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A COMPARATIVE STUDY OF USING FLY ASH AND MARBLE POWDER FOR FLUORIDE TREATMENT

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ABSTRACT

Fluoride is excessively available in groundwater is very common in India. To knowing the adverse effects of fluoride in water many adsorbents using time to time but due to many side effects of defluoridation we need a perfect method for this. In this research work, we have been applied a comparative study of two different cheap adsorbent. The using adsorbents are Fly ash and marble powder. Here used fly ash, generated from Chula. And pieces of marble collected from the construction area and crushed for use in the laboratory. In this research work Ash, Marble powder is mixed with fluoridated water. After the analysis, we found both of the adsorbents removes fluoride from drinking water but the results were found from fly ash is better than from marble powder.

KEYWORDS: *Adsorption, Defluoridation, Fluoride, Fly Ash, Marble Powder* P^h

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INTRODUCTION

Various Health Impacts of Fluoride

Fluoride is a positively charged ion like calcium (Ca^{++}). It is important for our bones and teeth but having the highest amount of calcium in the body attracts the maximum amount of fluoride. It is deposited in the form of Calcium Fluorapatite crystals in the body. Excess of fluoride above 1.5 mg/L may cause to serious manifestations, which are described below:-

- **Dental Fluorosis:** Problems related to teeth.
- **Skeletal Fluorosis:** Problem-related bones of hands, legs, and joints.
- **Non-Skeletal Fluorosis:** Excessive fluoride can cause several other problems and disorder like neurological, muscular, allergic, gastro-intestinal and urinary disorders.

Table 1: Concentration of Fluoride, Medium and Effects

Concentration of Fluoride	Medium	Effects
1	Drinking Water	Dental caries reduction
2 or < 2 8	Drinking Water	Mottled enamel (dental fluorosis) 10% osteosclerosis
20-80	Drinking Water or food	Crippling skeletal fluorosis
50	Drinking Water or food	Thyroid changes
100	Drinking Water or food	Growth retardation
125	Drinking Water or food	Kidney changes
2.5-5.0	Drinking Acute dose	Death

Note: Concentration given in mg/l or ppm

Common Defluoridation Methods of Drinking Water

Removal of excess fluoride from drinking water called defluoridation. The process of defluoridation can use by following methods:

- The treatment of fluoride water at the source and
- The treatment of fluoridated water at the household level.

Developed countries are using source treatment method (i). Defluoridation is a technical treatment method under the supervision of skilled persons here cost is a limiting factor. But in the less developed countries, the same approach may not be feasible, especially in rural areas, where settlements are scattered. Treatment at the point of use (ii) has several advantages over treatment at the community level. Treatment may only be possible at a decentralized level, for example at the community, village or household level. Costs are lower, as defluoridation can be restricted to the demand for cooking and drinking – usually less than 25% of the total water demand. Chemical treatment of the entire water demand would lead to the production of large volumes of sludge, which requires safe disposal.

It has been found that the defluoridation methods of drinking water are not sufficient when the initial concentration of fluoride in the water is very high and the pH of the untreated water is alkaline. Moreover, different degrees of hardness of water require different concentrations of alum.

Various limitations are found when we use of point of use treatment method like the reliability of the treatment units has to be assured, and that all users should be motivated to use only the treated water for drinking and cooking at the condition is untreated water is also available in the house

National Environment Engineering Research Institute in Nagpur, India (NEERI) has evolved an economical and simple method of defluoridation, which is referred to as the Nalgonda technique. UNICEF has worked closely with the Government and other partners in defluoridation programs in India, where excessive fluoride has been known for many years to exist in groundwater. In the 1980s, UNICEF supported the Government's Technology Mission in the effort to identify and address the fluoride problem: the Government subsequently launched a massive program, still underway, to provide fluoride-safe water in all the areas affected.

The Nalgonda technique has been repeatedly proven to be an economical and effective household defluoridation technique. (Lela Iyenger, March 2005, UNICEF, New Delhi) In this technique, fluoride is precipitated using 500 mg/L of alum and 30 mg/L of lime.

For removal of fluoride from drinking water three mechanisms can be applied:

- Addition of chemicals or Chemical additive methods
- Addition of solid media or Contact precipitation
- Adsorption/ion exchange methods by the help of sold surfaces

Addition of Chemicals or Chemical Additive Methods

Removal of fluoride with the help of addition of different soluble chemicals like lime (with or without magnesium or aluminum salts) into drinking water. In this method, precipitation and adsorption process can be used. The main demerit is when lime and magnesium used for treatment, water is unfit for drinking purpose due to high pH. Alum and a small amount of lime have been used for defluoridation of water.

From the past several years, a very common and popular method has been used for removal of fluoride is **Nalgonda technique** (RENDWM, 1993). Nalgonda is a town of state Madhya Pradesh where this methods first used so that the method is known as Nalgonda technology.

In this method addition of lime (5% of alum), bleaching powder (optional) and alum ($Al_2(SO_4)_3.18H_2O$) in sequence to the water, followed by coagulation, sedimentation, and filtration (L.Iyenger,2005). A higher dose of alum is required for fluoride removal (150 mg/mg F-). The dose of alum and lime to be added in raw water with different fluoride concentrations and alkalinity levels (G.Karthikeyan and A. Shunmuga Sundarraj, 1999).

Following reaction shows the methods of completion

$$2\ Al_2(SO_4)_3.18H_2O + NaF + 9Na_2CO_3 \rightarrow [5Al(OH)_3\ Al(OH)\ 2F] + 9Na_2SO_4 + NaHCO_3 + 8\ CO_2 + 45\ H_2O$$

$$3\ Al_2(SO_4)_3.18H_2O + NaF + 17NaHCO_3 \rightarrow [5Al(OH)_3\ Al(OH)\ 2F] + 9Na_2SO_4 + 17\ CO_2 + 18\ H_2O$$

The Nalgonda technology has been successfully used in India but due to some demerits of the technology has been stopped. (Dr. Shikha Modi and Ranjeeta Soni, IOSR, 2013)

Contact Precipitation

Contact precipitation means the addition of calcium and phosphate compound in fluoridated water. Saturated bone charcoal work as a catalyst for precipitation of fluoride. (Chilton, et al., 1999).

Adsorption/Ion-Exchange Method

Adsorption method can be used for the removal of fluoride from drinking water. For this process adsorption media or filter, bed has been used. In filter media bed containing Bauxite, magnetite, kaolinite, serpentine. Various types of clays and red mud are some of the naturally occurring materials can be used. The adsorbent gets saturated after a period of operation and requires regeneration.

The material retains fluoride either by physical, chemical or ion exchange mechanisms. Fluoride uptake capacity can be increased by certain pre-treatments like acid washing, calcination, etc. None of the mentioned materials generally exhibits high fluoride uptake capacities.

Activated alumina, activated carbon, bone char, defluoron-2(sulphonated coal) can also be used for defluoridation of drinking water as well as synthetic materials i.e.. ion exchange resins. Bone char, activated alumina, and calcined clays have been successfully used for defluoridation methods.

Materials, Methods and Experimental Design

Area of Work

Sitapura Industrial area, Tehsil Sanganer, District Jaipur (Rajasthan).

Sitapura Industrial Area has located 6.0 Km from Jaipur Airport along NH-12. This area is known as EPIP (Export Promotion Industrial Park). Jaipur city is 18 km from EPIP. The area is around 365.00 acres. The water quality is potable in this area. Water availability by tube wells. The Depth of tube wells is approximate 30m. Average discharge of water is 2,000 gallons per hour. The prominent industries of in this area are chemical and automobile industries. Thousands of residential flats are available in and around the area. ITI, Polytechnics, Engineering Institutes, Medical Institutes and Hospitals, Management, IT and Architectural colleges, Fashion Designing Institutes shopping complex, etc. are located in this area.

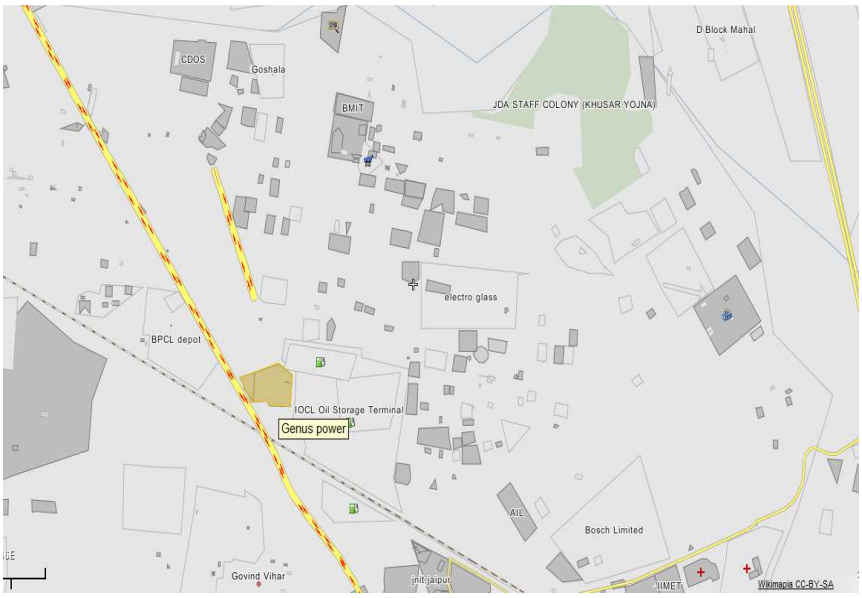

Figure 1: Map of Sitapura Industrial Area

Aim of Work

Due to proved health hazards, complicated procedure and expenditure, many popular defluoridation processes like Nalgonda, Activated alumina, etc. methods are in the phase-out process therefore, the aim of the present research work is to find out a best defluoridation method which is easy to use by illiterate villagers, requires minimal expenditure, involvement of less technical personal and effective methods for fluoride removal from drinking water so that these methods can be applied easily everywhere.

Importance of Defluoridation

Due to various health impacts of fluoride on human beings the treatment of fluoride is necessary.

Locations of Sample Collection

- Sample 1: Genus power Infrastructure, Sitapura
- Sample 2: Chevrolet Workshops, Sitapura
- Sample 3: Ratan Textiles, Sitapura
- Sample 4: Bharat petrol pump, Sitapura
- Sample 5: Hotel Amrapali Sitapura
- Sample 6: JNIT College Sitapura
- Sample 7: Sitapura Residential Area
- Sample 8: Near Chokhi Dhani Sitapura
- Sample 9: Sachiwalaya Nagar
- Sample 10: Laxhmipura

Time of Sample Collection

Based on the season cycle and water intake samples were collected in 3 phases.

- First Phase (Samples collection in Month of August)
- Second Phase (Samples collection in Month of December)
- Third Phase (Samples collection in Month of March)

Table 2: Applied Method for Defluoridation

S. No.	Methods	Details of Methods
1	Method A	Removal of fluoride by Fly Ash
2	Method B	Removal of fluoride by Marble powder

Defluoridation Methods used in Research Work

In view of the demerits of some defluoridation methods, in the present research work, we have tried to use some cheap, effective and easily available good adsorbent of fluoride for the purpose of defluoridation. These simple methods can be applied easily under all circumstances.

Method A: Defluoridation by Fly Ash (Based on Adsorption Process)

The fly ash is an effective adsorbent of fluoride. In this method we have used fly ash, generate from Chula. The 100 gm ash was mixed with 1-liter water having fluoride. Stir 5 to 10 minutes then keep it for settle at least for 2 hr. After 2 hours this solution was filtered (G-3 crucible)

Method B: Defluoridation by Marble Powder (Based on Chemical Treatment Process)

In chemical treatment processes, lime treatment is one of them. In this method, we collected marble stone and crushed into marble powder. Then it was used for removal of fluoride in place of lime.100 gm of marble powder was mixed with1 liter water having fluoride. Stir 5 to 10 minutes then keep it for settle at least for 2 hr. After 2 hours this solution was filtered (G-3 crucible)

Determination of Fluorides

Important fluorides bearing mineral are fluoride apatite, amphiboles, and micas. The concentration in groundwater is limited due to the low solubility of most fluorides, but in some areas, the concentration reaches above 5ppm.

- For the determination of fluoride, we use ions electrode method.
- A potential is established by the presence of fluoride ions across the crystal which is measured by an ion meter.

The ions selective electrode is a fluoride sensitive electrode of the solid-state type which consisting of a lanthanum fluoride crystal. It can be used in forms of a cell in the combination with a reference electrode, called a calomel electrode. The crystal contacts the sample solution at the one face and an internal reference solution at the other.

As per the **WHO (World Health Organization),** the permissible limit of Fluoride in drinking water should be 1.5 mg/l.

Observation Table

Table 3: Fluoride Removal Methods from Drinking Water in Various Phases

Sample Number	First Phase Concentration of F (Mg/L.)			Second Phase Concentration of F (Mg/L.)			Third Phase Concentration of F (Mg/L.)		
	Before Treatment	A	B	Before Treatment	A	B	Before Treatment	A	B
1	2.46	0.75	2.23	2.46	0.75	2.23	2.46	0.76	2.25
2	2.16	0.68	1.98	2.16	0.69	1.98	2.16	0.69	1.98
3	1.18	0.56	1.05	1.18	0.55	1.02	1.18	0.55	1.05
4	2.82	1.01	2.40	2.82	1.01	2.40	2.82	1.01	2.40
5	1.38	0.62	1.16	1.38	0.63	1.16	1.38	0.63	1.16
6	2.84	0.88	2.35	2.84	0.89	2.35	2.84	0.89	2.36
7	1.40	0.65	1.15	1.40	0.65	1.12	1.40	0.65	1.15
8	2.34	0.70	2.15	2.34	0.70	2.12	2.34	0.69	2.15
9	2.50	0.75	2.40	2.50	0.76	2.40	2.50	0.75	2.40
10	1.21	0.60	1.10	1.21	0.60	1.10	1.23	0.61	1.10

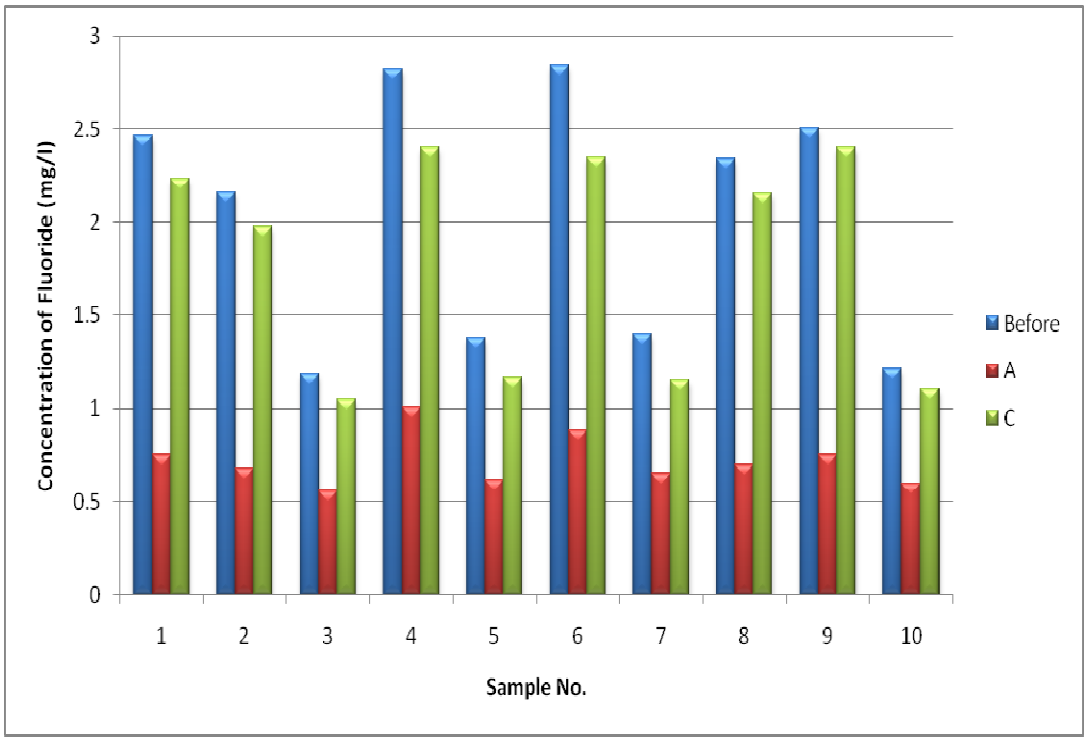

Figure 2: Comparative Fluoride Concentrations before and after using Adsorbents

Table: 4 Variations in Percentage (%) by Two Methods of Defluoridation

S. No.	Con of Fluoride Before Defluoridation (Mg/Lit.)	Fly Ash Method (Mg/Lit)	% Variation	Marble Powder Method (Mg/Lit)	% Variation
1	2.46	0.75	69.51	2.23	9.34
2	2.16	0.68	68.51	1.98	8.33
3	1.18	0.56	52.54	1.05	11.01
4	2.82	1.01	64.18	2.40	14.89
5	1.38	0.62	55.07	1.16	15.94
6	2.84	0.88	69.01	2.35	17.25
7	1.40	0.65	53.57	1.15	17.85
8	2.34	0.70	70.08	2.15	8.11
9	2.50	0.75	70	2.40	4
10	1.21	0.60	50.41	1.10	9.09

RESULTS AND CONCLUSIONS

To address the fluoridation problem in and around Sitapura Industrial Area, Jaipur 10 sample collection sites were undertaken for the present study. The study was conducted in three phases based on seasons and water usage. Two methods were employed on the samples for defluoridation in each phase. Different physical and chemical parameters along with Nitrate, Fluoride and Heavy metals were analyzed.

Physical and chemical analysis like P^H, Hardness, Chloride, Total Dissolve Solid (TDS), and Alkalinity has been done in the laboratory of Jagannath University, Chaksu while some parameters like Fluoride, Nitrate and Heavy metals were analyzed by Team Test Lab Sitapura, Jaipur.

In phase I, all samples was collected after the rainy season. During the analysis of parameters, we found that sample No.6 which is collected from JNIT College has maximum fluoride concentration (2.84mg/l) and minimum fluoride concentration found in sample No.3 which is collected from Ratan Textile (1.18mg/l). We also found the fluoride concentration exceeds from the permissible limit in maximum samples collected from the study areas.

When sample No. 6 was defluoridized with fly ash, the concentration of fluoride dropped from 2.84 to 0.88 mg/l. When we remove the fluoride by using fly ash method the concentration of fluoride goes down below the permissible limit. On using marble powder the amount of fluoride decreased to 2.35 mg/l.

In II and III phases, again we found the almost the same result for all the samples as in phase I.

We can see the variations of fluoride concentration in all samples of all phases before and after defluoridation methods (figure 2).

The variations in percentage (%) of Fluoride content by different defluoridation methods in all samples are given in Table 4. It has been observed from the percentage (%) variations, that fly ash can remove fluoride from drinking water more effectively than marble.

The aim of our study was to find out the best suitable defluoridation methods which could be employed easily and

effectively.

When samples were defluoridized with fly ash the concentration of fluoride dropped drastically in all samples. But due to the chemical nature of fly ash other Physico-chemical parameters showed an increase. All other parameters like TDS, Alkalinity, Hardness, Nitrate, and Chloride were controlled without hindering the ability of fly ash to defluoridize the water. No additional traces of heavy metals were detected in the samples after treatment with treated fly ash.

Marble was found to be not much effectively in the removal of fluoride comparative to fly ash.

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