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User perception study for performance evaluation of domestic defluoridation techniques for its application in rural areas

Sneha Lunge, Rajesh Biniwale, Nitin Labhsetwar, Sadhana S. Rayalu*

National Environmental Engineering Research Institute, India

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ABSTRACT

Fluoride concentrations in ground water have been monitored in rural areas of Dhar and Jhabua districts in Madhya Pradesh, India. A correlation of fluoride concentration with pH, TDS and conductivity has been estimated to identify surrogate monitoring parameter. Further, fluoride removal from drinking water has been achieved by using adsorbents specially developed for domestic applications. These adsorbents have been evaluated using three different methods namely; loose adsorbent, pre-packed sachet and packed bamboo column. Comparative evaluation of these methods has been demonstrated in the laboratory and field. The stringent limit of 1 mg/L for fluoride concentration in drinking water has been achieved by use of specially designed adsorbents. A feedback from end-users in Tarapur and Ukala villages of Dhar districts Madhya Pradesh regarding the adsorbents and its acceptability has been collected. User's perception regarding these household treatments reveals encouraging response for defluoridation methods. According to user's perception loose adsorbent approach emerged out as most simple, clean and safe household defluoridation method.

1. Introduction

Fluoride is one of the most abundant constituents occurring in groundwater worldwide and creates a major threat in safe drinking water supply. Widespread occurrence of fluoride above the prescribed limit in groundwater meant for human consumption has caused multidimensional health problems. The problem has engulfed many parts of the world and today many millions of people rely on groundwater with concentrations of contaminants such as fluoride above the WHO guideline value [1]. High fluoride concentrations in groundwater are found in many countries around the world, notably the United States of America, Africa, and Asia [2,3]. The most severe problem associated with high fluoride waters occurs in China [4], India [5], Sri Lanka [6] and Rift Valley countries in Africa. High fluoride ground waters have been studied in detail in Africa, in particular Kenya and Tanzania [7–11]. High fluoride groundwater is also found in the East Upper Region of Ghana [12]. In the early 1980s, it was estimated that around 260 million people worldwide (in 30 countries) were drinking water with more than 1 mg/L of fluoride [13]. In India alone, endemic fluorosis is thought to affect around 1 million people [14,15] and is a major problem in 17 out of the country's 22 states, especially Rajasthan, Madhya Pradesh, Andhra Pradesh, Tamil Nadu, Gujarat and Uttar Pradesh [16,17].

Fluoride in water derives mainly from dissolution of natural minerals in the rocks and soils with which water interacts. High fluoride concentrations can be built up in ground waters, which have long residence times in the host aquifers. The most important remedial action is prevention of further exposure by providing safe drinking water. However, in most of the areas hand pump and tube wells are the major source of water source in most of the rural areas, water source substitution may be impossible due to non-availability of alternate sources and therefore removal of excess fluoride is the only feasible solution. There are several defluoridation techniques, which can be categorized into four main categories namely precipitation, membrane processes and ion-exchange/adsorption onto various adsorbents [18].

Nalgonda process developed by NEERI is the most widely used defluoridation method particularly at community level [19–21]. The bucket defluoridation system based on Nalgonda technique has also been developed for domestic use [10]. Bucket defluoridation process is suitable for a daily routine, where one bucket of water is treated for one day's water supply of about 20 L. The process produces water with residual fluoride between 1 and 1.5 mg/L [22]. Calcium salts are also used in the process of precipitation [23]. When calcium salt is reacted with fluoride its gets removed by forming insoluble CaF_2 in a wide range of pH 4.0–10.4. Generation

* Corresponding author at: Environmental Materials Unit, National Environmental Engineering Research Institute, Nehru Marg, Nagpur 440020, India.
Tel.: +91 7122247828; fax: +91 7122247828.
E-mail addresses: s_rayalu@neeri.res.in, emu_neeri@yahoo.com, apcneeri@nagpur.dot.net.in (S.S. Rayalu).

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of a large amount of sludge is a major limitation associated with precipitation and therefore from environmental point of view it is not acceptable in cases where fluoride concentration is considerably high. Moreover skilled manpower is required and hence the technique is not suitable for rural areas where fluoride problem is more severe. Membrane processes such as reverse osmosis (RO), nanofiltration and electrodialysis have also been studied recently for fluoride removal from water [24–26]. However, RO membranes are subject to fouling and can also act as media for microbiological growth [27]. Moreover, RO systems produce concentrated brine discharges that must be disposed off safely. Reverse osmosis systems also result in significant water loss and are not suitable for arid regions where water scarcity is a big problem. The capital cost implications are not in favor of RO systems. Electrodialysis (ED) is a membrane process similar to RO, except that ED uses an applied D.C. (potential electric current), instead of pressure, to separate ionic contaminants from water. Lounici et al. [28] have also studied fluoride removal using electrodialysis. However, the ED process, besides having disadvantages associated with RO processes, is energy intensive and hence is not suitable for rural applications.

Therefore removal through adsorption is most promising in terms of cost of the medium and running costs, ease of operation, adsorption capacity, potential for reuse, number of useful cycles and the possibility of regeneration. In adsorption processes fluoride contaminated water is allowed to contact with an adsorbent where fluoride is removed by surface chemical reactions with the solid matrix followed by removal of the adsorbent. Fluoride removal through adsorbents can also be achieved using columns, which involves passage of the water through a contact bed where fluoride is adsorbed on the adsorbent and the treated water is collected at bottom. Several adsorbents reported for fluoride removal include alumina [29,30], flyash [31], clays [5,32,33], rare earth oxides [34], Zn/Al hydrotalcite [35], brick powder [16], red mud [36], bleaching earth [37,38], hydroxyapatite, fluorspar, calcite and quartz [18], bauxite [39], titanium rich bauxite [40] and Gypsum [41] and other low cost materials [42].

In this paper we report fluoride removal using specially designed adsorbents with fast kinetics and selectivity. These adsorbents have been used for development of household methods to treat small quantities of water, particularly used for drinking and cooking purpose. Fluoride levels in groundwater from study area were estimated and a statistical correlation is reported for identification of parameters for avoiding tedious and expensive methods for estimation of fluoride in field applications. These domestic methods of defluoridation were demonstrated at contaminated sites and feed back was collected from end users to understand user's perception regarding these methods. The questionnaires were designed to investigate the awareness and acceptance of fluoride mitigation methods, in general and acceptability of household methods in particular.

High concentrations of fluoride pose health problems when water is consumed either for drinking or for cooking purpose. Therefore treatment of small volumes of water at household levels is more appropriate than treatment attached to source of water (hand-pumps) to minimize the requirement of adsorbent. In this view three methods were developed for household treatment for fluoride removal. These methods were developed by using locally available materials for easy acceptance in rural areas.

2. Materials and methods

2.1. Water quality monitoring

Districts of Dhar and Jhabua in Madhya Pradesh, a state in central India were selected as study area wherein ground water contains fluoride in relatively high concentrations. Several cases of fluorosis both dental and skeletal have been reported from villages in these districts. Ground water drawn with the help of hand-pumps constitutes a major source of water consumption. Besides, there is paucity of well-water and surface water sources in this study area.

2.2. Synthesis of adsorbents

Synthesis of lanthanum treated chitosan granules with 22 and 10% lanthanum loading was carried out as follows: 5–9 g of chitosan (85% deacetylated) was dissolved in 200–400 ml of acetic acid (CH_3COOH) solution (5% v/v). 1.0–4.0 g of $LaNO_3 \cdot 9H_2O$ was dissolved in 100–150 ml of distilled water. The Lanthanum solution was then added to the polymer solution with stirring for 1–3 h. The resulting La-chitosan solution was drop wise added into NH_4OH solution (10–50% v/v) under vigorous stirring, using a syringe pump. The gel macro spheres formed were allowed to stabilize in NH_4OH solution for 0.25–6 h. The beads were separated from the NH_4OH solution and washed with deionized water and dried at 45–75 °C in oven for 8–10 h. Lanthanum treated chitosan granules were prepared with 10 and 20 wt% loadings and the samples with 10% and 22% lanthanum loadings were designated as Chito-La-10 and Chito-La-22, respectively. However, detail synthesis and batch adsorption studies of La-chitosan derivative have been studied in our previous publication [43,44].

2.3. Treatment methodologies

The treatment methods include controlled dose treatment using pre-packed sachets of adsorbents in porous cloth, loose adsorbent stirring in water followed by filtration through cloth and a bamboo column with a slow feed rate of water from top pot by dripping. Figs. 1 and 2 show the schematic diagram of treatment using sachet and bamboo column. The adsorbent preparation, regeneration and disposal may be handled at centralized facilities. A brief description of these defluoridation methods used is given in following paragraphs.

2.3.1. Loose sorbent

In this method a predefined dose of adsorbent is added to 1 L of water and stirred continuously by using a stick for about 10–15 min. Stirring ensures a good contact between adsorbent and water. After stirring, the adsorbent was separated by filtering treated water through a piece of cotton cloth. Treated water was then evaluated for its fluoride content and other physiochemical parameters.

2.3.2. Sachet technique

The method involves simple stirring of tea-bag type sachet, containing adsorbent material for application in the field. Pre-packed sachet technique using controlled dose for removal of fluoride from a pot of water would be appropriate to facilitate maintaining exact amount of adsorbent for treatment. Pre-defined controlled dose would facilitate the effective use of adsorbents in non-complicated manner by the end-user. For treatment of larger volumes of water an appropriate dose of adsorbent in multiple sachets may be used (Fig. 2(b)). This method involves stirring of adsorbent packed sachets for 10–15 min.

2.3.3. Bamboo column technique

Bamboo column as shown in Fig. 1(b) is closed at bottom with a piece of cloth. Bamboo is filled with a layer of fine sand and top layer of adsorbent. Water is fed through top from a pot with a flow rate of about 3 ml/min. This treatment may be suitable for overnight treatment for volume of water sufficient for a family. The residence time in column was about 10 min.

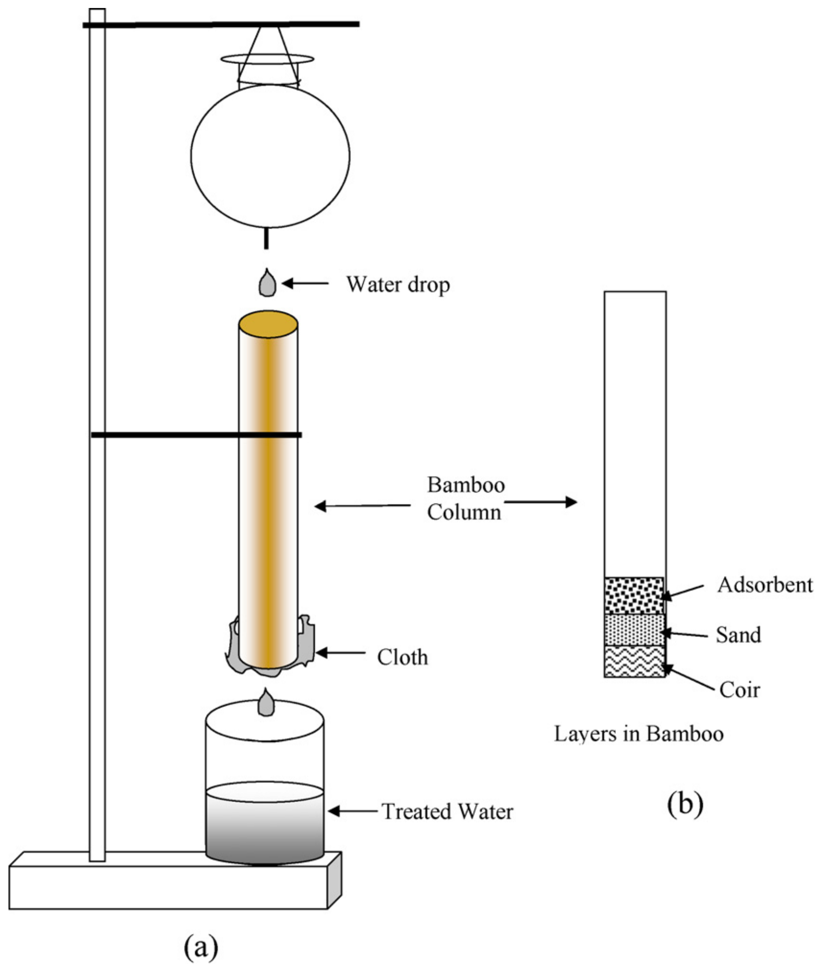

Fig. 1. Schematic of column treatment using bamboo.

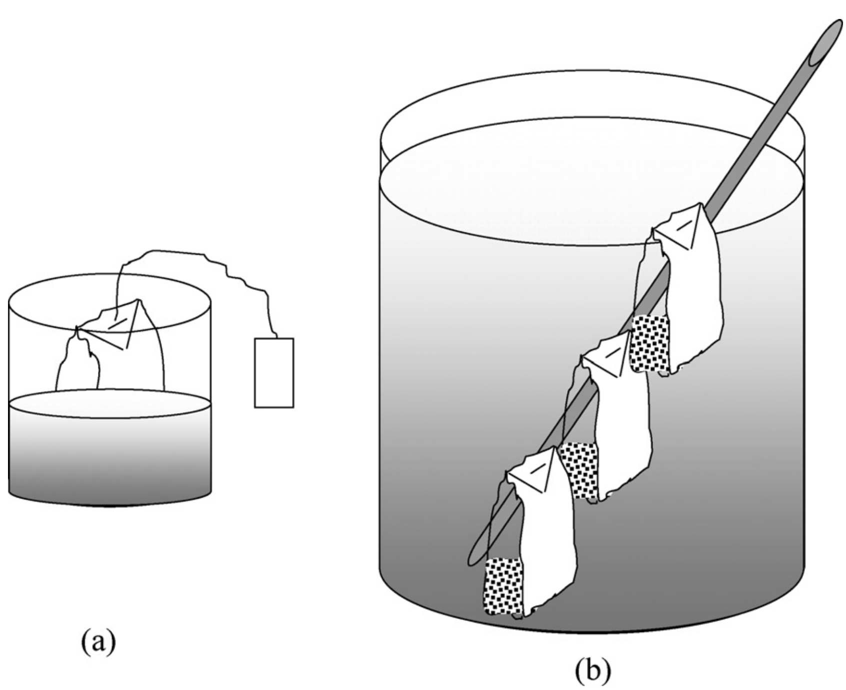

Fig. 2. Schematic of treatment method using pre-packed sachets (a) for small volumes and (b) with multiple sachets for larger volumes.

Table 1
Regression analysis between fluoride concentration and physico-chemical parameters of water.

Correlation equation	*P*-value
October 2005	
$F = -30.63 + 4.25$ pH	1.2×10^{-6}
$F = 1.30 + 0.002$ conductivity	0.128
$F = 2.14 + 0.0018$ TDS	0.99
December 2005	
$F = 48.85 - 5.57$ pH	3.48×10^{-6}
$F = 1.74 + 0.0023$ conductivity	0.021
$F = 1.721 + 0.020$ TDS	0.020

2.4. Method of fluoride estimation

The fluoride analysis was carried out using F^- selective electrode and ion meter (Euthech Model pH 2100). 10 ml of sample was used for F^- estimation after addition of 1 ml of total ionic strength adjusting buffer (TISAB-III) solution to release F^- in ionic form. Sample was stirred continuously during estimation using a magnetic stirrer at speed of 150 rpm.

2.5. User perception study

User perception study was conducted at two villages in Dhar districts. Three household treatment methods of fluoride removal described above were demonstrated to participants. Two questionnaires were designed to survey the user's perception and population characteristics. The participants to the demonstrations were local people of different age groups, education levels and occupations.

3. Results and discussions

3.1. Water quality monitoring

During October and December 2005 ground water samples were collected from more than 75 villages of Dhar and Jhabua district. These samples were analyzed for fluoride, pH, alkalinity, conductivity and TDS. The results of fluoride concentrations are given in Fig. 3(a), ca. 35% samples found to have fluoride concentration of 1 mg/L or lower. About 37% samples were in the range of 2–4 mg/L and 28% samples were having fluoride concentration more than 5 mg/L. Nearly 10% locations have acute problem of high fluoride concentration in the range of 9–14 mg/L. Histograms of pH, TDS and conductivity are shown in Fig. 3(b–d). Statistical correlations between fluoride and other parameters like pH, TDS and alkalinity were worked out and are presented in Table 1. Regression analysis of F^- concentration with pH, conductivity and total dissolved solids has been carried out separately for two seasons in October 2005 and December 2005. Total number of samples analyzed in October and December 2005 were 30 and 80, respectively. In case of correla-

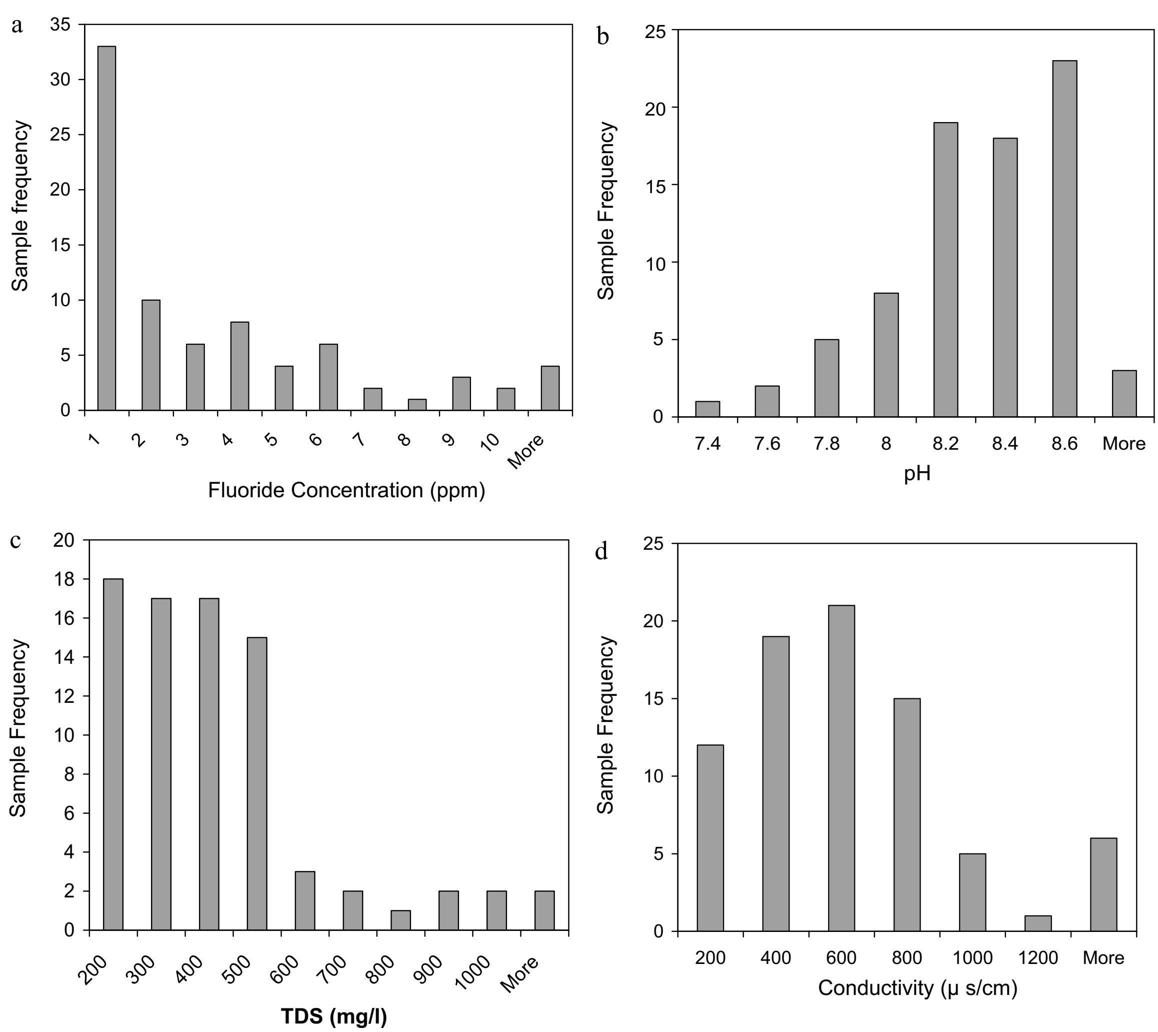

Fig. 3. Water quality assessment for Dhar and Jhabua (a) fluoride concentrations, (b) pH, (c) total dissolved solids and (d) conductivity.

tion of F^- with pH, a strong correlation was observed for both the sampling periods. For samples collected during October 2005 there was no strong correlation observed between F^- and conductivity or TDS. The regression analysis was conducted with 95% confidence level and $P < 0.05$ was used as criteria for establishing a correlation. One of the objectives of statistical correlation was to investigate for a possible surrogate parameter to identify fluoride levels in absence of expensive ion electrode or tedious SPANDS method in rural areas.

3.2. *Efficiency of different treatment methods*

Fluoride removal method by using loose adsorbent technique was evaluated using Chito-La-22. For comparison of adsorption capacity different doses of adsorbent were used and was stirred thoroughly after addition to water using a rotary shaker for 1 h. The adsorption capacities of various adsorbents are compared with a few reported adsorbents and are presented in Table 2. Generally the adsorption capacity of an adsorbent increases with increasing F^- initial concentration until saturation conditions. The isotherm studies performed for a higher range of fluoride concentrations will show higher capacity than those of lower ranges and the maximum adsorption capacity obtained from Langmuir isotherm will differ in both cases, so it is important to take in to consideration the initial fluoride concentration range, when performing comparative studies. It is apparent from the table that Chito-La-22 is having a high adsorption capacity of 8.07 mg/g when allowed to attain equilibrium. The adsorption capacity of Chito-La-22 is comparable to that of other reported chitosan based adsorbents. It has to be taken into account that some of the adsorbents reported presented high adsorption capacity at high F^- equilibrium concentration in water, but, in water treatment, the final concentration of F^- in the water solution must be below 1.0 mg/L. So it is desirable that the adsorbent presents high adsorption capacity at low fluoride equilibrium concentrations. The F^- adsorption capacity corresponding to F^- equilibrium concentration 1.0 mg/L is seldom reported. Table 2 also reports the amount of adsorbent required for reduction of fluoride concentration from 5 to 1 mg/L for 1 L of water. Most of the adsorbents developed in this study are of practical use in view of the relatively small amount of adsorbents required for treatment of 1 L of water. Particularly Chito-La-22 may be required about 0.5 kg for the same quantity of water and with its high fluoride uptake capacity was selected for treatment in field.

On the basis of adsorption capacity derived form batch adsorption studies the adsorption capacities work out to be 8.07 mg/g and 4.7 mg/g for adsorbents Chito-La-22 and Chito-La-22. Since L Chito-La-22 shows the high adsorption capacity, it was selected for further studies.

3.2.1. *Loose sorbent study*

Fig. 4 shows the results of effect of increasing dose of adsorbent from 1 to 10 g/L for loose sorbent method using three different adsorbents. Chito-La-22 is most active as compared to other two adsorbents and is used for further study. Initial concentration of fluoride was 5 mg/L. At the adsorbent dose of 1 g/L removal of 91.3% was observed which has increased to 97.7% at 2 g/L of adsorbent dose. Further increase in dose has not resulted in any significant improvement of fluoride removal efficiency therefore optimum dose selected for loose adsorbent treatment was 2 g/L. This loose adsorbent approach was demonstrated in the field at two different locations Tarapur and Ukala villages in Dhar districts. At Tarapur the ground water was having 14.3 mg/L of fluoride, which was reduced to 0.3 mg/L after treatment with 8 g/L adsorbent dose and stirring time of 20 min. Fluoride concentration at Ukala was 5.67 mg/L and was reduced to 0.6 mg/L after loose sorbent treatment.

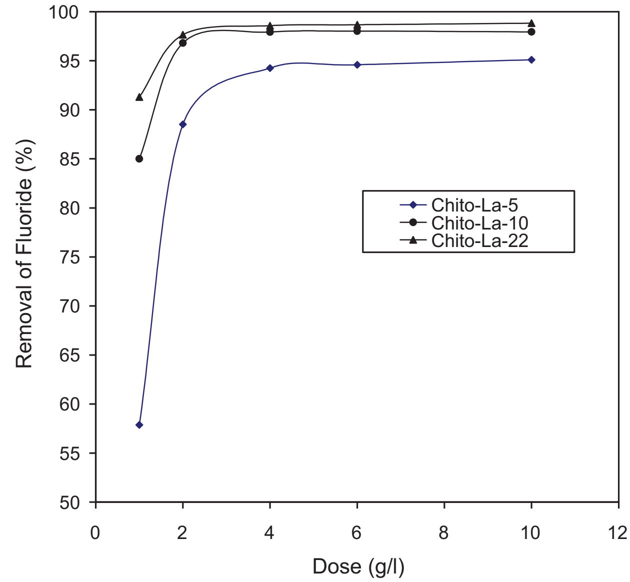

Fig. 4. Effect of dose of adsorbents on fluoride removal in loose adsorbent treatment method using various adsorbents.

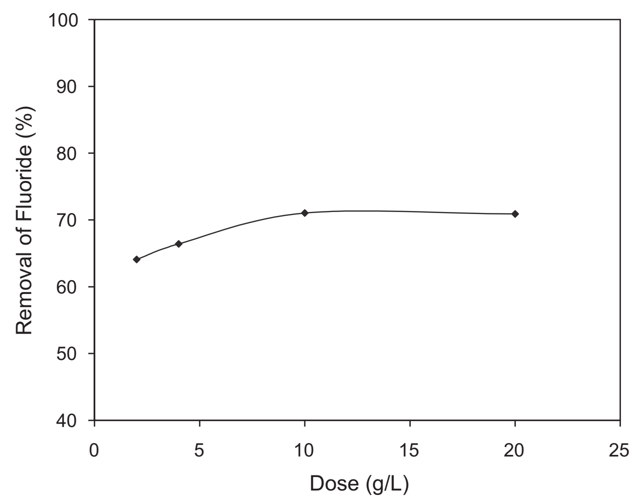

Fig. 5. Effect of dose of adsorbents on fluoride removal in sachet treatment method.

3.2.2. *Sachet technique*

In an alternate treatment method a membrane sachet typically used for preparing tea-bags has been used for packing adsorbent so as to apply controlled dose in treatment. Adsorbent packed sachet was stirred in water for 10 min and water was analyzed for final concentration of fluoride. Effect of variation of dose on fluoride removal is depicted in Fig. 5. At a dose of 2 g/L fluoride removal efficiency was 64% and increased to 71% at dose of 10 g/L. Further increase in dose has no effect on fluoride removal.

3.2.3. *Bamboo column*

As an alternative method for water treatment, water was allowed to pass through adsorbent filled column prepared from bamboo (Fig. 1). Using the adsorption capacity estimated in batch study a dose of 4 g was selected for treatment of 1 L of water with initial concentration of 10.2 mg/L. The results of column shown in Fig. 6 indicate that column works with fluoride removal efficiency of 94% and 97% for adsorbents Chito-La-10 and Chito-La-22, respectively. In about 1 h the removal efficiency for Chito-La-10 dropped to 89% and for Chito-La-22 it remained at 95% indicating more stability for later adsorbent.

Table 2
Comparison of adsorption capacities of different chitosan based adsorbents.

Adsorbents	Langmuir adsorption capacity (mg/g)	Initial fluoride conc. (mg/L)	Mass of adsorbent (kg)	Ref.
Chito-La-22	8.07	5	0.50	Present work
Chito-La-10	4.7	5	1.72	Present work
Treated alumina	1.22	5	3.28	This work
Alumina commercial	1.20	5	3.33	This work
POP	0.38	5	10.47	This work
Chitin	0.33	5	12.01	This work
La-chitosan flakes	1.27	5	–	[45]
La-CCB	11.905	10	–	[46]
Fe-CCB	13.69	10	–	[47]
Nd-modified chitosan	22.38	20	–	[48]
MgO chitosan composite	11.36	10	–	[49]
Protonated carboxylated chitosan beads	4.93	10	–	[50]

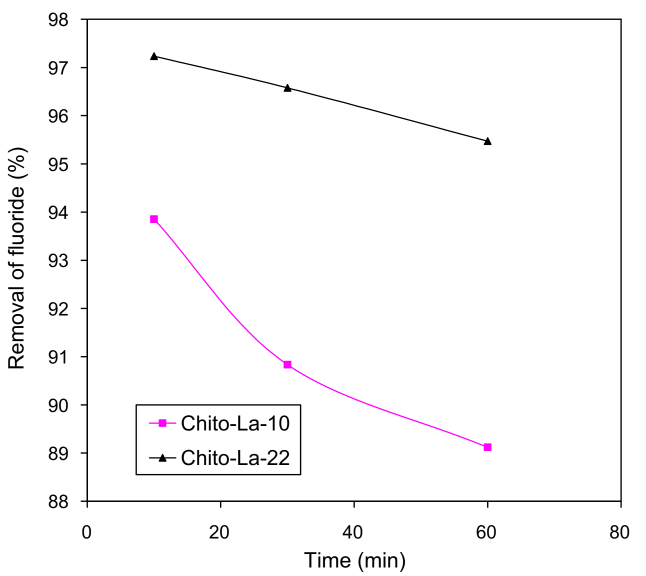

Fig. 6. Trend of removal of fluoride observed in continuous mode of column operation when flow rate is kept at 3 ml/min.

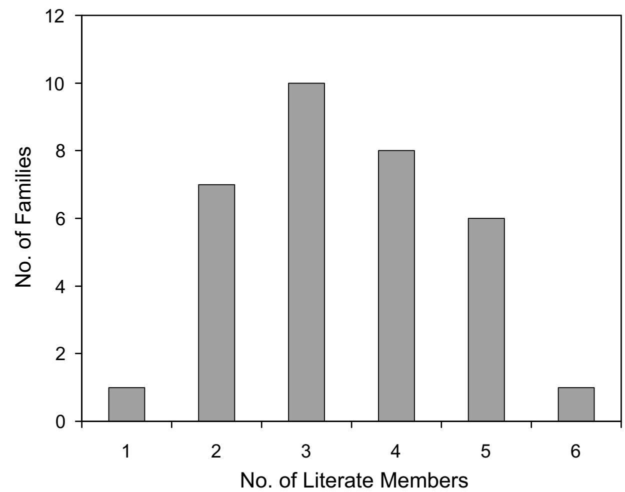

Fig. 7. Distribution of families in study area based on number of literate persons in a family.

3.3. Outcome of user's perception study

3.3.1. Population surveyed

User perception study was conducted at two villages in Dhar districts by demonstration of all three household methods of fluoride removal described above. Two questionnaires were designed to survey the user's perception and population characteristics. The study area was rural and the local population was mostly farmers or laborers on farms. Thirty eight families have participated in the survey on user's perception. Adults are mostly illiterate and children are students in school in most of these families. Fig. 7 shows distribution of families with number of literate members. Prevailing trend of education was significant and 95% of the families were of the opinion that they can afford education for children. 54% of the respondents expressed high satisfaction level with education facility whereas 46% were reasonably satisfied.

Information on source of water for the population was obtained. The major sources of drinking water include ground water from open well, hand-pumps and bore wells. Nearly 60% of the respondents are dependent on several hand-pumps near to their localities for water. Open wells are used by nearly 35% of the respondents whereas, only 5% of them expressed having access to bore wells fitted with water-lifting motorized pumps.

3.4. Acceptability of fluoride mitigation approach

Due to several awareness programs by multilateral organizations and NGOs in these areas, people are aware of health problems associated with high fluoride levels in ground water. The survey reveals that children are more severely affected (78%) as compared to adults (22%). A large section of the respondents (64%) expressed favorable opinion as regard acceptance of defluoridation technique if offered to them. The unwilling population was about 25% and 11% of the population did not express their opinion. The household methods developed for rural areas were demonstrated to the local population and feedback on suitability, cleanliness, simplicity and safe nature of methods was derived by personal interview with the respondents. Opinions expressed about methods are presented in Fig. 8. Wherein 38% of the respondents/users selected loose adsorbent treatment, 28% users opted for bamboo column and 21% opted for sachet method as most suitable method. About 3% of users did not express their opinion. About 10% of users had different opinion and were expecting alternate more suitable method for fluoride removal.

Similar analysis was carried out for assessing the simplicity of method, about 57% of the respondents were of the opinion that loose sorbent method is simplest approach, 24% opted for sachet and 8% opted for bamboo column method. Loose sorbent treatment was identified as cleanest method by nearly 47% of the user's in spite of direct addition of adsorbent to water. This may be attributed to subsequent step of filtration through cloth for separation of adsorbent from water. 27% of the respondents opined that the next cleaner method was bamboo column. No respondent was of the opinion that none of the method is clean. Only 9% of the respondents were of the opinion that all methods were clean to be used for drinking water treatment. Similarly, taking into account safety and health considerations, the respondents have identified loose sor-

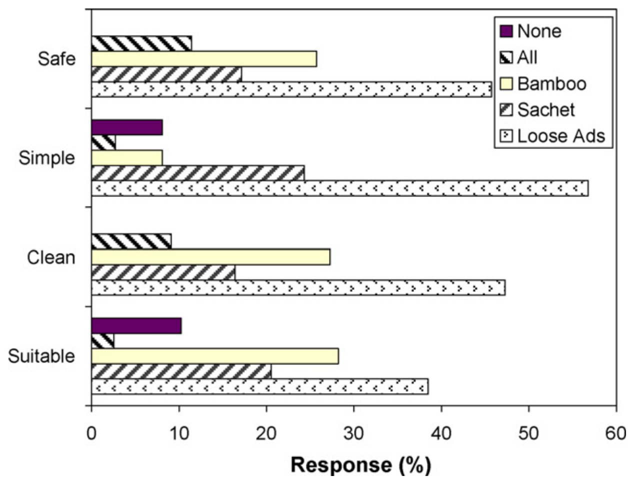

Fig. 8. Perception of user's regarding suitability, simple, clean and safe of methods for fluoride treatment.

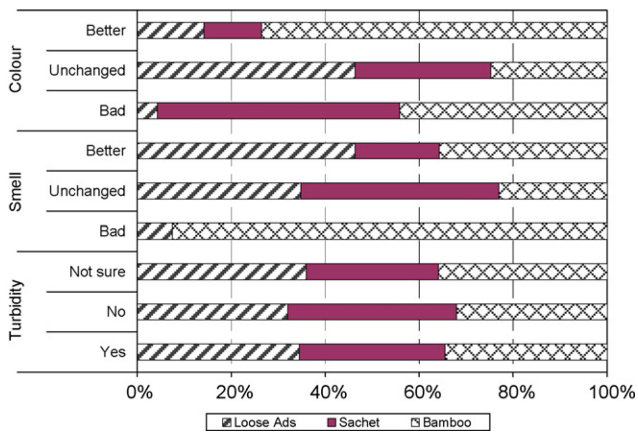

Fig. 9. Perception of user's regarding individual methods about smell, color and turbidity of treated water.

bent method followed by bamboo column method as safe method relatively free of health hazards.

According to respondents/user's perception loose adsorbent treatment emerges out to be the most favored method based on different criteria including suitability, simplicity, cleanliness and safety.

3.5. Perception about individual treatments

Respondent/user has also been interviewed for their perception regarding individual treatment method with specific reference to change in smell, color and turbidity in treated water as compared to raw water. Most of the respondents were of the opinion that the odour of treated water as compared to raw water was remained unchanged. As shown in Fig. 9 the percentage of respondents with this opinion were 76%, 92% and 50% for loose adsorbent, sachet and bamboo column methods, respectively. Some of the respondents/users opined improvement in odour in case of loose sorbent/sachet method whereas bamboo column treatment was imparting bad odour as expressed by about 33% of respondents/users. The observations pertaining to change in color by different respondents are presented in Fig. 9 wherein majority of the respondents (93.5%) were of the opinion that the color of water remained unchanged using loose sorbent treatment. A few users had unfavorable opinion regarding the color of treated water wherein 3.23% of the respondents/users opined that color was unacceptable (or bad). An equal percentage (3.23%) of users was of the opinion that treatment resulted in improvement in color. One of the major observations of the respondents was increased turbidity in treated samples as compared to untreated samples. Turbidity was major concern by most of the users for all three treatment methods. About 48–50% of the respondents/users were of the opinion that the treated water was turbid. This turbidity may be attributed to the presence of fines from adsorbent. These fines can be removed by filtration. The results pertaining to laboratory studies to decrease turbidity indicate that the treatment step has to be followed by filtration step using ordinary filter paper to attain turbidity limit of 10 NTU.

4. Conclusions

A high concentration of fluoride above prescribed limit of 1 mg/L is prevailing in the districts of Dhar and Jhabua of Madhya Pradesh state. The problem is acute at several locations with fluoride concentration in the range of 9–14 mg/L. Water quality assessment and subsequent statistical derivation of correlation of fluoride concentration with physicochemical parameter suggests a possible strong correlation with pH only. However identification of a surrogate parameter for fluoride may require more extensive monitoring of water quality.

For treatment of small quantities of water required for drinking and cooking, a household treatment is appropriate. Three potential methods have been developed using proprietary adsorbents for application in rural households. The user's perception study conducted on these method reveals that population in the study area is willing to accept the fluoride removal treatment and would prefer loose adsorbent method over other methods. Even considering criteria of suitability, simplicity, cleanliness and safe methods, according to user's perception loose adsorbent treatment emerges out to be the most favored method.

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