DEFLUORIDATION OF WATER BY COMPOSITE BED OF LOW-COST BIO-ADSORBENTS

Vinish V Nair[1], Anandu Aravind[2], Dayal Kurian Varghese[3]

[1]Associate Professor, Department of Civil Engineering,
Govt. Rajiv Gandhi Institute of Technology, Kottayam, Kerala, (India)
[2,3] Department of Civil Engineering,
Govt. Rajiv Gandhi Institute of Technology, Kottayam, Kerala, (India)

ABSTRACT

Drinking water contamination by fluoride is recognized as a major public health problem in many parts of the world. In fact, although fluoride is an essential trace element for animals and humans, excessive fluoride intake may cause adverse health effects. Present study has been conducted to investigate the fluoride removal efficiency of composite bed of bio-adsorbents such as Moringa Oleifera (MO), Rice husk Activated Charcoal (RHAC) using an experimental column study. With a constant initial F concentration, it was found that increasing the MO dose in composite bed resulted in increased fluoride removal. Prepared composite bed - 4cm thick consisting of 25% MO and 75% RHAC- shows an enhanced removal of fluoride by 90% at an equilibrium contact time of 30 minutes. The designed composite bed system provides a solution to the problem of high turbidity occurring in a single bed MO treatment system.

Keywords: Moringa Oleifera, Rice Husk Activated Charcoal, Bio-Adsorbents, Composite Bed.

I. INTRODUCTION

Water is most abundant and is an essential component of our life supporting system. In India, drinking water is polluted at many places by various pollutants such as fluorides, nitrates, iron etc. Fluoride is often considered as a “two edged sword” because deficiency of fluoride in drinking water leads to dental caries while excess consumption leads to dental and skeletal fluorosis (P. Gopalakrishnan et al, 1999). Fluoride is more toxic than lead, and just like lead, even in minute doses, accumulates in and is damaging to brain/mind development of children, i.e. produces abnormal behavior in animals and reduces IQ in humans (A.S. Parlikar et al, 2013).Workers exposed to high fluoride concentration areas are diagnosed with bladder cancer (Islam M et al, 2013). Various diseases such asosteoporosis, arthritis, brittle bones, cancer, infertility, brain damage, Alzheimer syndrome, and thyroid disorder can attack human body on excessive intake of fluoride.

Because of high toxicity of fluoride to mankind, there is an urgent need to treat fluoride contaminated drinking water to make it safe for human use. Health problems due to high fluoride content in ground water have also been reported worldwide and it is estimated that around 260 million people are adversely affected in 30 countries of the world (Neelo Razbe et al, 2013). According to World Health Organization norms, the upper limit of fluoride concentration in drinking water is 1.5 mg/l (WHO, 2008). The Bureau of Indian Standard which is the main regulatory agency for drinking water in India specifies that the desirable limit of fluoride in drinking water is 0.6mg/l to 1 mg/L. In Kerala, the condition of fluoride is reported to be endemic in the districts of Alappuzha and Palakkad (Tomas Blom et al, 2009; P. Gopalakrishnan et al, 1999).

The conventional method of fluoride removal includes: ion-exchange, reverse osmosis and adsorption. The ion-exchange, reverse osmosis are relatively expensive. Therefore, still adsorption is the viable method for the removal of fluoride. Defluoridation of drinking waters is usually accomplished by either precipitation or by adsorption processes. One of the well-known methods called 'Nalgonda Technique' was developed by National Environmental Engineering Research Institute; Nagpur, India (Bulusu et al, 1988) is precipitation processes employing alum followed by sedimentation and/or filtration. Disadvantage of this technique is that treated water has high residual aluminium concentration (2–7 mg/L) than the WHO standard of 0.2 mg/L (Vaishali Tomar et al, 2013). Alum coagulant can be used to remove fluoride selectively from aqueous solutions. Even at 60-70% removal, the left over fluorides were within the permissible limit for drinking water (V. Subhashini et al, 2012).Nowadays, biosorption method is very effect/attractive technique for removal of fluoride from water. This technique involves the low cost adsorbents (also called biosorbents) such as rice husk, saw dust, moringa olifera extract, red mud and goose berry etc.

Due to rich biodiversity of India, a large number of plant spices are available for treatment of various toxicities and musculoskeletal disorders. Research reports indicate that herbal and low cost plant products may be used for mitigation of fluoride toxicity (Stanley V A et al, 2000). Rice husk contains abundant floristic fiber, protein and some functional groups such as carboxyl, hydroxy and amidogen, etc. which makes adsorption processes possible(Runping Han, et al).Moringa oleifera consists chemical compounds like 4-(4'-O-acetyl-a-L rhamnopyranosyloxy)benzyl isothiocyanate, 4-(a-L-rhamnopyranosyloxy)benzyl isothiocyanate, niazimicin, pterygospermin, benzyl isothiocyanate, and 4-(a-L-rhamnopyranosyloxy)benzyl glucosinolate and several studies reported on the performance of Moringa oleifera seeds as a good adsorbent (Jed W). The objective of the present study is to investigate the effectiveness of naturally occurring and low-cost materials like Rice Husk and Moringa olifera for removal of Fluorides from water.

The efficiency of removal of fluoride by using various locally available natural plants were already investigated (Jain J K et al, 2013, Ravikumar K A et al, 2014) .Adsorption with Moringa oliefera seed powder reduced the fluoride concentration of fluoridated water below 1mg/L but the turbidity after adsorption was very high (>5mg/l) (Ravikumar K A et al, 2014). This study is an attempt to remove the fluoride from water by using composite bed of bio-adsorbents. In the present study, the efficiency of removal of fluoride by using composite bed of bio-adsorbents like MO, RHA was investigated. The study assesses the suitability of composite bed of in-expensive bio-adsorbents to effectively remediate fluoride contaminated water. The study investigated defluoridation capacities of the composite bed of bio-adsorbents like Rice Husk Ash (RHA), Moringa Oleifera (MO) and their optimum proportion. Effect of important controlling factors like dose of adsorbent, contact time and initial fluoride ion concentration on the fluoride removal efficiency was studied.

II. MATERIALS AND METHODS

2.1. Materials

In this study an attempt has been made to suggest certain low cost materials as effective adsorbents of fluoride. Naturally occurring and abundantly available materials like Rice husk, Moringa Oleifera seed were used as adsorbents.

International Journal of Advanced Technology in Engineering and Science
Vol. No.4, Issue No. 02, February 2016
www.ijates.com
ijates
ISSN 2348 - 7550

2.1.1. Preparation of mo seed powder

Dry Moringa oleifera pods were collected from Varkala, Trivandrum. Pod shells were removed manually; kernels were grounded in a domestic blender and sieved through 600 micron stainless steel sieve (See Figure 2.1, Figure 2.2).

Figure 2.1 Moringa oleifera seeds

Figure 2.2 Moringa oleifera seed powder

2.1.2. Preparation of rice husk activated charcoal (RHAC)

Rice husk was obtained from local mill. It was sieved through IS 600 micron and 300 micron sieves. Then the materials retained on IS 300 micron sieve was partially carbonized in laboratory oven at 110°C for 4 to 5 hours. The partially carbonized material was then completely carbonized in muffle furnace at temperature 250°C to 300°C for 1 hour. The material from muffle furnace was cooled to room temperature. Material was then repeatedly washed with hot boiling water so as to open the pores of carbon. Completely carbonized rice husk was used for column adsorption studies (See Figure 2.3, Figure 2.4).

Figure 2.3 Rice husk (Before Activation)

Figure 2.4 Rice husk activated charcoal

2.2. Methods

The adsorption studies for fluoride removal were carried out at room temperature, in a 2 inch diameter PVC pipe column using 500 ml water sample containing initial concentrations of fluoride 10 mg/l. The adsorbent was filled in this column to a sufficient bed depth. Then the incoming water is allowed to retain in this bed for a contact periods of 30 min, 1 hr and 2 hr. At the end of each period outflow sample was collected. The contents of the each sample were filtered using Whatmann's filter paper number 41. The initial and final concentrations of aqueous solutions of fluoride were determined by spectrophotometer and percentage removal of fluoride was determined. The experiments were conducted for varying depths of adsorbent beds (3cm, 4cm, 5cm) as shown in Table 3.1.

2.2.1. Preparation of standard curve

A stock solution of 100 mg/l of fluoride was prepared by dissolving 221 mg of unhydrous sodium fluoride (NaF) in1000 ml distilled water and fluoride standard samples in the range of 0.1 mg/l to 1.4 mg/l were then prepared from stock solution. Fluoride concentration was determined, at all stages of the process, by spectrophotometer (Figure 2.5) using SPADNS reagent. This method is based on the reaction between fluoride and zirconium dye. The fluoride dissociates a part of the dye, forming a colourless anionic complex. The amount of fluoride is inversely proportional to the colour of the solution, that is, colour becomes progressively lighter as fluoride concentration increases. Pipette 5 ml each of SPADNS solution and zirconyl acid solution and 2 drops of sodium arsenite solution to each standard and mixed well. Contamination avoided. The spectrophotometer was set to zero absorbance with reference solution and absorbance readings of standard were obtained (See Table 2.1). Reference solution was used as a blank solution. Spectrophotometer used at 570 nm wavelength was taken as per standard method procedure (See Figure 2.6)

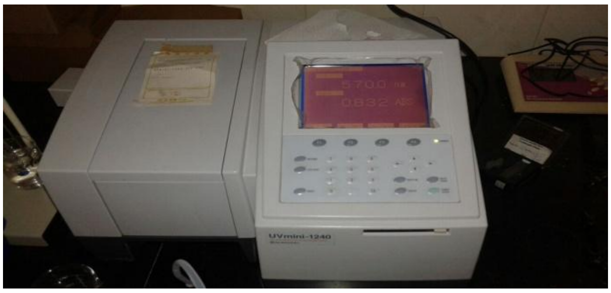

Figure 2.5 Spectrophotometer

Table 2.1 Standard curve

SL.NO	INITIAL FLUORIDE CONCENTRATION Mg/l	ABSORBANCE READING
1	Blank	0.832
2	0.1	0.804
3	0.25	0.782
4	0.5	0.783
5	1	0.769
6	1.4	0.752

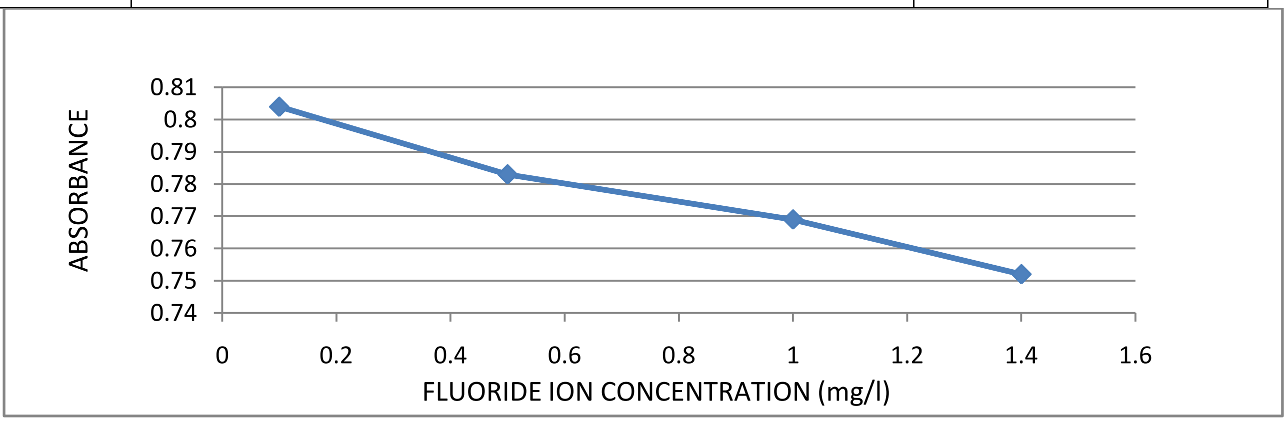

Figure 2.6 Calibration Curve

III. RESULTS AND DISCUSSION

Different bio-adsorbents were studied for testing the efficiency of fluoride removal and Moringa oleifera was found to be promising for the removal of fluoride (name et al, 2015). It was found that as the bed depth increases, the fluoride removal efficiency also increases. The effect of contact time on fluoride removal efficiency is shown in figure 3.1.The peak removal efficiency of 98.1% is obtained for a bed depth of 4cm and contact time of 30 minutes. Even though the efficiency obtained is high, the treated water is highly turbid and has some odour problems. Hence an attempt has been made to overcome the problems of turbidity and odour, a composite bed of MO and RHAC is designed and the optimum adsorbent bed depth is fixed as 4cm.

Table 3.1 2 Fluoride concentrations and fluoride removal efficiency after single bed MO treatment

Sample No	Depth of Adsorbent bed cm	Contact time (minutes)	Initial Fluoride Concentration mg/l	Final Fluoride Concentration mg/l	Fluoride Removal Efficiency %
1	3	30	10	0.26	97.4
2	3	60	10	0.41	95.9
3	3	120	10	0.65	93.5
4	4	30	10	0.19	98.1
5	4	60	10	0.35	96.5
6	4	120	10	0.64	93.6
7	5	30	10	0.17	98.3
8	5	60	10	0.33	96.7
9	5	120	10	0.62	93.8

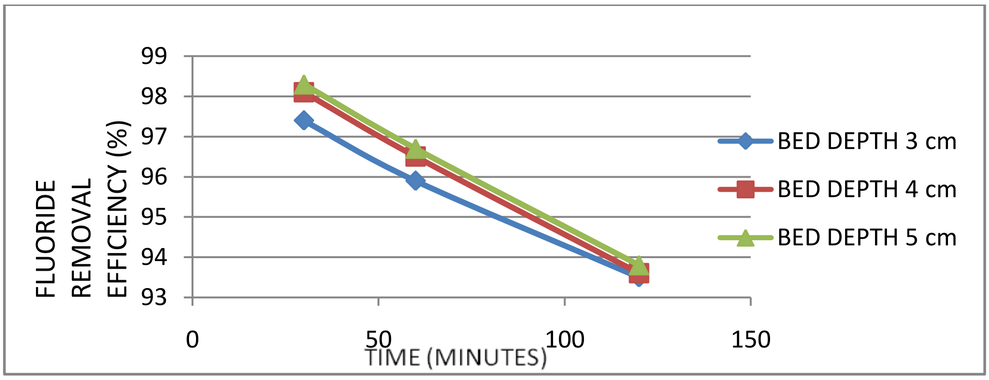

Figure 3.1 Effect of fluoride removal efficiency with contact time after treatment with MO seed powder for various adsorbent bed depths.

3.1 Study on Composite Bed of Bio-adsorbents

Composite bed of bio-adsorbent was prepared by adding RHAC and MO in various proportions that will reduce turbidity of defluoridated water after adsorption. From the adsorption study, the optimum depth of composite bed is fixed as 4 cm. Composite bed study was conducted in different trials with various proportions of MO and RHAC (Trial I- 15% MO and 85% RHAC; Trial II -25% MO and 75% RHAC; Trial III - 35% MO and 65% RHAC) in 4cm thick bed and the results are shown below.

3.1.1. Composite Adsorbent Bed (15% MO AND 85% RHAC)

The fluoride concentration and removal efficiency in all the five test samples of initial fluoride concentration 10mg/l after adsorption with composite bed of bio-adsobents (15% MO, 85% RHAC) are shown in Table 3.2.

Table 3.2 Fluoride concentration, fluoride removal efficiency and turbidity after composite bed (15% MO & 85% RHAC) treatment

Sample No	Contact time (minutes)	Initial Fluoride Concentration mg/l	Final Fluoride Concentration mg/l	Fluoride Removal Efficiency %	Turbidity NTU
1	10	10	3.6	64	3
2	20	10	3.45	65.5	3
3	30	10	2.75	72.5	4
4	40	10	3.3	67	5
5	50	10	3.65	63.5	6

Five samples were analyzed for different contact times; 10 minutes, 20 minutes, 30 minutes. It was found out that the turbidity of composite bed treated water is much lower than single bed MO treated water. The maximum fluoride removal efficiency is obtained for an optimum time of 30 minutes as shown in figure 3.2. Based on an equilibrium condition, for 72.5% fluoride removal, a contact time of 30 minutes is required and the results are presented in the figure 3.2.

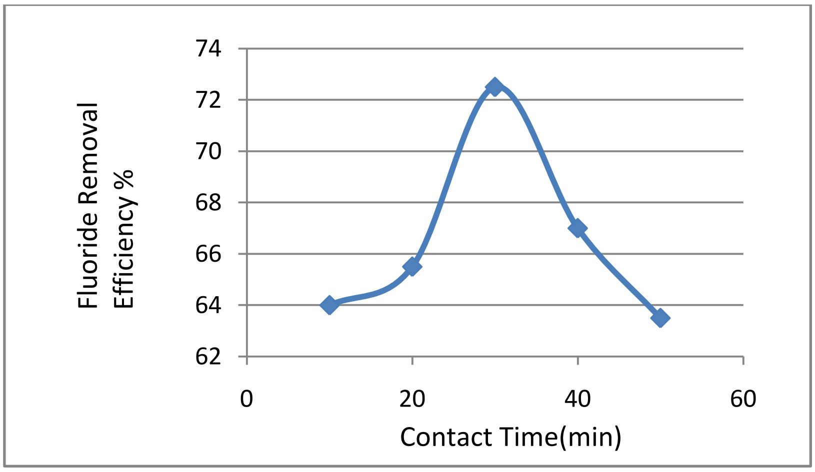

Figure 3.2 Effect of contact time on fluoride removal for composite bed of 85% RHAC & 15% MO.

3.1.2. Composite Adsorbent Bed (35% MO AND 65% RHAC)

The fluoride concentration and removal efficiency in all the five water samples of initial fluoride concentration 10mg/l after adsorption with composite bed of bio-adsorbents (35% MO, 65% RHAC) are shown in Table 3.3.

Table 3.3 Fluoride concentration, fluoride removal efficiency and turbidity of composite bed (35% MO & 65% RHAC) .

Sample No	Contact time (minutes)	Initial Fluoride Concentration mg/l	Final Fluoride Concentration mg/l	Fluoride Removal Efficiency %	Turbidity NTU
1	10	10	1.3	87	12
2	20	10	1.25	87.5	13
3	30	10	0.95	90.5	15
4	40	10	1.2	88	16
5	50	10	1.45	88.5	19

The maximum fluoride removal efficiency (90.5 %) is obtained for an optimum time of 30 minutes. However, the turbidity of treated water is found to be much higher than the standard limit of drinking water (> 5 NTU). This may be due to the high proportion of MO in the composite bed. Based on an equilibrium condition, for 90.5% fluoride removal of 10mg/l of fluoride, a contact time of 30 minutes is required and the results are illustrated in the figure 3.3.

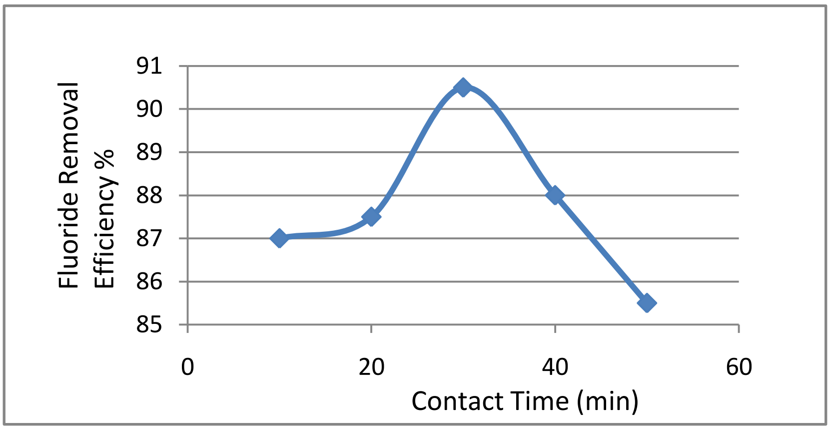

Figure 3.3 Effect of contact time on fluoride removal for composite bed of 65% RHAC & 35% MO

3.1.3. Composite Adsorbent Bed (25% MO AND 75% RHAC)

The fluoride concentration and removal efficiency in all the five water samples of initial fluoride concentration 10mg/l after adsorption with composite bed of bio-adsorbents (25% MO, 75% RHAC) are shown in Table 3.4.

Table 3.4. Fluoride concentration, fluoride removal efficiency and turbidity of composite bed (25% MO & 75% RHAC) .

Sample No	Contact time (minutes)	Initial Fluoride Concentration mg/l	Final Fluoride Concentration mg/l	Fluoride Removal Efficiency %	Turbidity NTU
1	10	10	1.7	83	2
2	20	10	1.52	84.8	2
3	30	10	1	90	3
4	40	10	1.4	86	3
5	50	10	1.65	83.5	4

In the present study (25% MO and 75% RHAC), the fluoride concentration falls below 1 mg/l (90% fluoride removal) and the turbidity is within the standard limit of drinking water (< 5 NTU). The maximum fluoride removal efficiency is obtained for an optimum time of 30 minutes as shown in the figure 3.4.The composite bed of proportion 25% MO and 75% RHAC is found to be more efficient in the de-fluoridation of drinking water. So this composite bed helps to remove fluoride effectively without any significant increase in turbidity of the de-fluoridated water

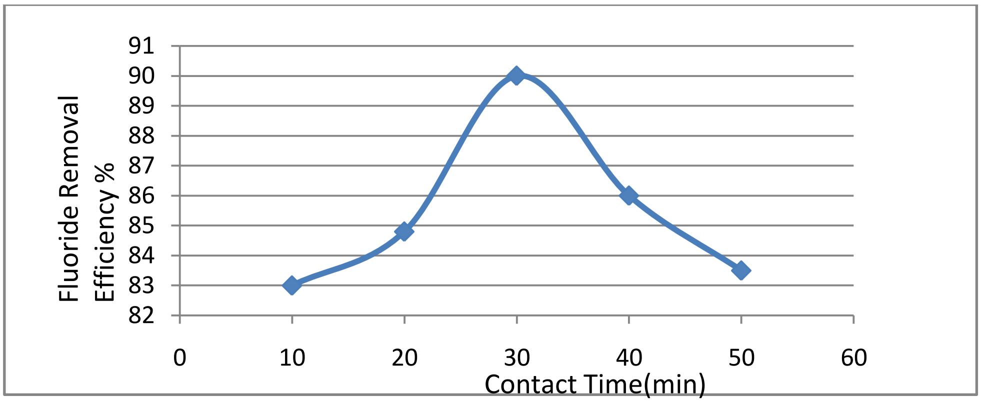

Figure 3.4 Effect of contact time on fluoride removal for composite bed of 75% RHAC & 25% MO

IV. CONCLUSION

An experimental column study was conducted to investigate the fluoride removal efficiency of composite bed of bio-adsorbents consisting of MO and RHAC. Prepared MO adsorbent shows enhanced removal of fluoride by 98.1% at equilibrium contact time of 30 minutes. However the turbidity of treated water is found to be very high .The fluoride removal efficiency increases with increase in MO dose.

Experimental column study was also conducted for different composite beds of bio-adsorbents. It was found that the composite bed consisting of 75% RHAC and 25% MO provided the best results with the fluoride concentration (< 1 mg/l) and the turbidity (<5 NTU) . Based on the study, a composite bed of 4 cm thick was designed for an equilibrium contact time of 30 minutes. The composite bed consisting of 75 % RHAC and 25% MO is found to be significantly reduces the turbidity along with 90 % removal of fluoride from water. The reagent cost of composite bed is low, and treatment effect is good. So the novel composite bio-adsorbents prepared in the present study can be used in drinking water treatment in the fluoride affected areas.

V. ACKNOWLEDGEMENT

The authors are highly thankful to Centre for Engineering Research and Development (CERD), Trivandrum, Kerala, India for providing financial support for the work.

REFERENCES

[1] Antu C D and Harikumar P S (2007), 'Genesis of fluo-ride in the shallow unconfined aquifer of muthalmada area in Bharatapuzha Basin', Proceedings of XIX Kerala Science Congress.

[2] Bulsu K R and Nawlakhe W G (1988), 'Defluoridation of water using Activated Alumina Batch operation', Journal of Environ., Health, Vol.30, p.262-264

[3] Anandu Aravind, Dayal Kurian Varghese,Arshik K, Meera S Prasad,Vinish V Nair(2015),'Defluoridation of Water Using Low Cost Bio-adsorbents: A comparative Study', International Journal of Engineering Technology Science and Research (IJETSR),Vol.2,Issue.5

[4] Runping H , Yuanfeng W , Weihong Y , Weihua Z , Jie S and Hongmin L, 'Biosorption of methylene blue from aqueous solution by rice husk in a fixed-bed column' , Water Supply, Vol. 2, p.311–317.

[5] Jed W, 'Moringa oleifera: A Review of the medical evidence for its nutritional, therapeutic and prophylactic properties', part I, Trees for life journal Vol.1, p.5.

[6] Stanley V A, Kumar T, Lal A A S, Pillai K S& Murthy P B K (2000) , 'Moringa oleifera seed extract as an antidote for fluoride toxicity' ,Vol.30, p.251.

[7] A.S. Parlikar, S.S. Mokashi (2013), 'Defluoridation Of Water by Moringa Oleifera- A Natural Adsorbent', International Journal of Engineering Science and Innovative Technology (IJESIT), Vol. 2, p. 245-252.

[8] Islam M, Patel RK (2011), 'Thermal activation of basic oxygen furnace slag and evaluation of its fluoride removal efficiency', Chem Eng J, Vol. 169, p. 68–77.

[9] Ravikumar K A & Sheeja A K (2014), 'Removal of Fluoride from Aqueous System using Moringa oleifera Seed Cake and its Composite Coagulants',Vol.4.

[10] WHO (2008), International Standards for Drinking Water, 3rd ed., Geneva.

[11] R. Rajendran, S. Balachandar, S. Sudha, Muhammed Arshid (2013), 'Natural Coagulants- An Alternative to Conventional Methods of Water Purification', International Journal of Pharmaseutical Research and Bio-science, Vol.2 (1), p.306-314.

[12] Vaishali Tomar, Dinesh Kumar (2013), 'A Critical Study on Efficiency of Different Materials for Fluoride Removal from Aqueous Media', Tomar and Kumar Chemistry Central Journal, Vol.7(1), p.1-15.

[13] V. Subhashini, A. V. V. S. Swamy, R. Hema Krishna (2012), 'Deflouridation from Aqueous Solutions using Alum', Chem Sci Trans, Vol.1(3), p.552-555.

[14] P. Gopalakrishnan, R. S. Vasan, P. S. Sarma, K. S. Ravindrannair, K. R. Thankappan (1999), ‘Prevalence of dental fluorosis and associated risk factors in Alappuzha district, Kerala’, The National Medical Journal of India, Vol.12(3).

[15] Tomas Blom, Elin Cederlund, Prosun Bhattacharya, P S Harikumar (2009), Fluoride Contaminated Groundwater in Palakkad and Alappuzha Districts of Kerala. CWRDM, Calicut, Kerala and Department of Land and Water Resources Engineering, KTH, Royal Institute of Technology, Sweden, TRITA LWR Master Thesis, p. 06-09.

[16] Neelo Razbe, Rajesh Kumar, Pratima, Rajat Kumar (2013), ‘Removal of Fluoride Ion from Aqueous Solution’, International Journal of Computational Engineering Research, Vol.3 (4), p.128-133.