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COMPARISON OF FLUORIDE UPTAKE BETWEEN MORINGA OLEIFERA AND TAMARINDUS INDICA

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ABSTRACT

Fluoride is one of the important minerals which is required by our body in order to maintain healthy bones and tooth but when provided in high amount it has many unfavourable effects such as dental fluorosis, osteoporosis or crippling skeletal fluorosis, bone deformities etc. One of the major sources of high fluoride intake is from fluoride contaminated water, so the removal of fluoride from the contaminated water is necessary in order to prevent the endemic of fluorosis. Several materials are tested for the fluoride uptake such as Moringa oleifera and Tamarindus indica, it was observed that Moringa oleifera have up to 93% of the uptake capacity at optimum pH and coagulant dose and Tamarindus indica dried seeds had up to 92.1% uptake at neutral pH but only when the particle size of coagulant Strictly at 75 micron and when the particle size is increased the uptake efficiency was decreased drastically, thus we can say fluoride uptake by Moringa oleifera gave better results as it does not require a particular particle size and also it have more fluoride uptake efficiency.

INTRODUCTION

Fluoride is a naturally occurring mineral which is generally present in the soil and water, most of the fluoride present in soil and water is due to the rock formation. It is present in the earth crust in the form fluorspar, rock phosphate, cryolite, apatite, muscovite, biotite, amphiboles and mica 17), and then fluoride leaches off into ground water from these minerals. Additional artificial fluoride is also present in the soil and ground water which comes from the fertilizers, mining industries, factories and different chemical industry waste discharge into rivers and landfills.

Fluoride is one of the minerals which is required by our body, essentially by our bones and tooth. Almost 95% of the fluoride is present in the bones and tooths. Every day when we eat, brush, drink minerals get lost from the tooth enamel and due to this enamel become vulnerable to the bacteria and plaque. Remineralization of enamel is necessary to prevent it from decaying. Fluoride is one such mineral which helps in the remineralization of the enamel and hence prevent tooth decaying. Thus, by using fluoride in water the epidemiology of the dental crisis can be controlled.

But the excessive intake of fluoride may cause toxic and harmful effects such as mild dental fluorosis which arise from drinking water which have fluoride concentration between 0.9 – 1.2 mg/L 15), osteoporosis or crippling, skeletal fluorosis 31), calcification of tendons and ligaments, bone deformities, damage to kidney 23), reproductive organs, nerve and muscles. Fluoride is found to more toxic then lead, and when accumulates even in minute concentration can cause mind damage, can hinder the development of the mind also associated with the less IQ in humans and abnormal behaviour in animals. 4) It was observed that it interferes with the thyroid glands and leads to hypothyroidism by increasing the concentration of TSH and a decreasing the concentration of T3 and T4 hormones. 13)27)**,** in some population it also affects the glycolysis by inhibiting the enzyme enolase which results into the high level of glucose in blood and it also been linked to the cancer 33). Water having concentration of fluoride more than 1.5mg/l is considered as unfit for drinking according to WHO guidelines 32). However, drinking water is considered to be safe for human consumption, if the concentration varies below 1.0 mg/l or remain little above 1.0mg/L. 21)

There are many underdeveloped and developing countries facing the high contamination of fluoride in ground water which leads the endemic. Syria, Jordan, Egypt, Libya, Algeria, Sudan and Kenya, Turkey, Iraq, Iran, Afghanistan, India, northern Thailand and China are the major countries going through endemic. The countries which are affected from fluoride contamination of water are generally under developing or developing countries and these countries cannot afford the expensive process for the defluoridation hence a cost-effective method is required with good fluoride removal efficiency.

REMOVAL OF FLUORIDE FROM CONTAMINATED WATER

Many techniques have been developed to remove fluoride from water such as chemical coagulation flocculation, adsorption, electro coagulation, and electro flotation and membrane separation. 34)35)16)10)5)20)

But most used techniques for defluoridation of drinking waters is usually accomplished by either precipitation or by adsorption processes. Another well-known method called 'Nalgonda Technique' which is developed by National Environmental Engineering Research Institute, Nagpur, India.11) Many effective techniques for fluoride removal are being develop at low-cost material such as Moringa oleifera (Drumsticks), Tamarindus indica (Tamarind), Azadirachta indica (Neem) and ocimum tenuiflorum (tulsi). The technique which utilises these natural absorbents is based on the principle of absorption. Adsorption is described as the one of the most practicable and cost-effective technique for fluoride removal but generally pH dependable.14) In adsorption method, fluoride of raw water is retained on the adsorbent due to physical, chemical or ion exchange interactions. Fluoride uptake by Moringa oleifera and Tamarindus indica are such methods which are easy to perform, affordable and shows good fluoride removal efficiency from contaminated water.

Moringa oleifera which is also known as drumsticks tree, miracle tree, the ben oil tree, or the horseradish tree is native to India but also grow in Asia, Africa and South America. Moringa oleifera is being used since ancient times for its medical properties and health benefits. It has antifungal, antiviral, anti-inflammatory and anti-depressant properties. It is rich in antioxidants and vitamins. Vitamin A, vitamin B1 (thiamine), B2 (riboflavin), B3 (niacin), B-6, Folate and ascorbic acid (vitamin C), calcium, potassium, Iron, magnesium, Phosphorus, zinc are the important vitamins and minerals which are present in Moringa oleifera. Now, it is being used in fluoride uptake and showing promising results. The general method by which fluoride is removed is by adsorption, where the fluoride present in water get adsorb to the minerals of Moringa oleifera such as calcium, magnesium, phosphorus, zinc etc. Adsorbent is prepared from the different parts of the tree such as drumstick leaves, leaves extract, dried seeds, and seeds after oil extractions etc. Fluoride gets adsorb to minerals by weak forces and thus reduce the concentration of fluoride in water. Then adsorb fluoride can be elute by using any eluent if needed. Also, precipitation of fluoride with these minerals results in fluoride removal from water. Moringa oleifera seeds are also been used in several experiments as a natural coagulant for the removal of different pollutants.25)9)29)24)6)8)7)

Tamarindus indica is a member of multipurpose, tropical evergreen leguminous species of Caesalpiniaceae subfamily. It is native to Africa and southern Asia. The fruit of Tamarind is very soft, succulent, juicy ripened pulp which is usually used in confectionery and household as an ingredient of chutnies, pickles, curries, preserves, beverages, and sherbets. Tamarind fruits have several nutrient values, electrolytes, phytonutrients, vitamins, and minerals. Also, Tamarind fruit exhibit antimicrobial, antibacterial, antioxidative, anti-inflammatory and anti-diabetes property. The major minerals found in Tamarind fruit is potassium, calcium, magnesium, sodium and iron. It is being used for the fluoride removal from water and it works on the same principle as drumsticks works, adsorption where adsorbent is prepared from different parts of the Tamarind tree such as leaves, roots, seeds, fruits etc and fluoride get adsorb to the minerals present in tamarind which are potassium, calcium, magnesium etc. And fluoride get adsorb to the minerals by weak forces, then fluoride can be eluted by using a suitable eluent. Also, precipitation can also take place between minerals and fluoride which leads to the removal of fluoride from water.

Here we are comparing different materials/methods of fluoride uptake by Moringa oleifera and Tamarindus indica.

MATERIAL AND METHODS

Sample preparation

Adsorbent preparation from Moringa oleifera

Dried Moringa oleifera seeds were obtained locally and kept in an oven at 500 C for 12 hrs. The seeds were powdered and sieved. And to obtain the extract of Moringa oleifera nut powder 10 g of Moringa powder was suspended in HCI and NaOH solution and dissolved. **Error! Reference source not found.**

The dried fruit of Moringa oleifera were collected from Lucknow, India. The seeds coat and wings were removed manually and grinded into a powered followed by sieving. Then the oil was extracted by rotatory evaporation distillation and left out seed residue was collected for experiment and then the seed residue was dried for 2-3 days and the powdered followed by sieving. 22)

The Moringa oleifera leaves were collected and then the leaf extract was used as adsorbent sample. 26)

Drumsticks were collected from the local trees and powdered, then powder sample was treated with HNO3 for acid treatment and NaOH for alkali treatment. The mixture was boiled for about 20 minutes the sample was washed with distilled water. Finally, it was dried again in an oven at 50°C for 6 hrs. 1)

Adsorbent preparation from Tamarindus indica

Samples were collected from two different sites from Bareilly district. The shell of tamarind was removed and washed with distilled water followed by drying in the sun. Then it was powdered (absorbent) and sieved to get fine powder. 2)

Tamarind seeds were collected from the kitchen waste and soaked water to remove any pulp followed by washing with water and double distilled water. Then the seed were dried in an air oven at 110 C for an hour followed by grinding and sieving. 19)

Tamarind seeds were collected from the kitchen waste and soaked in water to remove pulp followed by washing with water and then double distilled water. Then seeds were dried at 110 C and then grinded into fine powder. 40g of the powder sample was treated with HNO3 for acid treatment and with NaOH for alkali treatment and again dried. The acid adsorbent is washed with HNO3 and alkali adsorbent is washed with NaOH. 28)

Tamarind seeds were collected, washed, dried and powdered manually, then treated with 4N KOH solution. 30)

Fluoride uptake

Fluoride uptake by Moringa oleifera sample

Non-flow batch sorption studies without any agitation were performed and the effect of certain parameters like contact time of the sample with fluoride, dose of Moringa oleifera powder and pH of solution were also studied. **Error! Reference source not found.**

Jar test 18) which is a popular method to determine the efficiency of coagulant process was conducted. And the coagulant doses were optimised using initial fluoride concentration as 10mg/ml. Once test was performed the removal efficiency was calculated. 22)

5.0 ml of Alizarin red and zirconyl acid solutions were added to 10 ml of both standard and sample solutions. The solutions were mixed thoroughly and allowed to stand for 1 h for full colour development then Absorbance readings were taken at 520 nm. 26)

The fluoride removal studies by adsorption were conducted in by using 100 ml of synthetic water sample at different pH and initial concentrations of fluoride ion with varied dosage of adsorbent for different time period. After giving a required contact time, the contents of the flasks were filtered using and the filtrate was used for fluoride ion estimation using SPADNS method 3). The above procedure was repeated for different pH, contact times, adsorbent doses, particle sizes and different initial fluoride ion concentrations. The parameters were varied to find the maximum fluoride removal efficiency. 1)

Fluoride uptake by Tamarindus indica

Bags made up of cotton were filled with 25mg of the absorbent, then they were dipped in the sample of 100ml overnight. The brown colour given by shell powder was neutralized by using hydrogen peroxide. Then the

concentration of fluoride in the sample were determined by using SPADNS technique. The resulting colour complex from the SPADNS technique is measured by using spectrophotometer. Then statistical analysis was done to compare the reduction in the concentration of fluoride between pre-test and past test reading. 2)
Two methods were used to detect the defluoridation- Batch study and column study. 19)

1. Batch study-

0.1g of the adsorbent was mixed in 50 ml of the synthetic fluoride water and after a definite time interval the solution was centrifuged, and supernatant was analysed for fluoride by using SPADNS reagent and spectrophotometer. The same method was done for the sample which was collected locally. 19)

2. Column study-

5g of the tamarind seed powder was packed into a column and synthetic fluoride solution was drawn into the column, effluent was collected at interval of times then analysed with same method which is used in batch study. After column saturation, the adsorbed fluoride ion was eluted with HCl. Desorption studies were carried out by agitating 100 mg of fluoride-loaded Tamarind seed in 50 ml of 0.1 mol l21 HCl for a period of 2 h. 19)
The fluoride removal studies by adsorption were conducted in 250 ml conical flask using 100 ml of synthetic water sample containing different pH and initial concentrations of fluoride ion with varied dosage of coagulant. Then the contact period was given for different particle sizes. After giving a required contact time, the contents of the flasks were filtered, and the filtrate was used for fluoride ion estimation using SPADNS method. The above procedure was repeated for different pH, contact times, adsorbent doses, particle sizes and different initial fluoride ion concentrations. 28)
Column was prepared with an inlet and outlet at the bottom. The adsorbent was filled in the column and stock fluoride solution was flowed through the bed of the adsorbent. And as the principle states, fluoride get adsorbed to the adsorbent and thus defluorination of water takes place. The fluoride removed is calculated by marking the difference in the initial and final concentration of the fluoride in the water. And further analysis was done by using Fluoride selective electrode. 30)

RESULTS

Results for Moringa oleifera

It was observed that when the dose of Moringa oleifera adsorbent was increased the fluoride uptake was increased. The dose was increased progressively from 100 to 2000mg/l, fluoride uptake was also increased from around 8% at 300mg/l to 87% at 1000mg/l. But it was observed that after 1000mg/l there was no increase in fluoride uptake, it was constant which shows that the uptake was due to the formations of chemical complexes between fluoride and Moringa oleifera adsorbent. Also, it was observed when pH was increased from 3 to 6 the fluoride removal was also increased from 75% to 89% and after the pH of 6 -12 there was decrease in the fluoride removal from 89% to 77%, hence it was determined that pH 6 is the optimum for fluoride removal. And then it was observed that at optimum pH and coagulant dose the fluoride uptake was increased by 4%. **Error! Reference source not found.**

It was determined that when dose of adsorbent increased fluoride uptake increased. Since different dose of adsorbent at different concentration of fluoride were used it was found that till 16g/l dose of adsorbent there was significant difference between the fluoride uptake but at 18g/l of the adsorbent there was no significant difference hence 16g/l was the optimum dose of adsorbent for fluoride uptake. Also, the effects of the initial fluoride concentrations were also determined, and it was found that the removal of fluoride was increased when the concentration of initial fluoride were decreased. The maximum fluoride removal efficiency was observed at 18g/l that is 95.49% it was it does not have any significant change from the 16g/l which had 95.22% fluoride removal efficiency. 22)
When the results of Fluoride removal by Moringa oleifera was compared with the reference point, it was observed that there was a decrease level of fluoride from 3ppm to 0.015ppm, but after 6 hours fluoride level was found to be increased to 3ppmand it remained same even after 24hours. 26)
It was observed that when the Moringa oleifera seeds powder when treated with acid the optimum pH at which the maximum removal of fluoride (39%) took place is at 1 and for alkali treated seeds powder pH was 10

where it showed maximum (51%) fluoride removal efficiency. Also, the optimum contact time was determined for different particle size and it was observed that when the particle size decrease the contact time also reduce. Contact time of 2 hrs and 2.5 hrs were found optimal for adsorbents having particle size of 212 µ and 600 µ respectively and it was observed that after a time the fluoride removal efficiency remain same. The optimum adsorbent dose was also determined, and it was found that increase in adsorbent dose leads to increase in fluoride removal. At 400mg/l dose of adsorbent the maximum fluoride removal 68% and 76% was observed for both 212µ and 600µ, respectively. 1)

Results for Tamarindus indica

It was observed that there was 50% reduction in the fluoride concentration post treatment tamarind fruit shell powder. The initial fluoride concentration for site 1 was 1.53ppm and it was reduced to 0.76 ppm post treatment and the initial fluoride concentration for site 2 was 1.83 ppm pre-treatment and 0.91 ppm post treatment. 2)

The effect of pH was determined for fluoride removal and it was found that at pH 7 fluoride removal was more efficient also it was found temperature increase leads to the decrease in sorption hence decrease in fluoride removal. Effect of particle size and dose of adsorbent was also determined, and it was observed that when the particle size was increased sorption was decreased, when particle size increased from 75 to 300-micron fluoride removal efficiency reduced from 92.1% to 38.4% and for the adsorbent dose it was found that when there is increase in adsorbent dose fluoride uptake also increase. The sorption dynamics were also determined. 19)

The optimum pH was determined for the acid and alkali washed adsorbent and it was found that acid washed adsorbent gives maximum fluoride removal efficiency (37%) at pH 1 and alkali washed adsorbent gives maximum fluoride removal efficiency (51%) at pH 10. The relation between contact time and particle size was determined and it was observed that the contact time reduces with decrease in particle size and once a given particle size reach particular contact time the removal efficiency becomes constant. It was determined that contact time of 2.0hrs and 2.5hrs were optimal for adsorbents having particle size of 212 µ and 600 µ respectively. The optimum adsorbent dose was also determined, and it was found increase in adsorbent dose leads to increase in fluoride removal. The maximum fluoride removal i.e., 76% and 68% was observed at 400mg/l concentration of adsorbent dose for particle size 212 µ and 600 µ respectively. 28)

At the inlet of the column the fluoride concentration was 9.25 ppm and with the 3ml/min flow rate of adsorbent for 5.5 hours the fluoride concentration at the outlet was found to be 8.96 ppm and the adsorption efficiency was found to be 3.14%. 30)

DISCUSSION

For Moringa oleifera, dried Moringa oleifera seed powder **Error! Reference source not found.** gave the best results. The dose of 1g/ l gave the fluoride removal of 89% at pH 6 which is very close to the neutral pH hence easily achievable and when the optimum dose of the adsorbent and the pH was provided the removal efficiency was increased by 4%, so total 93% of the fluoride removal efficiency was given by this method, compare to the method in which drumstick powder 1) was used where optimum pH was found 1 for acid treated adsorbent and 10 for alkali treated adsorbent. Maintaining very low pH or very high pH is not cost efficient hence this method was expensive and not considered as a cost-effective treatment of fluoride water also the fluoride removal percentage was less compared to the method **Error! Reference source not found.**. Fluoride removal efficiency of adsorbent of seeds after the oil extraction 22) was also found to be good, 18g/l adsorbent gave 95.49% removal of the fluoride but for this method more adsorbent dose is required compare to the method **Error! Reference source not found.** which will lead to the increase in the cost hence this method is little expensive, also the optimum pH was not determined for this method. The least effective adsorbent was leaf extract of Moringa oleifera 1) which reduced concentration of fluoride from 3ppm to 0.015 ppm but after 3 hours fluoride level was again increased hence this method was considered the least effective to removal fluoride. So, the best, most effective and cheap method was found to be the dried Moringa oleifera seeds powder **Error! Reference source not found.**non-flow batch method where the adsorbent of dried seed powder gives the best result to combat the fluoride in water.

For Tamarindus indica, Tamarind seed powder gave the best result at neutral pH with the fluoride removal efficiency of 92.1% when the adsorbent particle size was 75 micron, this method 19) is better than other methods because it workers at neutral pH hence it is very cost effective and reduce the fluoride in water by good efficiency compare to the adsorbent of Tamarind fruit shell powder 2)which gave the fluoride removal efficiency of 50% and the optimum pH was not determined for this method 2). The acid and alkali treated adsorbent of the Tamarind powder 28) gave removal efficiency of 37% and 51% respectively. But this method cannot be considered as cost effective method because the it require the extreme pH for both acid and alkali which is pH 1 and pH 10 which leads to the increase in the cost and also the fluoride removal efficiency is not good when compared to the method 2)19).The least effective method was column adsorption method where adsorbent was prepared by Tamarind seeds powder which was treated with 4N KOH, and it gave the fluoride removal efficiency of 3.14% only. So, the best and cost-effective method was Tamarind seeds powder 19) where no treatment with acid or alkali was done.

When the best method of Moringa oleifera and Tamarindus indica was compared, the better results was shown by dried Moringa oleifera seeds powder **Error! Reference source not found.**with the fluoride removal efficiency of 89% at 6 pH and with the adsorbent dose of 1g/l also when the optimum pH and dose of adsorbent was provided the fluoride removal efficiency was increased by 4%, so the total of 93% removal efficiency was given by this method which better than the fluoride removal by Tamarind seeds powder 19) where 1g/l of the adsorbent was used for the batch study and 5g/l of the adsorbent was used for the column study and gave the fluoride removal efficiency of 92.1% at pH 7. But the removal efficiency decreased to 38.4% when the size of adsorbent particle was increased from 75 micron to 300 microns, so a very specific particle size is required to attain high fluoride removal efficiency and the advantage of this method was that it requires neural pH hence reduce the work of obtaining and maintain the optimum pH.

CONCLUSION

Fluoride is one of the minerals which is required by our body, it is necessary for our bones and tooths as it prevents fractures of bones and tooth cavities, it is also being linked as a prevention to osteoporosis. But the excessive intake of fluoride leads to fluorosis hence level of fluoride needs to be check in water. There are many under developing and developing countries which are facing the issue of high level of fluoride in drinking water and at the same time they cannot afford expensive process to remove the fluoride from water. So, the cost-effective ways are being developed for defluoridation such as use of Moringa oleifera and Tamarindus indica. And it was concluded that both Moringa oleifera and Tamarindus indica work efficiently to remove fluoride from water with the efficiency of 93% and 92.1% respectively. Thus, these are the cost effective and efficient ways to remove fluoride from water which can be easily adapted by under developing and developing countries.

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