European Journal of Environment and Public Health
2024, 8(2), em0158
e-ISSN: 2542-4904
https://www.ejeph.com

MODESTUM

Review Article

OPEN ACCESS

Defluoridation of drinking water by using low cost point of use treatment technologies: A review

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Citation: Tagar S, Rizwan M. Defluoridation of drinking water by using low cost point of use treatment technologies: A review. EUR J ENV PUBLIC HLT. 2024;8(2):em0158. https://doi.org/10.29333/ejeph/14739

ARTICLE INFO

Received: 20 May 2024

Accepted: 31 May 2024

ABSTRACT

Fluoride is an anionic pollutant found in surface or ground water in large quantities due to various human activities, for examples, disposal of industrial wastewater or geochemical reactions. The presence of fluoride in drinking water above certain limits has intense effects on human health. It strengthens the tooth enamel to a small level (1.0-1.5 mg/L). In drinking water, the presence of fluoride in the range of 1.5 to 4.0 mg/L may give rise to dental fluorosis at initial the stage, while the continuous exposure to high fluoride concentrations (4.0-10.0 mg/L) leads to skeletal fluorosis. In many countries of the world, including Pakistan, fluoride exists in ground water in high concentration, reaching above 30.0 mg/L on a large scale. The objective of writing this article is to offer accurate information on the efforts of a number of scholars who worked on fluoride removal from drinking water. The fluoride removal techniques have been categorized into two parts dealing with coagulation/precipitation and adsorption. Lime and alum (Nalgonda technique) and chitin have been discussed under coagulation technologies, while adsorption deals with a number of adsorbents, i.e., activated carbon, activated alumina, saw dust, bone char, rice husk ash, bauxite, tea-ash, and kaolin. Each technique discussed can remove fluoride under certain conditions. Each treatment technology has its limitations, and since there is no technology that can achieve its purpose in diverse conditions, the choice of fluoride removal techniques should be according to a specific site, depending on fundamental conditions and the needs of the local area.

Keywords: defluoridation, POU treatment, drinking water, adsorption, coagulation

INTRODUCTION

Clean water is scarce and not readily available everywhere. Deprived communities around the world rely on the consumption of contaminated water, which affects their health in a number of ways [1]. The quality of water can be undermined by various pollutants of which fluoride stands out as the first contamination of geogenic origin in many countries [2]. By adding a safe quantity of fluoride to drinking water, teeth can be protected from decaying and cavity risk can be minimized. However, it has been identified that overexposure to fluoride can lead to brain disorders, hyperactivity, and disordered muscles [3]. According to World Health Organization (WHO), in drinking water, allowable limits of fluoride are 1.5 mg/L as the maximum concentration [4].

Fluoride is naturally present in groundwater, soil, rocks, and biological chains, namely, human beings, fauna, and flora [5]. The presence of fluoride in ground water is based upon various factors such as, alkaline pH, bicarbonate ion (HCO_3), environmental conditions, water depth, geology, soil porosity and consistency [6]. In human body fluoride enters in many ways, such as breathing, the foodstuff we eat and drinking water. Drinking water is believed to be the main source of human contact with fluoride [7]. WHO estimates that 80% of all waterborne diseases are caused by the consumption of contaminated water and 65% of diseases occur due to drinking fluoridated contaminated water [8]. The disorders caused due to long term intake of fluoride are kidney and neurological disorders, thyroid and gland dysfunction. Other health issues include low intelligence in children and dental and skeletal fluorosis [9]. The details of the health impacts of excessive fluoride are given. However, at recommended levels of 1.5 mg/L in drinking water, fluoride has many positive impacts on our dental as well as skeletal tissues [10]. In many countries around the world, millions of people depend heavily on ground water, which is highly contaminated by fluoride. According to recent estimates, in 25 countries around the world, around 200 million people are destined for fluorosis. China and India, the two most populous nations in the world, are the most affected [11]. In Pakistan, the data on fluoride was analyzed for 29 major cities. Out of those 29 cities, 34% of cities show high levels of fluoride beyond the safe limit of 1.5 mg/l. The maximum values in Quetta, Lahore, and Tehsil Mailsi are 24.48, 23.60, and >5.5 mg/L [11].

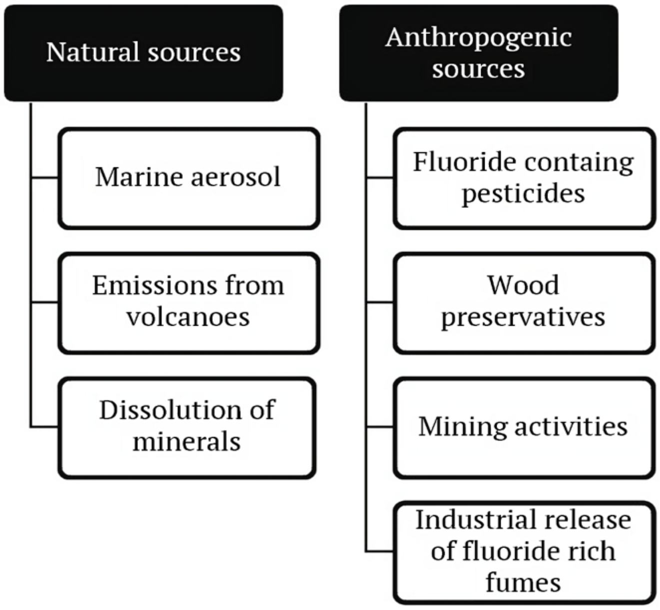

Figure 1. Anthropogenic & natural sources of fluoride in water (Source: Xu et al., 2022) [39]

Fluoride is found in both surface and ground waters since some fluorine-containing compounds present in the higher portion of the earth's crust easily dissolve in water. The average content of the earth's crust is 300 mg/kg in a widespread range of mineral deposits, i.e., apatite, fluorspar, hornblende, stone phosphate, mica, cryolite, and others [12]. The discharge of wastewater from different industries, such as the glass manufacturing industry, aluminum industry, and semiconductor industry, also contributes to fluoride pollution, especially in ground water [13]. Mainly, the sources of excessive fluoride in water are divided into two categories, i.e., natural sources and anthropogenic sources, as shown in **Figure 1**.

Since it has no cure, the only solution for fluorosis is prevention through the removal of fluoride from contaminated water. Routine techniques of fluoride removal, such as Nalgonda technology, activated alumina, reverse osmosis, etc., are widely used to supply safe water to communities affected by fluorosis [2]. However, some small and medium sized defluoridation plants do not work in most cases, possibly due to problems with operation and maintenance [14], and therefore, easy-to-use household filters designed for domestic use would be of great importance in providing fluorine-free water. As a result, this review focuses on point-of-use (POU) defluoridation techniques such as coagulation-precipitation and adsorption. Their working mechanism, advantages, and limitations are described later. The purpose of this review was to collect published material to provide readers with one publication explaining various ways to reduce fluorosis.

HEALTH IMPACTS OF WATER CONTAINING FLUORIDE

Dental Fluorosis

Dental fluorosis, another name for mottled enamel, may arise when levels of fluoride in drinking water slightly exceed to 1.0 mg/L [15]. During the process of tooth development, the intake of excessive fluoride results in an increase in the porosity of enamel. The fluorosis mechanism includes insufficient replacement of the organic enamel matrix with inorganic material that leads to hypomineralization of fluorotic enamel [16]. The severity of fluorosis depends on factors like dosage, age at which the exposure occurs, and duration of exposure. Typical dental fluorosis manifestations include loss of shinning of teeth and the appearance of thin yellow lines on the surface of teeth. The children born in or brought up in areas effected by fluorosis endemic are having high chances of developing dental fluorosis. Some of the signs of dental fluorosis are tooth extraction, inflammation, and toothache. The cure of dental fluorosis is possible with the help of some practices, such as tooth whitening or surface stain removal procedures [17]. When the above symptoms are seen in adults, it clearly indicates that the person in his or her childhood has been exposed to high fluoride levels [18]. Despite the research of many years, it is not possible to determine the correct limit of fluoride in drinking water [19]. Many aspects, including genetic predisposition, diet, individual metabolism, and regional attitude, influence the degree of fluorosis. In addition, it is assumed that nutritional status affects both dental fluorosis and fluoride metabolism [20].

Skeletal Fluorosis

This type of fluorosis impacts all age groups, including children. The symptoms of this disease do not show up until it reaches an advanced stage. The deposition of fluoride in the joints of the shoulder bones, joints of the bones of the neck, pelvis, and knee causes difficulty in movement and walking. Its signs and symptoms are similar to arthritis or spondylitis. At the initial stage, symptoms may include chronic fatigue, muscle weakness, sporadic pain, a sensation of burning, back stiffness, irregular deposition of calcium in the bones, and pricking and tingling in the limbs. Osteoporosis in long bones and bony outgrowths may develop later in the process. The vertebrae fuse with each other, and the affected person will eventually be crippled. Osteosarcoma, a rare bone cancer, may also occur, which in turn damages the spine, muscles, nervous system, and major joints [21].

Other Health Issues

This part of fluorosis is often ignored because of the concept that fluoride only damages teeth and bones [22]. Besides dental and skeletal fluorosis, excessive fluoride consumption may lead to deformities in RBCs, low hemoglobin levels, skin rashes, headaches, excessive thirst, nervousness, gastrointestinal problems, an itchy feeling in fingers and toes, depression, male sterility, repeated abortions or stillbirths, abdominal pain, reduced immunity, nausea, urinary tract malfunctioning, muscle fiber degeneration and neurological manifestations etc. It may also be responsible for causing damage to the reproductive system, respiratory system, excretory system, and central nervous system, as well as 60 enzymes. The effects of excessive fluoride in water have similar effects on animals to those on humans. The constant use of water with a high concentration of fluoride also negatively affects crop growth [21]. Different types of fluorosis

Table 1. Effects of excessive fluoride concentration on human health

Fluoride concentration in drinking water (mg/l)	Effects on human heath
<1.0	Allowable limits
1.0-3.0	Dental fluorosis (pitting, staining, & mottled patches on teeth)
3.0-4.0	Thickened and fragile joints and bones
4.0-6.0 & above	Deformations in hip & knee bones leading to paralysis, which makes a person walking or moving difficult

resulting from too much fluoride intake are described briefly in **Table 1** redraw from [21].

POINT OF USE TREATMENT TECHNOLOGIES

The purpose of treatment of fluoridated water is to reduce the fluoride concentration from drinking water and to maintain it to the standard limit. In fact, water defluoridation can be led at two organizational levels, i.e., domestic or household level and community level. As domestic defluoridation, it is done by a single household for their own consumption of water, while at the community level it is done at the village, city, and suburban level [23]. As it has been previously reported, in this review only household (POU) technologies are discussed. The developed regions of urban areas treat their drinking water in centralized treatment systems, and after treatment, water is directed to the residents through a widespread network of pipes. A centralized treatment system, while efficient in providing clean drinking water, is not applicable to all communities. If the houses are scattered from each other or if the government does not have enough funds for the necessary set-up, centralized treatment will not be available [24]. In such conditions, people may get water from improved sources, for example, public taps or covered wells, instead of tap water from a centralized source. However, even if the water is clean and free from contamination at the source level, it may be contaminated during transportation via contaminated pipes and while being stored in dirty containers [25]. In this situation, POU treatment systems could be a solution to overcome this problem.

In a POU treatment system, water is treated at the place, where it is used, for example, at home or school. By using these treatment systems, the contamination risk between the source of water and POU is reduced by providing treatment at home [26]. POU systems are cost-effective, durable, and simple to use. However, it should be designed, maintained, and operated properly to effectively remove contaminants from drinking water [24]. The design of POUs and their operation will be most effective if they address the contamination of the source of drinking water. They are divided in two categories: adsorption and coagulation technologies. Each of these technologies has its own advantages and disadvantages. **Figure 2** shows percentage of fluoride continent wise.

Coagulation Precipitation

In the coagulation process, the alum or any other coagulant used contains an opposite charge to that of fluoride, and it neutralizes the charge on fluoride. This technique incorporates fluoride precipitation or co-precipitation using some suitable reagents such as alum and lime coagulant

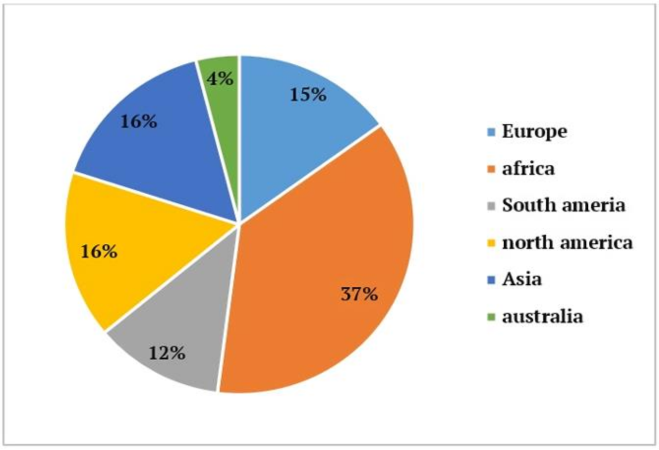

Figure 2. Percentage of fluoride continent wise (Source: Fawell et al., 2006) [40]

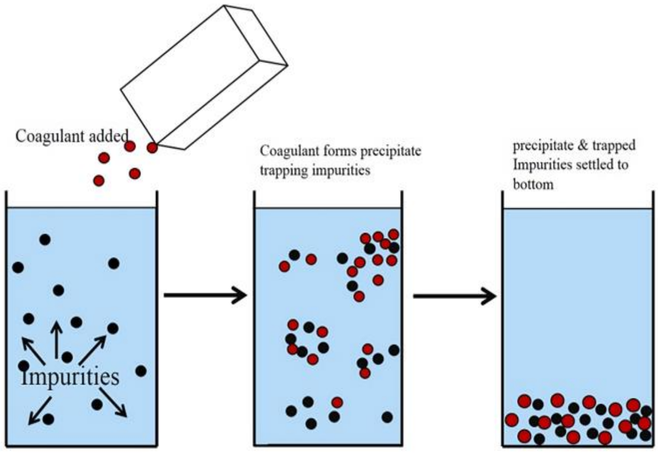

Figure 3. Mechanism of adsorption-1 (Source: Solanki et al., 2021) [41]

(Nalgonda technique) and chitin [27]. In **Figure 3**, the working mechanism of coagulation and precipitation is shown. It is a three-stepped process. In the first step, coagulant is added. In the second step, coagulant forms precipitate, trapping impurities present in the water. After that, precipitate and trapped impurities settled at the bottom of the container.

Nalgonda technique can be applied at different levels (**Figure 4**). It is introduced on a home scale in a bucket or drum and on a community scale in a fill-and-draw plant [28]. The most important coagulants for Nalgonda technique are lime and alum. At the moment we add alum to the water, essentially two reactions take place. In the first reaction, alum reacts with the alkaline component to form insoluble aluminum hydroxide (Al[OH]3). In the second reaction, the aluminum reacts with the ions of fluoride present in the water. Maximum removal of fluoride is possible at pH range of 5.5-7.5 [29]. At domestic

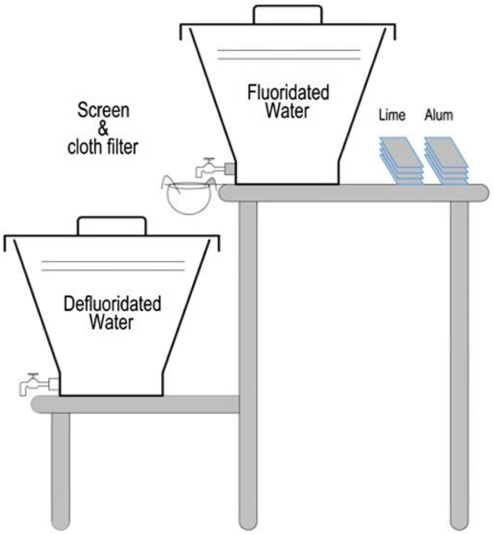

Figure 4. Nalgonda technique (Source: Choubisa, 2023) [42]

level, this technique is able to filter at least 20 liters of water per day [30].

Chitin and deacetylated chitosan, naturally rich biopolymers, have been used widely for the adsorption of metals from water. However, the use of these materials for the adsorption of fluoride from water has been rarely explored.

Despite its widespread usage, chitosan's ability as an adsorbent has not been understood to a satisfactory level yet, as it flakes or swells in water and crushes, which is why it is not suitable for use in an adsorption column [31]. However, we can increase its capacity by distributing chitosan on any physical support, which will rise availability of adsorbate-binding sites. To remove excessive fluoride concentration, applicability of chitosan, chitin, and 20% lanthanum-containing chitosan has been studied by many researchers. As compared to using bare chitin or chitosan, these adsorbents show much better removal efficiency in terms of fluoride. Nevertheless, coagulation methods in view of aluminum salts have some advantages and disadvantages, which are discussed here.

When compared with other defluoridation techniques, coagulation techniques are more practical. Coagulation techniques are easy to understand. However, in coagulation techniques, there is an issue of sludge transfer. The final concentration of fluoride in the purified water is largely dependent on the solubility of the calcium, aluminum salts, and precipitated fluoride. Due to the release of aluminum in treated water, Alzheimer's syndrome may be caused. The use of aluminum sulfate as a coagulant significantly increases the concentration of sulfate ion that can cause a cataractic effect on humans. In **Figure 4**, a domestic-level Nalgonda technique treatment plant is shown, redrawn from [27].

Adsorption

Adsorption is the bonding of molecules from a bulk solution to the surface of a solid by physical or chemical forces. Unlike various defluoridation techniques, the adsorption method is excellent due to its simplicity and accessibility to an extensive range of adsorbents [32]. Adsorption onto solid surfaces is a straightforward and flexible procedure for the removal of fluoride from drinking water, particularly at the

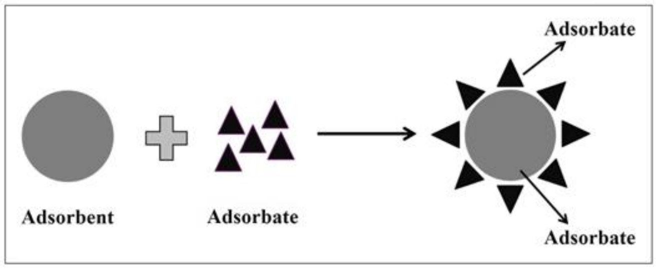

Figure 5. Mechanism of adsorption-2 (Source: Sadhu et al., 2021) [43]

domestic level. The mechanism includes the entry of water through the contact layer, where fluoride removal takes place either by surface chemical reaction with a matrix of the solid layer or by ion exchange method. **Figure 5** shows the mechanism of adsorption.

In the past, several adsorbents have been used to find an economical and effective defluoridation agent. Bone char, tea ash, red mud, activated alumina, bauxite, and rice husk ash are some of the adsorbents mentioned in various literatures. The most commonly used adsorbents are activated carbon and activated alumina. The efficiency of activated alumina for the removal of fluoride is influenced by hardness and surface loading (ratio of total fluoride concentration to the dose of activated alumina). The defluoridation capacity of activated alumina is not affected by chlorine. The process depends on pH, so the pH of solution should be between 5.0 and 6.0 [33].

The use of activated carbon in powdered form for the removal of fluoride was investigated by Mckee and Johnston 1934, and it gave good results [34]. This treatment method depends on pH and only produces good results below pH 3.0. Because it requires pH adjustment, the use of this material is costly. Many researchers have studied how fluoride can be removed from water by using original and activated forms of red mud [35]. It has been found that the adsorption capacity of the activated form of fluoride is higher than the original form of red mud. The process is dependent on pH. Studies have shown that the maximum fluoride adsorption occurs at pH 5.5 above. It has been found that the maximum required time to achieve adsorption equilibrium of fluoride ions is two hours.

Another effective adsorbent is bone char. The fluoride adsorption on the surface of the bone was one of the earliest methods proposed for defluoridating water supplies. It is an ion exchange in which the carbonate radical of apatite-containing bone, Ca(PO4)6.CaCO3, is replaced by fluoride to form insoluble fluorapatite. Bone char obtained by carbonizing bone at 1,100-1,600°C has superior qualities to untreated bone and therefore replaces bone as a defluoridating agent. A simple bone char defluoridation plant at domestic level is shown in **Figure 6**. The research in [36] concludes that sawdust has good properties for fluoride removal; under optimized conditions, including a contact time of 120 minutes, a pH value of 7.0, and a dosage of sawdust adsorbent of 2.0 g/l, it is possible to achieve a removal efficiency of more than 70% [36]. The new nanomaterial obtained from rice husk used in the study conducted by [37] showed that rice husk is able to act as an adsorbent material for fluorine removal in a cost-effective and promising way. In that study, it was found that the adsorption of fluoride was possible at neutral pH, which is usually

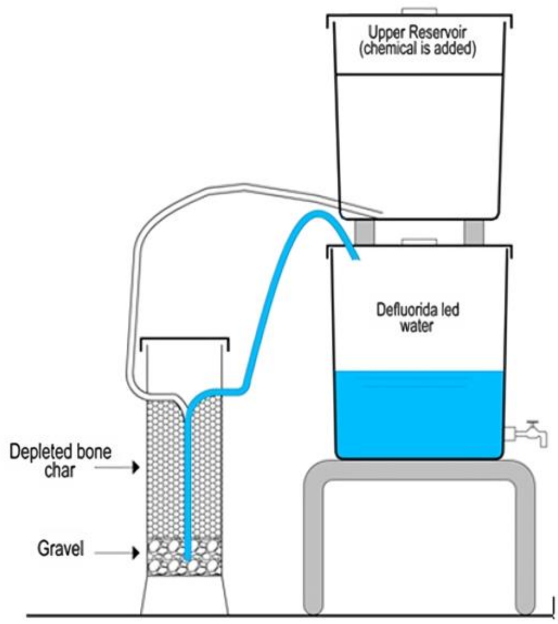

Figure 6. Defluoridation of fluoride by bone char at domestic level treatment plant (Source: Kanouo et al., 2020) [44]

challenging, and it was found that the removal of fluoride is a function of the contact time and the dose of adsorbent at a given initial concentration of solute. The removal percentage decreased with a higher initial concentration and increased with the adsorbent dose [37].

It was concluded that using tea ash powder as bio adsorbent for fluoride removal is reasonable [1]. At neutral pH the tea ach powder with acid treatment is proved better than tea ask treated with alkali. With the increase of pH value, the removal of fluoride also increases with this adsorbent, and the optimum adsorption is found at a dosage of 400 mg/L.

It is observed that the adsorption rate is greater for small particles (212 µm) of the bioadsorbent due to an increase in surface area than for large particles (600 µm) [1].

It was studied thermally activated titanium-rich bauxite (TRB) as an adsorbent for fluoride removal [38]. At a moderated temperature of 300-400 ^{o}C, the adsorption capacity of TRB is significantly increased by thermally activated TRB. The adsorption was rapid, and within 90 minutes the maximum level of fluoride removal was reached. With the increase in pH, the fluoride uptake also increases, it decreases after reaching the maximum at pH 5.5-6.5. In drinking water, the uptake of fluoride from aqueous solution was not affected by the occurrence of common interfering ions, which indicates the sorption behavior of TRB specific for fluoride.

Table 2 presents a typical initial and final concentration of each coagulation and adsorption technology and its corresponding removal efficiency [38]. There are some advantages and disadvantages of adsorption techniques, such as that they are easy to use, regeneration is possible, they produce high-quality water, and they are able to remove fluoride up to 90%. However, concentrated regenerated and lean adsorbents needed to be disposed, Interference due to the

Table 2. Coagulation & adsorption technologies for fluoride removal with corresponding initial & final concentration & removal efficiency

SN	Coagulation technologies	Initial concentration (mg/l)	Final concentration (mg/l)	Efficiency (%)	Citation
1	Lime & alum coagulant	50.00	33.500	18.00-33.00	[39]
		10.00	1.400	86.00	[40]
		7.90	2.800	35.00	
2	Chitin	50.00	4.800	90.40	[40]
		16.70	1.830	89.00	
		5.00	3.800	24.00	
	Adsorption technologies	**Initial concentration (mg/l)**	**Final concentration (mg/l)**	**Efficiency (%)**	**Citation**
3	Activated alumina	35.00	0.360	99.00	[41]
		13.80	0.138	88.00	[42]
		12.00	1.920	84.00	
		30.00	1.500	95.00	
4	Activated carbon	30.00	8.100	73.00	
		6.50	1.060	85.43	[43]
5	Red mud	21.46	2.720	87.30	
		5.00-150.00	10.000	97.60	
		100.00-100.00	5.000-50.000	95.00	
		10.00-70.00	-	-	
6	Bone char	17.43	6.620	62.00-66.00	
		15.00	4.500	70.00	
		30.00	7.500	75.00	
7	Saw dust	5.00	0.600	88.00	
		20.00	1.600	92.00	
		25.00	0.800	92.00	
8	Rice husk	10.00-60.00	1.000-6.000	90.00	
		50.00	25.000	50.00	
		50.00	6.700	80.60	
		3.00-10.00	0.300-1.000	90.00	
9	Tea ash	10.00	3.710	62.85	
		50.00	4.500	91.48	

proximity of various anion species can lead to competition for active sites on the adsorbent, adsorption techniques are highly pH subordinate, and after the regeneration phase, the removal effect is reduced. Moreover, contamination of the alumina layer is possible due to high concentrations of total dissolved solids. The uptake of fluoride from aqueous solutions was not affected by the occurrence of common interfering ions.

In **Table 2**, various coagulation and adsorption techniques along with their corresponding removal efficiency and initial and final concentration of fluoride removal are given. The **Table 2** was created after reviewing the research of various scholars in the field of fluoride removal. All the treatment technologies are effective in terms of fluoride removal. However, the percentage of fluoride removed depends on the initial concentration of fluoride in drinking water. On a similar initial concentration of 50.0 mg/l, chitin gave 90% efficiency, while Nalgonda technique gave 18%-33%. Among adsorption techniques, activated alumina and red mud are more efficient as compared to other adsorbents.

CONCLUSIONS

This review attempts to cover an extensive variety of procedures that have so far been used to remove fluoride from drinking water at the domestic level. Fluorine removal methods are broadly divided into two parts: coagulation and adsorption. Nalgonda technique and chitin are discussed under coagulation procedures. While the use of various adsorbents such as activated aluminia, activated carbon, bone char, red mud, rice husk, tea ash, and bauxite have been studied under adsorption techniques, many different techniques are used to remove additional fluoride from drinking water, but because each process has specific benefits and drawbacks, no method is suitable for defluoridation properly everywhere because some methods are expensive, while others may cause additional problems. Thus, depending on requirements like resource availability, concentration, area, etc., you can choose any suitable technique to remove additional concentrations of fluoride from drinking water.

Author contributions: All co-authors have involved in all stages of this study while preparing the final version. They all agree with the results and conclusions.

Funding: No funding source is reported for this study.

Ethical statement: The authors stated that the study does not require any ethical approval. It is based on existing literature.

Declaration of interest: No conflict of interest is declared by the authors.

Data sharing statement: Data supporting the findings and conclusions are available upon request from the corresponding author.

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