JETIR.ORG ISSN: 2349-5162 | ESTD Year : 2014 | Monthly Issue
JOURNAL OF EMERGING TECHNOLOGIES AND INNOVATIVE RESEARCH (JETIR)
An International Scholarly Open Access, Peer-reviewed, Refereed Journal

DEFLUORIDATION BY HOUSEHOLD METHODS - A REVIEW

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Abstract**:** Fluoride is such an element that is widely acknowledged for the benefits it provides at low concentrations. Fluoride ranks 13th with respect to the abundancy in the earth's crust. Fluoride is generally consumed through drinking water. The water may be tainted by organic materials, industrial wastes or natural sources. Fluoride is one of the contaminants which pollutes the water. The range of fluoride limit in drinking water is set around 1.5mg/l by WHO. Over 260 million people consume water with fluoride concentrations that is extensively high. In this review study, recent insights and comprehension of various defluoridation techniques have been highlighted along with the advantages and disadvantages of each technique. The study affirms that in spite of the development of several defluoridation techniques over the years, there has been no conclusive fluoride removal technique that can produce cost effective and sustainable results. Hence, a commercially sustainable, practical and economical technique is needed to protect people from the hazards and ill-effects of fluoride contamination.

Keywords: Fluoride, water, contamination, defluoridation, technique, household

INTRODUCTION:

Fluorine is said to be the most abundant element in nature. As fluorine is highly reactive, it is never found in gaseous state. Rather, it is always encountered in combined state in the form of fluoride. Fluorine is beneficial in many ways. It is essential for dental enamel formation and mineralization of bones. It prevents dental caries by curtailing the solubility of the enamel that may be caused by the acid production of the bacteria that reside in the mouth. The main sources of fluoride are certain foods like fruit juices, cheese, sea fish and drinking water. The other sources of fluoride are black salt, tea, tinned foods and suparis. The recommended fluoride level in the drinking water is 0.5-0.8 mg/L. Fluorine is also termed as "double edged sword" as it neither should be consumed in deficiency nor in excess. Excessive intake of fluoride through drinking water with a quantity more than 1 mg/L could result in skeletal and dental fluorosis whereas inadequate ingestion with a quantity below 0.5 mg/L could enhance the chances of dental caries. Fluorosis often marks the onset with non-skeletal changes. These changes can also be reversed by safe drinking water and nutritional supplementation but if left untreated, the disease progresses into dental and skeletal fluorosis which are incurable.

In India, fluorosis remains restricted to certain places depending on the fluoride content, hence, it is endemic in nature. 66 million people are at risk and 25 million are affected by this condition. High risk groups involve elders, children, lactating mothers, pregnant women and patients with cardiovascular and renal diseases. Dental fluorosis is often characterized by the "mottling" of the dental enamel when the intake is above 1.5mg/L. Eventually, the teeth lose the shiny texture and develop chalky white patches. This is taken as the first significant sign of dental fluorosis. With time, the patches turn brown or black. In severe cases, excessive enamel loss leads to a corroded appearance. Incisors and molars are commonly affected with deciduous teeth being an exclusion. Skeletal fluorosis, on other hand is caused when the daily intake is in the range 3.0 to 6.0 mg/L which is in fact a heavy deposition of fluoride in the skeletal parts. It is characterized by the calcification of tendons and ligaments. The person affected by this kind of fluorosis often feels pain in the joints of extremities and stiffness of back. Fluorosis remain confined to certain states like Andhra Pradesh, Karnataka, Kerela, Tamil Nadu, Punjab and Rajasthan. A new form of fluorosis known as genu valgum and osteoporosis involving the lower limbs has been reported in some places of Tamil Nadu and Andhra Pradesh. The syndrome was commonly found among people whose staple diet was jowar or sorghum. Further studies revealed that jowar based diets contain higher quantity of fluoride than the usual rice-based diets.

20th Century grasped the attention towards fluorosis for the very first time. Researchers were bewildered by the prevalence of "Colorado Brown Stain" on the teeth of residents of Colorado Springs. High levels of fluoride in the local water supply caused the stains. This fluoride was naturally found in the ground water. People with stains developed high resistance to dental caries. This eventually led to the formulation of policies to introduce fluoride into the various public water supplies. The fluoride level should be enough to prevent caries without causing fluorosis. One in every four Americans aged from 6 to 49 gets affected by fluorosis. As of April 2014, Government of India stated that fluoride prevalence was reported in almost 230 districts of 19 states with 25 million people who are impacted and another 66 million are at risk. Since fluoride occurs naturally in water, the levels above the recommended range for drinking water may increase the risk for causing severe fluorosis. The CDC recommends that the communities where the natural levels exceed 2 parts per million, the parents should provide water to their children from other sources.

Hence, fluoride intake has both beneficial and negative effects. On one hand, it reduces the incidence of dental caries and on other hand, it causes mottling of tooth structure and skeletal fluorosis following prolonged exposure that is unevenly high. Dental caries is highly prevalent all over the globe. 2.4 billion people suffer from caries of permanent teeth and almost 486 billion children suffer from primary teeth caries. In areas where fluoride intake is less, public health actions are required to provide sufficient fluoride in order to reduce tooth decay. This can either be done through drinking water, salt or milk fluoridation or by the usage of fluoride containing dental care products or by recommending a diet that is low in sugar. Fluoride intake in excess amount is likely to occur in warm climates when water consumption is greater and where high fluoride water is used to prepare food and irrigate crops. Such subjection generally leads to fluorosis of dental counterparts or crippling skeletal fluorosis in coalition with bone deformities, calcification of tendons and ligaments and osteosclerosis.

Although fluorosis is not labelled as a disease, the consequence it causes can be psychologically distressing and difficult to treat. Parental vigilance also plays an important role in preventing fluorosis. WHO has published certain guidelines for the communities to control fluoride exposures in addition to the establishment of an important balance between caries prevention and protection against adversities.

I. STATUS AND HEALTH IMPACTS OF DEFLOURIDATION

Fluoride was first detected in India in 1973 at Nellore district of Andhra Pradesh. At present, 19 states are impacted by the excess fluoride. The highest amount of fluoride is about 12mg/l to 35mg/l, found in the districts of Andhra Pradesh and Rajasthan (Jagtap et.al 2012). At present, fluorosis has affected around 1 million people (Jagtap et.al 2012). Besides, it causes lower haemoglobin levels, headache and RBC deformities. It may also alter digestive, central nervous system, excretory and respiratory system. The high fluoride problem is widely present across various states of India which has been depicted in Table 1.

Table 1: Reference range of fluoride in various states

States	Range of fluoride	No. of affected districts	Reference
Andhra Pradesh	0.60 to 1.80 mg/L	16	N. Subba Rao et al. (2017)
Assam	5 to 23 mg/l	2	A.B. Paul et al (2017)
Bihar	0.38 and 8.56 mg/l	6	Deepanjan Mridha et al (2021)
Chhattisgarh	0.1 to 7.1 mg/L	17	Chandrashekhar Azad Kashyap et al (2020)
Delhi	1 to 7.14 mg/L	7	Shekhar S et.al (2013)
Gujarat	1 to 6.53	24	Barot VV et.al (1998)
Haryana	1.65 mg/L	12	Shakir Ali et.al (2019)
Jharkhand	0.1 to 6 mg/L	5	Singh AK et.al (2008)
Karnataka	2.56 mg/L	16	Shakir Ali et.al (2019)
Kerala	5.75 mg/L	3	Shaji E et.al (2007)
Madhya Pradesh	5.98 mg/L	14	Shakir Ali et.al (2019)
Maharashtra	1.70 mg/L	10	Shakir Ali et.al (2019)
Orissa	16.4 mg/L	18	Das S et.al (2000)
Punjab	4.67 mg/L	17	Shakir Ali et.al (2019)
Rajasthan	0.2 to 13 mg/L	32	Vikas C et.al (2009)

Tamil Nadu	1.21 to 3.24 mg/L	9	Dar Ma et.al (2011)
Uttar Pradesh	0.1 to 2.5 mg/L	7	Avtar R et.al (2013)
West Bengal	0.3 to 17.6 mg/L	7	Mobarak Hossain et.al (2021)

II. TECHNIQUES ASSOCIATED WITH DEFLUORIDATION

1. Precipitation and Coagulation:

This method involves the use of lime and alum as coagulants. It is an efficient and economical method for water defluoridation where charged particles are neutralized and amalgamated, thereby left to settle down. Firstly, lime is added and when fluoride precipitates, alum is added to form coagulation. The temperature and pH of the solution are the prime aspects of the process involving precipitation. Therefore, when certain chemicals are added or solution temperature is reduced, it becomes unstable and leads to precipitation. According to Sujana et. al, 1998, appropriate pH range for defluoridation involving coagulation is 5.5 to 6.5. The commonly used chemicals for precipitation are ferric chloride, ferrous sulphate, sodium bicarbonates and potash alum. The major disadvantage is the inability to decrease fluoride content under the prescribed permissible limits of WHO. Researches have shown that the operational cost is extensively high and a lot of toxic sludge rich in aluminium is generated as the process involve high chemical demand. According to M.F Chanf et al. fluoride can be removed satisfactorily from semiconductor waste water too. At community level, Nalgonda technique is best applied in various villages of India. Lime and alum are used in a two-step process (Meenakshi et.al, 2006) in household or domestic level. Nowadays, plant-based materials are used as natural coagulants for being eco-friendly and sludge free. Gandhi et. al 2019, through their examination to remove fluoride used Passiflora foetida fruits as a natural coagulant. The efficiency of high fluoride removal decreased with increasing alkalinity at acidic and neutral pH mediums and the coagulant at low fluoride concentration worked best. On other account, this process removes a small quantity of fluoride, approx. 18 to 33%. At times, concentration of sulfate ion crosses the limit of permissibility which in turn causes a cathartic effect due to the usage of aluminium sulfate and residual aluminium. These elements in excess can cause dangerous disease.

2. Adsorption

A process that involves the ions transfer to the solid phase from the solution through various mechanisms and that helps in fluoride removal is known as adsorption. This process comprises of physical adsorption or chemical adsorption and is a surface phenomenon. Adsorbents are analysed on different scales and parameters. Activated carbon and alumina are the most commonly used adsorbents. UNICEF has initiated alumina strategy for fluoride removal in some places like Udaipur in order to ensure safe drinking water. The defluoridation efficiency gets simulated by different criterion and parameters like hydraulic loading factor, surface area, pH and hardness of water. Regeneration is required on the reduction of the adsorbent's effectiveness and that takes place in every 4 to 5 months. Porous materials are good enough for the process and thereby used for adsorption. The materials that are extremely microporous with ease of regeneration and higher adsorption capacity like activated carbon, silica, aluminium and zeolites are preferred. These elements have pores of desired quantity and quality. Adsorption is one of the most widely used defluoridation technique for households due to its effectiveness, efficiency, accessibility and reusability. One of the advantages is that any cheap adsorbent can be used which can provide high efficiency and at the same time can be cost benefit and effective. There are various low-cost adsorbents like kaolinite, lignite and charfines that are equally effective in fluoride removal (Kulkarni et. al, 1974). Lignite is a low category coal whereas charfines act as a byproduct during the formcoke production which is a more favourable and cleaner fuel than coal. In addition to this, when the indigenous materials are used, the process becomes more economical. The coherence of this process depends on multi-variants such as the initial fluoride concentration, dose of the adsorbent, loading capacity, physical and chemical properties of adsorbent and affinity to fluoride ions. With time and with every regeneration cycle, saturation takes place and reduction in fluoride removal capacity takes place. This generally results from the previous incomplete removal of adsorbed material during the process of desorption.

3. Ion Exchange

This is a technique to treat water that eventually can remove undesirable fluoride and chloride ions. This is often termed as conventional fluoride removal process. Materials involved in ion exchange are generally insoluble in water, that results in loose holding of the replaceable ions and exchanging of ions from solution. These materials may either be natural or synthetic. Natural materials comprise of cellulose, proteins, living cells and few soil particles. On the other hand, synthetic materials are classified as beaded polymer resins and membranes. Most often, the ion exchange resins are insoluble in water and most of the solvents which are organic in nature. Indion FR 10 and Ceralite IRA 400 have been found to be 95% effective for replacing chloride ions. The former is an ion exchange resin and the latter is an anion exchanger. Though ion exchange has proved to be effective in the removal of ionic contaminants, yet the resins are exhaustive, have longer reaction time, require longer time to react and produce a larger volume of waste water. The chloride ions of the resin are replaced by the fluoride ions. The process continues till all the resins are occupied. Then the resins are backwashed and supersaturated with dissolved sodium chloride salt. Eventually, fluoride ion is replaced by chloride ion. The resin gets recharged and the process starts over again. The main advantage of this process is that it has high removal efficiency and does not produce any change in colour or taste. Whereas, the limitation of this process is that it is not that effective or efficient in the presence of other ions like carbonates, sulphates and phosphates. It also generates fluoride rich waste which has to be treated separately. As a result, regeneration of resin is a hindrance and gives high level chlorides and low pH water.

4. Membrane Process

This is a highly efficient advanced defluoridation technique which provides pure and ultrapure water by using a semi-permeable membrane between adjoining phases for the removal of fluoride. In this method, very fine size particles are separated by a membrane technique. The membranes depending on the type of material may either be natural such as cellulose acetate or synthetic such as polysulphones. The segregated material decides the pore size of the membrane and the material. The process can further be classified based on techniques to separate fluoride using membranes into subtypes such as dialysis, electrodialysis, nanofiltration, ultrafiltration and reverse osmosis. The reverse osmosis method enables the removal of contaminants through a semipermeable membrane by applying pressure to water directly. Natural osmosis when reversed leads to reverse osmosis but it generates brine discharges that need to be disposed safely. According to Nidaye et.al, has been found that 98% of the fluoride ion can be removed by reverse osmosis. On another instance, electrodialysis is a technique involving the membrane but it consists of the application of direct current instead of pressure which is necessary in reverse osmosis to get the ionic contaminants separated. According to Martyna et. al, the technique showed appropriate removal of fluoride. One of the advantages is that it is highly efficient in removing fluoride without any external chemicals. Moreover, it gives results even under a wide pH range. On the other hand, this technique has certain limitations like the requirement of skilled labour, recovering of the membrane due to blockage or fouling after a certain period of analysis, high expenditure and incapability to act with high saline water.

III. CONCLUSION

The increasing effect and impact of fluoride is reviewed along with the standards it should maintain. Excess fluoride leads to fluorosis which is incurable, hence the best option is precaution. Numerous defluoridation techniques have been promoted. Every technique has its own advantages and disadvantages and each of this technique has been successful in removing fluoride but based on the state and requirement. This literature review manifests that the recent fluoride contamination in groundwater is very challenging and nowadays, fluorosis is impacting millions of people directly or indirectly. Various approaches have been used in different countries of the world to facilitate the fluoride removal. Though all the methods that are discussed have taken into account as efficient but there has not been a single defluoridation technique that can be claimed as the most viable solution for fluoride reduction.

Funding

Nil

Conflict of Interest

There are no conflicts of interest.

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