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Fluoride Removal Techniques from Groundwater: A Review

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Abstract—*In the present paper an attempt has been made to review the different techniques used for fluoride removal from drinking water. Among these techniques membrane and ion exchange process are not encouraged in India because of economic constraints and operation and maintenance problems. However Nalgonda technique is one of the well known strategies generally utilized for defluoridation of water as a part of developing nations like India. Various technologies are being used to remove fluoride from water but still the problem has not been rooted out. Fluoride being the lightest member of the halogen group and can interfere with the important biological processes as well as vital cellular constituents such as enzymes. Defluoridation of water is therefore one of the most important remedial measures in order to solve excess fluoride problem. However based on extensive literature survey a comparison of existing conventional and advanced treatment used for fluoride removal has been summarised. The efficiency and limitations of these techniques and methods has also been discussed.*

Keywords*: Defluoridation, Nalgonda technique, fluoride removal techniques*

1. INTRODUCTION

1.1 Fluoride in drinking water

Fluorine is one of the most reactive chemical elements and the lightest member of the halogen group, found as fluoride in the environment. Fluoride is the negatively charged ionic form of the element fluorine and present in all natural water in trace amounts. Fluoride is inorganic, monatomic anion of fluorine with its chemical formula as F^-. It is a mineral and is present in right amount in the drinking water, strengthens our teeth. Fluorides are found in a wide variety of minerals, including fluorspar, rock phosphate, cryolite, mica, hornblende and others. Fluorite (CaF_2) is a common fluoride mineral occurring in both igneous and sedimentary rocks having low solubility levels.

1.2 Guidelines

- As per WHO, the upper limit of fluoride in drinking water has been set up as 1.5 mg/lt.
- The Bureau of Indian standards (BIS) has laid down the maximum permissible limit of fluoride as 1.0 mg/lt.
- Intake of fluoride higher than the optimum level is the main reason for dental and skeletal fluorosis.

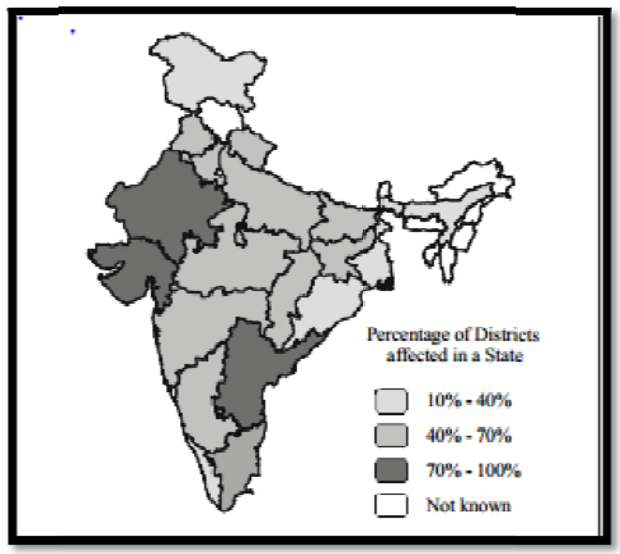

Figure1. Affected areas in India Fluoride

2. HUMAN HEALTH EFFECTS

Fluoride is considered to be very beneficial for teeth at low concentrations thus preventing decay, but with excessive exposure through drinking water, it results into many adverse effects. Fluoride exposure leads to acute problems following fluoridation overdosing i.e. gastrointestinal problems are common. The major effects involved with fluoridation ranges from mild dental fluorosis to severe skeletal fluorosis as well as bone fractures. Dental fluorosis is the most common manifestation of over-consumption of fluoride which results into mottling of teeth ranging from mild discoloration to severe dark patches on teeth. Exposure to very high fluoride over a prolonged period of time results in acute to chronic skeletal fluorosis. Fluoride increases bone volume, there is less strength per unit volume therefore Bone strength begins to decline leading to risk of Bone fractures.

Table1. Effects due to fluoride contamination

S.no.	Fluoride conc.	Health effects
1.	> permissible limit	Effects neuro development in children
2.	1-4 mg/lt	Skeletal fluorosis and fractures
3.	> 4 mg/lt / 12 mg/day	Kidney injury due to dehydration and polyuria
4.	> 1 mg/lt(range – 4 to 21 mg/lt)	Dental fluorosis
5.	50 mg/lt	Suppresses endocrine glands like thyroid

3. FLUORIDE MEASUREMENT

Addition of fluoride is certainly effective, but too much fluoride can be harmful; therefore, it is important to have a convenient Method for monitoring fluoride levels. Fluoride presence in water is odourless, tasteless and colourless; therefore the use of instrumentation is must for identifying its concentration in water. Fluoride selective electrode is considered to be the most reliable way of testing water for fluoride, though this method is difficult to do outside of a lab setting. Fluoride selective electrode consists of a lanthanum fluoride crystal (LaF_6) which in the presence of fluoride ions, experiences an electro-potential. Several methods have been developed, out of which most of them are colorimetric tests by which fluoride interacts with dyes and chemicals such as SPADNS method, where fluoride determination involves the reaction of fluoride with a red zirconium-dye solution. The fluoride combines with part of the zirconium to form a colourless complex, thus bleaching the red colour in an amount proportional to the fluoride concentration. Though these are less accurate than an electrode method, they can be used in order to evaluate if a body of water is safe to consume.

4. TECHNIQUES FOR FLUORIDE REMOVAL

Excessive fluoride level in drinking water in developing countries poses to be a serious problem and its detection demands laboratory equipment, skills and analytical grade chemicals. Preventing fluorosis through management is a tough task as it requires various favourable conditions. Several water treatment techniques are known to be used in order to remove fluoride from the water however, many of these techniques have failed to meet the requirements. In order to select an appropriate method of defluoridation to achieve a sustainable solution for fluorosis problem, the provision for safe, low fluoride water from alternate sources, either as an alternative source or for blending, should be investigated as the very first option. In cases where alternate sources are not available, then defluoridation is the only technique to prevent fluorosis.

This paper attempts to characterize the basics of removal techniques of fluoride from water, followed by discussing the most promising defluoridation techniques; bone char, activated alumina, Nalgonda and finally these methods are compared using indicators, which may be appropriate in developing countries.

5. DEFLUORIDATION TECHNIQUES

Defluoridation refers to methods of water treatment that reduce the concentration of fluoride in the water, normally, in order to make it safe for human consumption. The objective of Defluoridation is to prevent or to mitigate **endemic fluorosis.** Defluoridation technology has to be simple, affordable, reliable and operational. Defluoridation is defined as, "The downward adjustment of level of fluoride in drinking water to the optimal level." Various techniques of defluoridation can be categorized into four:

1. Adsorption technique
2. Precipitation technique
3. Ion-exchange technique
4. Other techniques, which includes reverse osmosis, electro- dialysis

Table2. Materials used in different defluoridation techniques

Adsorption	precipitation	Ion exchange	others
Carbon materials, Activated Alumina, Magnesia, Tricalcium phosphate, Calcite, Hydroxy apatite, Wood, Lignite, Activated char coal, Fish bone char, Processed bone, Nut shells, Avaram bark, Tea waste, Coir pitch, Fly ash, Bauxite, Serpentine	Anion exchange resins: NCL poly anion resin, Tulsion A27, Lewatit-MIH-59, Amberlite IRA-400, Deacedodite FF-IP, Polystyrene. Cation exchange resins: Defluoron-1, Defluoron- 2, Carbion.	Lime, Alum, Lime & Alum (Nalgonda technique), Alum flock blanket method, Poly Aluminium Chloride (PAC), Poly Aluminium Hydroxy Sulphate (PAHS), Brushite.	Electrochemical method (Aluminium electrode), Electro dialysis, Electrolysis, Reverse Osmosis.

5.1 conventional techniques

5.1.1 Nalgonda technique

Adsorption technique functions on the adsorption of fluoride ions onto the surface of an active agent. Activated alumina, activated carbon and bone char were among the highly tested adsorbing agents. After extensive testing of these materials; NEERI, Nagpur has evolved a simple and economical method for removal of fluoride that is referred to as Nalgonda technique in the year 1975 in response to fluorosis concerns. The methods are based on the addition of chemicals (coagulants and aids) and the subsequent formation of insoluble fluoride precipitates. Its involves the addition of lime, bleaching powder and aluminum salts followed by rapid mixing, flocculation sedimentation, filtration and disinfection. Aluminum salt is responsible for removal of fluoride from water.

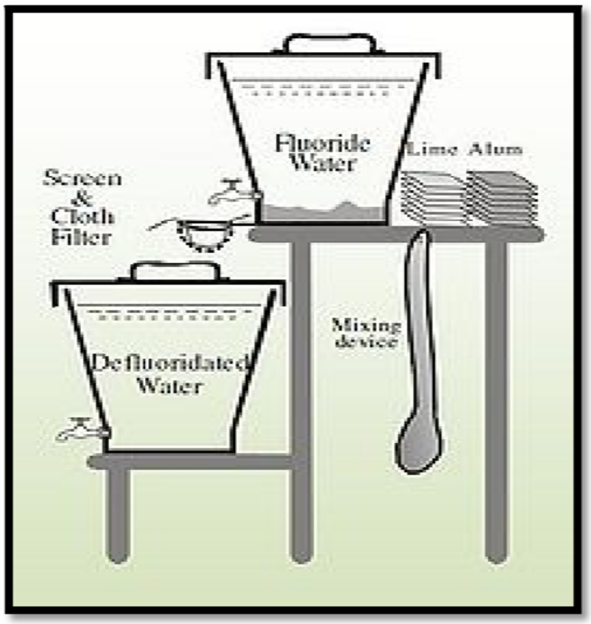

Figure2. Operational diagram of nalgonda technique

Operation of Nalgonda technique starts with the selection of either aluminium sulphate or aluminium chloride which depends upon sulphate and chloride contents of raw water to avoid them exceed the permissible limits. The test water and the required dose of aluminium sulphate is then mixed rapidly for a period of 30 to 60 seconds with a speed of 10-20 rpm. Lime facilitates formation of dense flocs for the rapid settling of insoluble fluoride salts, having the dose as empirically $1/20^{th}$ of that of aluminium salt dose. Bleaching powder is added in a dose of 3mg/lt to facilitate disinfection. Also alum is required in approximate doses to obtain water with acceptable limit of fluoride (<1.0 mg/lt). On chemical reaction tiny flocs are formed. Then mix for a period of 10-15 min with a speed of 2-4 rpm. Water is allowed to settle and filter through earth candles overnight. Nalgonda technique was preferable at all levels because of its low price, ease of handling and higher versatility having applications for both large and small communities.

5.1.1.1 Merits and Demerits

Merits:

- Regeneration of media is not required.
- No handling of caustic acids and alkalis.
- The chemicals required are readily available and are used in conventional municipal water treatment.
- Adaptable to domestic use.
- Economical
- Can be used to treat water in large quantities for community usage.
- Applicable in batch as well as in continuous operation to suit needs.
- Simplicity of design, construction, operation and maintenance.

Demerits

- Desalination may be necessary when the total dissolved solids exceed 1500 mg/l.
- Hardness of the raw water in the range of 200 mg/l to 600 mg/l requires precipitation softening and beyond 600 mg/l becomes a cause for rejection or adoption of desalination.
- Generation of higher quantity of sludge compared to electrochemical defluoridation.
- The large amount of alum needed to remove fluoride.
- Careful pH control of treated water is required. .
- The daily operations require a trained and conscientious operator.

Table3. Comparison between fluoride removal techniques based on different parameters.

Authors and year	Parameters	Activated alumina	Bone char	Nalgonda
Siddiqui A. H. 1955	no daily dose of chemicals; (no daily working load)	+	+	-
R. mehrotra et al 1999	Dosage designed for fluoride conc. Independent of unit or plant.	-	-	+
S. kumar 2000	No risk of false treatment due to breakpoint.	-	-	+
K. bailey et al 2000	Removal capacity of medium is independent of fluoride conc.	-	-	-

Siddiqui A. H. 1955	No regeneration or renewal of medium is required.	-	-	+
Sanghratna S. waghmare	High removal efficiency can be ensured.	+	+	-
Radheshyam et al 1999	Easy to construct, even by users.	+	+	++
Sushree swaroopa tripathy et al 2006	Construction materials are cheap and easily available.	+	+	++
Bersillon et al 2006	Can be sized for one or several families or e.g. a school	+	+	+
Radheshyam et al	No risk of medium chemicals unacceptability	+	-	+

6. CONCLUSIONS

Fluorosis is an important public health problem in india. Drinking water is the main source of ingestion of fluoride. The various manifestations of chronic fluoride toxicity are mild to severe dental fluorosis, skeletal fluorosis, crippling fluorosis, where visceral organs are involved. Defluoridation should be taken up when there is no alternative source of safe drinking water. So far nalgonda technique is the most suitable and appropriate technique for Indian rural communities. Where community defluoridation is not feasible, the residents of the endemic fluorosis areas should be educated and motivated to adopt domestic defluoridarion techniques. The techniques should be economic, adoptable and acceptable to the communities. Priority should be given to the techniques, which utilize locally available materials as defluoridation agents.

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