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RESEARCH ARTICLE

"ADVANCEMENTS IN DEFLUORIDATION TECHNOLOGIES FOR SAFE DRINKING WATER: A COMPREHENSIVE REVIEW"

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Abstract

Higher fluoride concentrations in groundwater have been reported in more than 20 developed and developing countries. In the quest for ensuring safe drinking water, the removal of fluoride has emerged as a critical concern globally. This comprehensive review paper delves into the realm of defluoridation technologies, exploring a myriad of methods employed to mitigate excess fluoride levels in water sources. By meticulously examining traditional techniques alongside cutting-edge advancements, this paper provides a detailed analysis of the evolution of defluoridation technologies. From adsorption processes to membrane technologies and electrocoagulation methods, diverse array of approaches are scrutinized for their efficacy in addressing the pressing issue of fluorosis. Through a systematic evaluation of the strengths and limitations of each technology, this review aims to offer valuable insights into the current landscape of defluoridation methodologies. By synthesizing existing knowledge with recent innovations, this paper highlights the progress made in combating fluoride contamination and also underscores the ongoing need for continuous research and development in this crucial field. It has been concluded that the selection of treatment process should be site-specific as per local needs and prevailing conditions as each technology has some limitations and no one process can serve the purpose in diverse conditions.

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Introduction:-

Access to safe drinking water is a cornerstone of public health, enshrined as a fundamental human right by the United Nations General Assembly [(OHCHR, n.d.)]. The World Health Organization (WHO) emphasizes that water intended for human consumption should be free from microbiological and chemical contaminants, ensuring its suitability for various domestic purposes [(Drinking-water Quality Guidelines, n.d.)]. According to the United Nations Children's Fund (UNICEF) and the World Health Organization (WHO) (2017), over 2 billion people globally lack access to safely managed drinking water services, exposing them to potential health risks associated with waterborne contaminants, including excessive fluoride. [(Water Supply, Sanitation and Hygiene Monitoring, n.d.)]. Over the past few decades, the ever-growing population, urbanization, industrialization, and unskilled utilization of water resources have led to degradation of water quality and reduction in per capita availability in various developing countries. The groundwater is getting polluted because of various anthropogenic activities and also natural geogenic compositions [Kass et al. (2005), Oren et al. (2004)].

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While advancements in water treatment technologies have significantly improved global access to clean water, challenges persist concerning the presence of naturally occurring elements like fluoride. Fluoride, at optimal concentrations, plays a crucial role in promoting dental health, particularly in preventing dental caries in children [(Advancing the Nation's Oral Health Through Research and Innovation | National Institute of Dental and Craniofacial Research, n.d.)]. The widespread presence of fluoride in groundwater creates a global health burden, particularly affecting developing countries with limited access to advanced water treatment infrastructure. Research by Fawzy and Gupta (2016) highlights the detrimental effects of excessive fluoride intake in these regions, leading to skeletal and dental fluorosis, a debilitating condition impacting bone health and causing dental enamel damage as shown in Table 1[Fawzy et al. (2016)]. Elevated fluoride concentrations are typically observed in regions near tall mountains or where geological deposits have formed due to seawater influence. Groundwater fluoride concentrations globally span from 0.01 to 48 mg/L [Mumtaz et al. (2015)]. The main source of fluoride in groundwater is considered to be fluoride-bearing minerals such as fluorite $[\text{CaF}_2]$, fluorapatite $[\text{Ca}_5(\text{PO}_4)_3\text{F}]$, cryolite, and hydroxyapatite in rocks [Farooqi et al. (2007)]. Studies by Ingebritsen et al. (2013) emphasize the geographical disparity in fluoride concentration within groundwater sources. Certain regions, particularly in Africa, Asia, and South America, exhibit naturally high levels exceeding the recommended WHO guideline of 1.5 mg/L as shown in Table 2[(Groundwater Quality | U.S. Geological Survey, 2018)]. This uneven distribution of fluoride necessitates the development and implementation of effective defluoridation strategies to safeguard public health.

Addressing this global challenge requires a multi-pronged approach. Research by Liyanage et al. (2015) advocates for improved water quality monitoring systems, particularly in vulnerable regions, to identify areas with excessive fluoride concentrations [Chatterjee et al. (2020)]. Furthermore, the development and implementation of cost-effective and sustainable defluoridation technologies are crucial for ensuring safe drinking water supplies in resource-limited settings. This review paper delves into the advancements made in defluoridation technologies, exploring both conventional methods and promising areas of research aimed at mitigating the challenge of excessive fluoride in drinking water.

Defluoridation Techniques:

Defluoridation techniques encompass a diverse range of methods aimed at reducing fluoride levels in water sources, including adsorption, membrane filtration, and chemical precipitation, reflecting ongoing advancements in water treatment.

Coagulation & Precipitation

Precipitation through coagulation is an economical and effective method for water defluoridation in which charged particles of suspension are neutralized and agglomerate to settle down. pH and temperature of the solution are the fundamental aspects of the precipitation process, and therefore, the addition of specific chemicals or reducing solution temperature makes the solution unstable and aids in precipitation [Adhikari et al. (2019)]. Chemicals such as ferric chloride, ferrous sulfate, lime, potash alum and sodium bicarbonates are commonly used chemicals for precipitation [Aziz et al. (2008)]. However, for fluoride removal, conventionally alum $(\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O})$ and lime $(\text{Ca}(\text{OH})_2)$ have been extensively utilized as coagulants [Nawlakhe et al. (1975)].

The lime is introduced to metal/fluoride polluted water initially, which triggers fluoride precipitates as insoluble CaF_2 or metal as CaM_2 (M represents any heavy metal), altering the pH of the treated water to 11-12. Currently, alum is incorporated, resulting in the production of insoluble $\text{Al}(\text{OH})_3$. In this procedure, lime, alum, and bleaching powder are added to the fluoride-contaminated water, followed by rapid blending. This leads to the co-precipitation of fluoride and the creation of insoluble aluminum hydroxide flocs at the base. This method is also relevant for eliminating heavy metals from wastewater [[Nawlakhe et al. (1975)]. Nevertheless, various modifications and novel technologies for coagulation are emerging that have demonstrated promising outcomes akin to plant-based coagulants, electrocoagulation, and metal ion-assisted electrocoagulation [Govindan et al. (2015)].

It has been noted that due to substantial chemical requirements, operational costs are generally high, and the process generates significant amounts of toxic sludge containing aluminum [Yadav et al. (2018)].

The Nalgonda technique devised by NEERI in India serves as a prime illustration of this method. It entails the addition of lime, alum, and bleaching powder followed by vigorous mixing, flocculation, sedimentation, and filtration. The inclusion of alum and lime aids in the formation and settling of aluminum hydroxide flocs while

bleaching powder is employed for disinfection. The entire operation necessitates approximately 2-3 hours with multiple batches treatable in a day [Ayoob et al. (2008)]. Over time, this technique has been widely adopted and refined with readily available and cost-effective chemicals. However, due to the necessity for consistent mixing, it demands significant labor. Additionally, concerns have been raised regarding an unpleasant taste in water and potential aluminum exposure since permissible limits are very low (0.2 mg/L), which could lead to adverse health effects like dementia [Karunanithi et al. (2018)].

Suspended and dissolved solids from a liquid can also be eliminated by passing an electric current through the solution, disrupting the solids and aiding in settling [Emamjomeh et al. (2011)]. Electrodes utilized in electrocoagulation consist of sacrificial metals like aluminum for fluoride removal that generate flocs of trivalent aluminum hydroxide upon electricity supply [Mouedhen et al. (2008)]. Unlike other coagulation methods that produce substantial sludge volumes, electrical conductivity (EC) results in minimal sludge production without requiring additional chemicals, making it a favourable defluoridation alternative

Adsorption

Adsorption emerges as a favoured defluoridation technology due to its perceived simplicity, efficiency, economic viability, and sustainability. Numerous investigations and reviews have explored adsorption, with a focus on developing various adsorbents. Overall, most adsorption studies are in early stages, primarily concentrating on material development and properties [Lacson et al. (2021)]. It includes physical adsorption or chemisorption by different processes such as chelation, complexation, ion exchange, etc. [Ayoob et al. (2008)].

Porous materials possess the ability to adsorb substances, making them valuable for adsorption purposes. Examples include activated alumina, activated carbon, activated alumina-coated silica gel, calcite, activated sawdust, activated coconut shell carbon, activated fly ash, groundnut shell, coffee husk, rice husk, magnesia, serpentine, tricalcium phosphate, bone charcoal, activated soil sorbent, carbion, defluoron-1, and defluoron-2, among others. The most frequently employed adsorbents are activated alumina and activated carbon. The fluoride removal efficiency of activated alumina is influenced by water hardness and surface loading, where the latter represents the ratio of total fluoride concentration to activated alumina dosage. Interestingly, chloride does not impact the defluoridation capacity of activated alumina [Sharma and Bhattacharya (2016)].

Due to its simplistic design, ease of operation, cost-effectiveness, efficiency, and reusability, adsorption is one of the most widely considered water defluoridation technique especially for small communities or even for household applications [Zendehdel et al. (2017)]. Furthermore, utilizing local materials enhances cost-effectiveness. The effectiveness of adsorption is influenced by factors like the physical and chemical characteristics of the adsorbent, the amount used, its attraction to fluoride ions, initial fluoride levels, and capacity for adsorption [Akbari et al. (2018), Akafu et al. (2019)]. In practice, activated alumina defluoridation techniques are actively disseminated in various villages through initiatives supported by UNICEF and other agencies, showcasing potential advantages such as a fluoride removal capacity of up to 90% and cost-effectiveness. However, these benefits must be weighed against the limitations and challenges posed by the method [Meenakshi and Maheshwari (2006)]. Besides, properties similar to higher adsorption capacity and ease of regeneration are also desired in an adsorbent [Ayoob et al. (2008)].

Ion Exchange

Ion exchange is a water treatment technique that can eliminate objectionable ions like fluoride along with some other ions such as chloride (having the same charge) which are not harmful or less objectionable. It has been a traditional fluoride removal process for many years. Ion exchange materials are insoluble in water and hold the replaceable ions loosely, which are used for exchanging ions from the solution [Sharma and Bhattacharya (2016)]. Metal/fluoride ions are swapped in this process with ions in dilute solutions held by electrostatic forces. This method is utilized to separate and purify metals. In this cycle, contaminated water continuously passes through a bed of ion-exchange resin in an up-flow or down-flow direction until the resin is exhausted [Punia et al. (2022)].

Ion exchange materials can be categorized as natural and synthetic. Natural materials include cellulose, certain soil particles, and proteins, while synthetic materials can be further classified as membranes and beaded polymer resins. Depending on the functional group attached to the matrix, ion exchange resins can be divided into anionic and cationic types. Anion exchangers such as inorganic metallic oxides exchange negatively charged ions (like fluoride), whereas cation exchangers such as zeolites exchange positively charged ions from the solution [Yadav et al.

(2018)]. Due to the weak binding force of the exchanged ion, it is loosely attached to the base and can be easily replaced by another chosen ion passing through a functional group [Shahid et al. (2023)].

Water flows down through an ion exchange packed column, which binds the desired ions to be removed. As the resin becomes saturated, it is back washed with a mild acid or alkali solution. In the case of fluoride, anion exchange resins with quaternary ammonium functional groups are used, which replace fluoride with chloride attached to these functional groups. Upon saturation, the resin is backwashed with a supersaturated sodium chloride salt [Raghav et al. (2019)]. The substitution of chloride in the resin with fluoride from the solution occurs due to the higher electronegativity of fluoride ions [Razbe et al. (2013)]. Indion FR, a commercial ion exchange resin and an anion exchanger like Ceralite IRA 400 for replacing chloride ions have shown efficiency of up to 95% [Wang et al. (2014)].

Ion exchange has proven to be an efficient process for fluoride removal due to its simplicity in eliminating ionic contaminants. Strong anion-exchange resins have been known to remove up to 95% of fluoride ions from aqueous solutions. However, ion exchange resins are exhaustive, require longer reaction times, frequent regeneration, and generate a large volume of wastewater, making it less attractive [Kumar et al. (2019)]. Moreover, the need for a large volume of regenerant for resin regeneration also restricts the use of this technique. While resins can be regenerated easily, they are costly and make the treatment uneconomical. Unfortunately, the regeneration process produces a significant amount of fluoride-loaded waste that requires disposal, which is a drawback of this method. Additionally, the process efficiency is relatively low and strongly influenced by the presence of other anions (such as sulphates, carbonates, nitrates, phosphates). [Yadav et al. (2018), Grzegorzec et al. (2020)]. Samadi et al. explained that the maximum capacity was achieved 13.7 mg/g at pH = 7 by ion exchange method [Samadi et al. (2014)]. Another study reported the maximum fluoride loading of 15.77 g/kg of resin [Millar et al. (2017)].

Membrane Filtration

Advanced defluoridation technologies that provide pure and ultrapure water use membrane processes, which involve a semi-permeable membrane to separate phases and remove water contaminants such as fluoride effectively [Velázquez-Jiménez et al. (2015)]. These membranes can be categorized as natural (cellulose acetate, cellulose triacetate) and synthetic (polysulfone, polyamide) based on the material used. The membrane's pore size and material selection depend on the substance to be separated. Depending on the methods employed to segregate fluoride using membranes, the process can be further subdivided into categories such as reverse osmosis, nanofiltration, ultrafiltration, dialysis, and electro dialysis.

The benefits of membrane processes include high fluoride removal efficiency, effective barrier against various pollutants, single-step treatment and disinfection, consistent water quality, minimal chemical requirements, low maintenance, extended membrane lifespan, broad pH range operability, and straightforward automated operation. However, drawbacks encompass the elimination of essential minerals necessitating remineralization, relatively higher costs compared to alternative methods, acidic water requiring pH adjustment, significant water wastage as brine, and complexities in brine disposal. Despite some limitations, the merits of membrane processes render them a compelling choice for drinking water production, particularly with enhanced management practices [Meenakshi and Maheshwari (2006)].

Reverse Osmosis (RO)

Reverse osmosis is a process where heavy metals or fluoride ions are isolated by applying pressure more than osmotic pressure on a semi-permeable membrane by the solids dissolved in wastewater. It is used for the desalination of seawater and brackish water. RO is a membrane method to eliminate molecules and ions from solutions. Reverse osmosis (RO) is a highly effective method for removing all inorganic pollutants from water. Numerous researchers have focused on RO technology in the past to address fluoride removal from source water [Schneiter and Middlebrooks (1983)]. Membranes used in reverse osmosis vary depending on the type of water to be treated, economic considerations, and working conditions such as temperature, pressure and membrane recovery. RO is affected by various parameters like ionic strength, type of ionic exchange membrane used, pH, presence of co-existing anions and applied potential, etc. [Kabay et al. (2008)]. Nevertheless, RO is not affected by initial concentration in water as up to 90% of fluoride can be removed using reverse osmosis [Waghmare et al. (2015)].

Electrodialysis (ED)

Electrodialysis is a form of direct current-driven electrochemical membrane technology used for separating ions, such as fluoride, without relying on pressure like reverse osmosis [Ahmed et al. (2019)]. Depending on the charge of the ions, membranes in electrodialysis can be categorized as anion and cation exchange membranes. Under a constant electric field, ions move through ion exchange membranes, with anions migrating to the anode and passing through anion exchange membranes, while cations cannot pass through and vice versa for cations, resulting in dilute and concentrate streams [Grzegorzec and Majewska-Nowak (2016)]. In a study by Ben Sik Ali Ali et al., it was demonstrated that the efficiency of the electrodialysis process was 86.2% for defluoridation [Ali et al. (2010)]. Another research indicated that electrodialysis could achieve fluoride removal rates ranging from 80% to 90% [Grzegorzec et al. (2020)]. Findings from a study showed that electrodialysis could eliminate 50–60% of fluoride within 6 minutes [Belkada et al. (2018)]. Additionally, one study reported that the fluoride removal efficiency through electrodialysis ranged from 50% to 90% [Chibani et al. (2019)]. While another study claimed a fluoride removal rate of 92% from drinking water using the electrodialysis method [Gmar et al. (2015)].

Nanofiltration

Nanofiltration is a process that falls between the upper end of reverse osmosis (RO) and the lower end of ultrafiltration. The permeability of nanofiltration membranes is higher than that of RO membranes. Nanofiltration membranes exhibit a high retention of charged particles. It necessitates lower pressure and investment compared to RO and finds broad application, particularly in drinking and wastewater treatment, and is utilized in studies on fluoride removal [Harma et al. (1999), Tahaiqt et al. (2007), Bejaoui et al. (2011), Hoinkis et al. (2011)]. This approach seems to be the most effective among all membrane techniques for eliminating fluoride due to its high and specific membrane selectivity. Some challenges of this method that require improvement include membrane fouling, insufficient separation and rejection, chemical durability, and limited membrane lifespan [Yadav et al. (2018)]. Among various defluoridation methods, nanofiltration is a successful technique for water treatment when compared to other membrane methods like RO and electrodialysis (ED) [Dhillon et al. (2016)]. One research indicated that the retention of fluoride anions by nanofiltration was around 60% [Bejaoui et al. (2011)]. In a study, Chakraborty et al. demonstrated that the composite polyamide nanofiltration membrane employed in the cross-flow method effectively removed 98% of fluoride from contaminated water [Chakraborty et al. (2013)]. Another study highlighted that the retention of fluoride by the HL membrane surpassed 80%.

Electrolytic Defluoridation

The Electrolytic Defluoridation (EDF) method, developed by the National Environmental Engineering Research Institute (CSIR-NEERI) in India, aims to combat high levels of fluoride in water sources. This technique operates on the principle of electrolysis by directing Direct Current (DC) through aluminum plate electrodes submerged in water containing fluoride. Throughout the process, the aluminum plate connected to the anode dissolves, forming polyhydroxy aluminum species that eliminate fluoride through complex formation, adsorption, and settling. In an electrolytic defluoridation process the 'in situ' generation of coagulating ions occurs in three consecutive stages viz. (i) electrolytic oxidation of anode resulting into formation of coagulants; (ii) destabilization of fluoride ions and (iii) aggregation of the destabilized phases resulting into floc formation.

Electrocoagulation has a well-established track record as a water treatment technology utilized for the removal of a diverse array of pollutants. However, electrocoagulation has never become accepted as a mainstream water treatment technology. The absence of a structured methodology for designing and operating electrocoagulation reactors, along with concerns about electrode reliability, such as electrode passivation, has restricted its widespread adoption. Nevertheless, recent advancements in technology, coupled with an increasing demand for small-scale decentralized water treatment systems, have prompted a reassessment of electrocoagulation [Holt et al. (2005)]. Various operational factors like initial pH, current strength, influent fluoride levels, flow rate, and residual aluminum were taken into account. The pH level was identified as a crucial factor significantly impacting fluoride removal. The optimal influent pH range for effective defluoridation was determined to be 6.0–7.0, with 6.5 being the preferred value. The EDF plants were shown to produce treated water with fluoride concentrations below 1 mg/L. It was observed that the current intensity minimally affects fluoride removal, while the required residence time increases with higher initial fluoride concentrations [Holt et al. (2005)].

Phytoremediation

Phytoremediation involves the use of specific plants to cleanse contaminated water, soil, and sediment. It employs living green plants to eliminate pollutants from polluted water, air, soil, and sediments. Specially selected or engineered plants are employed in this process. Plants are exposed to a metal solution, and their roots and stems accumulate metal ions, thereby purifying the water. However, this remediation process is time-consuming, requiring the regeneration of plants for further use [Baunthiyal and Ranghar (2013)]. Limited research has been conducted on fluoride removal through phytoremediation. Typically, hyperaccumulator plants that exhibit significant fluoride accumulation with minimal toxicity are preferred for fluoride removal [Sharma et al. (2014)]. Additionally, plants with a fibrous root system and high biomass are well-suited for phytoremediation, such as trees over herbs and shrubs. They absorb contaminants through their roots and transport them to other above-ground parts for storage, Khandare [Khandare et al. (2017)] attempted fluoride removal using garden ornamentals like Nerium oleander, Portulaca oleracea, and Pogonatherum crinitum, noting positive results, particularly with Nerium oleander (92%) compared to other plant species. In a comparative study by Karmakar [Karmakar et al. (2015)] on the effectiveness of three aquatic plants for fluoride removal under low fluoride contamination levels, Pistia stratiotes exhibited the highest efficiency (19.87%), followed by Spirodela polyrhiza (19.23%) and Eichhornia crassipes (12.71%). Similarly, Baunthiyal and Sharma [Baunthiyal and Sharma (2017)] assessed the defluoridation potential of various hydrophytes in aquatic environments, including Cladophora glomerata, Hydrilla verticillata, and Chara coralline, with Chara coralline demonstrating superior performance compared to other macrophytes.

Conclusion and Recommendation:-

In conclusion, this review has explored the growing challenge of fluoride contamination in drinking water and the importance of defluoridation technologies in ensuring safe water supplies. We have examined a range of established and emerging defluoridation techniques, each with its advantages and limitations.

Adsorption techniques, particularly those utilizing low-cost, naturally occurring materials, offer a promising solution for decentralized and community-based defluoridation. Membrane filtration technologies like nanofiltration demonstrate high efficiency but require significant infrastructure investment. Coagulation-precipitation methods are well-established and cost-effective but require careful monitoring and sludge disposal strategies.

Overall, the optimal defluoridation approach depends on several factors, including the severity of fluoride contamination, community size, infrastructure availability, and operational costs.

Based on the reviewed advancements, future research efforts should focus on:

Developing low-cost, sustainable, and easily deployable defluoridation technologies suitable for resource-limited settings. Enhancing the efficiency and selectivity of existing methods to achieve optimal fluoride removal while minimizing waste generation. Exploring the potential of hybrid or combined defluoridation systems that leverage the strengths of different approaches. Investigating the applicability of nanotechnology for developing novel and highly effective defluoridation materials.

By pursuing these research avenues, we can ensure continued progress in providing safe drinking water free from excessive fluoride for communities worldwide.

Tables:-

Table 1:- Adverse consequences of consuming fluoride on human health.

Fluoride concentration, mg/L	Health Outcome
< 0.5	Encourages the development of tooth decay.
0.5-1.5	Achieving optimal oral health and fostering the growth of robust bones and teeth.
1.5-4.0	Dental fluorosis
>4.0	Fluoride-related issues affecting dental and skeletal health.
>10.0	Encourages the onset of debilitating skeletal fluorosis and increases the risk of cancer.

Table 2:- Amount of fluoride level in groundwater of several nations.

Country	Region/province/city	Fluoride level (mg/L)	Ref.
Ethiopia	South Ethiopian	Shallow wells: 0.5–1.29; deep wells: 0.48–5.61	[Haji et al. (2018)]
Malawi	South Malawi	1.5–6	[Addison et al. (2020)]
Iran	West Azerbaijan	Warm seasons: 0.01–3; cold seasons: 0.01–4	[Asghari et al. (2017)]
India	Telangana	0.4–2.2	[Adimalla and Rajitha (2018)]
Thailand	Lamphun and Northern Thailand	0.01–14.12	[Chuah et al. (2016)]
China	Northern Anhui Province	0.55–2.06	[Hao et al. (2021)]
Sweden	Kalmar	0.1–15.0	[Augustsson and Berger (2014)]
United States	-	0.7–4.0	[McMahon et al. (2020)]
Italy	Aosta Valley Region	0.03–1.14	[Tiwari et al. (2017)]
Nigeria	Southwestern Nigeria	Mean: 1.23	[Emenike et al. (2018)]
Ghana	Upper East Region of Ghana	0.5–4.6	[Craig et al. (2018)]
Mexico	Central region in Mexico	0.56–1.60	[Irigoyen-Camacho et al. (2016)]
Pakistan	Sindh and Punjab	0.1–3.9, and 0.1–10.3	[W. Ali et al. (2019)]
Sudan	Tiraat El-Bijah and Um Duwanban	0.45–1.36	[Bhattacharya et al. (2016)]
Tanzania	East African Rift	0.5–10	[Zabala et al. (2016)]
Argentina	Del Azul Creek basin	Above 1.5	[Zabala et al. (2016)]
Benin	Central Benin	1.5–3.02	[Avoceföhoun et al. (2018)]

Table 3:- A comparison between the fluoride removal methods.

Process (name of the methods)	Type of environment	Removal performance	Advantages	Disadvantages	Ref.
Adsorption (Exhaustive coffee grounds and iron sludge)	Groundwater	Iron sludge 62.92% and exhausted coffee grounds 56.67%	Cheap and easily available adsorbents	Low efficiency removal	[Melak et al. (2019)]
Adsorption (porous starch loaded with common metal ions)	Drinking water	Maximum adsorption capacity of porous starch with Zr (PS-Zr) of 25.41 mg/g	Use of commercial scale	-	[Xu et al. (2017)]
Adsorption (nepheline from alkali hydrothermal)	Aqueous solutions	Maximum adsorption capacity of 183 mg/g	Cheap adsorbents	High efficiency and adjustment of pH	[H. Wang et al. (2017)]

NF and RO	Groundwater	Fluoride rejection: 98% for RO and 90% for NF	High efficiency	Membrane fouling, decreased membrane lifetime and chemical persistence, high capital operation and maintenance costs, and hazardous effluent generation	[Brião et al. (2019), Dubey et al. (2018)]
Ion exchange, membrane filtration, and EC	Aqueous solutions	90%–95%, 99%, and 85.5%	High efficiency	Costly techniques, production of waste, and recommended for small community systems	[Kumar et al. (2019b)]
Adsorption (purolite A520E resin)	Aqueous environments	64.6%	Good stability and flexibility	Expensive processes	[Nasr et al. (2014)]
NF	Groundwater	98%	High efficiency	High capital and running and maintenance costs	[Chakraborty et al. (2013)]
Adsorption (CuO NPs)	Aqueous solutions	97%	High efficiency	–	[59]
Adsorption (Earth modified alumina)	Aqueous solutions	Adsorption capacity of F ⁻ : 26.45 mg·g ⁻¹	Easy utilization and high efficiency	Limited yield and long exposure time	[He et al. (2019)]
Adsorption (fungus hyphae-supported alumina)	Aqueous solutions	Nearly 90%	Economical and effective technique	Long exposure time	[Yang et al. (2017)]
Freezing temperature	Water solutions	Deionized water spiked with fluoride 85% and salinity 75%	High efficiency and little contamination	More susceptible to the freezing temperature	[Y. Yang et al. (2017)]
Adsorption (diatomite modified with aluminum hydroxide)	Aqueous solution and natural groundwater	89%	Low-cost	Leak of soluble alumina	[Akafu et al. (2019)]
Adsorption (Zirconium onto tea powder)	Drinking water	Adsorption capacity of 12.43 mg/g	Effective, and safe biosorbent	A slight functional pH span	[Cai et al. (2016)]
Adsorption (activated carbon: banana peel and coffee husk)	Aqueous solution	80% to 84%	Cheap, simple, and environment friendly	Limited efficiency and long exposure time	[Tilahun et al. (2014)]
Adsorption (singlewalled carbon nanotubes)	Aqueous solution	87%–100%	Low cost	Generation of toxic waste	[Balarak et al. (2016)]

Adsorption (Mg/Ce/Mn oxide-modified diatomaceous Earth)	Aqueous solution	>93%	Low cost and simple operation	High yield often demands adjustment of pH	[Gitari et al. (2020)]
Adsorption (aegle marmelos)	Aqueous solution	52%	Low cost	Low efficiency	[Singh et al. (2017)]
Precipitation/coagulation (lime and alum)	Aqueous solution	-	Simple process and little energy requirement	High cost of maintenance and production of hazardous waste	[Waghmare et al. (2015)]
MOFs	Aqueous solution	Adsorption capacity of 41.36 mg/g	High surface area and high porous	-	[Zhao et al. (2014)]
EC: electrocoagulation; NPs: nanoparticles; NF: nanofiltration; MOFs: metal organic frameworks.					

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