Comparative Evaluation for Defluoridation of water, using three low cost household materials: Tamarind us Indica (T.I), Moringa Oleifera (M.O) and Tinospora Cordifolia (T.C) – 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 because health hazards, deflouridationis the technique used to remove fluoride from water. Household methods of defluoridation by using adsorbent materials are cost effective than other methods used to remove fluoride from water.

Aim: To determine the efficacy of low cost household materials Tamarind us Indica (T.I), Moringa Oleifera (M.O) and Tinospora Cordifolia (T.C) for defluoridation of water

Material & Methods: An in-vitro study was done using materials like Tamarind us Indica, Moringa Oleifera, Tinospora Cord folia. The materials were obtained in powder form of particle size of 75 micron. Standard solution was prepared by using sodium silica-fluoride (NaSiF6), which was weighed and dissolved in distilled water and diluted to 10ml. The solution was then diluted to make 10 parts per million (ppm) standard solutions. The standard solution was filtered through the 2 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 Tamarindus Indica was found to have Defluoridation capacity of 80% at neutral pH as compared to other household materials like Moringa Olieferaand Tinospora Cordifolia 70% &67% respectively reduction in fluoride concentration at acidic pH.

Conclusion: The present study concluded that amongst the aforementioned plants, Tamarind us 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, 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 etc. Turmeric is good source of fluoride[3].According to 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]

De fluoridation was the conventional and widely tested method for supplying safe water to the fluoros is affected communities. De fluoridation 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, Miscellaneous methods. Adsorption technique of de fluoridation 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 de fluoridation plant, based on activated alumina, was launched by UNICEF in rural India.[9]The basic principle of functioning of Brick piece column is the same as that of activated alumina. The soil used for brick manufacturing contains Aluminum oxide. During burning operation in the kiln, it gets activated and adsorbs excess fluoride when raw water is passed through [7]. The mud pot also will act as adsorbent media. De fluoridation by bone char as the ion exchange and adsorption between fluoride in the solution and carbonate of the apatite comprising bone char. The efficacy of the plant depends upon temperature and pH of raw water; duration for which the bone-char is in contact with raw water. If Calcium is present in the raw water, it precipitates out the fluoride [10].

De fluoridation by ion exchange techniques is done through synthetic chemicals, namely, anion and cation exchange resins have been used for fluoride removal. Some of these are Polyanion (NCL), Tul-sion a - 27, Deacedite FF (IP), Amberllte IRA 400, Lewatit MIH - 59, and Amberlite XE – 75. These resins have been used in chloride and hydroxy form [11]. Precipitation methods are based on the addition of chemicals (coagulants and coagulant aids) 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 de fluoridation [12]. It involves addition of Aluminum salts, lime and bleaching powder followed by rapid mixing, flocculation, sedimentation, filtration and disinfection. Aluminium salt may be added as aluminum sulphate (alum) or aluminum chloride or combination of these two. The Modifications for Nalgonda technique are Poly Aluminum Chloride (PAC) and Poly Aluminum Hydroxyl Sulphate (PAHS)[13].

In reverse osmosis, the hydraulic pressure is exerted on one side of the semi permeable membrane which forces the water across the membrane leaving the salts behind. The relative size of the pollutants left behind depends on the pressure exerted on the membrane. In electro dialysis, the membranes allow the ions to pass but not the water. The membranes are very sensitive to pH and temperature [11].De fluoridation by electrolysis is the basic principle of the process is the adsorption of fluoride with freshly precipitated aluminum hydroxide, which is generated by the anodic dissolution of aluminum or its alloys in an electro chemical cell [13].

Natural adsorbents from various trees were tried as de fluoridation 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 (Vetiveriazizanoides) are another product that traditionally been used for water purification. Tamarind seeds were successfully tested for de fluoridation by sorption. Since maximum de fluoridation is achieved at an optimum pH of 7, post de fluoridation pH adjustment is not required [5].

Amongst 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, biologically friendly method is required for removal of fluoride in drinking water. Moringa Oleifera seed, vetiver grass roots, Tinospora Cordifolia, tamarind seed, tea ash and egg shell powder is native materials available as effective fluorosis migitation 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 de fluoridation of water by using three low cost household materials,Tamarindus Indica, Moringa Oleifera and Tinospora Cordifolia. Thus the present study was conducted to assess the household materials efficacy.

Aim & Objectives

Aim

- To determine the efficacy of low cost household materials Tamarindus Indica, Moringa Oleifera and Tinospora Cordifolia for de fluoridation of water

Objectives

- To assess reduction in percentage of fluoride content after filtering the standard solution at neutral, acidic and alkaline pH through natural adsorbents Tamarind us Indicia, Moringa Oleifera and Tinospora Cord folia.
- To assess the reduction in pH value after filtering the standard solution at neutral, acidic and alkaline pH through natural adsorbents Tamarind us Indica, Moringa Oleifera and Tinospora Cord folia.
- To assess the comparison between the capacity of natural adsorbents (Tamarind us Indica, Moringa Oleifera and Tinospora Cord folia) to remove fluoride from standard solution.

Material and Methods

Study Setting : An in vitro study was carried out on three natural adsorbents (*Tamarindus Indica, Moringa Oleifera and Tinospora Cord folia)* purchased from online market (Amazon) in Spectro research lab ventures (P) Ltd., Kanpur (U.P) to assess the efficiency of the materials in de fluoridation of the water using two tire filtration method.

Brief profile of the study area: Spectro research lab 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: An in vitro study was conducted in Spectro research lab ventures (P) Ltd. Kanpur (U.P) in month of 10 August 2019 – 10 September 2019. The duration of the full study was 1 month.

Materials and supplies

Tamarindus Indica seed powder, Moringa Oleifera seed Powder, Tinospora Cord folia, Sodium Silicofluoride, Disposable gloves, Disposable mouth masks, Distilled Water, Drip set, Measuring Cylinder, Filter Cylinder, Electrode filling solution, TISAB III, Mcllvaine's Buffer

Training and calibration

Before the commencement of the study, the examiner was standardized and calibrated in the Spectro research lab 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 *Tamarindus Indica* seed powder, *Maringa Oleifera* seed Powder, *Tinospora 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: 10 ppm solution of sodium Silico fluoride

Preparation of Stock Solution

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

$$Weight\ in\ Grams = \frac{Molecular\ weight\ of\ Sodium\ Silicofluoride}{Molecular\ weight\ of\ Fluoride}$$

10gm (approx) of Sodium Silicofluoride is dissolved in the 1 liter of distilled water to get the 1000 parts per million (ppm) concentration of fluoride solution. The stock solution was further diluted to get the lower concentration of 10ppm 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 9 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 3 solutions is kept as a control group having its neutral pH of 7.0.

Filtration: Continuous down flow column method

A two tire filtration system was used to filter the standard solution. Standard solutions were passed through filters containing 20gm of adsorbents in between sand and cotton plugs so the adsorbents not get flow out with filter at the rate of **1.5 ml/minute**. After the completion of filtration process all the 9 samples are analyzed for pH and Fluoride concentration. The treated water gets collected in the storage vessel, which is then taken for the analytical evaluation of fluoride and pH. To determine the percentage of fluoride reduction and pH variation after the treatment of standard solution with different adsorbent powders at an acidic, basic and neutral pH.

Analytical Method

The method used for treated water analysis is Fluoride ion selective electrode method. The machine is called ion meter. The components of ion meter are: 1) Electrode filling solution.2) TISAB III – Total Ionic Strength Adjustment Buffer Solution. Ion meter was started 15 minutes before analysis, first the ion

electrode is filled with electrode filling solution to check for the air bubble after which the electrode is washed with water and wipe out to make it clean. Now 50 ml of sample is taken in the beaker with the help of measuring cylinder and 5 ml of TISAB III is added with the help of pipette.

Sample beaker is kept upon the magnetic stirrer and a stirring bar is placed in the beaker for moderate stirring so the distribution of ion is homogeneous. Electrode is dipped in the sample solution and channel for fluoride is selected. Now the reading starts stabilizing, it takes around 1-3 minutes, reading is noted down as the ion meter gets stabilized.

Analysis of Fluoride level was done before and 24hr after the filtration of stock solution with three natural adsorbents powder at (2% w/v) solutions.

1). Moringa Oliefera – 20g/litre of stock solution.
2). Tamarindus Indica – 20g/litre of stock solution
3) Tinospora cord folia – 20g/litre of stock solution.

Analysis of pH variation in water at baseline and 24 hr after treatment with three natural adsorbents.

Statistical Analysis

The data obtained was compiled systematically. The total data was distributed meaningfully and presented as individual tables.

Results

When all three adsorbents *Tamarindus Indica, Moringa Oleifera* and *Tionospora cordifolia* are compared at different pH with similar concentration of fluoride it was found that Tamarindus Indica was efficient in removal of fluoride from water

Table1. Distribution of pH and Fluoride variation in water after exposure to different adsorbents at Neutral pH

At Neutral pH 7.0 the fluoride concentration was 10 ppm. After the filtration process completed there were three filtered samples with *Tamarind us Indica, Moringa Oleifera* and*Tionospora cordifolia* were analyzed separately in an fluoride ion electrode it was found that *Tamarind us Indica* Shows reduction in Fluoride level from 10ppm to 2 ppm which was 80% reduction with decrease in pH from 7.0 to 6.8 whereas *Moringa Oleifera* shows decrease in fluoride level from 10ppm to 3 ppm which was 70% reduction with decrease in pH from 7.0 to 6.2, similarly *Tinospora Cord folia* Shows decrease in fluoride level from 10ppm to 4.6 ppm which was 54% with decrease in pH from 7.0 to 6.5

Table2. Distribution of pH and Fluoride variation in water after exposure to different adsorbents at Acidic pH

At Acidic pH 3.0 the fluoride concentration was 10 ppm. After the filtration process completed there were three filtered samples with *Tamarind us Indica, Moringa Oleifera* and *Tionospora cordifolia* were analyzed separately in an fluoride ion electrode it was found that *Tamarind us Indica* Shows reduction in Fluoride level from 10ppm to 3 ppm which was 70% reduction with increase in pH from 3.0 to 3.1 whereas *Moringa Oleifera* shows decrease in fluoride level from 10ppm to 4 ppm which was 60% reduction with decrease in pH from 3.0 to 2.9, similarly *Tinospora Cord folia* Shows decrease in fluoride level from 10ppm to 3.3 ppm which was 67% with increase in pH from 3.0 to 3.2

Table3. Distribution of pH and Fluoride variation in water after exposure to different adsorbents at alkaline pH

At Alkaline pH 10, the fluoride concentration was 10 ppm. After the filtration process completed there were three filtered samples with *Tamarindus Indica, Moringa Oleifera* and*Tionospora cordifolia* were analyzed separately in an fluoride ion electrode it was found that *Tamarindus Indica* Shows reduction in Fluoride level from 10ppm to 2.5 ppm which was 70% reduction with increase in pH from 10 to 8.4 whereas *Moringa Oleifera* shows decrease in fluoride level from 10ppm to 3.4 ppm which was 60% reduction with decrease in pH from 10 to 8, similarly *Tinospora Cordifolia* Shows decrease in fluoride level from 10ppm to 6 ppm which was 67% with decrease in pH from 10 to 8.5

Table 1: Distribution of pH and Fluoride variation in water after exposure to different adsorbents at 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	**Tamarind us Indica**	10	2	7	6.8
2	**Maringa Oleifera**	10	3	7	6.2
3	**Tinospora cordifolia**	10	4.6	7	6.5

Table 2: Distribution of pH and Fluoride variation in water after exposure to different adsorbents at 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	**Tamarind us Indica (T.C)**	10	3	3	3.1
2	**Maringa Oleifera (M.O)**	10	4	3	2.9
3	**Tinospora cord folia (T.C)**	10	3.3	3	3.2

Table 3: Distribution of pH and Fluoride variation in water after exposure to different adsorbents at 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	**Tamarind us Indica**	10	2.5	10	8.4
2	**Maringa Oleifera**	10	3.4	10	8
3	**Tinospora cord folia**	10	6	10	8.5

Discussion

Successful application of the adsorption techniques demands innovation of cheap, non – toxic, easily and locally available material. Tamarind seed mainly contains polysaccharides with fat, tennis, proteins and amino acids in minimum portions. The factors affecting the fluoride ion adsorption are pH and nature of the adsorbent, contact time, particle size and fluoride ion concentration. There were several studies conducted on the natural adsorbents to reduce the fluoride ion concentration through adsorption techniques like filtration process, Batch sorption process [24].

There were various studies on natural adsorbents which showed that *Tamarind us Indica* had higher percentage of fluoride reduction as compared to other natural adsorbents like *Tinospora Cord folia* and *Maringa Oleifera*. In the present study two tire filtration processes is used as an adsorption technique. It was also found that *Tamarind us Indica* shows the higher percentage of fluoride ion reduction at neutral pH whereas *Tinospora Cord folia* shows the lesser percentage of reduction in fluoride concentration

Murugan et al 2006[5] demonstrates that the maximum de fluoridation achieved by Tamarind seed was 90% whereas, de fluoridation capacity decreases with increase in temperature. They showed that temperature is an unfavourable factor for fluoride adsorption on tamarind seed powder. The present study showed that there is 80% reduction in fluoride concentration by *Tamarind us Indica* at neutral pH which is approximate to the previous studies.

Kumar NP et al 2012[19] demonstrates 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 de fluoridation occurs at pH 6.0, the amount of adsorbent significantly influence the fluoride adsorption. It can also be seen that the fluoride removal markedly increased upto adsorbent dose of 0.7g, however further increase in adsorption does not show any appreciable improvement in fluoride removal.

In the study by Parlikar et al 2013[20] demonstrates that *Moringa Oleifera* adsorbent in acidic pH had maximum removal efficiency of fluoride ion was 39%whereas in case of alkali it was 51%. They observed that maximum removal efficiency of fluoride removal was 76% at 400 mg/liter and 40% at 50 mg/liter. The present study showed that there is 70% reduction in fluoride concentration by Moringa Oleifera at neutral pH which is nearly similar to the previous studies.

Dobaradaran S et al (2015) [22] demonstrates that *Moringa Oleifera* seed ashing is done at 650 0C. 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 *Moringa Oleifera* ash can be used as an environmental friendly, cheap and effective adsorbent from aqueous solutions.

Sontakke et al 2017[15] demonstrated that *Moringa Oleifera* lowers the fluoride concentration from 3ppm to 0.015ppm at baseline after 6 hr fluoride concentration raised to 0.3 ppm and persisted same after 24 hr whereas Tamarind us Indica showed decrease in fluoride concentration from 3ppm to 0.18 ppm after 6 hr, *Tamarind us Indica* was found to be more effective in de fluoridation showing reduction upto 0.18 parts per million.

Pandey P. K. et al[29] demonstrates that *Tinospora Cord folia* had maximum fluoride removal efficiency of 70% at the fixed contact time of 120 minutes at pH 7. This is due to the lack of active sites on the adsorbents surface, the percentage removal of fluoride removal remain constant after 5 mg/ L. The present study showed that there is 54% reduction in fluoride concentration at neutral pH whereas it was found that there is 67% reduction in fluoride concentration at acidic pH 3 which is in contrast with the other studies.

In this study *Tamarind us Indica* was found to be most efficient amongst the natural Adsorbent like *Moringa Oleifera* and *Tinospora Cord folia*. It reduces the fluoride level from 10ppm to 2 ppm at neutral pH. The down flow column filtration process was used in the present study as the drop rate of 1.5ml/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 de fluoridation of water via various methods. 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. It was observed that *Tamarind us Indica* seed powder shows maximum reduction in fluoride concentration from 10 ppm to 2ppm, 3ppm and 2.5 ppm at Neutral, Acidic and alkaline pH, Fluoride removal is favored at neutral pH.

Therefore from the present study it was concluded that *Tamarind us Indica* seed powder has a potential to be an efficient de fluoridation agent at domestic level as compared to *Moringa Oliefera* and *Tinospora Cord folia*. . Amongst the aforementioned plants, *Tamarind us indica* powder was found to be the best domestic material for de fluoridation property. It can be adopted for the de fluoridation method in villages as it is cheaper and easily available bio sorbent in Indian households.

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To cite this article: Comparative Evaluation for Defluoridation of water, using three low cost household materials: Tamarind us Indica (T.I), Moringa Oleifera (M.O) and Tinospora Cordifolia (T.C) – An in vitro study: Dr. Surbhit Singh, Dr. Nidhi Pruthi, Dr. Jaspreet Singh Tuteja, Dr. Devina Pradhan, Dr. Deepika Singh, Dr. Swati Parsai, Rama Univ. J. Dent. Sci. 2020 December; 7 (4): 11-17