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Fluoride in groundwater: Causes, impacts and their potential remediation techniques

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Abstract
Groundwater is used worldwide. Groundwater fluoride is one of the world's largest toxicological environmental threats. Groundwater fluoride primarily results from weathering and leaching of fluoride-bearing minerals from rocks and sediments. Fluoride reduces dental caries at 0.5 mg/L, but at 1.5 mg/L or higher, it can cause fluorosis. 200 million people throughout 25 countries have fluorosis. There are several low-cost and simple defluoridation methods, but rural populations have yet to benefit from them due to barriers. Rural communities require fluoride-safe drinking water. The study compared fluoride geochemistry, groundwater contamination, human exposure, adverse effects and possible fluoride toxicity treatment options. This review examines fluorosis treatment options, water fluoride reduction technologies, and protective methods. This study also examines national, regional, and Indian fluorosis prevention efforts.

Keywords: Chemistry of fluoride, defluoridation methods, fluorosis, mobilization of fluoride, mitigation measures

Introduction
Heavy agricultural and industrial demand and weather and hydrologic cycle changes has increased water stress (Gleick, 2003) [24]. Fluorides impact 62 million, arsenic 300 million (Jha, and Mishra, 2016) [31]. Heavy metals, organics, inorganics and fertilizers contaminate water (Singh *et al*. 2020). Electrocoagulation, precipitation, floatation, anion exchange, filtration with nano-filtration (NF) and reverse osmosis (RO) membranes, electro-dialysis, and adsorption are fluoride remediation techniques (Li *et al*. 2011) [36].

Cost-effective technologies are required to remove excess fluoride from ground water worldwide. Many countries, particularly China, India, Sri Lanka and the Rift Valley countries in East Africa, Turkey, and parts of South Africa, contain groundwater with high fluoride concentrations and risk of fluorosis. Liming and fluoride precipitation removed fluoride from contaminated water (Harrison, 2005) [26]. Ion-exchange, precipitation with iron-III (Tressaud, 2006) [65], calcium (Huang and Liu, 1999) [29], alum (Sujana, *et al*., 1998) [59] and activated alumina (Ghorai and Pant, 2005) [23] are also employed to defluoridate water. Similarly, electro coagulation (Hu *et al*., 2003) [28] and reverse osmosis (Simons, 1993 and Sehn, 2008) [57, 56]. Due to high costs of operation and maintenance pollution, and complex treatment, many of these methods were not widely used. Coagulation provides the most effective defluoridation method, however it doesn't decrease fluoride concentration. Membrane processes are expensive to build and operate and more likely to clog, scale or deteriorate. Electrochemical techniques are unpopular due to high maintenance and installation costs.

Chemistry of fluoride
This element fluorine, which really is capable of reacting, belong to this halogen family. The ninth element on the periodic table, fluorine, has an atomic weight = 18.9984 and is member of the group VII A. It is an odourless, chemically reactive gas with a pale yellowish colour (Cotton and Wilkinson 1988 and Mackay and Mackay 1986) [38].

Sources of fluoride pollution and its causes
The fluoride issue is made more severe by the unsustainable discharge of mining and agricultural wastes, as well as the excess use of fertilizers and agrochemicals. Fluoride is element which gets into the soil, air, vegetation, and animals by anthropogenic and natural activities. Fluoride occurs naturally as a result of the breakdown of fluoride-containing minerals such volcanic rocks, fluorspar, coastal flooding, fluorapatite, weathering and cryolite

(Dharmshaktu 2013) [16]. Fluoride was released into the environment in as solid and water waste during chemical manufacturing and processing that manufacture phosphate fertiliser, sodium fluoride, hydrogen fluoride, sodium hexafluoro silicate calcium fluoride, and fluoro-silicic acid, burning coal, applying pesticide, fluoridating drinking water, and irrigation using fluoridated water were example of other artificial sources (Dharmshaktu, 2013) [16].

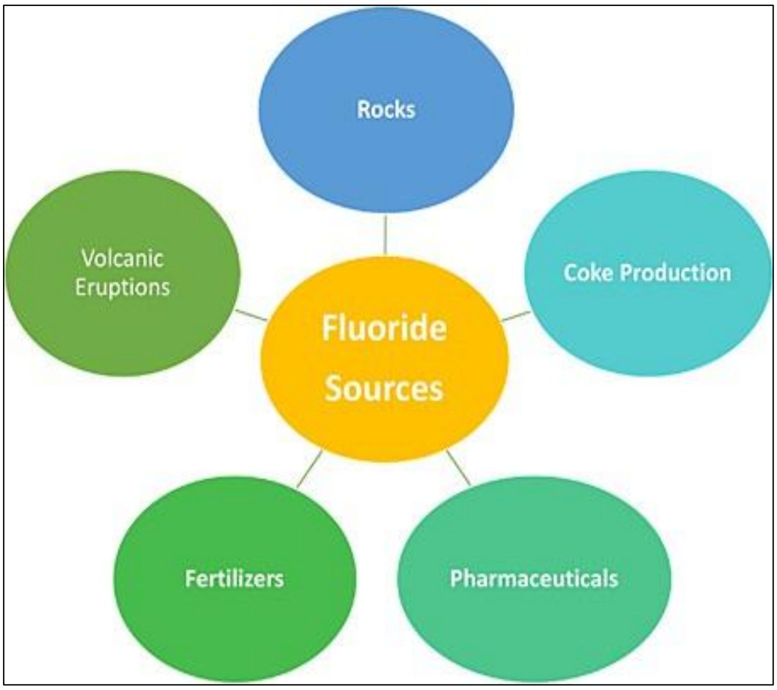


Fig 1: Different Sources of fluoride in groundwater

Hydro-geochemistry of fluoride

Fluoride levels in surface waters were influenced through volcanic emission, geographical dispersion and industrial discharges (WHO, 2004) [67]. Between 0.01 and 0.3 parts per trillions of fluorides found in the surface water, according to ASTDR (1993) [4]. Fluoride concentrations were 1.2 to 1.5 ppm higher in seawater than surface or fresh water. Groundwater chemistry is affected by chemistry of an aquifer matrix. Groundwater quality is determined by aquifer matrix, weathering, mineral dissolution, evaporation, and ion exchange between water and rock. Acidic rainfall absorbs atmospheric CO_2 and wash off alkali soil salts like sodium bicarbonate, sodium sulphate, and sodium chloride. Fertilizers produced form rock phosphate absorb fluoride into agricultural soil. Soil cations can undergo an anion exchange reaction at the same time (clay minerals). The ASTDR 1993 [4] document notes the inactive volcanic eruptions contribute an extra 10% of hydrogen fluoride to the stratosphere, with only an emission ranging of 60 to 6000 kilo tonnes. Fluoride retention was influenced by soil's type, pH, and organic content. Fluoride which is readily dissolves in water was essential for the well-being both plants and animals equally. According to Davison (1983), a g/g for contamination of soil can vary from tens to hundreds of thousands. Fluoride emissions come through brick manufacturers, aluminium smelters, coal power plants, tile, and, phosphate processors. Inorganic fluoride as in air is affected through evaporation, aerosolization, hydrolysis and dry and wet deposition (Environment Canada, 1994) [20].

Fluoride and Diseases

Fluoride levels over 1.5 ppm have indeed been connected to skeletal and dental fluorosis, a disease that may induce calcified ligaments, discoloured teeth, temporarily incapacitate bone abnormalities, and even death (Fawell *et al*. 2006) [22]. Skeletal fluorosis, which requires exposure to above 10ppm to manifest, is more severe than dental fluorosis and causes bone deformation that may be identified radiologically. Fluoride in drinking water has been linked to "non-ulcer dyspeptic" symptoms. The most common symptoms are nausea, vomiting, diarrhoea, constipation, bloating, gas and a headache (Susheela, 2001) [63]. There is documentation indicating fluoride exposure causes lung cancer, lowered IQ and behavioural and cognitive abnormalities in small kids (Yiamouyiannis, 1993 and Li *et al*. 1994) [68, 35].

Fluoride standards for drinking water adopted in different nation

Fluoride standards in drinking water of different countries illustrated in table 2.

Table 2: Fluoride standards of drinking water

Country and Standards	Particulars	Standards (ppm)	References
Indian standards	Allowable limit	1	
Maximum Allowable limit when no other sources are available		1.5	BIS (IS-10500-2012)
Australia	Maximum concentration of impurities	1.5	NRMMC (2011) [53]
WHO	Allowable limit	1.5	WHO (2006) [66]

Canadian standards	Maximum allowable limit	Upto 1.5	Ministry of Health, Government of Canada (2010) [42]
US EPA	Allowable limit	0.7	NRC (2006) [52]
	Allowable limit	1.2	
	Maximum amount of contamination	4	
Japan	Standard value	0.8	MHLW (2010) [41]
South Korea	Allowable limit	1.5	ECOREA (2013) [19]
Malaysia	Allowable limit	1.5	ESD (2004) [21]
Ireland	Allowable limit	1.5	NEIA (2018) [51]
Singapore	Allowable limit	0.7	NEA of Singapore (2008) [50]
Switzerland	Allowable limit	1.5	Bucheli *et al*. (2010) [8]
New Zealand	Maximum Allowable limit	1.5	MH (2008) [40]
UK	Allowable limit	1.5	DWI (2009) [17]

(Source: Kisku and Sahu, 2020) [33].

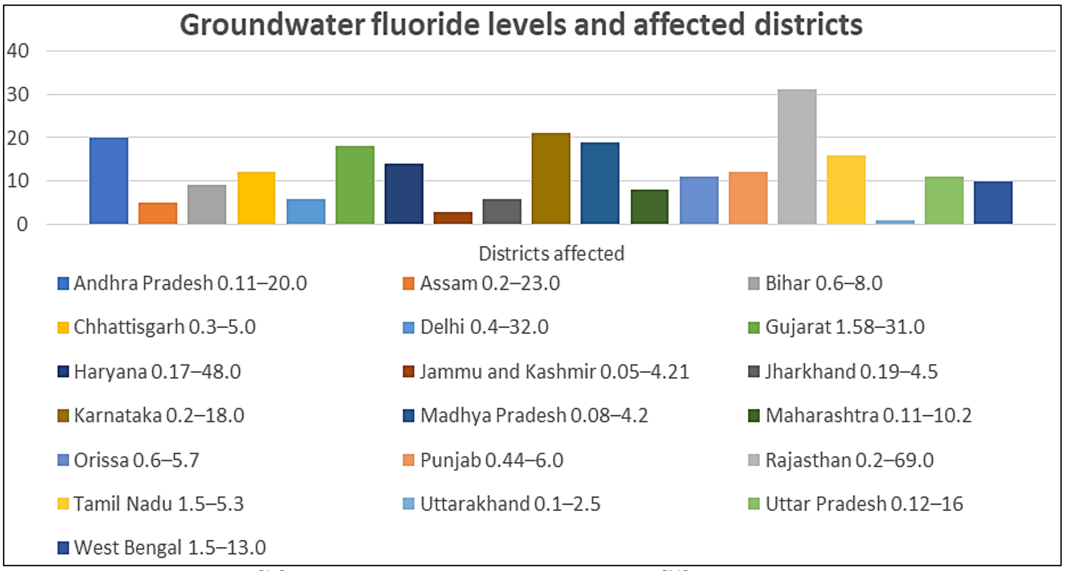
Worldwide Scenario of fluoride

The geology of different areas of the world influences fluoride concentrations in groundwater these are most common in midlatitude areas and accounting for a total deposit of 85 million tonnes (Sahu *et al*., 2018) [55]. Fluoride-rich groundwater is found in the Middle East, China, Southern Asia (India and Sri Lanka) and Africa. According to WHO, major fluoride belts extended from Eritrea through India, Syria, Malawi, Afghanistan, China and Turkey. There are also similar fluoride zones also observed in Iraq, United States, Japan, Iran and Kenya (Sahu *et al*., 2018) [55].

WHO recommended about 1.5 ppm fluoride in drinking water, but it has not been adopted worldwide because it depends on how much water is consumed, climate, and diet (Dharmshaktu 2013) [16]. Intake water fluoridation and associated endemic problems affect around 200 million people in 29 countries worldwide. The concentration of fluoride has changed with respect to the environmental and geographical conditions. Kenya has the greatest levels of fluoride in groundwater about 2800 ppm in Nakuru Lake and 1640 ppm in Elementaita, followed by Ethiopia 177 ppm (Nair and Manji 1982; Haimanot *et al*. 1987; Kloos and Teklee-Haimanot 1993; WHO 2006; Dharmshaktu 2013) [48, 25, 66, 16]. Fruit and vegetable bioaccumulation of fluoride varies between 0.1 to 0.4 mg/kg, and adds to normal exposure. Nevertheless, higher concentrations are now being studied at in foodstuffs include rice and barley 2-8, pulses about 13, fish protein 370, fish 2-5, radish 63 mg/kg (Bhattacharya *et al*. 2017; Mumtaz *et al*. 2015; Murrey 1986) [6, 46, 47].

Indian Scenario of fluoride

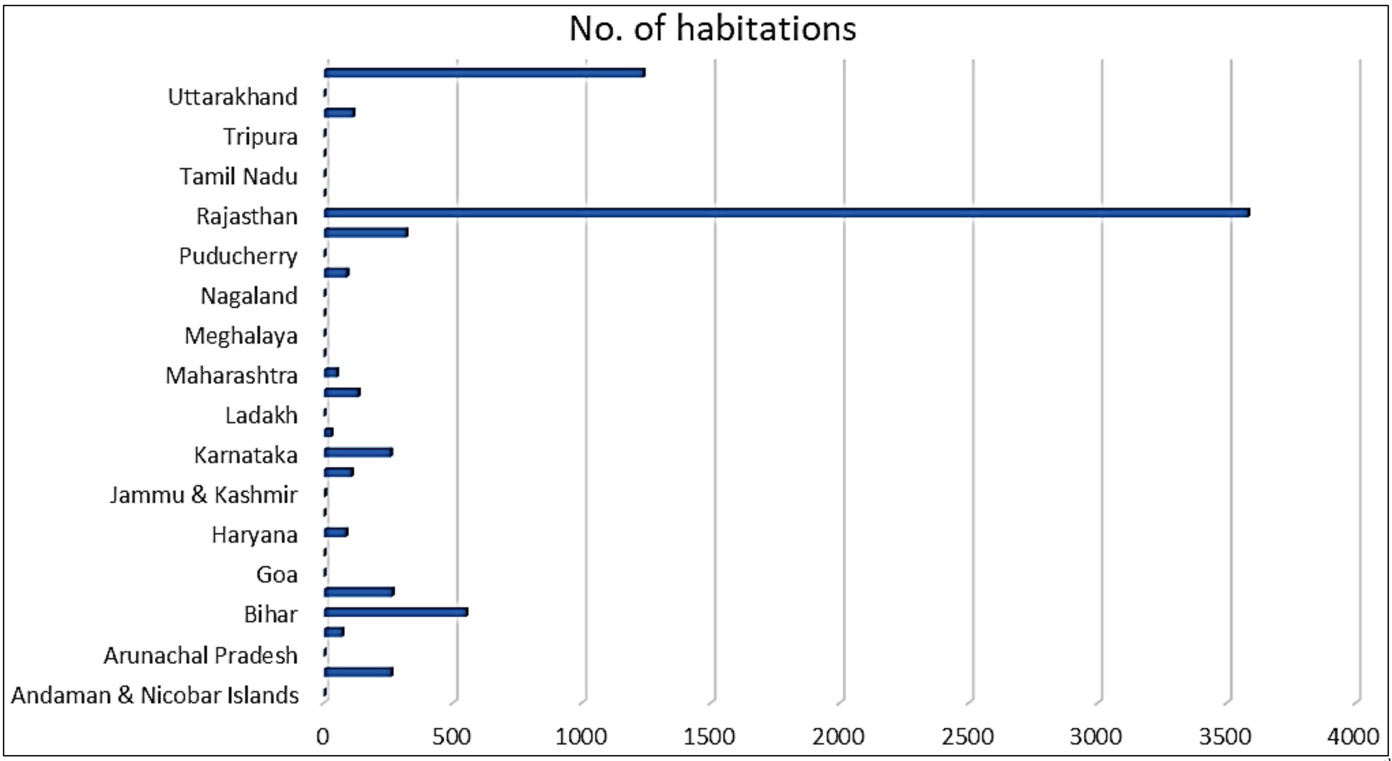
India seems to have the second largest demographic. By 2020, it is projected that there will water consumption will increase by 20 to 40%. India only has 4% of a world's water resources, however its population contributes up 16% of that as well, according to the Planning Commission's reports from 1996 and 2002. Ayoob and Gupta (2008) [5] reported the very first incidence of fluoride in drinking water was reported in 1937 in the Nellore district of Andhra Pradesh. Fluorosis symptoms were only found in four states before the 1930s, but since 1986, 1992, 2002, 2013 and currently, respectively, they have become prevalent in 13, 15, 17, 18 and 19 states of India.



Source: Susheela (1999) [62], Meenakshi and Maheshwari (2006) [39], State of Environment Report (2009), CGWB (2014) [10] and Mumtaz *et al*. (2015) [46]

As documented by Susheela (2003) [61], the states of Andhra Pradesh, Gujarat and Rajasthan have the highest endemic rates of long-term fluorosis. The fluoride concentration in groundwater was studied, which was found to be greater than 10 ppm in 9 of India's 19 states, namely Rajasthan, Haryana, Andhra Pradesh, Gujarat, Maharashtra, Delhi Assam Madhya Pradesh, West Bengal and Karnataka, (Dharmshaktu 2013) [16]. Sahu *et al*. (2018) [55] observed more than 10 ppm fluoride in the groundwater of Raebareli, Uttar Pradesh. Odisha, Chhattisgarh, Bihar, Tamil Nadu and Punjab all have had levels that were greater than 5 ppm but less than 10 ppm. According to Susheela (2002) [60], 66 million of people in 250 districts of India were at risk of endemic fluorosis, out of these 25 million of people have suffering from dental fluorosis, primarily among the below 18 age group.

Teotia and Teotia (1994) [64] reported approximately 12 million tonnes of fluoride was accumulated inside the earth surface of the India. According to report issued by Ministry of Environment and Forest and Climate Change, Government of India in 2009, fluorosis is believed to be occurs in 19 states of India, influencing 65 million of people overall, 6 million of which are children. As a reason, it is a subject of serious concern for the health and well-being of the general population. The Ministry of Health and Family Welfare, Government of India, supports the National Programme for Prevention and Control of Fluorosis (NPPCF) in 100 out of 230 endemic fluorosis districts in India during the 11th five-year plans (Dharmshaktu 2013) [16].



(***Source***: Ministry of Health and family Welfare, Govt. of India, New Delhi, Lok Sabha unstarred question No.1093 to be answered on 7th February, 2020 (Fluorosis)

Fig 2: State-wise number of habitations affected fluoride-contaminated ground water

History of DE fluoridation

Despite the fact that water defluoridation was not addressed until the 20th century, fluorosis remains a long-standing issue (Littleton, 1999) [37]. Many tribes in fluorotic areas were formerly isolated and never thought of dental fluorosis as odd; however, this perspective has now changed as a result of greater interaction. The first cases of dental fluorosis were reported in Mexico and Italy in 1888 and 1891. Fluorosis was linked to drinking water in the 1920s by Dr. Fedrick S. McKay in Colorado (Rajchagool and Rajchagool, 1997) [54]. In the 1930s, many countries started looking into the harmful effects of fluoride and how to eliminate it from drinking water. An aluminium-sand fluorine filter was developed in 1933 by Dr. S. P. Kramer. In 1945, M. Kenneth was granted a patent in France for defluoridation water. An activated alumina community defluoridation facility was established in 1952 at Bartlet, Texas, in the United States (Rajchagool and Rajchagool, 1997) [54]. Groundwater in several locations had too much fluoride by the middle of the 1980s. 25 million people in 8700 villages were reportedly using fluoridated water in 1987, according to the Rajiv Gandhi National Drinking Water Mission. Many states started testing of fluoride in all water sources and adopting changes in technology.

Integrated approaches in defluoridation

It seems promising to combine fluoride-rich groundwater treatment with industrial effluent treatment. To detoxify wastewater and remove inhibiting dissolved contaminants, in addition to enhance microbial activity and treatment response, biological treatments are often combined with electrocoagulation (Al-Qodah *et al*. 2019) [2]. The most efficient method for removing fluoride was simultaneous treatment, which has been followed by bioaccumulation and adsorption (using the leaves plants *Citrus limetta* and *Ficus religiosa*) and Gram-negative bacteria *Shewanella putrefaciens* (Dwivedi *et al*. 2017) [18]. Adsorption *Actinobacter* were tested by Mohammad and Kumar (2019) [43] on the peel of a sweet lemon to evaluate potential removal for fluoride through adsorption and bio-accumulation. More fluoride is removed by bioaccumulation (*Actinobacter*) than through adsorption (lemon peel), but bio-removal is slower. Chee *et al*. (2020) [11] showed how combining chemical (Ecogent F-Loc) and natural (*Moringa oleifera* seed and eggshell) coagulants improved fluoride removal. Gypsum plaster and electrocoagulation were employed in Jadhao *et al*. (2019) [30] study on wastewater defluoridation. Ca-F interactions enhanced effective removal for fluoride.

Review of defluoridation technology used globally

To purify fluoride-contaminated groundwater, various defluoridation methods have been used by various nations. In South Africa, fluoride was extensively eliminated in underground mine water in the 1980s with activated alumina. Fluoride levels decreased from 8 ppm to 1 ppm. (Schoeman and Botha 1985). In the 1980s, Ethiopia, Kenya, and Tanzania started using alum in fluoride treatment after note taking from the 1940s and 1930s US and India. Since 1500 BC, Egypt has used it (1970s-1980s). For about 40 years, two Nalgonda community units operated throughout central Ethiopia. Their efficiency declined to 60% due to old age and maintenance, and purchasing imported activated alumina was costly. Because of the limitations of the Nalgonda method and the simplicity of using bone char defluoridation in these countries, Mueller *et al*. (2006) [45] moved their attention to the bone char technique (Dahi, 2016) [14]. Due to religious and cultural beliefs, bone char is unsanitary and undesirable since it contains microbes. In Tanzania's rural Arusha region, contact precipitation-based plants have indeed been successful in households, schools, as well as other public areas. Bone char generates fluoride after joining calcium and phosphate compounds (Dahi, 2016) [14]. Using one of the earliest techniques, bone charcoal, the US field defluoridated from 1940 to 1960. (AWWA, 1971) [3]. There is defluoridation plant in Britton, South Dakota, with a 102 g fluoride/m3 exchange capacity and a 5-ppm initial fluoride concentration (Maier, 1953).

Fluoride is removed by reverse osmosis systems in even more than 1200 Thai communities. Because of the expensive installation and maintenance costs, these are unsustainable, therefore a large number of rural areas always have high fluoride water. Furthermore, 83 Thai bone char-based systems operating. Up-flow clay column filters in Sri Lanka eliminate fluoride. Over a period, about 80% of a 1,400 clay bricklet-filled domestic filters that have been installed in 60 communities were still working. Nanofiltration has evolved into a major water purification method that aids defluoridation. A 380-600 m^3/day nanofiltration system in Finland was 76% efficient in removing fluoride from groundwater. Numerous alternative fluorides involve a range are successfully utilized globally at different sized because of a lack of information.

Defluoridation methods adopted in India

Indian researcher have developed many novel techniques which have cost-effective, affordable and shown promising results.

Algona Technique

The Algona technique is developed by the National Environmental Engineering Research Institute (NEERI) using polymeric aluminium hydroxide and calcium salts. As compared with Nalgonda, Algona requires less aluminium hydroxide and poly aluminium chloride improves the both performance and cost.

KRASS Technique

This technology developed by Public Health Engineering Rajasthan (PHE) and Council of Scientific and Industrial Research (CSIR). Recharging the column using 10% alum solution is adsorption-based. This method removes 4 gm of fluoride per kg of alum. The method is most effective at 11 to12 ppm fluoride and 7 to 8 pH. (Agarwal *et al*. 1999) [1].

Prasanti Technique (Activated Alumina)

Indian communities are using a technique to defluoridation. In 1997, Satya Sai University (Bioscience Department) was formed in Prasanti Nilayam, Andhra Pradesh. In Udaipur, Rajasthan, Sarita Sansthan distributes buckets with micro-filters and activated aluminium to remove fluoride with UNICEF's support. Its drawbacks also include necessity for trained staff to reactivate micro-filters, the presence of aluminium by product residue and maintenance cos and operating costs high (Agarwal *et al*. 1999) [1].

IISc Method

It utilizes magnesium oxide, sodium bisulphate, and lime for precipitated fluoride. The Indian Institute of Science (IISc), Bangalore developed it. Magnesium fluoride precipitates in water as fluoride interacts with magnesium oxide. Sodium bisulphate adjusts pH, while magnesium oxide affects it. Lime removes bicarbonate interference (Karunanithi *et al*. 2019) [32].

Nalgonda technique

After testing many materials, the National Environment Engineering Research Institute, Nagpur developed a precipitation-coagulation method in 1961. It started at the community level in Kadri town (Nalgonda), Andhra Pradesh. It has been adapted from the lab to community and household levels both domestically and internationally. Lime, alum, bleaching powder, rapid mixing, flocculation, sedimentation, and filtration are being used. Bleaching powder disinfects and alum and lime form and precipitate aluminium hydroxide flocs. Purification several batches in a day requires 2-3 hrs. This method has been widely used and adapted, and chemicals are cheap and readily available. However, regular mixing makes it difficult. There also issues concerning water flavour and the risk of aluminium exposure, which can cause dementia at extremely low levels (0.2 ppm) (Karunanithi *et al*. 2019) [32].

The worldwide fluorosis mitigation programmes

There are a variety of steps that can be undertaken to reduce the levels of fluoride in the water, depending on the situation changing to an alternative water source with more appropriate concentration, combining the existing water supply with one that contains lesser fluoride, supplying bottled water, treating the water at the point-of-use on domestic level in small treatment devices (domestic defluoridation unit), or treating water on a bigger scale (Cummins, 1995) [13].

Global occurrences of fluorosis have reduced as a result of defluoridation programmes. The ICOH Mobile Bus Unit Project in Thailand (Nasakolnakorn, 2004) [49] focused on having to raise awareness of the health risks of excessive fluoride consumption and assisting people with the on defluoridation using a bucket defluoridator produced from bone char (Jacobsen and Dahi, 1997 and Bravo *et al*., 2000) [7].

Fluorosis mitigation programmes-The Indian Scenario

Fluoride mitigation may also include any of following methods, depending on the specific conditions: introducing a new or alternative source of water with acceptable levels; blending the established water supply with one that contains less fluoride; providing bottled water; treating the water at the point-of-use at the domestic level in small treatment devices (domestic defluoridation unit); or treating water on a larger scale (Cummins, 1995) [13].

Fluorosis elimination has been the focus of several mitigation programmes conducted by Indian government and state government for several decades. The tables 1 and 2 illustrate the Indian fluorosis mitigation programmes.

Table 5: Overview of fluorosis mitigation programmers in India

Program and Year	State	Method	Outcome
Project SARITA 1996	Rajasthan	Precipitation mechanism	Considerable reduction in non-skeletal symptoms
Sachetana plus drinking water project, 2006	Karnataka	Rainwater harvesting	There have been built 5600 rainwater collection structures. Borewells are recharged and an artificial catchment is created.
Nuapada Fluorosis Mitigation, 2005	Orissa	EQMG, Domestic defluoridation kits, Rainwater harvesting.	6995 recipients
Sonbhadra fluorosis Mitigation Project, 2004	Uttar Pradesh	EQMG Nutritional intervention.	2146 families, 970 kids.
Fluorosis Mitigation in Nalgonda district 2004	Andhra Pradesh	Rainwater harvesting Bone char defluoridation techniques.	Decrease of urinary fluoride up to 38%. 6, 5 and 8 (%) increase of serum calcium, serum phosphorus, and serum alkaline phosphatase respectively.
Dhar district, Madhya Pradesh, 2008 A Pilot study	Madhya Pradesh	Removal of Hand pumps which were source of high fluoride water content. Providing Safe water through collaborative effort with EQMG.	Implemented in 24 clusters from 8 panchayats
Madhya Pradesh. Integrated Fluorosis Mitigation, 2005	Madhya Pradesh	Integrated Fluorosis Mitigation	Reversal of skeletal fluorosis. 86, 77 and 60 (%) reduction in I, II and IV grade fluorosis respectively.
Tamilnadu, Hogenakkal water supply and fluorosis mitigation project, 2008	Tamil Nadu	Health delivery outlets, schools and community-based approaches	Ongoing Project
Prasanti Technology using Activated Alumina, 1978	Andhra Pradesh	Adsorption (Activated Alumina)	Evidence of improvement within a short period
UNICEF in India using household based defluoridation 1996-2002	Andhra Pradesh, Rajasthan	Adsorption (Activated Alumina)	By June 2003, 60 dealers in AP and 20 in Karnataka. By mid-2003, 24000 DDUs distributed in 5 districts of Rajasthan
Fluoride removal by IISc method, 2005.	Karnataka	Magnesium oxide based, precipitation, sedimentation and filtration technique of defluoridation	Scaled up to treat fluoride contaminated water at community level (500-2000 litres/day).
Nalgonda Technique, 1961.	Andhra Pradesh	Precipitation (Activated Alumina)	Fluoride, turbidity, colour, odour, pesticides and organic impurities are removed. Reduction in bacterial contamination.

Recent development in mitigation programme in India

The Integrated Fluorosis Mitigation (IFM) Program was initiated in 2019 in selected fluorosis-endemic villages of Nawada district, Bihar by the Centre for Fluorosis Research at Anugrah Narayan College, Patna, CSIRNEERI, Nagpur, NIRTH, Jabalpur, the PHED, Government of Bihar, and UNICEF, Patna. In addition to ensuring that all residents of the study villages have direct connections to fluoride-free water all the time, the programme as well involves comprehensive awareness-cum-interaction programmes with the villagers to educate them on the hazards of consuming fluoride-contaminated water and the benefits of drinking fluoride-safe water. Through the use of adsorbent-based defluoridation units supplied the CSIR-NEERI, Nagpur, the villagers have now accessible the drinking water using the pumping stations (Singh and Singh, 2019) [58].

Conclusion

Fluoride in public sources of water should not above the WHO's recommended dosage of 1.5 ppm. Various studies performed by experts across the world have demonstrated that fluoride in groundwater does have the potential to be a problem for human society. The primary source of fluoride in groundwater is found in fluoride-rich rocks. Several different types of rocks include sellaite, fluorite, cryolite, fluorapatite, apatite, fluor-mica, biotite and amphibole, contain fluoride. High groundwater fluoride concentrations were produced by weathering of these rocks and prolonged residence. Next main source of fluoride are combustion of coal and volcanic ash. Other sources of fluoride include alumina smelting, cement production, ceramic and brick firing, infiltration of agricultural runoff containing chemical fertilisers, improper disposal of liquid waste from industries, and improper liquid waste disposal.

Defluorination techniques are employed to remediate groundwater that contains a high amount of fluoride. They comprise mainly electrodialysis, reverse osmosis, ion exchange, adsorption, coagulation and precipitation, and ion exchange. A most modern technology amongst which has been reverse osmosis. On-site treatment includes artificial recharge methods like rainwater harvesting, constructing check dams, percolation ponds, or facilitating rainwater recharge through existing wells, among many other factors. The initial fluoride concentration, source, and commercial feasibility of an area significantly influence which approach is chosen. The government has to increase public awareness of the problem in order to prevent the development of diseases related to fluoride.

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