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Physicochemical Process for the Reduction of Excessive Fluoride Contents in Potable Water Using Indigenous Materials

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Abstract. The chemical examination of drinking water samples colleted from twenty one sampling sites in a village of Khyber Agency (Pakistan) have been reported, described and discussed. It was noted that the quality of drinking water was poor in more than 60% of the samples collected, in respect of its excessive fluoride contents, which was found in the range of 0.27-5.03 mg/l, and was therefore many times higher in most of the samples than the WHO recommended limits of 0.5 mg/l. The higher concentration of fluoride in the potable water is considered to be the main cause of dental decay among the inhabitants of the area. Certain naturally occurring materials like plastic clay, bauxite, and high alumina clay were applied to remove the excessive fluoride contents from the potable water of the area. Fluoride removal from the potable water was effectively achieved using a mixture of indigenous plastic clay, bauxite, and high alumina clay in different ratios with sand. The process is cheap and simple, with 86% removal of fluoride contents achieved from the drinking water.

Keywords: fluoride removal, water fluoride, fluoride sorption, fluoride water treatment

Introduction

Most of the diseases in the developing countries are caused by the consumption of water of poor quality (Dix, 1981; Holden, 1970; Feachem *et al*., 1970). Poor quality of water may be either due to the presence of harmful bacteria or lack/over abundance of certain chemical entities (Holden, 1970). Increasing agricultural activities, industrial development, poor sanitation, and unhygienic practices by the public in general are regarded as the main factors responsible for deteriorating the quality of water.

Excessive concentration of fluoride in water causes fluorosis, a disease in which the teeth first change to brown and then their decay occurs gradually (Jolly *et al*., 1968). Studies have also shown that long-term consumption of fluoridated water increased the risk of hip fracture, especially in women aged 65 years and older (Jacobsen *et al*., 1990). It has been reported that fluorosis may occur when the fluoride level in water exceeds the limit of 3.5 mg/l (Holden, 1970). The high fluoride content may also cause weakening of bones, weight loss, and thyroid and kidney injury. It has been also revealed that fluoride is a mutagen (an agent causing genetic damage), a carcinogen (an agent causing cancer), and a cancer promotor on the basis of laboratory and clinical studies on the cells of animals and humans (Jacqmin-Gadda *et al.,* 1995).

Fluoride occurs naturally in many water supplies. Studies on the assessment and control of nitrates and fluorides in some cities of southern Punjab (Pakistan) have been carried out (Ahmad, 2000), which indicate excessive fluoride content in 29.7 % of the total samples analyzed. These studies have also reported defluoridization with alumina.

The high fluoride levels in drinking water and its impacts on human health, in many parts of India have been investigated (Bower and Hatcher, 1967), which have indicated that the fluoride-bearing minerals in the rocks and soils are the cause of high fluoride content in the groundwater. Studies have been, therefore, conducted to develop a suitable low-cost method for the removal of fluoride present naturally in the groundwater. The ability of red soil to adsorb fluorine from solutions has also been reported by some researchers (Fluhler *et al.,* 1982; Gupta *et al.,* 1982). Some studies have also attempted to remove fluoride by using columns of natural materials, such as red soil, charcoal, brick powder, fly ash, and serpentine (Chidambaram *et al.,* 2003).

The inhabitants of the Federally Administered Tribal Areas (FATA) of Pakistan face serious problem of tooth decay during their early age. The cause of early tooth decay was noted to be due to excessive fluoride content. The conclusion was based on the chemical analysis of the potable water consumed by these inhabitants. The presence of fluoride in the potable water bodies is probably due to the occurrence of fluoride bearing rocks and minerals, such as phosphorite (fluroapatite), calcite, dolomite, etc., in the area. The present study deals

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with the quality assessment of the drinking water of the area, and attempts to develop a procedure for the reduction of excessive fluoride content to the desired WHO level using natural materials, such as plastic clay, bauxite, high alumina clay, and apatite.

Materials and Methods

Sampling. Water sampling was done from Sher Bridge village, located 15 km away from Warsak Road, Peshawar, towards North in the Mula Gori Khyber Agency, Pakistan. The village is located on the side of a straight road of 4½ km long. It consists of about 200 houses constructed adjacent to each other. A canal, Warsak Canal, passes near the village, however, the main source of drinking water are the traditional wells. Every household has a well, and people use the water of these wells for drinking, bathing and washing purposes. Since the main source of drinking water in the area under study are the wells, these were randomly selected for the collection of water samples. Plastic bottles (1.5 liter capacity), used for sampling, were carefully cleaned with a non-ionic detergent, followed by rinsing with HCl and finally with distilled water. All necessary standard precautions were taken while filling up the sample bottles with the tube-well water and in their transport and storage. The wells selected for sampling in different localities were well-used by the inhabitants of the area. Details of sampling locations are given in Table 1.

Chemical reagents and analytical procedures. All the reagents used were of Analar grade (Merck/BDH). Chemical evaluation of each sample was carried out in accordance with the standard methods (APHA/AWWA/WEF, 1998). pH was determined on the spot with a potable pH meter (Corning). Sodium and potassium were determined by flame photometer (model PFP-7, Jenway, England). The samples were subjected to the instrument for direct readings from the digital display of the machine and stable values were reported. Conductivity was determined by a conductivity meter (Jenway 4060, England). The fluoride contents were determined by ion selective electrode (ion meter Jenway 3345, England), using certified reference materials in the form of standard solution of fluoride (1000 ppm) and ionic strength adjusting (ISA) solution for calibration of the equipment.

Results and Discussion

General physicochemical analysis of the studied water samples. Twenty one samples were chemically evaluated for 13 parameters each (Tables 2 and 3). It is evident from the chemical analysis that some of the parameters exceed the permissible limits as suggested by World Health Organization

Table 1. Details of sampling location

Sample number	Sampling point	House identity	Nature of source	Water collection depth
S-1	2 km from Masjid-e-Noor towards North	House of Haji Iftikhar Mir	well	45 ft
S-2	1½ km from Masjid-e-Noor towards North	House of Hatiat Subedar	well	45 ft
S-3	1½ km from Masjid-e-Noor towards North	House of Haji Kanzal Gul	boring	30 ft
S-4	1¼ km from Masjid-e-Noor towards North	House of Razi Gul	boring	30 ft
S-5	1 km from Masjid-e-Noor towards North	House of Haji Aziz Muhammad	boring	70 ft
S-6	½ km from Masjid-e-Noor towards North	House of Qayyum	well	60 ft
S-7	near Masjid-e-Noor	House of Haji Zair Gul	well	60 ft
S-8	Masjid-e-Noor	Masjid	well	60 ft
S-9	¼ km from Masjid-e-Noor towards South	House of Haji Shah Nawaz	well	60 ft
S-10	½ km from Masjid-e-Noor towards South	House of Bhai Khan	well	60 ft
S-11	1 km from Masjid-e-Noor towards South	House of Shah Wali Khan	well	75 ft
S-12	1¼ km from Masjid-e-Noor towards South	House of Siar Muhammad	well	45 ft
S-13	Warsak Canal; 2 km from Masjid-e-Noor towards North	Warsak Canal	-	-
S-14	2½ km from Masjid-e-Noor towards North	House of Mulk-e-Awan	well	55 ft
S-15	Sheikhan Killey	House of Siarullah	well	75 ft
S-16	1½ km from Siarullah well	House of Muhammad Tariq	well	45 ft
S-17	1 km from Siarullah well	House of Azizullah	well	60 ft
S-18	1 km from Siarullah well	House of Daud Khan	well	135 ft
S-19	Government High School, Paindi Lalma	School	well	30 ft
S-20	Warsak Canal; 2 km from Sher Bridge village	Warsak Canal	-	-
S-21	2½ km from Sher Bridge towards East	House of Noor Islam	well	105 f

(WHO, 1999). Sample numbers 17 and 18 contained calcium content of 500 mg/l and 332 mg/l, respectively, which are above the WHO limits for drinking water. Similarly, the magnesium content in sample numbers 17 and 18 also exceeded the permissible limits of WHO. Both the samples had the magnesium content of 240 mg/l and 168 mg/l, respectively. The excess of magnesium causes gastrointestinal problems. Sample numbers 6, 7, 8 and 18 contained chloride contents of 283 mg/l, 262 mg/l, 380 mg/l and 284 mg/l, respectively, which render these samples unfit for drinking purpose, since their chloride contents were above the WHO permissible level of 250 mg/l. Similarly, sample numbers 6, 7, 8, 9, 12, 16, 17, 18, 19 and 21 had sulphate contents of 387 mg/l, 271 mg/l, 272 mg/l, 261 mg/l, 376 mg/l, 368.8 mg/l, 314.88 mg/l, 384 mg/l and 280.32 mg/l, respectively, which made these samples unfit for drinking purpose, as the sulphate contents of these samples exceeded the WHO permissible limits of 250 mg/l. The sodium content in almost all the samples was within the WHO permissible limits of 200 mg/l. The potassium content in all the samples, with the exception of sample numbers 9, 13, 14, 19, 20 and 21, were much higher than the permissible level of WHO (75 mg/l). Nitrate contents, with the exception of sample number 11, were within the permissible WHO limit of 45 mg/l (WHO, 1999).

The fluoride content is considered to be the major issue in respect of the present studies as the inhabitants of the area were observed to have tooth decay problems due to excessive fluoride content in their drinking water supplies. The fluoride content in the samples varied from 0.27-5.03 mg/l. All the samples, except sample numbers 15, 17, 20 and 21 had higher fluoride content as compared to the recommended international standards (WHO, 1999), with the maximum limit of 0.5 mg/l, as shown in Table 3. Among the samples collected and analyzed, 60% were found to have much higher fluoride content, which is the main reason of the dental decay problem among the inhabitants.

Fluoride reduction in potable water. Attempts were made during the present study to remove fluoride by the column procedure, using indigenous natural raw materials. The removal of fluoride was tested using different materials, such as plastic clay, bauxite, high alumina clay, and their mixtures in different ratios. The materials used for defluoridization were not pre-

Table 2. pH, conductivity, total hardness and alkalinity in the water samples collected from Sher Bridge village of Khyber Agency, Pakistan (results are expressed in mg/l)*

Sample number	pH	Conductivity ($\mu S/cm^3$)	Hardness	Calcium	Magnesium	Total alkalinity
S-1	7.24±0.01	1468±1	288±2	244±2	44±2	356±2
S-2	7.15±0.01	755±1	224 ±2	140±2	84±2	356±2
S-3	7.48±0.01	738±1	148±2	80±2	68±2	276±2
S-4	8.08±0.01	1144±1	148±2	76±2	72±2	308±0
S-5	7.90±0.1	1309±1	208±2	112±2	96±2	400±2
S-6	7.76±0.01	2000±1	152±2	84±2	68±2	384±2
S-7	7.36±0.01	983±2	228±0	144±0	84±0	252±0
S-8	7.71±0.01	1195±1	204±2	136±2	68±2	268±2
S-9	7.67±0.01	418±1	232±2	140±2	92±2	212±2
S-10	7.67±0.01	820±1	180±2	116±2	64±2	248±2
S-11	7.54±0.01	962±1	240±4	168±4	72±4	236±0
S-12	7.91±0.10	1362±1	176±2	124±2	52±2	304±2
S-13	8.14±0.00	385±1	180±2	96±2	84±2	176±2
S-14	7.69±0.01	589±0	200±2	140±2	60±2	184±2
S-15	7.18±0.01	1016±1	232±2	128±2	104±2	338±2
S-16	7.57±0.01	1428±1	328±2	192±2	136±2	324±0
S-17	7.43±0.01	2000±1	740±2	500±2	240±2	292±2
S-18	6.81±0.01	1742±1	500±2	332±2	168±2	272±2
S-19	7.63±0.01	954±1	340±2	200±2	140±2	192±2
S-20	8.16±0.00	403±1	180±2	88±2	92±2	156±2
S-21	7.58±0.01	624±1	260±2	140±2	120±2	184±2
WHO****	**6.5-9.20**	**-**	**500**	**250**	**150**	**500**

* values are mean ±SD for readings analyzed individually in triplicate; ** recommended/permissible limits (WHO, 1999)

treated and the size fractions taken were of 2.5µm mesh. The surface characters of the solid materials used were not considered here. The mixture of plastic clay, bauxite and sand in the ratio of 1: 1: 2 was found to be most effective for fluroide removal (Table 4). As evident from Fig. 1 and 2, the removal of fluoride was reduced with higher retention time in the column, probably caused due to saturation of the anion exchange sites. The flow rate of 2.5 ml/min was found to be satisfactory (Fig. 3), whereas higher flow rates gave high fluoride values in the effluent water. In general, alumina compounds were found to be good fluoride removers because of the interaction between aluminum and flouride molecules (Bullusu and Nawlakle, 1998). The indigenous bauxite ore used in these studies contained 70% of alumina, which provided an additional source for fluoride adsorption over its surface.

As shown in Table 4, the mixture of plastic clay and sand in the ratio of 1 : 1 resulted in 78.33% fluoride removal, whereas 79.52% of fluoride removal was noted with a mixture of bauxite and sand. The mixture of high alumina clay and sand had a negligible effect on the removal of fluoride from potable water. The maximum removal of fluoride (86.08%) was observed with a mixture of plastic clay, bauxite and sand in the ratio of 1 : 1 : 2 with 5 min retention time.

Fluoride reduction in standard fluoride solutions. The effect of different materials on the removal of fluoride from standard fluoride solutions is illustrated in Table 5. Standard fluoride solutions of 10 mg/l were treated using the same process as was used in the case of collected water samples from wells in the study area for the reduction of fluoride content. It is evident from Table 5 that a mixture of high alumina clay and sand (1 : 1) resulted in 30% removal of fluoride, whereas with a mixture of plastic clay and sand in the ratio of 1 : 1, 84.5% reduction in fluoride content was observed. It was also noted that the mixture of bauxite and sand (1 : 1) gave somewhat better results with 92% removal of fluoride. Different retention times were also studied, and it was observed that the retention time of 5 min resulted in maximum removal of fluoride content (Fig. 2). The observation also revealed that after a period of 30 min retention time, the removal of fluoride was markedly reduced due to saturation of the anion exchange sites as shown in Table 5. The best results were achieved using a mixture of bauxite, plastic clay and sand in the ratio of 1 : 1 : 2, with 94.7%

Table 3. Chlorides, sulphate, nitrates, nitrites, alkali metals (Na^+, K^+) and fluoride contents in water samples collected from Sher Bridge village of Khyber Agency, Pakistan (results are expressed in mg/l)*

Sample number	Chloride	Sulphate	Nitrate	Nitrite	Sodium	Potassium	Fluoride
S-1	104±2	161.28±2	2.81±0.01	traces	2.5±0.1	170±0	1.12±0.01
S-2	43±2	119.00±2	7.95±0.01	traces	3.0±0.1	90±1	2.13±0.01
S-3	42±2	103.68±2	10.25±0.01	nil	38.4±0.0	88±1	2.23±0.01
S-4	89±2	180.48±2	23.70±0.01	nil	0.8±0.1	178±0	2.88±0.01
S-5	155±2	230.40±1	5.76±0.01	nil	2.8±0.1	196±0	3.77±0.01
S-6	283±0	387.84±2	2.01±0.01	nil	2.8±0.1	293±1	5.03±0.01
S-7	120±2	271.48±2	16.03±0.01	nil	3.4±0.1	131±0	2.95±0.01
S-8	262±0	272.64±0	5.46±0.01	nil	2.4 ±0.1	163±0	2.86±0.01
S-9	40±2	261.12±2	1.21±0.01	traces	2.0±0.1	19±1	0.55±0.00
S-10	96±2	234.24±2	29.41±0.01	traces	2.0±0.0	110±0	3.23±0.01
S-11	122±2	249.60±2	48.29±0.01	0.3±0.0	6.6±0.1	112±1	2.61±0.00
S-12	110±2	376.32±1	28.12±0.01	nil	1.4±0.1	218±1	2.89±0.01
S-13	44±2	203.52±2	3.35±0.01	nil	4.3±0.1	19±1	0.57±0.00
S-14	62±2	241.92±2	21.67±0.01	nil	2.1±0.1	69±1	0.70±0.00
S-15	86±2	203.52±2	27.45±0.01	nil	11.5±0.1	150±1	0.30±0.00
S-16	178±2	330.24±1	nil	traces	13.5±0.1	161±1	1.05±0.01
S-17	380±0	364.80±2	nil	traces	30.6±0.1	228±1	0.45±0.01
S-18	284±2	314.88±2	nil	til	20.0±0.0	150±1	4.00±0.01
S-19	62±2	384.00±1	nil	nil	7.7±0.1	71±0	0.54±0.01
S-20	36±2	199.68±2	nil	nil	6.5±0.1	18±1	0.27±0.00
S-21	48±2	280.32±2	nil	nil	4.1±0.1	43±0	0.35±0.01
WHO**	**250**	**250**	**4.5**	**0.1**	**200**	**75**	**0.5**

* values are mean ±SD for readings analyzed individually in triplicate; ** recommended/permissible limit (WHO, 1999)

fluoride reduction in the standard fluoride solution for a retention time of only 5 min.

Methodology for fluoride reduction. Out of the 21 samples collected from the area, sample number 6 had the highest fluoride content of 5.03 mg/l. This sample was studied for the reduction of fluoride by the column process developed presently. Sample number 6 was therefore passed through the columns containing different adsorption materials, namely, plastic clay, bauxite, high alumina clay, and sand. The materials used for defluoridization were not pre-treated and the size fractions taken were of 2.5 µm mesh. The flow rate of the columns was adjusted at 2.5 ml/min on the basis of maximum removal of fluoride with this flow rate as evident from Fig. 3. The size of the bedding materials, plastic clay, bauxite and sand were kept in the column at 2 cm, 2 cm and 4 cm, respectively. The effluents were collected at intervals of 5, 10, 20, 30, 40, 50 and 60 min. The collected effluent samples were analyzed for fluoride content by ion selective electrode using a series of fluoride standards for calibration of the equipment. Four columns were set up in proportion with the adsorption capacity of the four materials mentioned above. Different retention times were studied as reported in Tables 4 and 5, and a retention time of 5 min was found to be satisfactory for maximum reduction of fluoride content, both in the potable water sample and in the standard fluoride solutions. Higher retention time gave higher fluoride in the collected water due to saturation of anion sites of the materials. The standard fluoride solution of 10 mg/l was also passed simultaneously through all the columns and the collected effluent samples were analyzed for fluoride content. The results achieved are summarized in Table 5. The chemical analysis of the effluent sample after treatment was also carried out for parameters other than fluoride and it was noted that there was no significant difference in the concentration of such parameters as pH, Ca, Mg, SO_4, Cl, NO_3.

Table 4. Reduction of fluoride content in water samples* collected from Sher Bridge village of Khyber Agency, Pakistan on passing through columns using different indigenous materials

Retention	Bau + PC + Sand			PC + Sand			Bau + Sand			HAC + Sand		
time (min)	F^- removed (mg/l)	F^- retained (mg/l)	F^- removed (%)	F^- removed (mg/l)	F^- retained (mg/l)	F^- removed (%)	F^- removed (mg/l)	F^- retained (mg/l)	F^- removed (%)	F^- removed (mg/l)	F^- retained (mg/l)	F^- removed (%)
5	4.33	0.70	**86.08**	3.94	1.09	**78.33**	3.90	1.13	77.53	zero	5	**zero**
10	4.18	0.85	83.10	3.92	1.11	77.93	4.0	1.03	**79.52**	zero	5	zero
20	3.75	1.28	74.55	1.00	4.03	19.88	3.92	1.11	77.93	zero	5	zero
30	3.35	1.68	66.60	0.10	4.93	1.99	3.55	1.48	70.58	zero	5	zero
40	3.18	1.85	63.22	-0.03	5.06	zero	3.09	1.94	61.43	zero	5	zero
50	3.20	1.83	63.61	-0.52	5.55	zero	3.12	1.91	62.03	zero	5	zero
60	3.20	1.83	63.61	-1.07	6.10	zero	3.05	1.98	60.64			

Bau = bauxite; PC = plastc clay; HAC = high alumina clay; * volume of water sample used was 10 ml

Table 5. Reduction of fluoride content in the standard fluoride solution* on passing through columns using different indigenous materials

Retention	Bau + PC + Sand			Bau + Sand			PC + Sand			HAC + Sand		
time (min)	F^- removed (mg/l)	F^- retained (mg/l)	F^- removed (%)	F^- removed (mg/l)	F^- retained (mg/l)	F^- removed (%)	F^- removed (mg/l)	F^- retained (mg/l)	F^- removed (%)	F^- removed (mg/l)	F^- retained (mg/l)	F^- removed (%)
5	9.47	0.53	**94.7**	9.2	0.8	**92.0**	8.45	1.55	**84.5**	3	7	**30.0**
10	9.28	0.72	92.8	8.98	1.02	89.8	8.32	1.68	83.2	2.79	7.21	27.9
20	8.82	1.18	88.2	8.76	1.24	87.6	8.09	1.91	80.9	0.2	9.8	2.0
30	8.68	1.32	86.8	8.68	1.32	86.3	7.94	2.06	79.4	zero	zero	zero
40	8.54	1.46	85.4	8.40	1.60	84.0	7.93	2.07	79.3	zero	zero	zero
50	8.39	1.61	83.9	8.33	1.67	83.3	7.79	2.21	77.9	zero	zero	zero
60	7.99	2.01	79.9	8.28	1.72	82.8	7.68	2.32	76.8	zero	zero	zero

*volume of water sample used was 10 ml containing standard fluoride (10 mg/l)

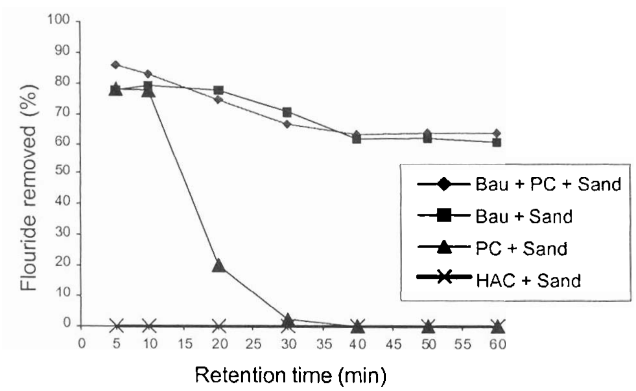

Fig. 1. Fluoride removal (%) from the collected water sample using different indigenous materials; Bau = bauxite, PC = plastic clay, HAC = high alumina clay.

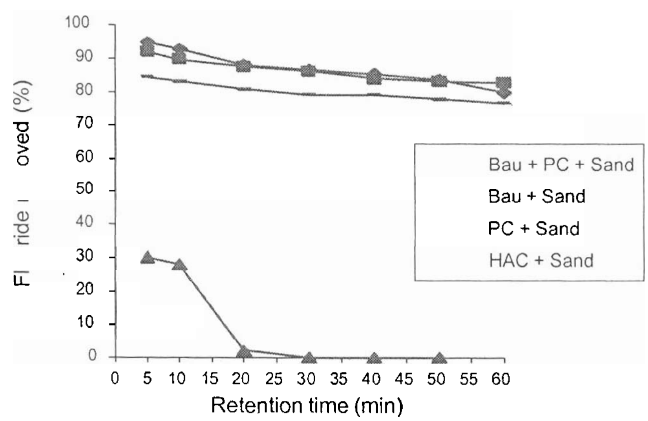

Fig. 2. Fluoride removal from the standard fluoride solution (10 mg/l) using different indigenous materials; Bau = bauxite, PC = plastic clay, HAC = high alumina clay.

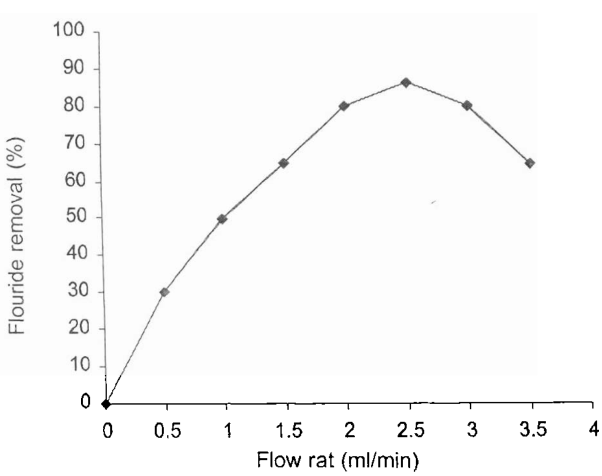

Fig. 3. Fluoride removal as a function of flow rate (ml/min).

Conclusion

The study reveals that among the four materials used for defluoridization, a mixture of plastic clay, bauxite and sand with a ratio of 1 : 1 : 2 had the best defluoridization capacity as compared to the others, because of the fact that both the bauxite and plastic clay materials had oxides of aluminum as their major component and were therefore considered to be good adsorbers of fluoride content.

The present studies further highlight the advantage of using low cost and locally available natural raw materials for the removal of excessive fluoride contents from drinking and domestic water. Moreover, the process is economically viable for most of the population living in the area below the poverty line that cannot afford sophisticated treatment equipment or bottled water for their daily consumption. The simple low technology process may be utilized by domestic consumers of the area for their daily use, and as such the problem of their early teeth decay may be prevented.

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