbents on pH of the water.

pH of the intial water sample	Adsorbent dosage (grams)	pH of the adsorbent water samples					
		Pineapple peel		Banana peel		Ginger	
		pH	Effect	pH	Effect	pH	Effect
9.5	0.5	7.46	As adsorbent dosage was increased the pH decreased	5.67	As adsorbent dosage was increased the pH increased	8.62	As adsorbent dosage was increased the pH increased
9.5	1.0	6.40		5.72		8.82	
9.5	1.5	5.88		5.76		9.00	
9.5	2.0	5.76		5.95		9.14	

Figure 1: (a) Collection of ground water sample from Wadloor village of Raichur, (b) Ion selective electrode used for the estimation of fluoride level and (c) Estimation of pH of water sample.

Figure 2 (a and b): Evaluation of dental fluorosis incidence by Dean's Fluorosis Index by oral screening of 200 children for evaluating the incidence of dental fluorosis.

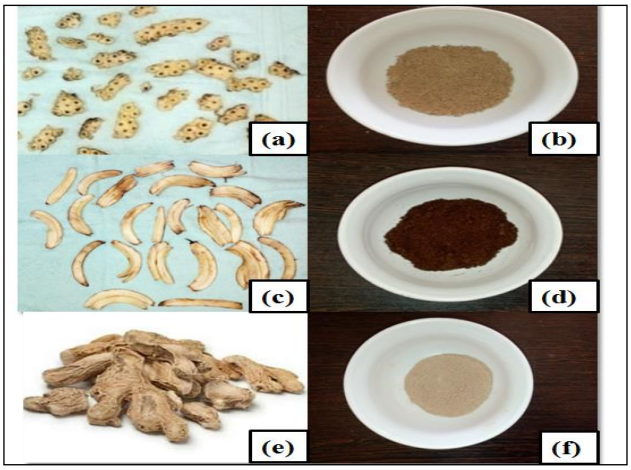

Figure 3: Preparation of natural adsorbents from Pineapple peel to powder (a, b), Banana peel to powder (c, d), and Ginger to powder (e, f).

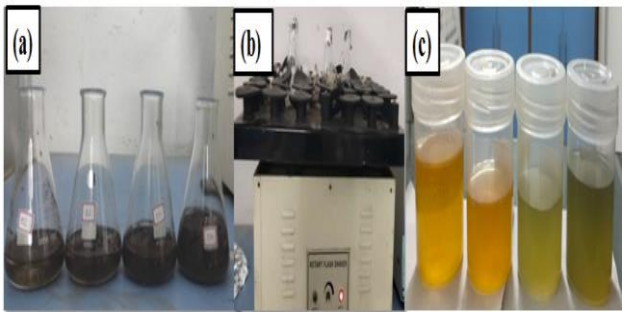

Figure 4: 100 ml of water samples to which natural adsorbents of 0.5, 1, 1.5 ,2 grms each were added and placed on rotary shaker for 24hr.

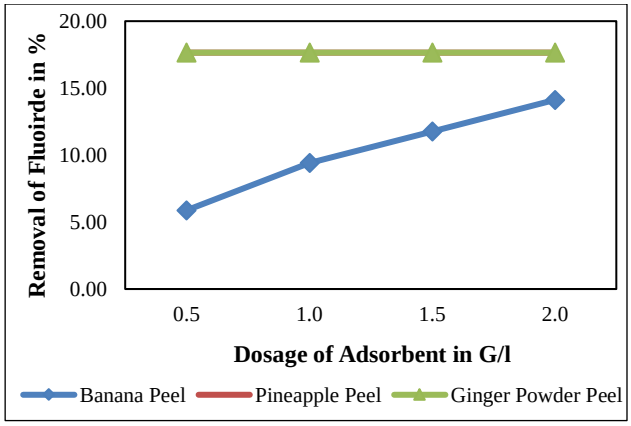

Figure 5: Variation of final fluoride concentration (showing values of pineapple peel and ginger samples coinciding with each other).

DISCUSSION

Human survival relies on three essential elements: food, water, and air. Groundwater, found in aquifers beneath earth's surface, is a crucial source of potable water.[2] Fluoride, an ionic form of fluorine, is found in large quantities in nature and can be harmful to human health due to its high concentrations in groundwater, affecting teeth, bones and neurological tissues.[10,2]

A recent investigation by Prasad et al, shed light on a significant oral health issue in Raichur district, Karnataka, where 46% of children aged 6-12 in 17 villages of Manvi and Devadurga talukas are affected by dental fluorosis. The study's findings indicate a sharp increase in the likelihood of dental fluorosis with age, underscoring the need for effective prevention and treatment strategies to address this growing concern.[5] The present study also found that one of the highly fluoridated village in Raichur district with 8.5ppm had 66% incidence of dental fluorosis (Table 2). NPPCF laboratory data of the fluoride level in water in Wadloor village Raichur district over the years is depicted in Table 5.

Dental fluorosis is a tooth enamel defect caused by excessive fluoride exposure during amelogenesis.[13] Skeletal fluorosis thickens bones and causes ligament calcification.[9] Acute fluorosis symptoms include nausea, vomiting, diarrhea, and abdominal pain.[14] Long-term fluoride consumption can reduce milk production, cytotoxic, and impact fetal brain development in turn hampers the quality of the life.[14,15,9]

Table 5: Report of drinking water fluoride level of Wadloor village.

Taluka	Village	Habitation	Source of water	Place of source	Result 2015	Result 2018	Result 2021	Result 2023
Raichur	Wadloor	Wadloor	Hand pump	Near Kannada school	7.2	7	7.2	6.4
Raichur	Wadloor	Wadloor	Borewell	Near Govt Kannada school	7	7.1	7	6.6
Raichur	Wadloor	Wadloor	Borewell	Basavana Temple	6.2	8	7.8	8.6

To combat these health hazards a wide range of methods and substances were employed to defluoridate potable water. Defluoridation is the process of removing excess, naturally occurring fluorides from drinking water to reduce the prevalence and severity of dental fluorosis.[16] The various methods used to remove fluoride include precipitation, ion-exchange, membrane process and adsorption technique.

The Nalgonda technique, developed by NEERI in 1961, is a precipitation process using lime, bleaching powder, and aluminum salts.[17] The Nalgonda technique relies on the flocculation principle.[18] It involves quick mixing, flocculation, sedimentation, filtration, and disinfection.[19,20] However, daily operations require trained operators and excess aluminum contamination may occur.[20] The ion exchange technique effectively removes fluoride maintaining water taste and color but is expensive, has high chloride content and low pH in treated water.[21] The membrane filtration technique is a non-chemical, pH-neutral method with minimal ion-obstruction, but not suitable for highly salinized water, and requires higher costs and skilled labor.[22] The limitations of the aforementioned technique have led to a search for a natural, chemical-free alternative method for defluoridation, resulting in the opting of adsorption technique as a viable method for removing fluoride from water.

Adosrption is a process where ions, atoms, or molecules adhere to a solid material's surface.[10,22] The greater the surface area per unit mass, the higher its adsorption capacity at specific pressure and temperature.[10] Adsorption of fluoride in solid particles involves three fundamental steps: molecular diffusion, where F- diffuses through the boundary layer, adsorption on the particle surface, and intra-particle diffusion where F- exchanges with the substance in the adsorbent particle or transfers to the inner surface.[27] It is widely used for small communities and household applications as it is noninvasive, most effective, simple, efficient, and can be recycled.[10,22-26]

Different natural adsorbents had been used for fluoride removal from drinking water at small scale. The present study used pineapple peel, banana peel and ginger powder as natural adsorbents and results shown that relative decrease in fluoride content in all the 3 natural adsorbents water sample, with highest being pineapple peel and ginger powder with removal of 17.65% of fluoride irrespective of the adsorbent dosage.

The present study demonstrated that irrespective of adsorbent dosage used the percentage of fluoride removal using Pineapple peel as adsorbent was constant of 17.65% (Table 3). Gandhi et al reported that *Ananas comosus* (pineapple) peel powder and *Citrus sinensis* (orange) peel powder showed remarkable fluoride removal efficiency, exceeding 90% at specific pH levels (pH 4 for pineapple and pH 6 for orange) after a 60-minute exposure period.[29] This study was in accordance with the present study. Rani et al investigated the defluoridation potential of pineapple in drinking water, achieving satisfactory results of 91% removal of fluoride under various parameters.[12] The present research's result was contradictory to this study as it focused on the effect of varying adsorbent dosage, demonstrating the percentage of fluoride removal for pineapple peel powder was found to be 17.65% (Table 3).

Banana peels, a widely available and underutilized resource, have been found to possess significant defluoridation potential. Rich in fiber (20-30%) and protein (6-9%), ripe banana peels contain more starch and less free sugar than their green counterparts. The current study's findings of fluoride removal efficiency was [range of 5.88-14.11%]demonstrated in (Table 3) were in accordance with those of the study done by Saranya et al, who observed increased fluoride removal efficiency with higher adsorbent doses using powdered banana peel (range of 9%-40%).[28] Research by Mondal et al demonstrated that banana peel powder can remove up to 86.5% of fluoride content from highly fluoridated water.[9] While the current study found a lower defluoridation capacity of 14.11% using banana peel powder, this discrepancy can be attributed to differences in adsorbent preparation methods.

Ginger, a versatile member of the *Zingiberaceae* family, has been utilized for centuries not only as a culinary ingredient but also as a natural remedy for various ailments, including flu, stomachaches, and headaches. Recent research has also explored its potential in reducing fluoride levels in water.[1]The present study demonstrated that irrespective of adsorbent dosage used the percentage of fluoride removal with Ginger adsorbent was constant of 17.65% (Table 3). Study done by Prasad et al have consistently demonstrated ginger as a defluoridating agent reduced the fluoride content to 72%, outperforming other adsorbents like turmeric, spinach, and orange peel.[1] Further investigation by Anuja et al revealed that dry ginger powder can achieve significant fluoride reduction, ranging from 9% to 49.2%, by optimizing physical parameters like pH, contact time, and adsorbent dose.[10] The present study's investigation is in accordance with these above studies, as the present study results show a notable 17.65% reduction in fluoride levels using ginger adsorbent (Table 3).

The color of the water after defluoridation for pineapple peel was brownish colour, for banana peel was light brown colour and for ginger was dark brown colour. Notably, researchers have suggested the use of activated charcoal, rice husk, rice bran, and their combinations to effectively remove color in defluoridation of water.[30]

This study has few limitations. The study only evaluated the efficacy of the adsorbents at a single initial fluoride level and did not consider varying concentrations. The study did not investigate the effects of other water quality parameters on the adsorption process. The study only evaluated the adsorbent's efficiency over a 24-hour period, which may not be representative of long-term performance. The study did not compare the results with standard defluoridation methods.

CONCLUSION

This research aimed to develop cost-effective adsorption methods for defluoridating water in communities with high fluoride levels. The use of natural materials, such as ginger, pineapple peel, and banana peel, offers an economical and eco-friendly solution for small-scale water treatment. The results showed that ginger and pineapple peel powder were the most effective adsorbents among the natural materials tested. Hence, adsorption technique using naturally available materials can be an alternative for water defluoridation which can be employed on daily basis at home in the villages affected by high fluoride water levels.

Funding: No funding sources
Conflict of interest: None declared
Ethical approval: The study was approved by the Institutional Ethics Committee of *AME'S Dental College and Hospital, Raichur. (Ref.No.AME/DC/67/22-23)*

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Cite this article as: Havale R, Shrutha SP, Revati K, Raj S, Quazi N, Kumar AY, et al. Natural solution for fluoride removal: a comparative analysis of three adsorbents. Int J Community Med Public Health 2024;11:4354-60.