INTERNATIONAL JOURNAL OF APPLIED BIOLOGY AND PHARMACEUTICAL TECHNOLOGY

Volume: 2: Issue-4: Oct - Dec -2011 IJABPT *ISSN 0976-4550*

FLUORIDE POLLUTION IN GROUND WATERS OF KANDUKUR REVENUE SUB-DIVISION OF PRAKASAM DISTRICT IN A.P., INDIA AND BATCH MODE DEFLUORIDATION USING ACTIVE CARBONS OF SOME PLANT BYPRODUCTS AS ADSORBENTS

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ABSTRACT : Fluoride is an acute toxin and is deemed to be slightly more dangerous than even lead. 48 water samples collected from hand pumps and bore wells belonging to 24 gram panchayats (villages) of Kanduku revenue sub division of Prakasam district in Andhra Pradesh, were chemically analyzed for fluoride ion concentrations. High and low fluoride containing regions were identified on the basis of fluoride levels in the water samples and also on the prevalence rate of dental and skeletal fluorosis of the study area. Further, water samples containing high fluoride levels were tried for defluoridation by employing Active Carbons of abundantly available low-cost plant byproducts. These materials under optimum conditions of adsorbent dosage, time of equilibration and pH, were found to be successfully decreasing the fluoride ion concentration below permissible limits without disturbing drinking water quality standards.

Key Words: Kandukur mandal, Field survey, Defluoridation of water, Plant byproducts, Carbon adsorbents

INTRODUCTION

Good drinking water quality is essential for the well being of all people. Unfortunately in many countries around the world, including India, some drinking water supplies have become contaminated with respect to hazardous ions, which have impact on the health and economic status of the populations. Fluoride is a ubiquitous element present in earth's crust and is also being added to the environment anthropogenically. It is the most electronegative of all elements. Fluorine is found in the soil and the content of Fluorine in the lithosphere varies between 100 and 1500 g/ton[1]. Fluoride has gained importance due to its dual influences on human beings. In lower concentrations, Fluoride is an essential nutrient which aids in the formation of bones, prevents tooth decay, etc., whereas in higher concentrations it causes fluorosis, brittling of bones, curvature of bones, dwarfishness, mental derangements, cancer, etc. and in extreme cases even death.

It is estimated that around 260 million people worldwide (in 30 countries) are drinking water with Fluoride content more than 1.0 mg/L. In India alone, endemic Fluorosis is thought to affect around one million people and is a major problem in 17 of the 25 states, especially Rajasthan, Andhra Pradesh, Tamil Nadu, Gujarat and Uttar Pradesh. In India totally 25 states have been reported as fluoride affected areas but severe problem occurred in the states of Andhra Pradesh[2-5]. Fluoride could be found in a number of minerals, of which fluorspar, cryolyte and fluorapatite are the most common. Many epidemiological studies of possible adverse effects of the long-term ingestion of fluoride via drinking water have clearly indicated that fluoride primarily produces effects on skeletal tissues (bones and teeth)[6].

According to WHO standards, in temperate regions, where water intake is low Fluoride level up to 1.5 mg/L is acceptable[7]. The Bureau of Indian Standards, BIS[8], has prescribed a desirable limit and permissible limit of Fluoride in drinking water as 1.0 and 1.5 mg/L respectively.

Several methods are in vogue to remove fluoride from drinking water[9]. Defluoridation of drinking waters is usually accomplished by either precipitation or by adsorption processes. One of the well-known methods called 'Nalgonda Technique' developed by National Environmental Engineering Research Institute, Nagpur, India[10] is a precipitation processes employing alum followed by sedimentation and/or filtration.

In the present work the revenue sub-division of Kandukur in Prakasham Districe of Andhra Pradesh has been chosen for our study as the ill-effects of Fluoride contamination of ground waters such as dental fluorosis, is predominant and there is a huge public cry for protect them from the problems of fluorosis. The present work endeavors to find high and low fluoride containing areas in the revenue sub-division of Kandukur using the isopleth mapping technique and proposing remedial methods in bringing the fluoride concentration within the permissible concentrations by exploring the sorption potentialities of some of Active Carbons prepared from dry fruits of various plants as adsorbents.

Geography

Kandukur revenue subdivision comprises of Kandukur town (a municiplaity) and mandal having more than 24 villages in the Prakasam district in the Indian state of Andhra Pradesh. Kandukur town is located 52.5 km distance from its District Main City Ongole (Figure 1). It is located 281 km distance from its State Main City Hyderabad. Kandukur (Earlier known as Skandapuri) is located at 17°04′N 78°29′E / 17.07°N 78.49°E[11]. It has an average elevation of 632 meters (2,073 ft), Latitude 17.0664 , Longitude 78.4892, Altitude (feet) 2073, Lat (DMS) 17° 3' 59N,Long (DMS) 78° 29' 21E, Altitude (meters) 631. Telugu, the regional language of the state of Andhra Pradesh is spoken in Kandukur. As of 2001 India census[12], Kandukur town had a population of 1,50,084. Males constitute 51% of the population and females 49%. Kandukur has an average literacy rate of 63%, higher than the national average of 59.5%: male literacy is 72%, and female literacy is 55%. In Kandukur, 11% of the population is under 6 years of age. Kandukur has a historic background. Skandapuri which was an area ruled during Srikrishna Devaraya period was renamed as Kandukur. It is now one of the fastest developing towns in the state.

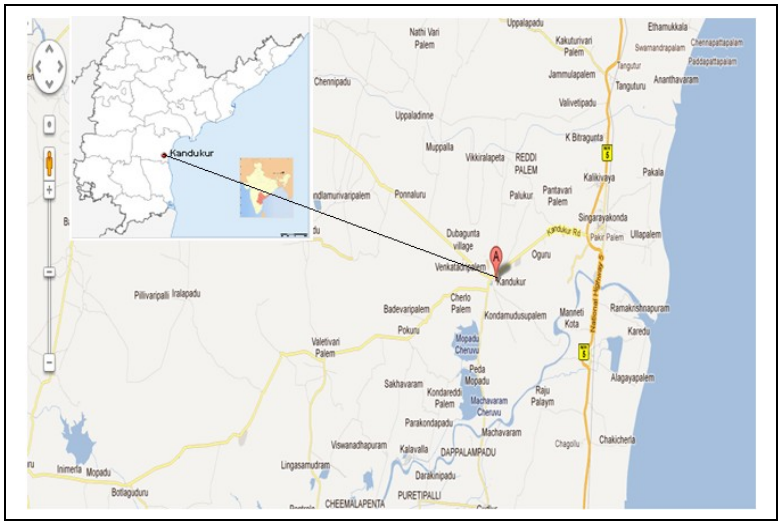

Figure 1: Location of Kandukur mandal in Andhra Pradesh

Methodology

A door-to-door survey was conducted on the residents of the selected villages or locations, in the study area along with the registered medical practitioner. In this survey, people were broadly divided into three categories depending on the age limits between 5 to 15, 15 to 25 and above 25 years of age. In each group totally 25 persons were examined and prepared statistical report. After conformation of fluorosis presence, totally 48 samples were collected from all drinking water sources from all places in the division and water samples were analyzed for fluoride ion concentration Spectrophotometrically using well-known SPADNS method[13].

Sample collection

Water samples have been collected from all the existing sources of drinking water in the study area for investigation and chemical analysis. For the present investigation, separate sets of samples were collected for chemical analysis from the source. The bottles for sample collection have been thoroughly cleaned by rinsing with 8M HNO_3 solution, followed by repeated washing with deionized distilled water. They are further rinsed with sample water before collection. Physico-chemical analysis was done by standard procedures [13].

Material preparation

Defluoridating materials were prepared from the dry fruits, collected from the plants *Typha angustata* (TAC) belongs to *Typhaceae family (Figure 2a)*; *Lagenaria siceraria* (LSSC) *(Figure 2b) belongs to* cucurbitaceae family and *Acacia farnesiana* (AFC) *(Figure 2c)* : from Mimosideae family in the plant kingdom. These materials are available as agricultural wastes and were carbonized at 400 to 500ºC in muffle furnace. The prepared carbons were chemically treated with 0.5 M HNO_3 solution and then washed with distilled water, dried and finally sieved in to 70 μ□ particles size.

Figure 2: a) *Typha angustata* *b)Lagenaria siceraria*

c) Acacia farnesiana

Defluoridation method

1.0 g of adsorbent carbon sample was mixed with 100 ml of water samples and stirred at 200 rpm speed on Remi shaker for 30 minutes. Solution was filtered through Whatman No 42 filter paper and the filtrate was examined for further fluoride ion concentration by SPADNS method[13] using U.V- Visible Spectrophometer (Model No: Elico U.V-2600). Using the three prepared active carbons as adsorbents, the optimization of experimental conditions such as agitation time, adsorbent dosage, and pH have been studied for maximum removal of fluorides adopting batch mode studies. The optimum condition for maximum extraction fluoride was found to be: 45-60 minutes agitation time; 3-6 g/L as adsorbent concentration; effective pH range is 6.5-7.5. The same conditions were applied in defluoridation of drinking water samples in batch mode study.

RESULTS AND DISCUSSION

Ground water is the only source of potable water for majority of people in the study area. However, the inhabitants here are averse to drink bore well water or water from public water system. They say that water drawn from great depths is not tasty and hence their preference to hand pump water. A survey of residents of the selected villages in the study area on the impact of water used for drinking on health of the habitants revealed that, most of the residents suffer from dental discoloration, early tooth decay and bone deformations. The practicing physicians of the study area also confirmed our observations. A perusal of the results would reveals that dental discoloration in female population was more common than the male population. Moreover, the ill-effects were observed to be more prevalent between 5 and 15 years of age and found to be decreasing with increase in age.

Source of fluoride and distribution in the study area

The main source of fluoride and their concentrations in ground waters is geological source. A colorless and odorless natural pollutant fluoride comes in to contact with ground water from its source of origin, the rock minerals. The fluoride-affected area of Kandukur revenue sub-division is full of geological futures, such as joints fracture and faults. Besides, ancient and younger sedimentary rocks, unconsolidated materials, like gravel, sand, sand stone and clay with intrusive granite and quartz shale, which are known to have high fluoride content, are present in the area. From the Table No. 1, it is evident that the concentration of fluoride in all samples of study area, has varied from 1.07 to 2.62 in the case of hand pump samples and 1.25 to 2.8 mg/L with bore well water samples. Further it is can infer that, the samples of the study area can be broadly classified into three categories depending upon the concentration of fluoride ion: between 0.0 to 0.50, 0.51 to 1.50, 1.51 to 2.00 and above 2.00 mg/L. It is observed that 17 water samples from bore wells and 17 samples from hand pump waters, have more than permissible limit of F^- ion (1.5 ppm) concentration. But only 7 water samples each from bore well and hand pump have less than 1.5 mg/L concentration and so, these seven sample station waters have been relatively less contaminated with respect to Fluoride ion and they may be used as water source for drinking purpose with some reservation as non of these samples have less than 1.0mg/L of fluoride.

Table No. 1: Classification of fluorotic areas basing on the concentration limit

S.No	Conc. Limit (mg/L)	Hand pump	Bore well
1	0.0-0.50	0	0
2	0.51-1.50	7	7
3	1.51-2.00	4	12
4	2.01-above	13	5

Defluoridation studies of potable water samples

The results of fluoride analysis of all the water samples collected from various villages of Kandukur sub division have been presented in the Table Nos: 2 and 3. The data indicates that most of the samples contain excess of fluoride beyond the permissible World Health Organization limit (1.5 mg/L). Hence in the present work, the defluoridation studies have been carried out on these particular samples using Active Carbons prepared from waste materials of plant origin adsorbents namely: Typha angustata carbon (TAC), Lagenaria siceraria shell carbon (LSSC) and Acacia farnesiana carbon (AFC). In order to reduce the fluoride content below the permissible limit, optimum experimental conditions for dosage of adsorbent, fluoride ion concentration and equilibration time, reported in our previous works have been used. For water samples, which contain fluoride range between 2.0 and 3.0 mg/L, the dose of adsorbent used was 6.0 g/L and for those water samples, which contain fluoride, ranging from 1.5 to 2.0 mg/L, the dose of adsorbent was 4 g/L. The contact time was fixed between 45-60 minutes with constant stirring at 200 rpm speed. The concentrations of fluoride ion in these samples after defluoridation was analyzed and reported in the same Table Nos:2 and 3.

Table No:2 :Fluoride ion concentration (before and after defluoridation) of hand pump water samples

Sample Station No.	Location of Sample Station	C_i (mg/L) :before defluoridation	C_f (mg/L):after defluoridation		
			TAC	LSSC	AFC
1	Kammarivari palem	2.51	0.83	1.00	1.13
2	Chirrikurapadu	2.38	0.79	0.95	1.07
3	Balijapalem	1.86	0.61	0.74	0.84
4	Pandalapadu	2.62	0.86	1.05	1.18
5	Jillelamudi	1.65	0.55	0.66	0.74
6	*Vikkiralapeta	1.40	::	::	::
7	Palukuru	2.39	0.79	0.96	1.08
8	Narisetty varipalem	2.26	0.75	0.90	1.02
9	Kondi kandukur	2.06	0.68	0.82	0.93
10	Kovuru	2.56	0.84	1.02	1.15
11	Mahadevapuram	2.36	0.78	0.94	1.06
12	Dubagunta	2.10	0.69	0.84	0.94
13	Ananthasagaram	2.45	0.81	0.98	1.10
14	Oguru	2.08	0.69	0.83	0.94
15	Venkatadripalem	2.35	0.78	0.94	1.06
16	Ganigunta	2.30	0.76	0.92	1.03
17	*Kancharagunta	1.25	::	::	::
18	Gallavaripalem	1.58	0.52	0.63	0.71
19	*Kondamudusu palem	1.07	::	::	::
20	*Anandapuram	1.25	::	::	::
21	Mopadu	1.62	0.53	0.65	0.73
22	*Paluru	1.43	::	::	::
23	*M.G.Puram	1.24	::	::	::
24	*Machavaram	1.44	::	::	::

Table No:3: Fluoride ion concentration (before and after defluoridation) of bore well water samples

		C_i (mg/L)(before defluoridation)	C_f (after defluoridation)		
			TAC	LSSC	AFC
1	Kammarivari palem	1.92	0.63	0.77	0.86
2	Chirrikurapadu	2.01	0.66	0.80	0.90
3	Balijapalem	1.51	0.50	0.60	0.68
4	*Pandalapadu	1.42	::	::	::
5	Jillelamudi	1.66	0.55	0.66	0.75
6	Vikkiralapeta	2.1	0.69	0.84	0.95
7	Palukuru	2.25	0.74	0.90	1.01
8	Narisetty varipalem	2.02	0.67	0.81	0.91
9	Kondi kandukur	2.23	0.74	0.89	1.00
10	Kovuru	1.86	0.61	0.74	0.84
11	Mahadevapuram	1.98	0.65	0.79	0.89
12	Dubagunta	1.9	0.63	0.76	0.86
13	Ananthasagaram	1.54	0.51	0.62	0.69
14	Oguru	1.28	0.42	0.51	0.58
15	Venkatadripalem	1.88	0.62	0.75	0.85
16	Ganigunta	1.34	0.44	0.54	0.60
17	*Kancharagunta	1.44	::	::	::
18	Gallavaripalem	1.82	0.60	0.73	0.82
19	*Kondamudusu palem	1.23	::	::	::
20	Anandapuram	1.44	0.48	0.58	0.65
21	Mopadu	1.86	0.61	0.74	0.84
22	Paluru	1.64	0.54	0.66	0.74
23	*M.G.Puram	1.43	::	::	::
24	Machavaram	1.66	0.55	0.66	0.75

*C_i- initial concentration, C_f- final concentration of fluoride ion in mg/L.
*no need to reduce fluoride concentration

A comparative study of the results of some physico-chemical analysis of water from bore well and hand pump before defluoridation and after defluoridation, indicate that water quality parameters like pH, EC, TDS, PO_4^{3-}, SO_4^{2-}, Cl^-, K^+ etc. values were marginally effected in many water samples when the adsorbents TAC,LSSC and AFC were used for the defluoridation. From the data presented in the Table No.2 and 3, it may be inferred that TAC, LSSC and AFC effectively decreasing the fluoride content in water samples to a considerable extent without affecting the permissible limits of other water quality parameters. Further, it was observed that the fluoride concentration after treating the water samples with the TAC,LSSC and AFC, was of the order: TAC<LSSC<AFC and hence the adsorption capacities of the adsorbents is of the order :TAC>LSSC>AFC

CONCLUSION

The results of the present study indicate that the area under study is fully affected with endemic fluorosis and the concentration of fluoride ion in all water sources derived from bore well and hand pumps, varies from place to place. The fluoride contamination may be due to the nature of rock and soil formation. The surface sorption characteristics of TAC, LSSC and AFC have been explored in reducing the fluoride concentration below permissible limts. It is found that these active carbons are effective in the control fluoride ion under optimum, experimental conditions. Further, the low cost adsorbents TAC and LSSC are found to remove fluoride content from potable water to a larger extent compared with the other adsorbent, AFC. Hence these adsorbents can be used for the defluoridation of potable water at house hold level. Finally the results also suggest that the area is fully contaminated with fluoride and the ground waters are not suitable for drinking purpose unless properly treated before consumption.

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