Original Article

Comparative Evaluation for Defluoridation of Water, Using Three Low-Cost Household Materials: *Tamarindus indica*, *Moringa oleifera*, and *Tinospora cordifolia* – An *In Vitro* Study

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Abstract

Background: Fluoride is like a double edge sword, when ingested in lower amount benefits the health while ingested in larger amounts cause health hazards, defluoridation is the technique used to remove fluoride from water. Household methods of defluoridation using adsorbent materials are cost effective than other methods used to remove fluoride from water. The aim of the study is to determine the efficacy of low-cost household materials *Tamarindus indica*, *Moringa oleifera,* and *Tinospora cordifolia* for defluoridation of water. **Material and Methods:** An *in vitro* study was done using materials such as *T. indica, M. oleifera, and T. cordifolia.* The materials were obtained in powder form of particle size of 75 micron. Standard solution was prepared using sodium silicofluoride ($NaSiF_6$), which was weighed and dissolved in distilled water and diluted to 10 ml. The solution was then diluted to make ten parts per million standard solutions. The standard solution was filtered through the two tire filtration process and analytical determination of fluoride was done after treatment of water with the 2% (W/V) adsorbents powder filtration. **Results:** The results of the study showed that *T. indica* was found to have defluoridation capacity of 80% at neutral pH as compared to other household materials such as *M. oliefera* and *T. cordifolia* 70% and 67%, respectively, reduction in fluoride concentration at acidic pH. **Conclusion:** The present study concluded that among the aforementioned plants, *T. indica* powder was found to be the best domestic material for defluoridation property. It can be adopted for the defluoridation method in villages as it is cheaper and easily available biosorbent in Indian households.

Keywords: Defluoridation, Biosorbents, Drumstick, Tamarind, Removal of fluoride.

Introduction

Fluoride is an essential microelement for human health.[1] Fluorine does not occur free in nature and was extremely difficult for scientists to isolate. In its pure form, it is a poisonous, pale, and yellow-green gas. Like other halogens, molecular fluorine is highly dangerous; it causes severe chemical burns on contact with skin.[2] Fluoride intake by human being is generally by two ways: (1) By water and non-dairy Beverages: Fluoride intake of adults is obtained from water and other beverages. (2) By dietary sources: such as meat, fish, jawar, banana, potatoes, taro, yams, and cassava. Turmeric is good source of fluoride.[3] According to the World Health Organization (WHO 1984), the maximum acceptable fluoride concentration in drinking water is 1.5 mg/L.[4] However, drinking water is considered to be safe for human consumption, if the concentration does not exceed 1.0 mg/L (Indian Standard 1996).[5]

Defluoridation is defined as, "the downward adjustment of level of fluoride in drinking water to the optimal level".[6] The different methods so far tried for removal of excess fluoride from water can be broadly classified into four categories[7] – adsorptive methods, ion exchange methods, precipitation methods, and miscellaneous methods. Adsorption technique of defluoridation functions on the adsorption of fluoride ions onto the surface of an active agent. Activated alumina, activated carbon, and bone char were among the highly tested adsorbing agents.[8] Application of domestic defluoridation plant, based on activated alumina, was launched by UNICEF in rural India.[9] The efficacy of the plant depends on temperature and pH of raw water; duration, for which the bone char is in contact with

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raw water. If calcium is present in the raw water, it precipitates out the fluoride.[10]

Defluoridation by ion exchange techniques is done through synthetic chemicals, namely, anion and cation exchange resins have been used for fluoride removal.[11] Precipitation methods are based on the addition of chemicals and the subsequent precipitation of a sparingly soluble fluoride salt as insoluble fluorapatite. The best example for this technique is the famous Nalgonda technique of defluoridation.[12] It involves addition of aluminium salts, lime, and bleaching powder, followed by rapid mixing, flocculation, sedimentation, filtration, and disinfection. Aluminium salt may be added as aluminium sulfate or aluminium chloride or combination of these two. The Modifications for Nalgonda technique are poly aluminium chloride and poly aluminium hydroxy sulfate.[13]

Natural adsorbents from various trees were tried as defluoridation agents. Seeds of the drumstick tree, roots of vetiver grass, and Tamarind seeds were few among them. The seeds of the drumstick tree (*Moringa oleifera*) adsorb fluoride from water. They have long been a traditional method for purification of turbid water in both India and Africa. The roots of vetiver grass (*Vetiveria zizanoides*) are another product that traditionally been used for water purification. Tamarind seeds were successfully tested for defluoridation by sorption. Since maximum defluoridation is achieved at an optimum pH of 7, post defluoridation pH adjustment is not required.[5]

Among all these methods, the adsorption method is more practicable. Most adsorbents and methods have some limitations such as low adsorption capacity, poor integrity, and need for pre-treatment. Therefore, a suitable, low cost, and biologically friendly method is required for removal of fluoride in drinking water. *M. oleifera* seed, Vetiver grass roots, *Tinospora cordifolia*, tamarind seed, tea ash, and egg shell powder are native materials available as effective fluorosis mitigation tools.[14] However, the applicability of these low-cost methods is limited either due to their low efficiency or lack of public acceptance. The successful and cost effective removal of contaminants from waste water, by adsorption techniques, demands the optimal operation of the adsorption units.[15]

Nevertheless, very few studies were conducted to assess the efficacy of defluoridation of water using three low-cost household materials, *Tamarindus indica, M. oleifera, and T. cordifolia*.[16] Thus, the present study was conducted to assess the household materials efficacy.

Aim and objectives

The aims of this study were as follows:

1. To determine the efficacy of low-cost household materials *T. indica, M. oleifera, and T. cordifolia* for defluoridation of water.
2. To assess reduction in percentage of fluoride content after filtering the standard solution at neutral, acidic, and alkaline pH through natural adsorbents *T. indica, M. oleifera,* and *T. cordifolia.*
3. To assess the reduction in pH value after filtering the standard solution at neutral, acidic, and alkaline pH through natural adsorbents *T. indica, M. oleifera,* and *T. cordifolia.*
4. To assess the comparison between the capacity of natural adsorbents *(T. Indica, M. oleifera,* and *T. cordifolia)* to remove fluoride from standard solution.

Material And Methods

Study setting

An *in vitro* study was carried out on three natural adsorbents (*T. indica, M. oleifera, and T. cordifolia)* in Spectro research laboratory ventures (P) Ltd., Kanpur (U.P) to assess the efficiency of the materials in defluoridation of the water using two tire filtration method.

Brief profile of the study area

Spectro research laboratory ventures (P) Ltd., is located in South Kanpur (U.P), it was well established and sophisticated laboratory having comprehensive facilities for testing, inspection, training, and consultancy services.

Study duration and ethical clearance

An *in vitro* study was conducted in Spectro research laboratory ventures (P) Ltd. Kanpur (U.P) in month of 10 August 2019–10 September 2019. The duration of the full study was 1 month. No ethical clearance is applicable to this type of study.

Materials and supplies

T. indica seed powder, *M. oleifera* seed Powder, *T. cordifolia* stem powder, Sodium Silicofluoride, Disposable gloves, Disposable mouth masks, Distilled Water, Drip set, Measuring Cylinder, Filter Cylinder, Electrode filling solution, TISAB III, and Mcllvaine's Buffer were used.

Training and calibration

Before the commencement of the study, the examiner was standardized and calibrated in the Spectro research laboratory ventures (P) Ltd. Kanpur (U.P) by chemical analysts to ensure uniform interpretations, understanding, and application of the procedure and analytical methods.

Test agent

Test agents are *T. indica* seed powder, *M. oleifera* seed Powder, and *T. cordifolia* in powder form purchased from the market and sun dried so that all the moisture gets evaporated from the powder. Now, the powder is sieved through ASTM E11 No. 200 to get the desired 75 micron particle size.

Stock solution

Ten parts per million (ppm) solution of sodium silicofluoride.

Preparation of stock solution

Amount of sodium silicofluoride in gram required for making 1000 ppm solution is calculated by the formula.

$$Weight\ in\ grams = \frac{Molecular\ weight\ of\ sodium\ silicofluoride}{Molecular\ weight\ of\ fluoride}$$

10 gm (approx) of sodium silicofluoride is dissolved in the 1 L of distilled water to get the 1000 ppm concentration of fluoride solution. The stock solution was, further, diluted to get the lower concentration of 10 ppm of fluoride. The pH of the solution is adjusted to 7.0 by adding a Mcllvaine's Buffer solution drop by drop to the sample solution using pH analyzer. The obtained standard solution is divided in to nine parts equally. Three solution is adjusted to pH of 3.0 by adding HNO_3 (0.1 mol^{L-1}), other three solution pH are adjusted to pH 10 by adding NaOH (0.1 mol^{L-1}) solution. Remaining three solutions are kept as a control group having its neutral pH of 7.0.

Results

When all three adsorbents *T. indica, M. oleifera* and *T. cordifolia* are compared at different pH with similar concentration of fluoride, it was found that *T. indica* was efficient in removal of fluoride from water. Table 1 shows pH and fluoride variation in water after exposure to different adsorbents (Neutral pH).

At Neutral pH 7.0, the fluoride concentration was 10 ppm. After the filtration process completed, there were three filtered samples with *T. indica, M. oleifera,* and *T. cordifolia* were analyzed separately in an fluoride ion electrode it was found that *T. indica s*hows reduction in fluoride level from 10 ppm to 2 ppm which was 80% reduction with decrease in pH from 7.0 to 6.8, whereas *M. oleifera* shows decrease in fluoride level from 10 ppm to 3 ppm which was 70% reduction with decrease in pH from 7.0 to 6.2, similarly, *T. cordifolia* shows decrease in fluoride level from 10 ppm to 4.6 ppm which was 54% with decrease in pH from 7.0 to 6.5. Table 2 shows pH and fluoride variation in water after exposure to different adsorbents (Acidic pH).

At acidic pH 3.0, the fluoride concentration was 10 ppm. After the filtration process completed, there were three filtered samples with *T. indica, M. oleifera,* and *T. cordifolia* were analyzed separately in an fluoride ion electrode, it was found that *T. indica s*hows reduction in fluoride level from 10 ppm to 3 ppm which was 70% reduction with increase in pH from 3.0 to 3.1, whereas *M. oleifera* shows decrease in fluoride level from 10 ppm to 4 ppm which was 60% reduction with decrease in pH from 3.0 to 2.9. Similarly, *T. cordifolia* shows decrease in fluoride level from 10 ppm to 3.3 ppm which was 67% with increase in pH from 3.0 to 3.2. Table 3 shows pH and fluoride variation in water after exposure to different adsorbents (Alkaline pH).

At Alkaline pH 10, the fluoride concentration was 10 ppm. After the filtration process completed, there were three filtered samples with *T. indica, M. oleifera,* and *T. cordifolia* that were analyzed separately in an fluoride ion electrode that it was found that *T. indica* shows reduction in fluoride level from 10 ppm to 2.5 ppm which was 70% reduction with increase in pH from 10 to 8.4, whereas *M. oleifera* shows decrease in fluoride level from 10 ppm to 3.4 ppm which was 60% reduction with decrease in pH from 10 to 8. Similarly, *T. cordifolia* shows decrease in fluoride level from 10 ppm to 6 ppm which was 67% with decrease in pH from 10 to 8.5.

Discussion

Adsorption is the defluoridation technique use in the present study.[17] The factors affecting the fluoride ion adsorption are pH and nature of the adsorbent.[18] Tamarind seed mainly contains polysaccharides with fat, tannis, proteins, and amino acids in minimum portions.

Murugan and Subramanian, in 2006,[5] demonstrates that the maximum defluoridation achieved by Tamarind seed was 90%,

Table 1: pH and fluoride variation in water after exposure to different adsorbents (Neutral pH)

S. No.	Adsorbents samples	Fluoride in ppm at baseline	Fluoride in ppm after filtration	pH Score at baseline	pH score after filtration
1.	Tamarindus *indica*	10	2	7.0	6.8
2.	Moringa oleifera	10	3	7.0	6.2
3.	*Tinospora cordifolia*	10	4.6	7.0	6.5

Table 2: pH and fluoride variation in water after exposure to different adsorbents (Acidic pH)

S. No.	Adsorbents samples	Fluoride in ppm at baseline	Fluoride in ppm after filtration	pH Score at baseline	pH score after filtration
1	Tamarindus *indica*	10	3	3.0	3.1
2.	Moringa oleifera	10	4	3.0	2.9
3.	*Tinospora cordifolia*	10	3.3	3.0	3.2

Table 3: pH and fluoride variation in water after exposure to different adsorbents (Alkaline pH)

S. No.	Adsorbents samples	Fluoride in ppm at baseline	Fluoride in ppm after filtration	pH Score at baseline	pH score after filtration
1.	Tamarindus *indica*	10	2.5	10	8.4
2.	Moringa oleifera	10	3.4	10	8
3.	*Tinospora cordifolia*	10	6	10	8.5

whereas defluoridation capacity decreases with increase in temperature. They showed that temperature is an unfavorable factor for fluoride adsorption on tamarind seed powder. In the present study, it was found to be 80% at neutral pH which is similar to the present study.

Kumar *et al*., in 2012,[19] demonstrate that the tamarind fruit cover in its natural and acidic form could be used as a potential defluoridating agent for the removal of fluoride ion from water. The maximum defluoridation occurs at pH 6.0, the amount of adsorbent significantly influences the fluoride adsorption. It can also be seen that the fluoride removal markedly increased up to adsorbent dose of 0.7 g; however, further, increase in adsorption does not show any appreciable improvement in fluoride removal.

In the study by Parlikar and Mokashi, in 2013,[20,21] demonstrate *Moringa Oliefera* In acidic pH maximum removal efficiency Was 39% whereas it was 51% in case of alkaline pH. They observed that maximum removal efficiency of fluoride removal was 76% at 400 mg/L and 40% at 50 mg/L which was similar to the resent study.

Dobaradaran *et al*.[22] demonstrate that *M. oleifera* seed ashing is done at 650°C. The highest removal adsorption was at 64 g/L adsorbent, 10 min contact time and initial F concentration at 8 mg/L the fluoride removal efficiency was 81.14%. The results showed that the *M. oleifera* ash can be used as an environmental friendly, cheap, and effective adsorbent from aqueous solutions.[23]

Sontakke *et al*., in 2017,[15] demonstrated that *M. oleifera* lowers the fluoride concentration from 3 ppm to 0.015 ppm at baseline after 6 h fluoride concentration raised to 0.3 ppm and persisted same after 24 h, whereas *T. indica* showed decrease in fluoride concentration from 3 ppm to 0.18 ppm after 6 h, *T. indica* was found to be more effective in defluoridation showing reduction up to 0.18 ppm.

In this study, *T. indica* was found to be most efficient among the natural adsorbent such as *M. oleifera* and *T. Cordifolia*. It reduces the fluoride level from 10 ppm to 2 ppm at neutral pH. The down flow column filtration process was used in the present study as the drop rate of 1.5 ml/min in two tire filtration system. This technique was easy and cost effective and can be adopted in rural areas as these adsorbents are easily available. Furthermore, *in vitro* studies needed to be conducted to demonstrate the fluoride adsorption at different particle size and doses.

Conclusion

Fluorosis is an important pubic heath problem in India as there is no cure for the disease but only prevention is the solution. Prevention can be achieved by defluoridation of water through various methods.[24] The present study represents a way of reducing fluoride content in water using locally available natural adsorbents.

The removal of fluoride ion from the prepared stock solution using adsorption technique through two tire filtration system is done.[25] It was observed that *T. indica* seed powder shows maximum reduction in fluoride concentration from 10 ppm to 2 ppm, 3 ppm, and 2.5 ppm at neutral, acidic, and alkaline pH, fluoride removal is favored at neutral pH.[26,27]

Therefore, from the present study, it was concluded that *T. indica* seed powder has a potential to be an efficient defluoridating agent at domestic level as compared to *M. oliefera* and *T. cordifolia*. Among the aforementioned plants, *T. indica* powder was found to be the best domestic material for defluoridation property.[28] It can be adopted for the defluoridation method in villages as it is cheaper and easily available biosorbent in Indian households.

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