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Occurrence, Detection and Defluoridation of Fresh Waters

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Abstract The fluoride is an essential nutrient for human beings which occur in the surface as well as in groundwater. In surface water, it reaches due to both geogenic and anthropogenic sources but in groundwater, it mainly comes from geogenic sources. Authorities like World Health Organization (WHO), United State Environmental Protection Agency (USEPA), and Bureau of India Standard (BIS) have provided guidelines regarding the concentration of fluoride in drinking water. A higher fluoride concentration in drinking water results in fluorosis. Therefore, the understanding of fluoride occurrence, its detection and removal from drinkable water is the urgent requirement. The chemical behavior of fluoride, the reasons for fluoride concentration in groundwater, the fluoride detection methods, and some case studies on the occurrence of fluoride in fresh water bodies of Uttarakhand are summarized. The effectiveness of different techniques for removal of fluoride from water samples has been reviewed.

Keywords: *fluoride, freshwater, detection methods, Uttarakhand, India, defluoridation, material*

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1. Introduction

In halogen family, fluorine occurs in the form of gas due to its high reactivity and electronegativity in nature. Fluoride is an anionic form of fluorine and makes different organic and inorganic compounds due to its smaller size and greater tendency to behave as a ligand. A few compounds of fluoride are soluble in water and occurs in water resources [1,2,3]. High fluoride concentrations have been found in ground waters of more than 20 nations. Fluoride is the main inorganic pollutant of natural origin found in more than permissible limit in groundwater of 19 states of India viz. Andhra Pradesh, Assam, Bihar, Chhattisgarh, Delhi, Karnataka, Kerala, Madhya Pradesh, Gujarat, Haryana, Jammu & Kashmir, Jharkhand, Maharashtra, Orissa, Punjab, Rajasthan, Tamil Nadu, Uttar Pradesh, West Bengal [4]. However, the groundwater contributes only 0.6% of the total water resources on earth and the preferred source of drinking water both in rural and urban areas. Ground water provides 80 % of the total drinking water need in India. At present, surface water and ground water sources both are being deteriorated due to increasing population, industrialization, and urbanization. Ground water flows through a variety of geological structures in shallow aquifers of the earth. Some researchers have reported that various factors either natural or anthropogenic are also responsible for groundwater deterioration [5,6]. The fluoride occurs mainly in the form of fluorspar, cryolite, sellaite and fluorapatite. Fluorspar occurs in sedimentary rocks whereas cryolite in igneous rocks. Higher fluoride concentration in groundwater (up to more than 30 mg/L) has occurred broadly, remarkably in Africa, Asia, and the USA [7-12].

Typically, the water sources have become hazardous not only for consumption but for other activities like irrigation and industrial requirements. Accordingly, there is a need for attention on the future impact of water resources in planning and developmental issues. The usual amount of fluoride essentially depends on the geological, chemical and physical characters of the concerned aquifer, temperature, the porosity as well as acidity of the soil and surrounding rocks. Therefore, the present article describes the fluoride sources, its effects, and methods existing for fluoride removal from water and wastewater.

2. Chemical Behavior of Fluoride

Naturally, fluorine is found in the form of CaF_2. The fluoride enters the soil through weathering of rocks and precipitation or waste runoff. However, the endemic fluorosis in India is principally caused by hydro-geochemical reasons. It has been established that the lower calcium and higher bicarbonate alkalinity increase the high fluoride content in groundwater [12,13]. Elrashidi and Lindsay, 1986 [14] explained that the fluoride content of soils varies from less than 20 mg/L to an unevenly high-level concentration in highly affected areas. However, fluoride may present in the soil in the form of fluorapatite, fluorspar etc. [15,16]. The fluoride ion is substituted by hydroxyl ion due to redox reactions in soil or rocks and finally increases the concentration of fluoride ions in circulating water. Fluoride amount may also increase in ground water where cation exchange of sodium for calcium occurs [17]. The large-scale withdrawal of water for agriculture purpose, low rainfalls, poor recharging and pollution due to industrial effluents are few important reasons to increase the fluoride content in groundwater.

Fluoride ion is strongly electronegative in character. Therefore, fluoride ion is attracted by positively charged Ca^{++} ions in teeth and bones. Islam and Patel, 2011 [18] told that the high fluoride concentration exposure in workers of furnace create the problem of bladder cancer. The too much ingestion of fluoride may cause dental [19] and skeletal disorders [20]. Fluoride displaces the OH^- ions from hydroxyapatite $[Ca_5(PO_4)_3OH]$, which is the main mineral constituent of teeth particularly in the enamel and bones, finally forms the harder and tougher fluorapatite $[Ca_5(PO_4)_3F]$.

As a strong ligand and in various pH ranges of water, fluoride may form soluble complexes with polyvalent cations like Ca^{+2}, Mg^{+2}, Fe^{3+}, Al^{3+}etc.[21]. Under acidic conditions, fluoride forms metal complexes. Some researchers have reported that fluoride (CaF_2) and fluorapatite $[Ca_5(PO_4)_3F]$ have been considered to be the main minerals responsible for ground water fluoride contamination [22,23,24]. Several researchers have explained that the dissolution of these two minerals may be increased by calcite precipitations which withdraw the calcium ions from the solution as given below in equation (1) and finally decreases the aqueous calcium ion activity [22,25,26,27,28].

$$CaCO_3 + 2F^- + H^+ = CaF_2 + HCO_3^- . \qquad (1)$$

3. Reasons for Natural Fluoride Concentration in Ground Water and Its Health Implications

The occurrence of a higher amount of fluoride in ground water of developing nations is mainly due to the lack inappropriate infrastructure for water treatment. The ground water sources are linked with different types of rocks, volcanic activities, industrial and agricultural activities which are directly responsible for higher fluoride concentration in groundwater. However, the geology of the area plays an important role to increase the amount of fluoride in water resources. The amount of fluoride in ground water mainly depends on geological settings and types of rocks. Crystalline rocks, sedimentary rocks, and metamorphic rocks play a key role to increase the fluoride concentration in groundwater through several geological activities. Besides this, the reaction time of ground water with aquifer minerals is also a key factor to increase the natural fluoride concentration in groundwater [29].

Fluoride possesses the interesting qualities related to human health especially in the prevention of dental caries. The calcium and phosphorus metabolism in the human body is also affected by the higher concentration of fluoride in drinking water. Generally, fluoride in lower quantity is an essential component for normal mineralization of bones and formation of dental enamel [30]. The excessive intake of fluoride may create the problem of fluorosis. López *et al*. 2006, [31] told that about five million people are affected with fluorosis in Mexico which covers about 6% of the population. In the Indian context, fluorosis was first reported in Nellore district of Andhra Pradesh in 1937 as reported by Shortt in 1937 [32]. Susheela, 1985 [33] reported that the industrial fluorosis leads to inhalation of fluoride dust or gas or fumes in humans. Central Pollution Control Board (CPCB) under Ministry of Environment & Forest, Govt. of India (1998), has given a permissible limit of industrial emission of fluoride as less than 25 mg/Nm^3 [34]. The prolonged ingestion of high fluoride concentrations *i.e.* >1.5ppm in drinking water causes severe health problems like dental and skeletal fluorosis. BIS 10500 [35] and WHO [36] have determined the standard maximum value of fluoride in drinking water as 1.5 mg/L.

4. Fluoride Detection Methods for Fresh Water Samples

Several lower level detection methods are available to determine the fluoride in freshwater samples (Table 1).

5. Occurrence and Detection of Fluoride in Fresh Water Sources of Uttarakhand, India: Case Studies

Uttarakhand is a Himalayan state of India with prosperous biodiversity [51] and also possess a large number of freshwater resources including rivers, springs, lakes, ponds, glaciers and locally available several traditional water resources [52]. The water quality of surface and ground water sources of the state has been monitored [47,48,49,53]. During the study of 2014, our research group has detected the presence of fluoride in Alaknanda river, Madhuganga river, Gadoligadhera, Kevrugadhera, Gazaldgadhera, Adwani gadhera at six selected sites of Pauri districts of Uttarakhand state with maximum values as 0.43 and 0.23 mg/l during pre-monsoon and post-monsoon seasons respectively [49]. In another study, we have described the spatial and temporal variations in surface water quality of Pithoragarh district in Kumaun region of Uttarakhand. Under this study, the average fluoride values were found as 0.38 and 0.31 mg/l in pre-monsoon season and post-monsoon season of 2011 in the monitoring of Ram Ganga river, Thuligad, Raigad, Thalika Gadhera, Kaliyapatal and Ghurghatiya water sources of Pithoragarh [47].We have also studied the drinking water quality of 18 selected locations of Dehradun district in Uttarakhand and found the average fluoride values as 0.52 and 0.46 mg/l in pre – and post-monsoon seasons respectively [48]. Recently, our research group have assessed the groundwater quality of Bhagwanpur Industrial Area of Haridwar district in the state and found the mean values of fluoride as 0.1225 and 0.3275 mg/l during March and April months of 2015 respectively, which were within the limit of BIS [54].

6. Fluoride Determination in Natural Water Samples by SPADNS Method

Fluoride ions possess dual importance in water supplies. Fluoride concentration less than 0.8 mg/l causes dental cavities while higher concentration causes dental fluorosis. Therefore, it has become necessary to maintain its concentration within limits. Among several methods of fluoride determination in water and waste water samples, Colorimetric i.e. SPADNS method is one of the most popular methods in laboratories. Under acidic conditions, fluoride ions react with Zirconium-SPADNS [Sodium 2-(parasulphophenylazo)-1,8-dihydroxy-3,6-naphthalene disulphonate] solution. Its dissociation and formation of colorless complex anion take place. As soon as a number of fluoride ions increases, the color becomes lighter [46,55]. The chemical reaction of such method is given below:

$$\begin{aligned} &\text{Zirconium} - \text{SPADNS} + 6F^{-} \\ &= \text{SPADNS Red Bleached} + ZrF_6^{-}. \end{aligned} \quad (2)$$

7. Fluoride Removal Methods

Defluoridation is a method of removal of fluoride from drinking water. The defluoridation method may generally be categorized into two categories such as adsorptive methods and additive methods. In adsorptive methods, a bed of better surface activity is preferred and fluoride contaminated water passes through the bed. Thus, preferentially the fluoride ions get adsorbed on the surface bed material due to greater surface activity and complete a decline in fluoride concentration in the out-stream water whereas, in additive methods, suitable reagents are added for the defluoridation purpose. The fluoride ions in water react with the added chemical reagents and form an insoluble complex [56]. In other words, the defluoridation of drinking water can be achieved mainly by two options such as the treatment of water at the source and the treatment of water at the household level. The treatment at the water source is the preferred method in most of the developed countries whereas the treatment of the water at household can be preferred in less developed countries due to several advantages over treatment at the community level [57,58]. Mostly, the fluoride removal techniques from drinking water work on the principles of precipitation, use of lime softening, alum-lime adsorption methods with a diversity of materials like activated alumina, bone char, synthetic calcium hydroxylapatite and bauxite. Defluoridation of water and wastewater can be achieved mainly by the important techniques as given in Figure 1.

Table 1. Fluoride Detection methods

Sr. No.	Detection Methods	References
1.	High-performance liquid chromatography with hydride generation-atomic fluorescence spectrometry (HPLC-AFS)	[37]
2.	High-performance liquid chromatography-inductively coupled plasmamass (ICP) spectrometry	[38]
3.	Non-chromatographic hydride generation-atomic fluorescence spectrometry	[39,40]
4.	Electro-analytical methods	[41,42,43]
5.	Chemically modified electrodes (CMEs)	[44,45]
6.	Ion Selective Electrode Method	[46]
7.	Sodium 2-(parasulphophenylazo)-1,8-dihydroxy-3,6- Naphthalene Disulphonate (SPADNS) Method	[46]
8.	Spectrophotometric determination	[47,48,49]
9.	Ion Chromatography	[50]

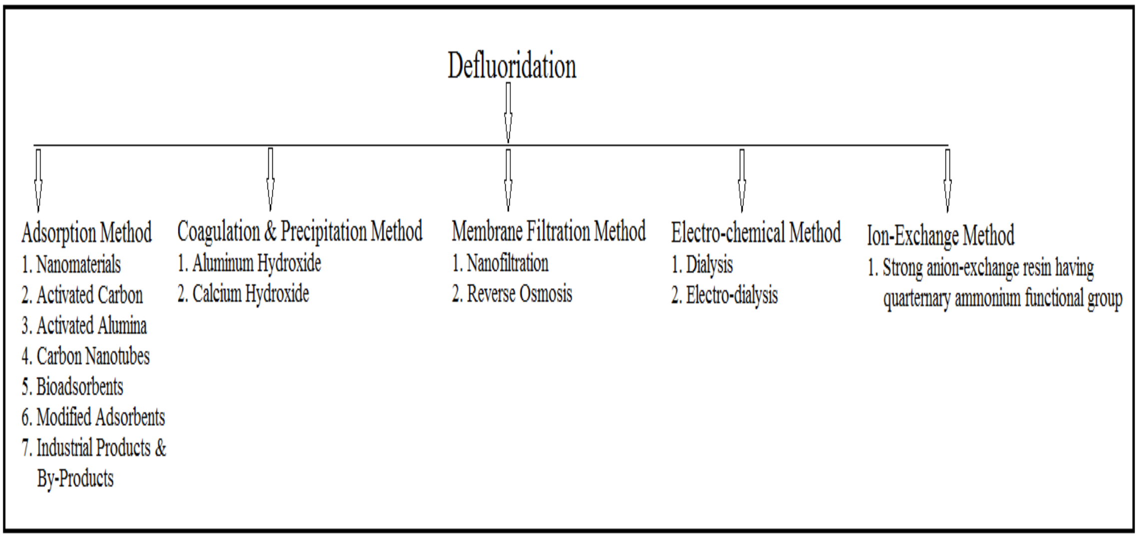

Figure 1. Different Defluoridation methods [1,5,53,58,59]

The efficient elimination of water pollutants especially fluoride from fresh water is a major issue globally. The techniques like membranes separation, adsorption, ozonolysis, an advanced oxidation process; photocatalysis etc. are some of the important techniques for the treatment of water. Nowadays, nanoscience has emerged as an indispensable tool for the treatment of waste water. At the nanoscale, the material represents the remarkable variation in their properties because their surface area gets increased with a decrease in particle size. The particles of 1 to 100 nm size are called as nano-particles. Nano-sized materials possess numerous novel and interesting physical, chemical and biological properties like small size, porosity, ease of separation, high reactivity, large surface area, catalytic potential, chemical, thermal, electromagnetic, mechanical, optical and huge number of active sites to interact with pollutants etc. Several types of nano-materials have been prepared and being used for water treatment [1]. Usually, the nanoparticles of alumina, maghemite, iron, iron oxide, iron hydroxide, titanium oxide, nickel oxide, silica, stannous oxide, titanium oxide, zinc sulfide, zinc oxide, zirconia, anatase, akageneite, cadmium sulfide, cobalt ferrite, copper oxide etc. are found useful in water treatment.

Adsorption is a surface process in which contaminants are adsorbed on the solid surface. Generally, adsorption occurs by physical forces but, weak chemical bondings also contribute in the adsorption process. The process of adsorption is controlled by some factors like nature of the adsorbent and adsorbate temperature, pH, the concentration of pollutants, contact time, particle size etc. At equilibrium, the correlation between the amounts of contaminant adsorbed and in water is called as adsorption isotherm. There are several important renowned models such as Freundlich, Langmuir, Halsey, Henderson, Smith, intra-particle diffusion, Lagergren and Elovich liquid film diffusion etc. which were used to explain the results of adsorption process [60,61].

After the comparative study, it has been found that the adsorption method as the better and cost effective water treatment method among several water treatment methods like evaporation, aerobic, anaerobic, ion exchange, solvent extraction, electro-dialysis, micro- and ultrafiltration, reverse osmosis, precipitation, distillation, oxidation *etc* [62]. For defluoridation by adsorption, the method covers the adsorption of fluoride ions onto the surface of an active material. In adsorption method, the contaminated water is passed through a defluoridating material which retains the fluoride ions either by physical, chemical or ion exchange procedures. After a definite period, the adsorbent needs the replacement with the newer one. Numerous adsorbent materials have been employed for defuoridation of water as per their specific character of adsorption. The parameters considered for the removal of fluoride from water by adsorption method were adsorbent dose, contact time, thermal pre-treatment of adsorbent, initial fluoride concentration, and pH value. Initially, several adsorbent materials have been used to find out an effective and cheap defluoridating agent.

Nowadays, the most commonly used adsorbents are activated alumina and activated carbon. Mainly defluoridation methods involve the utilization of aluminum compounds due to the similarities of the charges on the fluoride ions with hydroxyl ions and the ease of replacement of the fluoride ions with that of hydroxyl ions to form fluoride-aluminum hydroxide complex. Researchers have used the acidic alumina [63], amorphous Al $(OH)_3$, gibbsite or alumina (Al_2O_3) [64] for defluoridation. Usually, the defluoridation ability of activated alumina gets affected due to the hardness of water as well as surface loading also. Farrah and co-workers, 1987 [64] studied the interaction of fluoride ions with amorphous aluminum hydroxide, gibbsite and aluminum oxide over a range of pH from 3 to 8 and Fluoride ion concentrations about 1.9–19 mg/L. A research group has established the efficiency of alum-impregnated activated alumina in defluoridation of water and it was 92.6% at pH 6.5 with a contact time of 3 hours when 25 mg/L fluoride was initially present in water [65]. Sometimes in the cases of acidic solutions, the adsorption of fluoride was found to be retarded due to the electrostatic repulsion whereas at higher pH, the adsorption of fluoride ion takes place due to electrostatic repulsion of fluoride ion to the negatively charged surface of alumna as well as the competition for active sites by excessive amount of hydroxyl ion [66]. Moreover, Langmuir and Freundlich isotherm models have been described for the equilibrium behaviors of the adsorption method. In 1975 Nawlakhe [67] and his research team reported Nalgonda procedure, which was named after a village of Andhra Pradesh in India, is based on adsorption of fluoride on flocs of aluminum hydroxide, in which two chemicals, alum, and lime are rapidly mixed with the fluoride-contaminated water followed by succeeding gentle stirring. The developed flocs of aluminum hydroxides take the majority of the dissolved fluoride and are removed later. Afterward, this Nalgonda method was adopted in other nations.

When the fluoride contaminated water passes through a packed column of activated alumina i.e. through the grains of aluminium oxide, contaminants in the water were adsorbed onto the surface of these grains of activated alumina [68,69,70]. Higher defluoridation capacity was reported over a pH range of 4.0–9.0 by using both untreated and thermally treated hydrated alumina [71] (Shimelis *et al*. 2006). The pH plays a valuable role in defluoridation of drinking water. Modification increases the adsorption ability of adsorbent material especially alumina. In this direction, numerous studies have been completed using the manganese oxide in fluoride removal like MnO_2 coated alumina [72,73,74,75] and MnO_2 coated activated carbon [76,77]. Magnesia modified activated alumina [78] was also found suitable for the removal of fluoride from water. When the surface of alumina was impregnated with alum, its defluoridation capacity was found 92.6% at pH 6.5 although it decreased further at higher pH conditions [66]. A research group has found that the aligned carbon nanotubes which were prepared by the decomposition of xylene using ferrocene as a catalyst, absorb 4.5 mg/g fluoride from 15 mg/L fluoride at pH 7 [79]. Li and his group have explained the adsorption of fluoride on alumina supported on carbon nanotubes [80,81,82] and these nanotubes were prepared by the pyrolysis propylene–hydrogen mixture using Nickel as a catalyst. Furthermore, a range of adsorbents have been used for the removal of fluoride from drinking water by the researchers such as activated carbon [83,84,85,86,87], activated clay [88] and impregnated alumina [69,89-94], solid industrial wastes such as red mud, spent catalysts and fly ash [95-101], sorbents[102-104], alum [105,106] and other materials [107-113] etc.

Ion exchange process removes the fluoride from drinking water up to 90-95% maintaining the taste and color of water. Strongly anion-exchange resin having quaternary ammonium functional groups was employed for the defluoridation purpose [5]. Moreover, there are some restrictions with this method like the efficiency of ion-exchange method is reduced due to the presence of other interfering ions and expensive in nature [1].

Coagulation and Precipitation method of defluoridation of water have high efficiency and are commercially available methods. Calcium hydroxide and aluminum hydroxide were used in this method [1,114] but this method is found as expensive, pH dependent with the presence of other co-ions [115].

Membrane filtration technique of defluoridation of water includes the reverse osmosis and nano-filtration methods. Reverse osmosis is a physical process and is capable of removing the contaminants through the semi-permeable membrane after the application of high pressure. Nano-filtration is a comparatively low-pressure technique having slightly bigger pores than membrane process. Although, these are highly selective and efficient methods but have some disadvantages like higher running cost with the production of toxic wastewater [1,116,117,118,119].

The dialysis method separates the solutes through the membrane which possesses the pores as less restrictive in comparison to nano-filtration [42,43,120,121]. This technique is reported as highly selectively, efficient with higher installation and maintenance cost [1,115,118].

8. Programmes Adopted for Fluoride Mitigation in India

Definitely, Government may play an important role in fluorosis mitigation. Besides this, multi-institutional linkages will be helpful in implementing the fluorosis mitigation programin the country. Moreover, different projects can be implemented by the government with community participation. The government of India has launched a program named as “National Drinking Water Mission” to provide safe drinking water for the community and also to combat the problem of fluorosis in the country. Fluorosis mitigation project in Dhar District in Madhya Pradesh; Fluorosis mitigation project at Sonbhadra, Uttar Pradesh; Integrated Fluorosis Mitigation, Madhya Pradesh; Fluorosis Mitigation in Nuapada District, Orissa; Sachetana Plus Fluoride Mitigation Project in Karnataka; Project SARITA in Dungarpur of Rajasthan; Mitigation of Fluorosis in Nalgonda District’s Villages etc. are some important programmes which have been conducted by the government.

9. Conclusion

The occurrence, detection and the removal methods of Fluoride in fresh water samples have been studied in the paper. Primarily, the detection of fluoride in water samples either fresh or waste water samples is the most important point which should be considered. Therefore, movable water testing laboratories covering fluoride analysis including cheaper fluoride testing kits are required in rural areas especially in hilly parts which are the places having fresh water resources without any water testing facilities.

The chemical and geographical conditions impact the defluoridation efficiency accordingly. The selected method for defluoridation may vary as per the geological conditions of the area. Therefore, the technology should be adopted using the actual water sample which is to be treated before implementation in the particular area. Fluoride contaminated water creates the health difficulties globally including India. In this paper, we emphasized the various techniques to resolve the problems of higher concentration of fluoride ions in fresh water. Among several defluoridation technologies, adsorption method possesses many advantages over the other methods such as more accessibility, cheaper method, availability of a broad range of adsorptive materials, simple operation etc. Fluoride removal from water samples by activated alumina is accepted treatment technology by the researchers. Therefore, various International Organizations have classified the activated alumina adsorption as one of the best established available technologies for fluoride removal. In the view of above facts, it is proposed that adsorption based techniques are better than other contemporary techniques for fluoride removal from fresh water in urban as well as rural areas. This technique is not only a cost effective method but also don’t need specific instruments and power supply.

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