Low Cost Deflouridation of Water Using Brick Pieces

S. Manjunath, R. Santhosh, S. Raja and V. Modi Jemishkumar

Abstract--- Excess fluoride in water causes health hazards to the natural environment. The removal of fluoride was attempted using natural brick pieces of two sizes 6.3-4.75 mm and 16-12 mm. Each material was set up in a PVC column and defluoridation capacities of these materials were studied with respect to initial concentration of fluoride and time. Concentration of fluoride in water is estimated for corresponding contact time using kinetic up flow equations. The study reveals that the estimated concentrations satisfactorily fit the measured concentrations and bricks of size 6.3 - 4.75 mm have good fluoride removal capacity of 90.33% for an initial concentration of 3mg/l, 61.85% for removing fluoride of 7mg/l and a brick of size 16-12 mm has an efficiency of 79.33% and 48.25 % respectively.

Keywords--- Excess Fluoride, Environment, Deflouridation, PVC Column, Bricks

I. INTRODUCTION

Widespread occurrence of fluoride, above the prescribed limit, in groundwater consumed by human beings has caused multi-dimensional health problems. Most common of these are dental fluorosis and skeletal fluorosis. Fluoride is one of the most abundant constituent occurring in groundwater in India and creates a major problem for safe ground water supply. Fluoride exists fairly abundantly in the earth's crust and can enter groundwater by natural process; especially soil at the food of mountains is particularly likely to be high in fluoride from the weathering and leaching of bedrock with high fluoride content. Fluorine is so highly reactive that it is never encountered in its elemental gaseous state except in some industrial processes [1] [16].

An optimum concentration can reduce the incidence of dental caries. Low levels of fluoride are required for humans as it has beneficial effects on tooth and bone structures. However ingestion of excessive fluorides, mainly through drinking water causes dental and skeletal fluorosis. Long- term ingestion of excessive fluoride has a chronic effect on the kidneys as well. The optimum level suggested by WHO is 0.7 mg/l from infancy to 16 years. According to the WHO, the maximum acceptable concentration of fluoride ions in drinking water is 1.5 mg/l to prevent tooth and bone problems. Concentration of fluoride below 1.5 mg/l is helpful in prevention of tooth decay and such level of fluoride also assists in the development of perfect bone structure in human and animals. However, a dose of fluoride above 1.5 mg/l increases the severity of tooth mottling and induces the prevalence of osteoporosis and collapsed vertebrae. Fluorosis, resulting from excessive consumption of fluoride, has no treatment and is considered to be deadly disease. It is considered that probable source of high fluoride in Indian waters is that during weathering and circulation of water in rocks and soils, fluorine is leached out and dissolved in ground water [12]. Tile fluoride content of ground water varies greatly depending on the type of rocks from which they originate. Among the various minerals responsible for high concentration of fluoride, the Fluor-apatite, CaF_2 and fluorite, CaF_{2-} is important. However, the most important being the fluorite (CaF2-) and the leaching of fluoride from

S. Manjunath, Assistant Professor, Acharya Institute of Technology, Bangalore-560107
R. Santhosh, Assistant Professor, Acharya Institute of Technology, Bangalore-560107
S. Raja, Lecturer, Acharya Institute of Technology, Bangalore-560107
V. Modi Jemishkumar, Lecturer, Acharya Institute of Technology, Bangalore-560107

the metamorphic rocks hornblende gneiss of proterozoic age. Concerned with the magnitude of health problems due to excess concentration of fluoride in drinking water several methods of defluoridation of drinking water have been developed. The ion-exchange, adsorption and precipitation are the usual means of defluoridation. However, in India precipitation and adsorption methods are most preferred. The adsorption method involves the contact of the fluoride containing water with a suitable adsorbent. Precipitation process is based on the addition of chemicals and removal of insoluble compounds as precipitates. In adsorption method, different types of adsorbents are being used for defluoridation and other minerals dyes and heavy metals e.g., activated alumina, coconut shell carbon, bagasse, chemically activated carbon, bone charcoa, natural zeolites, hydroxyapatite, burn clay, clay pots and crushed clay pots, membrane technologies i.e., donnan dialysisand other low cost bioadsorbents like saw dust, used tea leaves, cows dung have been found to be highly effective, cheap and eco-friendly. The shortcomings of most of the preferred methods are high operational and maintenance costs, low fluoride removal capacity, lack of selectivity for fluoride, undesirable aftereffects on water quality, generation of large amount of sludge and complicated procedure. The most common available method in India, Nalgonda Technique of community defluoridation, is based on precipitation process and is very efficient and cost effective. The main limitation of Nalgonda technique are daily addition of chemicals, large amount of sludge production, least effective with water having high total dissolve solids and high hardness. Besides it converts a large portion of ionic fluoride (67-87 %) into soluble aluminium complex and practically removes only a small portion of fluoride in the form of precipitate (18-33 %). Therefore, this technique is erroneous [2][3]. Residual aluminium ranges from 2.01-6.86 mg/l was also reported in Nalgonda technique, which is dangerous to human health as aluminium is a neurotoxin and concentration as low as 0.08 mg/l in drinking water has been reported to cause Alzheimer's disease17 and has strong carcinogenic properties. Adsorption methods are effective on both terms i.e., fluoride removal and cost for removal. Hence the need to find locally available defluoridation media for less expensive and technically feasible in rural communities in India and for easy use at both household and small community level is desirable. In present study, an attempt has been made for defluoridation of drinking water by using brick as a new feasible, suitable, effective and low cost adsorbent [5].

A. *Health Impacts of Fluoride in Portable Water*

Low dental caries incidence rates demonstrate that fluoride concentrations of up to 1.0 mg/l in potable water are beneficial to the oral health of children and, to a lesser extent, adults. In several developed countries fluoridation of water supplies is practised if the natural concentration is below the desired level. Recently, fluoridation of drinking water has been questioned and many countries have expressed concerns over this practice due to the adverse health effects of fluoride [6].

a. Dental Fluorisis

Dental fluorosis, also called “mottled enamel”, occurs when the fluoride level in drinking water is marginally above 1.0 mg/l. A relationship between fluoride concentration in potable water and mottled enamel was first established in 1931. Typical manifestations of dental fluorosis are loss of shining and development of horizontal yellow streaks on teeth. Since this is caused by high fluoride in or adjacent to developing enamel, dental fluorosis develops in children born and brought up in endemic areas of fluorosis. Once formed, the changes in the enamel are permanent. When the above manifestations are seen in an adult, they clearly indicate that the person has been exposed to high fluoride levels during her or his childhoodas shown in fig 1.1.

Fig 1.1: Image of Tooth Affected by Dental Fluorisis

b. Skeletal Fluorosis

Skeletal fluorosis affects both adults and children and is generally manifested after consumption of water with fluoride levels exceeding 3 mg/l. Fluoride is retained in the bones and induces hardening of all the bones, including the spine hypertrophy of the joints and a bone is seen, similar to osteoarthritis. Sclerosis or ossifications of the posterior longitudinal ligament can occur with resulting mydopathy and radiculopathy from root comparison. The entire spine cab by ossified with flulorises. Typical symptoms of skeletal fluorosis are pain in the joints and backbone. In severe cases this can result in crippling the patient. Recent studies have shown that excess intake of fluoride can also have certain non-skeletal health impacts such as gastro-intestinal problems, allergies, anaemia and urinary tract problems. Nutritional deficiencies can enhance the undesirable effects of fluoride as shown in fig 1.2.

Fig 1.2: Children Who are Affected by Skeletal Fluorosis in India

c. Information on the Occurence of Excessive Fluoride in Ground Water in India

India is among 23 nations in the world, where fluoride contaminated ground water is creating problems. Sixty-two million people including 6 million children in the country in 17 states are affected with dental, skeletal and non-skeletal fluorosis. The extent of fluoride contamination in the ground water varies from 1.0 to 48.0 mg/1. In the absence of perennial rivers, surface and canal systems; ground water remains the main source of drinking water for about 95% of population. Ground water of 18 districts content has fluoride (2 – 20 mg/1) affecting approximately 3 million populations. Table 1.1 represents total no of habitation with excess fluoride and total percentage of people affected in each state of India.

Table 1.1: Summarized Information on the Occurrence of Excessive Fluoride in Ground Water in India

State	No. of habitation with excess fluoride	State	No.of habitation with excess fluoride
Andhra Pradesh	7548	Meghalaya	33
Bihar	12	Maharashtra	39
Gujrat	2376	Orrisa	1138
Karnataka	860	Punjab	700
Kerala	287	Rajasthan	16560
Madhya Pradesh	201	Tamilnadu	570
Haryana	334	Uttar Pradesh	1072
Himachal Pradesh	488	West Bengal	21
Delhi	46		

II. EXPERIMENTAL INVESTIGATIONS

The objective of this study is to determine the suitability of broken bricks as filter media for the low cost domestic defluoridator. This is achieved by determining the following model parameters related to the broken bricks:

- Capacity parameter for fluoride in broken brick pieces.
- Rate of absorption.

Another important factor in the choice of clayware as defluoridating agent/absorbing media is its firing temperature. The firing of clays causes a series of physical and chemical changes in the material. Free water in clay is driven off by gently increasing the temperature to about 100ºC. Chemically bonded water namely water of crystallization escapes at 400ºC to 550ºC. Change in crystalline structure of clay mineral take place about 350ºC.Carbonaceous matters in clay is burned out at 800ºC to 900ºC. The efficiency of clay pots and crushed clay pots in removing fluoride from water. In the experiments fluoride adsorption declined as the firing temperature increased and when heated above 800ºC pots were rendered unfit for defluoridation purpose.

A. *Mechanism of Absorption of Bricks*

The brick pieces are a mixture of oxides of silica, aluminium, iron, calcium, magnesium etc. In the presence of water these compounds can be hydroxylated. The specific adsorption reaction can be explained by two step mechanism as follows:

$$\equiv \text{S-OH} + \text{H} \longleftrightarrow \text{S-OH2}^{+} \quad (1)$$

$$\equiv \text{S-OH2}^{+} + \text{F} \longleftrightarrow \text{S-F} + \text{H2O} \quad (2)$$

B. *Model Description*

There have been various attempts to establish mathematical models to describe the fluoride uptake in a defluoridating agent, in the past five decades. Initially let us consider following three models from research studies done on various defluoridating agents. -Larsen (1974), using dental enamel and Bhargava et al (1991), using bone char, described the uptake as a simple first order process kinetics to fluoride concentration in water (S) as described by:

$$\frac{ds}{dt} = -k1\, S \quad (3)$$

Christoffersen et al (1984), described the uptake as first order with respect to the fluoride concentration in water and with respect to fluoride saturation deficit (fm –f) in defluoridating agent namely, Synthetic Hydroxy Apatite (Hap) as given by:

$$\frac{ds}{dt} = -k1\, Xda\, (fm - f)S - k2\, Xda\, F \text{ --------- } (4)$$

Stumm (1992) used a semi-infinite linear diffusion limited sorption in porous media in general, where uptake is proportional to concentration of fluoride in water and dosage of defluoridating agent and also the reciprocal of square root of contact time as given by:

$$\frac{\mathrm{ds}}{\mathrm{dt}} = k\, Xda\, S\, t^{-.5} \text{ ------------------------ } (5)$$

These tested models fit some of the data collected, but none of them seem to fit in general, for all the different experimental conditions. Therefore, the development of a general model to describe the change in fluoride concentration in water in batch as a function of time by mean of an explicit mathematical equation.

a. *A Kinetic Model to Describe fluoride Uptake in Batch*

Bregnhøj et al (1995) developed a model to address the situation of treatment of water in fluoride affected areas in developing countries. So the number of experimental variables and rate parameters were to be kept as few as possible and advanced characteristics of the filter media such as grain size, initial concentration.The following differential equation to describe concentration of fluoride.

$$\beta = \frac{Xda\, fmb}{\mathrm{So}} \text{------------------------------------ } (6)$$

$$S = \frac{So\,(\beta-1)}{(\beta e^{2(\beta-1)So\,k\,t^{0.5}} - 1)} \text{-------------------------------- } (7)$$

Where ,

X_{DA} is Dosage of Defluoridating Agent. (DA)

S is the final concentration of fluoride in mg/l

$S_o$is the initial concentration in mg/l

t is the time in hours

*k*is the constants which depend on initial concentration of fluoride (S_o)

f_{mb} The capacity parameter f_{mb} mainly depends on quality of bricks, especially the firing temperature of bricks and the optimal range is between 500°C and 700 oC.

C. Materials and Methods

Raw water is poured in through the funnel attached to 110mm diameter PVC pipe which runs to the bottom compartment. Thus raw water enters the defluoridator through the bottom compartment and moves upward as the water was removed from the top. The water collected can be used for household purpose.

a. Reagents and Stock Solutions

Preparation of stock solution using sodium fluoride

Equivalent weight of sodium fluoride =41.9g

Equivalent weight of fluoride=18.9g

18.9g of fluoride is present in 41.9 g of sodium fluoride

Therefore, Quantity of sodium fluoride to be added to preapre 1mg /liter standard fluoride solution

=(molecular wt of NaF/ molecular wt of F) =41.9/18.9

=2.21mg/l

All of the reagents used in this study were of analytical grade. Fluoride stock solution (1 000 mg·ℓ−1) was prepared, by dissolving 2.21 mg of NaF in distilled water and filling to 1 ℓ. This was then diluted with distilled water to get the required subsequent concentrations.Two samples of 3mg/l and 7mg/l are prepared

b. Sorbent Preparation (Broken Bricks)

Two sizes of bricks are prepared i.e. 12– 16mm and 6.3-4.75mm is prepared with respective sieves and was washed several times with distilled water till clear water was obtained and dried in oven at 105^{o}C for 12 h .The following parameters are determined and tabulated in Table 2.1 - 2.3.

- Specific gravity.
- Matter soluble in water.
- Weight of bricks taken

Fig. 2.1: Preparation of Sorbent Media Using Bricks

1. Specific Gravity Test(G)

Table 2.1:Results of Specific Gravity Test

Size	16-12mm	6.3-4.75mm
weight of pycnometer W1 (grams)	680	680
weight of pycnometer+1/3 of sample W2 (grams)	1078	990
weight of pycnometer+1/3 of sample + water W3(grams)	1803	1730
weight of pycnometer +water W4(grams)	1550	1550
G =(W2- W1) / ((W2- W1)-(w3-w4))	2.54	2.78

2. Matter Soluble Test

Table 2.2: Results of Matter Soluble Test

Size	16-12mm	6.3-4.75mm
Initial weight (w1)g	1000	1000
Weight after 24 hrs soaking in water (w2)g	989.43	983.36
Matter soluble in water (%) ((w1-w2)/w1)X100	1.057	1.664

Table 2.3: Characteristics of Broken Bricks Used in Filter

Size	16-12mm	6.3-4.75mm
Specific gravity	2.45	2.78
Matter soluble test(%)	1.057	1.667
Weight of bricks taken (g)	5900	7300

Fig. 2.2: Prepared Filter Media of size 16-12mm and 6.3-4.75mm Respectively

D. Column Fabrication

Small-scale column tests are carried out to evaluate the capacity of the media for removing fluoride from water under continuous flow conditions. A P.V.C tube of diameter 110 mm and height 1000mm was used to conduct the adsorption tests. The column was packed with the desired amount of the media to obtain the desired bed height. A schematic diagram of the column. Influent water is poured through the column with the help of funnel. Upward flow of fluid was chosen to minimize channeling inside the column. Samples of the outlet bulk solution were collected at definite intervals of time and examined for fluoride concentration in fig 2.3-2.4.

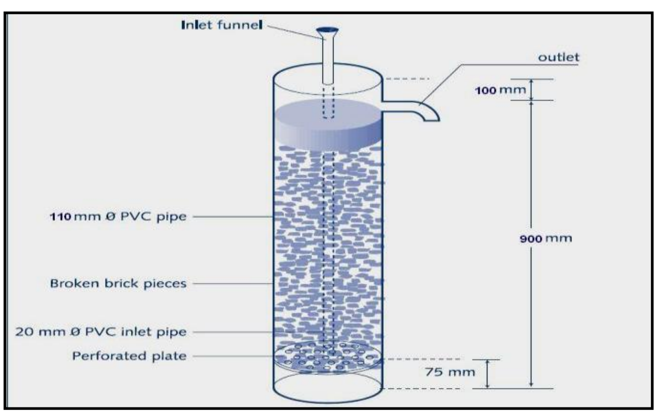

Fig. 2.3: Schematic Diagram of the Up Flow Filter

Fig. 2.4: Actual Model of Filter

Table 2.4: Representing Specifications of Filter

Specifications	Model 1 12-16mm	Model 2 6.3-4.75mm
Diameter (mm) (top/bottom)	110	110
Maximum bed height(mm)	1000	1000
Sorbent bed height (mm)	825	825
Volume of filter (liters)	8	7.4

E. Methodology

Two sizes of brick pieces were used for the experiment in two separate filters. The size of particles, initial concentrations and weight of the batch of brick pieces is given in Table 2.5. Two trials were down using both filters. The fluoride concentration and contact time were recorded. Fluoride in water is estimated using Kinetic upflow formula.Table 2.5represents Kinetic model parameters for broken brick pieces.The Fluoride in water was analyzed using the direct reading DR / 2000 HACH spectrophotometer.

Table 2.5: Parameters Considered for Experiment

Parameter	Filter	
	F1	F2
Particle size (mm)	16-12mm	6.3-4.75mm
Volume of Raw Water (L)	8	7.4
Initial concentration of fluoride(mg/l)	3&7	3&7
Weight of the batch of bricks (kg)	5.9	7.3

Estimating Fluoride Concentration using Kinetic model Equation

The following equation is used to estimate concentration of fluoride.

$$\beta = \frac{Xda\, fmb}{So} \text{------------------------------ (8)}$$

$$S = \frac{So\ (\beta - 1)}{(\beta e^{2(\beta-1)So\ k\ t^{0.5}} - 1)} \text{ ---------------- (9)}$$

where ,X_{DA} Dosage of Defluoridating Agent. (DA)

S is the final concentration of fluoride in mg/l

S_o is the initial concentration in mg/l

t is the time in hours

*k*is the constants which depend on initial concentration of fluoride (S_o)

f_{mb} The capacity parameter f_{mb} mainly depends on quality of bricks, especially the firing temperature of bricks and the optimal range is between 500°C and 700°C.

The values of k and β are adopted in table

Table 2.6: Showing the Estimated Final Concentrations of Flouride and Measured Final Concentrations of Flouride

Sl. No.	Brick Size (mm)	Time (Hrs)	Initial Concentration of Flouride (mg/L)	K	β	Final Concentration of Flouride- Estimated (mg/L)	Final Concentration of Flouride- Measured (mg/L)
1	6.3-4.75	2	3.0	0.00165	54.91	1.39	1.43
2		4				1.02	1.10
3		8				0.65	0.69
4		12				0.46	0.51
5		24				0.21	0.29
6		2	7.0	0.00064	23.53	5.32	5.32
7		4				4.75	4.76
8		8				4.05	3.91
9		12				3.59	3.48
10		24				2.73	2.67
11	16-12	2	3.0	0.00165	54.91	1.84	1.92
12		4				1.50	1.61
13		8				1.13	1.25
14		12				0.91	1.06
15		24				0.56	0.62
16		2	7.0	0.00064	15.27	5.77	5.83
17		4				5.34	5.41
18		8				4.77	4.85
19		12				4.38	4.44
20		24				3.62	3.71

Note:_The vaules of K and β obtained from Kinetic model parameters for broken brick pieces

Fig. 2.6: Measuring Fluoride Concentration using the Direct Reading in Spectrophotometer

Table 2.7: Final Results of Estimated and Measured Fluoride Concentration

Description	Trial 1	Trial 2	Trial 3	Trial 4
Broken Brick Size (mm)	6.3-4.75	6.3-4.75	16-12	16-12
Initial concentration I_0 (mg/L)	3	7	3	7
Measured final concentration F_0 (mg/L)	0.29	2.67	0.62	3.71
Estimated final concentration F_0 (mg/L)	0.21	2.53	0.56	3.62
Percentage reduction $=((I_o-F_o)/I_o)*100$	90.33%	61.85%	79.33%	48.25%
Absorption capacity of bricks= $\frac{(Io-Fo)}{(weight\ of\ bricks)}$ (mg/kg)	0.4875		0.4803	

III. Results and Discussion

The fluoride removing capacity of the bricks pieces for size 6.3-4.75mm has good absorption capacity of fluoride since it has larger surface area. Also the bricks exhibited poor fluoride removal capacity for higher concentrations of fluoride in water. The kinetic up flow formula results were similar to measured concentration. The maintenance cost of this defluoridator is low and has simple design which can be easily operated.

Two sizes of bricks were considered such as 6.3-4.75 mm and 16-12mm for initial concetrations of flouride (3 and 7 mg/l), in both cases the flouride content was decreasing linearly as the time increased as shown in Fig. 3.1-3.4. The estimated results from kinetic up flow formula and measured results were almost same.

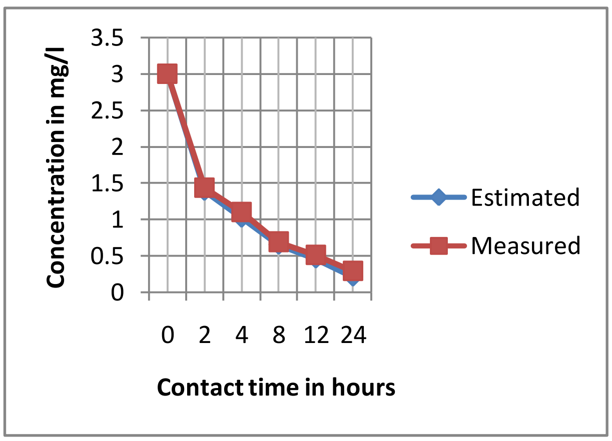

Fig. 3.1: Comparison of Measured and Estimated Values for Filter with 6.3-4.75mm with 3mg/l of S_o

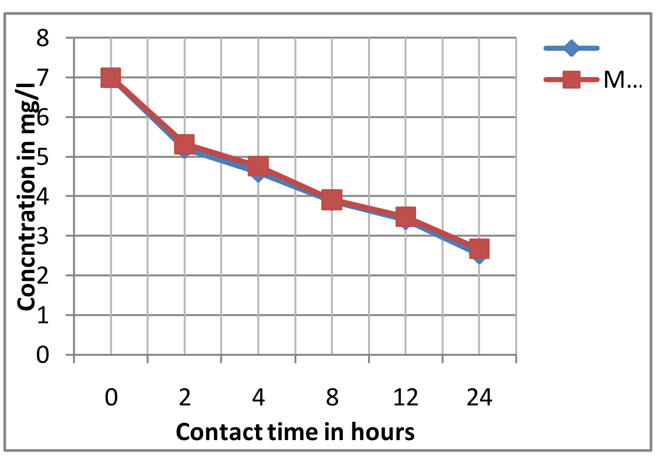

Fig. 3.2: Comparison of Measured and Estimated Values for Filter with 6.3-4.75mm with 7mg/l of S_o

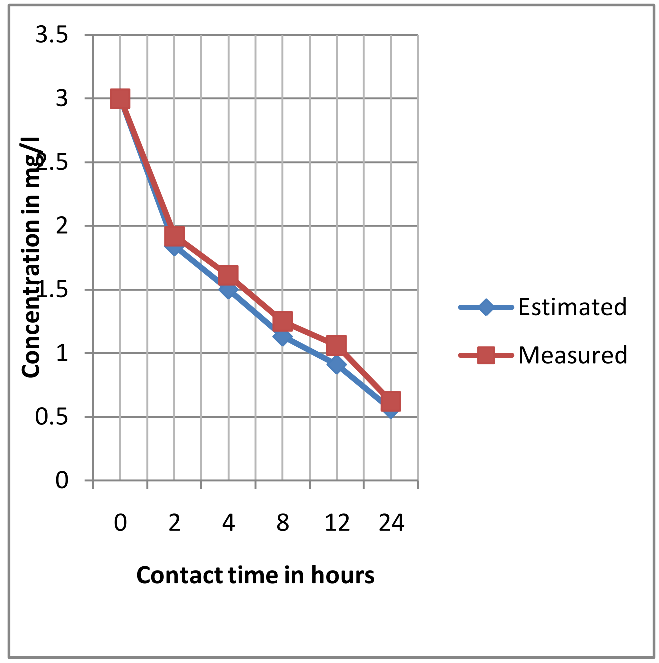

Fig. 3.3: Comparison of Measured and Estimated Values for Filter with 16-12mm with 3mg/l of So

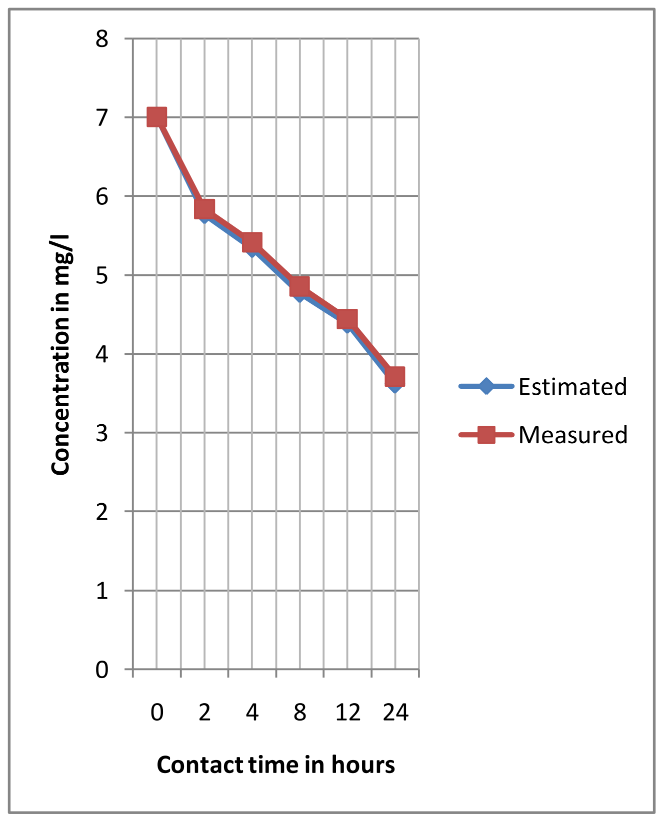

Fig. 3.4: Comparison of Measured and Estimated Values for Filter with 16-12mm with 7mg/l of $S_{o\backslash}$

IV. CONCLUSION

The following conclusions were drawn from above study:

- Bricks demonstrate poor fluoride removal quality for raw water containing high concentration of fluoride; they are not suitable as filter media to achieve the desired level of fluoride in the effluent under these conditions. Although the rate of removal of fluoride is better in smaller size of brick pieces as it has larger surface area for adsorption, water storage capacity decreases in the defluoridator with the decrease in porosity. .
- It was assumed that the raw water was retained in the defluoridator for a day before consumption. Predicted quality of the effluent in the defluoridators for broken brick pieces as defluoridating agents seems to tally reasonably well with the measured data, in spite of various limitations encountered.
- The required capital investment is modest and can be affordable by the affected community.
- The maintenance cost of the method is low.
- It is simple in design and the villagers would be able to operate it.
- The method is able to reduce the fluoride content to low levels within a day or so and could improve the quality of water in general.

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