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Studies On The De-Fluoridation Efficacy Of Chrysopogon Zizaniodides, Ocimum Tenuiflorum And Their Combinational Use

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Abstract: Fluoride in drinking water can be detrimental to health and causes many health problems to human beings. Excess of fluoride in drinking water has become a threat in recent days due to the lesser availability of potable groundwater resource especially in rural areas. Hence there is a pressing demand for the solution of fluoride contamination using natural bioadsorbants. In the present study, a cost effective method of de-fluoridation using two medicinal plant species Chrysopogon zizaniodides and Ocimum tenuiflorum have been investigated which are cost effective bio adsorbents. The investigation was carried out on the adsorption of fluoride ion to analyze the effect of various parameters like contact time, adsorbent dose, particle size and the combination of the plants as adsorbents. The Hydrophonic study revealed that the plants of Chrysopogon zizaniodides reduced 3.5 mg L^{-1} of fluoride after treatment when compared to 1.2 mg L^{-1} reductions by Ocimum tenuiflourum. The combinations of Chrysopogon zizaniodides: Ocimum tenuiflorum in the ratio of 2:1 revealed the maximum defluoridation efficiency ranging from 65 to 89 % correspondingly from 1 to 5mg L^{-1} fluoride concentration. The maximum percentage removal of fluoride ion occurs at 24 h contact time and with least particle size (1mm). The defluoridation increases with the increases in the adsorbent dose due to increase in surface area. Thus, a cost effective method of de-fluoridation using Chrysopogon zizaniodides and Ocimum tenuiflorum will be much beneficial especially in rural areas.

Key words: Chrysopogon zizaniodides, Ocimum tenuiflourum, hydroponics, defluoridation.

1. Introduction

Fluoride pollution of groundwater in lesser extent surface water, mostly from geogenic sources is a global concern. Excess of fluoride in groundwater has become a threat in recent days due to the lesser availability of potable groundwater resource especially in rural areas (Chidambaram et al. 2013). It is often described as a double-edged sword as inadequate ingestion, less than or equal to 0.5 mg L^{-1} in drinking waters, which could be associated with dental caries whereas excessive intake, greater than 1.5 mg L^{-1}, leads to dental, skeletal and soft tissue fluorosis. High or low concentrations of fluoride can occur depending on the nature of the rocks and the occurrence of fluoride bearing minerals [1, 2]. Fluoride could be common in groundwater, which calcium are found to be poor in purification. Fluoride pollution in the environment occurs through two different channels which are natural and anthropogenic sources [1, 3]. During weathering of alkali, igneous and sedimentary rocks and circulation of water in fluoride bearing rocks and soils, fluorine can be reached out and dissolved in groundwater [4, 5]. A large number of fluoride contaminated areas are found throughout the world where ground waters contain exceeded levels of fluoride. In the worldwide, India is most severely fluoride affected country. Of the 85 million tons of fluoride deposits in the earth's crust, 12 million are found in India [6]. In India in 2002, 17 states have been identified as endemic for fluorosis [7] and now the problem exists in more than 17 states [1]. In India, concentrations of fluoride in drinking water in different parts of the country vary from 0.5 to 50 mg L-1it and the extent of fluoride contamination in groundwater varies from 1.0 to 48 mg L^{-1} [8]. In India alone, a total of 60-70 million people including children are affected with dental and skeletal fluorosis [9]. Several areas of composite state of Andhra Pradesh have fluoride concentration greater than the permissible limit by WHO [10]. Defluoridation of drinking water is the only solution to sort out the problems of excessive fluoride in drinking water. Various defluoridation technologies have been developed to mitigate excess fluoride from drinking water before consumption. Defluoridation can be carried out by reverse osmosis, ion exchange, precipitation-coagulation [11], membrane separation process [12], electrolytic defluoridation, electro dialysis and adsorption. Most of these techniques are relatively expensive than adsorption [13]. Adsorption is an efficient process to defluoridate water, in which contaminated water is passed through an adsorbent bed where fluoride is removed through ion exchange/surface exchange with the adsorbent. Adsorption mechanism using waste plant material (biosorbents) takes place either by ion exchange or surface complex formation. Besides being ecofriendly, absorption also has of being cost effective, reduces sludge disposal and high efficiency in dilute effluents. The literature survey shows that plant materials accumulate fluoride and therefore it is used for defluoridation of water [14, 15, and 16]. Recently, researchers have devoted their study on different types of low-cost but effective materials, clay [17], brick powder [18], cotton cellulose [19], spent bleaching earth [20], Rice straw [21], activated alumina [22]; zeolites [23], red mud [3] etc. Some of the inexpensive defluoridation techniques developed to control fluoride content in water are reverse osmosis, adsorption using sunflower plant dry powder, steam of phytomass, Holly Oke, neem bark powder, activated cotton, jute carbon, bagasse ash, burnt bone powder, phosphate-treated saw dust, bone char, etc. as adsorbents, Nalgonda technique, activated alumina process and ion exchange process. However, due to high cost or lower efficiency or nonapplicability on mass scale these techniques are not much in use [24]. In the present study, a cost effective method of de-fluoridation using two medicinal plant species Chrysopogon zizaniodides and Ocimum tenuiflorum have been investigated which are cost effective and natural adsorbents. The study also deals on the adsorption of fluoride ion to analyze the effect of various parameters like contact time, adsorbent dose, particle size and the combination of the plants as adsorbents.

2. Materials and Methods

2.1. Collection of plant materials

Twelve healthy plants each (Ocimum tenuiflorum and Chrysopogon zizaniodides) of the same height and dimensions were procured from medicinal plant Nursery, Agastya International Foundation, Kuppam, Andhra Pradesh. The roots of Chrysopogon zizaniodides were collected from live plants and shade dried, and fresh Ocimum tenuiflorum leaves were collected 15 min before experimentation (Figure S1).

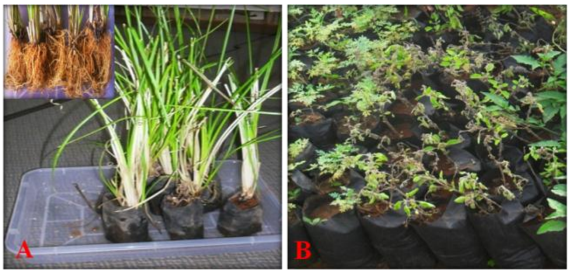

Figure S1. *Plants used for the defluoridation assay; A. Chrysopogon zizaniodides, B. Ocimum tenuiflorum*

2.2 Preparation of Fluoride standards

The Standard fluoride solutions were prepared using Sodium fluoride (NaF) in distilled water. Different concentrations of fluoride standards were prepared as 1.0, 2.0, 3.0, 4.0 and 5.0 mg L^{-1} fluoride.

2.3 Hydroponic system of defluoridation

The roots of Chrysopogon zizaniodides were washed and cleaned using distilled water to remove any trace elements attached to it. They were adapted to the hydroponic system in specially designed trays, before subjecting to actual experimentation (Figure S2). Plants were subjected to 5 mg L^{-1} fluoride water for 24 h in the trays. Water (5 mg L^{-1} fluoride solution) in the trash without the plants was served as control. The treated water sample after incubation period was collected for lab for water analysis to study the concentration of fluoride degraded in treated as well as control water.

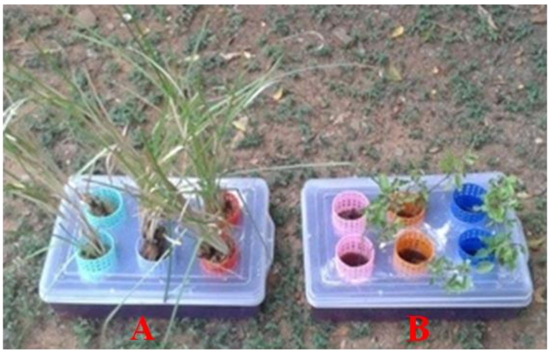

Figure S2. *Hyrophonic System for the determination of defluoridation efficiency of Chrysopogon zizaniodides (A) and Ocimum tenuiflorum (B).*

2.4 Defluoridation using Chrysopogon zizaniodides and Ocimum tenuiflourum plants as adsorbents

Different concentrations of fluoride solution (1mg L^{-1}, 2mg L^{-1}, 3mg L^{-1}, 4mg L^{-1} and 5mg L^{-1}) were collected in 5 beakers. Chrysopogon zizaniodides roots (1g) were placed in a beaker of respective fluoride concentrated water. Similarly 5g fresh Ocimum tenuiflorum leaves were placed in beakers of respective fluoride concentrated water. These adsorbents were subjected to contact time of 24 h and 48 h. Water (5 mg L^{-1} fluoride solution) in the trays without the adsorbents was served as control. Water samples treated after respective time periods were collected for the concentration of fluoride degraded in treated as well as control water.

2.5 De-fluoridation of water using a combination of adsorbents - Chrysopogon zizaniodides roots and Ocimum tenuiflorum fresh leaves

Experiments were conducted to investigate the defluoridation efficiency of the different combinations of Chrysopogon zizaniodides roots and Ocimum tenuiflorum fresh leaves (1:1, 2:1 and 1:2). Different concentrations of fluoride solution (1mg L^{-1}, 2mg L^{-1}, 3mg L^{-1}, 4mg $L^{-}1$ and 5mg L^{-1}) were collected in 5 beakers. The combination of Chrysopogon zizaniodides roots (1 g) and fresh Ocimum tenuiflorum leaves (5g) were placed in beakers of respective fluoride concentrated water. These adsorbents were subjected to contact time of 24 h and 48 h. Water (5 mg L^{-1} fluoride solution) in the trays without the adsorbents was served as control. Water samples treated after respective time periods were collected for the concentration of fluoride degraded in treated as well as control water.

2.6 Determination of fluoride

All the samples were subjected to water analysis carried out at a water testing lab in Kuppam, Andhra Pradesh. The analysis was carried out by adding SPADNS Fluoride reagent solution (Sigma Aldrich) and quantified using a portable colorimeter (DR/850, Preiser Scientific, China) (Figure S3)with a detectable range of 0.0 -2.5mg L^{-1} concentration of fluoride solution. Percentage of heavy metal removal (%) = [(Fc – Ft) /Fc] × 100 Here, Fc- initial fluoride concentration of test solution, mg L^{-1}; Ft was a final concentration of test solution mg L^{-1}.

Figure S3. *DR-850 Colorimeter- Instrument used for the quantification of fluoride in treate*

2.7 Effect of adsorbent contact time of De-fluoridation of water

Adsorbent was added in 100 ml of the fluoride solution of known concentration 1mg L^{-1}, 2mg L^{-1}, 3mg L^{-1}, 4mg L^{-1} and 5mg L^{-1} in each flask. The absorbent suspensions in 100 ml test solution were taken in each conical flask at different time interval 12, 24 and 48 h. The fluoride removal percentage was determined using the following equation: Percentage of Fluoride removal (%) = [(Fc – Ft) /Fc] × 100 Here, Fc- initial fluoride concentration of test solution, mg L^{-1}; Ft was a final concentration of test solution mg L^{-1}.

2.8 Effect of adsorbent particle size of De-fluoridation of water

Experiments were conducted to evaluate the influence of adsorbent particle size for a constant weight on the removal of fluoride ions. Particle size analysis was conducted on treating bioadsorbents and the percentage composition of particle size was investigated. The defluoridation experiments were conducted using Chrysopogon zizaniodides roots with three different particle sizes viz. 1mm, 1.4mm and 2mm. Percentage of Fluoride removal (%) = [(Fc – Ft) /Fc] × 100 Here, Fc- initial fluoride concentration of test solution, mg L^{-1}; Ft was a final concentration of test solution mg L^{-1}.

2.9 Effect of adsorbent dosage on defluoridation efficiency

The defluoridation efficacy of the adsorbent based on the dosage was determined by using a series of adsorbent dosages viz; 0.5g L^{-1}, 1g L^{-1}, 3g L^{-1}, 5g L^{-1},7gL^{-1},9g L^{-1} and 10g L^{-1} while the initial fluoride concentration was fixed at 5mg L^{-1}. The fluoride removal percentage was determined using the following equation: Percentage of Fluoride removal (%) = [(Fc – Ft) /Fc] × 100 Here, Fc- initial fluoride concentration of test solution, mg L^{-1}; Ft was a final concentration of test solution mg L^{-1}.

3. RESULTS AND DISCUSSION

Successful application of the adsorption technique demands innovation of cheap, nontoxic, easily and locally available material. Bio adsorbents meet these requirements. Knowledge of the optimal conditions would herald a better design and modeling process. Thus, the effect of some major parameters like pH, contact time, and particle size of adsorbent and materials was investigated from kinetic perspective. Adsorption studies were performed by a batch technique to obtain the rate and equilibrium data. The removal of fluoride from drinking water by using two medicinal plant species Chrysopogon zizaniodides and Ocimum tenuiflorum have been investigated which are cost effective and natural adsorbents and determined by various parameters such as contact time, adsorbent dose, particle size etc. which were represented graphically.

3.1 Efficacy of Chrysopogon zizaniodides and Ocimum tenuiflourum plants as adsorbents

Hydroponics system of defluoridation technique was used to study the efficacy of Chrysopogon zizaniodides and Ocimum tenuiflourum plants as bio-adsorbents. The samples treated with the plants of Chrysopogon zizaniodides and Ocimum tenuiflourum exhibited a varied degree of defluoridation concentration. The plants of Chrysopogon zizaniodides reduced 3.5 mg L^{-1} of fluoride after treatment when compared to 1.2 mg L^{-1} reductions by Ocimum tenuiflourum (Figure 1). It is evident that, the efficacy of defluoridation was high in the treatment by Chrysopogon zizaniodides indicates the significant capability of using this plant as a bio-adsorbent. Studies have been carried out on phytoremediation of Chrysopogon zizaniodides (Vetiveria zizanioides) to use as efficient bio-adsorbents to remove chromium [25], lead [26], tetracycline from aqueous media [27]. To the best of our knowledge, this is the first report on the use of hydroponics to study the defluoridation efficacy of Chrysopogon zizaniodides and Ocimum tenuiflourum in India.

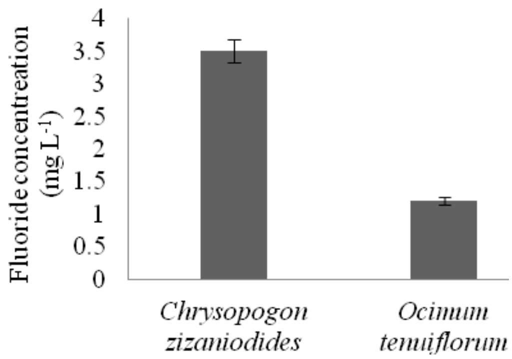

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Figure 1. *De-fluoridation of water using Chrysopogon zizaniodides and Ocimum tenuiflourum plants as adsorbents*

3.2 Defluoridation using the roots of Chrysopogon zizaniodides and the leaves of Ocimum tenuiflourum as adsorbents

Experiments conducted to evaluate the efficacy of the roots of Chrysopogon zizaniodides and the leaves of Ocimum tenuiflourum to use as defluoridating agents. The results revealed that treatment of water with the roots of Chrysopogon zizaniodides exhibited a noteworthy diminution of fluoride after 24 and 48 h of treatment. The reduction percentage of different fluoride concentrations are represented in Figure 2. The percentage of adsorption has no significant change after 24 h incubation. The reduction percentages of fluoride concentrations were 70, 65, 58, 52 and 43%, corresponding to 1 mg L^{-1}, 2mg L^{-1}, 3 mg L^{-1}, 4 mg L-1 and 5 mg L^{-1} respectively after 24 h treatment. The roots of Chrysopogon zizaniodides (Vetiveria zizanioides) are efficient bio-adsorbents that aid in the remediation of fluoride polluted water as evident in the study by Harikumar et al. [13], revealed the highest defluoridation percentage when compared to other bio-adsorbents support the present study. In another study by Guptha et al. [28], highlighted the use of Chrysopogon zizaniodides (Vetiveria zizanioides) for the treatment of water due to its efficiency and adaptability to several ecological factors and it is very efficient when compared to other adsorbents.

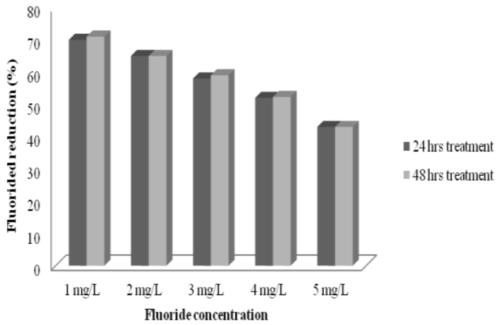

Figure 2. *De-fluoridation of water using the roots of Chrysopogon zizaniodides as an adsorbent*

On the other hand, the defluoridation efficacy of the leaves of Ocimum tenuiflourum revealed the varied percentage reduction of fluoride in different concentrations. The percentage reduction of fluoride after 24 h treatment were 68, 65, 49, 43 and 38 %, corresponding to 1 mg L^{-1}, 2mg L^{-1}, 3 mg L^{-1}, 4 mg L^{-1} and 5 mg L^{-1} respectively (Figure 3). In this case, the result may be due to hindrance at the surface of the treated biosorbents by higher fluoride concentrations. The importance of Ocimum tenuiflourum as water cleanser and use in defluoridation has been investigated by many researchers. A study by Sudheer and Ahmed, [29] revealed that more than 50 % of the fluoride has been effectively removed from the fresh leaves of Ocimum tenuiflourum. In addition, Bhattacharjee, [30] used the leaves of Ocimum tenuiflourum in reduction of fluoride content in water. He also confirmed that, as the concentration increases, the defluoridation efficiency increases. Thus, all the studies confirmed the defluoridation ability of Ocimum tenuiflourum leaves supporting the present investigation.

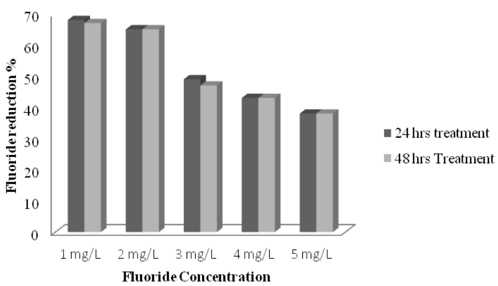

Figure 3. *De-fluoridation of water using Ocimum tenuiflorum fresh leaves as an adsorbent*

3.3 De-fluoridation of water using a combination of adsorbents - Chrysopogon zizaniodides roots and Ocimum tenuiflorum fresh leaves

Adsorption efficiency of fluoride concentration by using different combinations of Chrysopogon zizaniodides: Ocimum tenuiflorum (1:1, 2:1 and 1:2) have been investigated. The results showed that the combinations of Chrysopogon zizaniodides: Ocimum tenuiflorum in the ratio of 2:1 revealed the maximum defluoridation efficiency ranging from 65 to 89 % correspondingly from 1 to 5 mg L^{-1} fluoride concentration. The defluoridation efficiency ranged from 58 to 75 % since 1:1 ratio of combinations of Chrysopogon zizaniodides: Ocimum tenuiflorum. The least defluoridation percentage was observed with the 1:2 combinations of Chrysopogon zizaniodides: Ocimum tenuiflorum with efficiency ranging from 43 to 67% (Figure 4). The maximum defluoridation efficiency may be due to the adsorption contribution by Chrysopogon zizaniodides when compared to Ocimum tenuiflorum. Chrysopogon zizaniodides (Vetiver) root is found to be an efficient defluoridating agent, the sorption process of fluoride ion on activated Chrysopogon zizaniodides (Vetiver) root was found to be influenced by temperature, pH, agitation time and solvent dosage [14]. In another study, by Harikumar et al. [13] the absorption of fluoride was confirmed by scanning electron microscopy (SEM) micrographs of activated Chrysopogon zizaniodides (Vetiver) adsorbent after fluoride loading is evident and supports the present study.

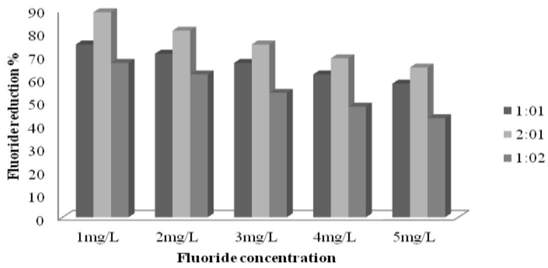

Figure 4. *De-fluoridation of water using a combination of adsorbents - Chrysopogon zizaniodides roots: Ocimum tenuiflorum fresh leaves*

3.4 Effect of adsorbent contact time of De-fluoridation of water

Adsorption of fluoride ion by using 2:1 combination of Chrysopogon zizaniodides: Ocimum tenuiflorum at different contact times 12, 24, and 48 h are graphically presented in Figure 5. It is found that during the experiment the removal of fluoride ion percentage increased up to 85 % with a contact time till 24 h after that it reached at a constant value. The result shows maximum percentage removal of fluoride ion occurs at 24 h contact time and 10 mg L^{-1} initial concentration. Our findings were significant when compared with the results of the previous studies by Harikumar et al. [13] who reported maximum defluoridation of 54 % using the roots of Chrysopogon zizaniodides.

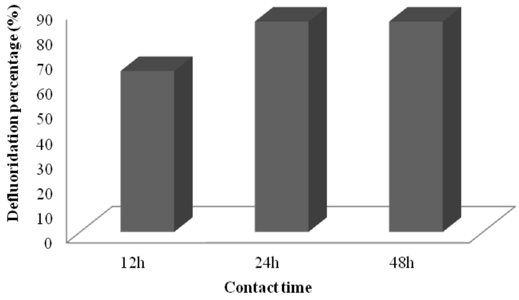

Figure 5. *The effect of adsorbent (combination of adsorbents - Chrysopogon zizaniodides roots: Ocimum tenuiflorum fresh leaves, 2:1) contact time on De-fluoridation of water*

The changes in the strength of adsorption might be due to the fact that initially all the adsorbent sites were open and the solute concentration was relatively high. Later, the fluoride uptake by the adsorbent decreased significantly, due to decrease in the active sites. It indicates that the possible monolayer of fluoride ions on the outer surface pores of the adsorbent and pore diffusion onto the inner surface of adsorbent particles through the film due to continuous mixing maintained during the experiment. The combination of Chrysopogon zizaniodides: Ocimum tenuiflorum has a significant effect on the defluoridation efficiency.

3.5 Effect of adsorbent particle size of De-fluoridation of water

Experiments were conducted to evaluate the influence of adsorbent particle size (2, 1.4 and 1mm) for a constant weight on the removal of fluoride in water. Particle size analysis was conducted on treating bio-adsorbents and the percentage composition of particle size was determined. The results obtained with the variation of adsorbent particle size and the percent of the fluoride ions are graphically represented in Figure. 6.

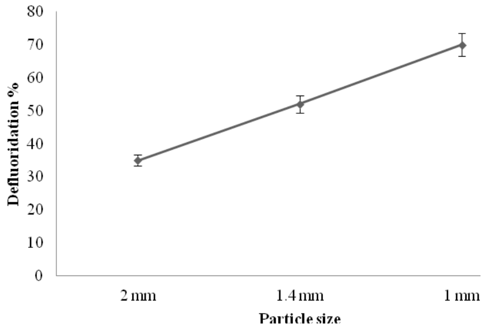

Figure 6. *Effect of adsorbent particle size of De- fluoridation of water*

The uptake of fluoride ions at different adsorbent particle sizes increased with decrease in Sorbent particle diameter. Maximum defluoridation activity (70%) was observed with least particle size (1mm) in contrast with the minimum defluoridation (35%) with 2mm adsorbent size. The presence of a large number of smaller particles affords the sorption system with a larger surface area available for fluoride ion removal and it also reduces the external mass transfer resistance. Also, the time required for 50% of the total absorption is less with the particles of smaller size. This also gives some idea of the rate-limiting step of the adsorption process. The removal of fluoride ions has been studied at a room temperature of 27 ± 0.5 °C. Small particle size provides more active surface area and hence such results were observed. Our results are in correlation with the results of Goswami et al. [31] who reported the maximum defluoridation efficiency with smaller adsorbent size.

3.6 Effect of adsorbent dosage on defluoridation efficiency

The results in adaptive removal of fluoride ion with respect to adsorbent dose are shown in Figure 5. Studies on effect of adsorbent doses are conducted by varying adsorbent doses between 0.5 g to 10 g L^{-1}. The initial fluoride ion concentration is fixed at 10mg L^{-1} and contact time is kept as 24 h. Increase the adsorbent dose the removal efficiency will also increase due to increase in surface area, there is more active sites are available for the absorption of fluoride. The assay revealed maximum 94 % percentage removal of fluoride ion occurs at 10mg L-1 concentrations, followed by 90 % defluoridation at 9mg L^{-1} fluoride concentration (Figure 7). The adsorbent dose has a significant role in the defluoridation efficiency. Recently, Panchor et al. [32] used brick powder as an adsorbent and found that as the dosage increased, the defluoridation efficiency increased relatively.

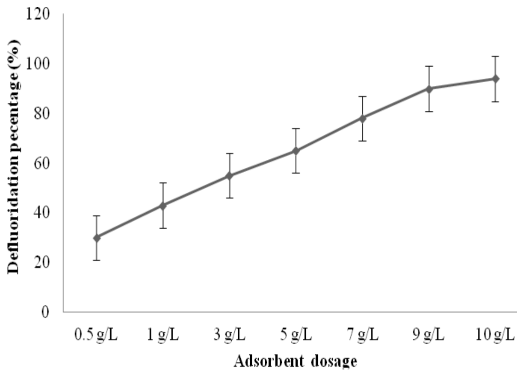

Figure 7. *Effect of adsorbent dosage on De-fluoridation of water*

4. CONCLUSION

The results demonstrate that the combination of Chrysopogon zizaniodides and Ocimum tenuiflorum has exhibited economical and effective adsorbents in removing fluoride from water to acceptable levels. Results also showed that these low-cost bio adsorbent could be fruitfully used for the removal of fluoride over a wide range of concentrations. The percentage of fluoride removal was found to be a function of adsorbent particle size and time at a given initial fluoride concentration. It increased with time, and higher initial solute concentration decreased with time. Treated basements are locally available and hence involve no expenditure on transportation and have a very low cost for pretreatment. There is no need to regenerate the exhausted treated basements as they are available abundant, easily, cheaply and locally. The experimental investigations by using low cost adsorbents for defluoridation would be useful in developing countries and also useful in rural areas for domestic purpose.

5. ACKNOWLEDGEMENTS
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