

Chapter 11

Fluoride Contamination and Health Effects: An Indian Scenario



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Abstract Uncontaminated water is the prime requirement of drinking water and lifeline of living organisms. Although some of the minerals are essential for growth and healthy development, excessive buildup of these minerals, viz. fluoride, can pose a great health risk to flora and fauna. Fluoride is a vital element for developments and strengthens of teeth and bones, but its elevated levels are associated with many incurable negative health effects reported on human and animals in all over India. The dental caries causes due to very low concentration of fluoride < 0.5 mg/L in drinking water and excessive fluoride (> 1.5 mg/L) also causes to dental fluorosis, skeletal fluorosis and neurotoxin effect, muscles degeneration, gastrointestinal system, reproductive system, etc. The effects of fluoride illness are more prevalent in children than adults. Therefore preliminary knowledge becomes necessary for everyone to know about the fluoride concentration in the drinking water, daily diets and other environmental matrix to avoid chronic fluoride diseases. The foremost sources of fluoride in groundwater are geogenic in origin, but the secondary sources are industrial activity like phosphate fertilizers, coal-based power plant and ceramic and glass industry. The higher concentrations of fluoride have been reported in Rajasthan (0.2–69.0 mg/L), Haryana (0.17–48 mg/L), Delhi (0.4–32 mg/L), Gujarat (1.58–31 mg/L) and Assam (0.2–23 mg/L). This chapter covers the fluoride-related issues and discusses the magnitude of problems those occurring in India and possible preventive measures and preventive programmes initiated by the Government of India.

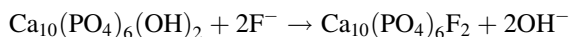
Keywords Fluoride · Groundwater · Dental and skeletal fluorosis · Environmental matrix · Hydrogeochemistry

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11.1 Introduction

Water resource has played a critical and very important role throughout the history in the growth and development of human civilization. In modern times, water resources have key role in the economic growth of all contemporary societies. Therefore, water resource assessment and sustainability consideration is of utmost importance in developing countries such as India, where water is commonly has economic and social significance (Kumar 2005; Kumar et al. 2018). The inaccessibility of uncontaminated drinking water and scarcity is the major problem in the world including India, where groundwater is main sources for drinking purpose (Mumtaz et al. 2015). It contains dissolved ions (As, Hg, U, F, NO_3^- SO_4^{2-} and heavy metals) beyond the permissible limit which is harmful and creates lots of waterborne disease. WHO have reported worldwide 748 million people are exposed to contaminated groundwater resources crisis for drinking purpose in 2012, 200 million human population in 27 nations all over the world facing critical issue of fluoride contamination while 66.64 million people in India (Mumtaz et al. 2015).

Merriam-Webster's Collegiate Dictionary (2003) defines that "Fluorosis is an abnormal condition (as mottling of the teeth) caused by fluorine or its compounds" (Dharmshaktu 2013). Fluorine is an element of the halogen groups and is among the highly reactive elements. The fluorine does not found free form in nature because of its strongly electronegative nature. Naturally it is found in the rocks, coal and clay soil. It is essential and chiefly abounded in earth crust. Fluorosis is an incurable disease mediated by the intake of F^- -rich water by humans and animals. Fluoride (F) within the body form highly insoluble and stable compound calcium appetite and react with calcium phosphate hydroxide. The absorbed fluoride deposited in bones and teeth which can results in major problems in the normal functioning of bones and teeth.



One lakh fifty thousand villages are affected by dental fluorosis in India mostly from state of Bihar, Andhra Pradesh, Rajasthan, Gujarat, Madhya Pradesh, Uttar Pradesh, Tamil Nadu and Punjab (Pillai and Stanley 2002). Rajasthan is one of the fluoride zones identified as it is in arid region and has acute water crisis. The state is unique as all the 32 districts were found to have highly contaminated groundwater with elevated level of fluoride. Vikash et al. (2009) and Jacks et al. (2005) both wrote in their research article that 10% villages of Rajasthan have excessive fluoride in water supply for domestic purpose. The chief source of fluoride was taken by humans through drinking water, particularly in fluoride-contaminated region. The release of fluoride in groundwater largely depends on geochemical process, where fluoride-containing minerals such as biotites, basalt, fluorite, shale, topaz, syenite, etc. are present. High fluoride-containing groundwater has low level of calcium due to poor solubility of CaF_2 in water. Dissolution of fluoride is dependent on pH, sodium bicarbonate, evapotranspiration, residence time of groundwater, soil types,

etc. The UNICEF (1999) mentioned in its report that 20 states of India have high level of fluoride in groundwater and 65 million peoples (included six million children) were affected by fluorosis.

11.1.1 Chemobiokinetics of Fluoride

Fluoride present in water is a simple covalent bonding with cations (Na, K). Fluoride is absorbed by the gastrointestinal tract and passes through the placenta, but some amount can be also excreted as sweat and lacteal secretion and saliva. The excretion of ingested fluoride via renal system in 3 years children were found more than 4–6 h is 50% of total absorbed fluoride, but children over 3 years to adults excreted about 90%. The total absorbed fluoride in the body, just about 90%, set down in teeth and bones. Absorption capacity of soluble fluoride (inorganic) compound is high as compared to other compounds of fluoride, and biological fluoride have several years half-life. Excessive concentration of inorganic fluoride has more harmful effects than organic fluoride. Fluoride is mainly excreted with urine and other faeces (Rani 2006).

11.2 Occurrence

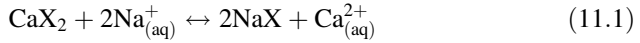
Fluoride is member of halogen groups and the 13th most abounded elements in earth's crust. 611 mg/kg and 285 $\mu\text{g/gm}$ of fluoride were found in continental crust and soil, while various rocks such as basalt, granites, limestone, sandstone and shale contain 360, 810, 220, 180 and 800 $\mu\text{g/gm}$, respectively. Endemic fluorosis is prevalent in India since 1937 (Shortt et al., 1937). The important fluoride-bearing minerals are fluorite (CaF_2) which is less soluble in water, and it occurs in igneous and sedimentary rocks. The fluoride rich minerals such as apatite, amphiboles, hornblende and micas are replacing to OH^- and Cl^- ions of groundwater by releasing F^- . The occurrence of fluoride is not limited to only rocks but also present in soil and accumulated by plants (CGWB 2014). Beryllium, ferric ions, boron and aluminium form sturdy complexes with fluoride, while solubility of fluoride in natural water increases with binding with silica (CGWB 2014).

11.2.1 Hydro-geochemistry of Fluoride

Chemistry of groundwater principally depends on chemical composition of aquifer matrix. The quality of groundwater is generally controlled by natural processes such as aquifer matrix, weathering of rocks, dissolution of minerals, evaporation of groundwater and exchange of ions between water and rock. The rainwater is acidic

in nature due to dissolution of atmospheric CO_2 and percolates downward through the soil with leached out secondary salt from soil such as sodium bicarbonate, sodium sulphate and sodium chloride.

Phosphate fertilizer is additive sources of fluoride-bearing compounds in agricultural land. Simultaneously, anion exchange reaction occurs with cations of soil (clay minerals):

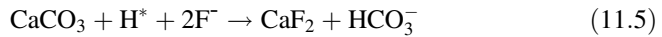


Here, X is clay minerals.

The hydrogen ion concentration increases during dissolution of CO_2 and CaCO_3 (Saxena and Shakeel 2003; Subbarao and John 2003; Handa 1975):



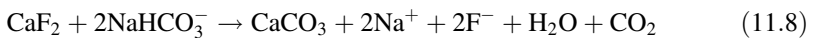
Bicarbonate dissociates as in hydrogen and carbonate ions (Arveti et al. 2011):



In the presence of alkaline water, fluoride is highly mobilized, and CaF_2 settles down as calcium carbonate:



In the presence of excessive sodium bicarbonates in groundwater, the dissolution activity of fluoride is high and can be expressed as (Ayoob and Gupta 2006):



Solubility product of fluorite is low:

$$\text{K}_{\text{sp}}^{19} = (\text{F}^-)^2(\text{Ca}^{2+}) = 4.0 \times 10^{-11} \quad (11.9)$$

11.2.2 Global Scenario

The distribution of elevated fluoride in groundwater depends upon the geology of different parts of the world; mostly in midlatitude areas, it has total deposit of 85 million tons in earth crust (Sahu et al. 2018 and Teotia and Teotia 1994). High fluoride-containing groundwater occurs in enormous parts of Africa, China, the Middle East and southern Asia (India and Sri Lanka). WHO reported that major fluoride belts extended from Eritrea through Malawi, Syria to Turkey, Afghanistan, India and China. Other similar fluoride belt is also found in America, Kenya, Iraq, Japan and Iran (Sahu et al. 2018).

Symptom of fluorosis is observed in population of a particular area having fluoride concentration in groundwater between 1.5 and 10 mg/L. 1.5 mg/L fluoride in drinking water has been recommended by WHO (2011), though it is not adopted in all over the country because it depends on volume of drinking water taken, climate and diet (Dharmshaktu 2013). The magnitude and sternness of fluoride has vary with references to environmental sitting of geography and approximately 200 million people in 29 countries around the world faces the endemic problems of fluoride in intake water or drinking water. The highest concentration of F^- in the groundwater has been observed in Kenya (1640 mg/L in Elementaita and 2800 mg/l in Nakuru Lake) followed by 177 mg/L in Ethiopia and 69.7 mg/L in India (Nair and Manji 1982; Haimanot et al. 1987; Kloos and Teklee-Haimanot 1993; WHO 2006; Dharmshaktu 2013). Bioaccumulation of fluoride found in fruit and vegetables is 0.1–0.4 mg/kg contributing normal exposure. But elevated levels are investigated in rice and barley (2–8 mg/kg), protein of fish (370 mg/kg), pulses (about 13 mg/kg), fish (2–5 mg/kg), radish (63 mg/kg), etc. (Bhattacharya et al. 2017; Mumtaz et al. 2015; Murray 1986) (Table 11.1).

11.2.3 Indian Scenario

India is the second extremely populated country in the world, but according to area, it counts under the seventh biggest country. It is predicted that by 2020, water consumption rate will increase by 20–40% and population will reach up to 1550 million. According to the Planning Commission 1996 and 2002, India has only 4% water resources, but population contributes 16 percent in the world. Ayoob and Gupta (2006) mentioned in his research article that fluoride in drinking water was first noticed in Nellore district of Andhra Pradesh (India) in 1937 (Dharmshaktu 2013). Before the 1930s, only in four states symptoms of fluorosis were observed, while it is increasing in 13, 15, 17, 18 and 19 states of India during the years 1986, 1992, 2002, 2013 and now, respectively.

Table 11.1 Range of fluoride in groundwater and affected district in different states of India

India	Range of fluoride (mg/l)	Number of affected district
Andhra Pradesh	0.11–20.0	20
Assam	0.2–23.0	5
Bihar	0.6–8.0	9
Chhattisgarh	0.3–5.0	12
Delhi	0.4–32.0	6
Gujarat	1.58–31.0	18
Haryana	0.17–48.0	14
Jammu and Kashmir	0.05–4.21	3
Jharkhand	0.19–4.5	6
Karnataka	0.2–18.0	21
Madhya Pradesh	0.08–4.2	19
Maharashtra	0.11–10.2	8
Orissa	0.6–5.7	11
Punjab	0.44–6.0	12
Rajasthan	0.2–69.0	31
Tamil Nadu	1.5–5.3	16
Uttarakhand	0.1–2.5	1
Uttar Pradesh	0.12–16	11
West Bengal	1.5–13.0	10

Source: Susheela (1999), Meenakshi and Maheshwari (2006), State of Environment Report (2009), CGWB (2014) and Mumtaz et al. (2015)

Susheela (2003) mentioned that Rajasthan, Gujarat and Andhra Pradesh are the extreme endemic areas that are affected by chronic fluorosis. The concentration of fluoride in groundwater was measured beyond 10 mg/L in 9 states out of 19 states in India, which are Andhra Pradesh, Rajasthan, Madhya Pradesh, Haryana and Maharashtra, Assam, Delhi, Gujarat, Karnataka and West Bengal (Dharmshaktu 2013), while Sahu et al. (2018) found more than 10 mg/L fluoride in groundwater of Raebareli district of Uttar Pradesh. The concentration below 10 mg/L and more than 5 mg/L was found in Bihar, Chhattisgarh, Odessa, Punjab and Tamil Nadu. Susheela (2002) has mentioned that 66 million population in 250 districts of India were at risk of endemic fluorosis, while 25 million population has been affected by problem of dental fluorosis, mainly in population below 18 years of age.

According to Teotia and Teotia (1994), deposition of fluoride in Indian earth's crust was 12 million tons. The report released from Ministry of Environment and Forest and Climate Change (Government of India) during 2009, it has been estimated that fluorosis is prevalent in 19 states of India, where around 65 million populations were affected, out of which 6 million children was reported. It is, therefore, a matter of high concern from the point of view of public health and welfare. The Ministry of health and family welfare (Government of India) had released funds during 2008-2009 in 11th Five Year Plan under NPPCF (National

Programme for Prevention and Control of Fluorosis) in 100 districts out of 230 endemic fluorosis districts in India (Dharmshaktu 2013).

11.3 Causes/Sources of Fluoride Contamination

Without environmental considerations, reckless dumping of mining and agricultural wastes and excessive use of fertilizers and agrochemicals are adding to the present fluoride crisis. Overexploitation of groundwater can cause hyperaccumulation of contaminants in the soil (Subramanian 2000; Singh et al. 2000; CPCB 2008). Fluoride present in various environmental media such as water, soil, air, flora and fauna is a contribution of both anthropogenic and natural activities. Natural presence of fluoride in the environmental matrix is because of weathering and dissolution of fluoride-containing minerals such as cryolite, fluorspar, fluorapatite, marine aerosol, volcanic ash, etc. (Dharmshaktu 2013; ASTDR 1993). The anthropogenic activities release fluoride into environmental matrix such as waste water, solid waste and stack gases releases from chemical manufacturing and processing industries of calcium fluoride, phosphate fertilizers, sodium fluoride, hydrogen fluoride, fluorosilicic acid and sodium hexafluorosilicate. Others possible manmade sources like burning of coal, application of pesticide, fluoridation of drinking water and irrigation of fluoride-containing water contributing to fluoride into environmental matrix (Dharmshaktu 2013).

11.3.1 Exposure/Potency of Fluoride in the Environment

11.3.1.1 Surface Water

In surface waters, level of fluoride is dependent upon source of volcanic emission, geographic distribution and discharge of industrial effluents (WHO 2004). According to ASTDR (1993), the concentration of fluoride in surface water is usually between 0.01 and 0.3 ppm. Fluoride concentration in sea water is 1.2–1.5 ppm higher than as compared to fresh water or surface water.

11.3.1.2 Air

Aluminium smelter plants, still mills, tiles and bricks manufacturers, phosphate processors and coal power plants are major contributors of fluoride contamination in the environment. Formation of aerosol, hydrolysis, vaporization and dry and wet deposition influence the destiny of atmospheric inorganic fluoride (Environment Canada 1994). The gaseous fluoride (hexaethane, carbon tetrafluoride, silicon tetrafluoride and hydrogen fluoride) are adsorbed on the particulate matters

which are not easily hydrolysed and remain suspended in the atmosphere (US NAS 1971); this process is observed near coal-based thermal power plants and other emission sources (Sidhu 1979). Fluoride-containing airborne particles are depending upon meteorological condition, chemical reactivity, emission strength and particle size (WHO 2002).

11.3.1.3 Soil

According to ASTDR 1993, 60–6000 kilo tonnes of hydrogen fluoride is emitted via passive eruption of volcanic sources, which contributes approximately 10% to the stratosphere. Retention of fluoride in soil depends on types of soil, pH and organic contents. Water-soluble fluoride is biologically important for animal and plant. According to Davison (1983), 20–1000 µg/g is estimated in uncontaminated area while several thousands in contaminated soil.

11.3.2 Major Sources of Fluoride Ingestion by Human

Air, water, soil, drugs, food and cosmetics are various sources of fluoride ingestion by humans, but water and dietary intake are chief sources.

11.3.2.1 Water

Only drinking water contributes more than 60% of total intake of fluoride. Inorganic fluoride is the most available form, but higher concentration can cause highly toxic effect on human health. The natural fluoride content in groundwater is influenced by lithology, depth of aquifer and physical and chemical characteristics of the aquifer.

11.3.2.2 Air

The significant concentration of fluoride in and around the industrial area where workers and near inhabitant exposed to inhalation of fluoride. The fluoride pollution causing industries are aluminum smelter, glass, coal-based operated thermal power plants, fertilizers and ceramic industries.

Table 11.2 Global and Indian standard for fluoride in drinking water

Types of standards	Description guideline	Guideline value (mg/L)	References
Indian standards	Allowable limit	1	
	Permissible limit in the absence of other sources	1.5	BIS (IS-10500-2012)
WHO guidelines	Guideline value	1.5	WHO (2006)
US EPA	Desired limit	0.7	
	Permissible limit	1.2	
	Maximum contaminant level	4	NRC (2006)
Canadian guidelines	Maximum acceptable (MAC)	1.5	Ministry of Health Government of Canada (2010)
South Korea	Maximum permissible limit	1.5	ECOREA (2013)
Japan	Standard value	0.8	MHLW (2010)
Singapore	Maximum prescribed quantity	0.7	NEA of Singapore (2008)
Malaysia	Permissible limit	1.5	ESD (2004)
Ireland	Permissible limit	1.5	NEIA (2018)
UK	Permissible limit	1.5	DWI (2009)
Switzerland	Permissible limit	1.5	Bucheli et al. (2010)
Australia	Maximum impurity concentration	1.5	NRMMC (2011)
New Zealand	Maximum acceptable value	1.5	MH (2008)

11.3.2.3 Food

According to Jagtap et al. (2012), food items contained fewer amounts of fluoride in uncontaminated area, but huge amounts of fluoride have been reported in dietary product of contaminated area. Concentration depends upon fluoride contents in irrigated water and growing media such as soil. The enormous contents of fluoride are found in Indian tea leaf (39.8–68.59 mg/L) that contributes chief intake of fluoride after drinking water. Similarly fluoride concentrations found in various dietary items range between 3.27 and 14.03 mg/L (wheat), 5.6 mg/L (legumes and pulses), 1.28 and 2.29 mg/L (cabbage), 4.0 mg/L (lady figure) and 2.8 mg/L (potato).

11.3.2.4 Drugs and Cosmetics

Inorganic fluoride is applied for the prevention of tooth decay through drinking water and tablets or other drugs as a fluoride supplements. The niflumic acid is anti-inflammatory agent applied to cure of rheumatoid arthritis. Sodium fluoride used in mouthwash for preventing of oral cavity. The high concentration of fluoride also

Table 11.3 Effect of fluoride on plants and laboratory animal in India

Test animals/ plants	Fluoride in diet/dose	Effect of fluoride on test animals	References
(a) Developmental, reproductive and genotoxicity effect in test animals			
		<i>Effect on reproductive and developmental system</i>	
Female mice (<i>Mus musculus</i>)	10 mg/kg	Reduced protein contents in muscles, small intestine and liver with significant accumulation of glycogen in liver	Chinoy et al. (1994)
Mice (<i>Mus musculus</i>)	5 mg/kg	Damaged works of protective enzymes and damaged manufacturing of glutathione with reduced calcium under ovaries	Chinoy and Patel (1998)
(b) Effect of fluoride on human systems			
		<i>Effect on reproductive system</i>	
Indian pregnant women	0.12–0.42 µg/ml	Passive transfer of fluoride passing through mother to foetus (25 women)	Malhotra et al. (1993)
Indian men	1.5–14.5 mg/L	Symptoms of skeletal fluorosis and reduced testosterone serum (30 men)	Susheela and Jethanadani (1996)
		<i>Effect on hepatic and renal system</i>	
Indian men and women	3.5–4.9 mg/L	Kidney stone because of malnutrition in 18, 706 people	Singh et al. (2001)
		<i>Effect on human bone</i>	
Indian children	1.5–25 mg/L	Deformities and metabolic bone disease due to chronic ingestion of high fluoride and low dietary calcium (ninety percent in 45725 child)	Teotia et al. (1998)
(c) Effect on aquatic plants			
Macrophyte (<i>Hydrilla verticillata</i>)	20 mg/L	Reduced protein content and chlorophyll after 7 days	Sinha et al. (2000)

used in toothpastes, the concentration of fluoride varies from ~1000 to 4000 mg/L depending on the brand of the toothpaste.

11.3.2.5 Other Anthropogenic Activities

The source of anthropogenic activities includes discharge of municipal waste water and the effluents from ceramic industry, bricks manufacturing and chemical industries into water body like rivers results in accumulation of fluoride in plants and aquatic animals (Tables 11.2 and 11.3) (Jagtap et al. 2012).

11.4 Health Effect on Human

Fluoride intake has both advantageous and harmful effects; if the concentration is less than 0.5 mg/L, then its deficiency causes dental caries and weakness of bones, while their excessive intake (>1.50 mg/L) may cause fluorosis.

11.4.1 Dental Caries

Dental caries is the scientific term for tooth decay or cavities. Dental caries is demineralization of teeth due to bacterial decomposition. *Lactobacilli* and *Streptococcus mutans* bacteria attached onto the dental plaque and consume the organic matter and produce propionic, lactic and acetic acid that incise the enamel, thus forming cavity (Ayoob and Gupta 2006 and CDCP 1999).

11.4.2 Dental Fluorosis

Excessive intakes of fluoride can cause deformation of teeth during developmental stage or mottling of enamel called “dental fluorosis”. The fluoride deposits into ameloblasts (primary cell for formation of tooth enamel) and spreads out over the enamel with changing the colour from white to radish brown resulting enamel decay and loss of luster.

Dean has been measuring for quantifying the sternness of dental fluorosis in 1934. The classification of dental fluorosis based on nutritional status is given below:

- Class 0 – Normal teeth
- Class 1 – Very mild fluorosis (25% area of tooth surface covered by opaque white)
- Class 2– Mild fluorosis (50% area of tooth surface covered by opaque white)
- Class 3 – Moderate fluorosis (tooth surface affected by red brown dark)
- Class 4 – Severe fluorosis (widespread around whole tooth by brown stains) (Ayoob and Gupta 2006)

Dr. McKay first noticed symptoms of the fluorosis (mottled enamel) in the tooth enamel and began his research on association of fluoride and drinking water (McKay 1925; McKay 1928). Fluorosis was initially noticed in cattle of Nellore District (Andhra Pradesh), India, before the 1930s and foremost published in Indian Medical Gazette by Shortt et al. 1937. The dental health survey was conducted in nine states of India during 1987–1992, and dental fluorosis were observed 5–20% and 2–30% in children (6–14 years) and adult (Susheela 2003). The health survey was carried out on school children of 18 districts in Gujarat where children’s pretentious by dental fluorosis was found between 2.60% to 33%. The percent of dental fluorosis affected

districts of Gujarat were observed 17.75% in Ahmadabad, 11.43% in Gandhinagar, 24.90% in Mehsana, 17.78% in Banaskantha, 14.50% in Sabarkantha, 16.87% in Baroda, 12.16% in Kheda, 8.4% in Panchmahal, 14.9% in Bharuch, 7.9% in Surat, 2.6% in Vasad, 33.0% in Junagarh, 16.6% in Amreli, 22.0% in Surendranagar, 15.5% in Jamnagar, 14.1% in Bhavnagar, 14.7% in Rajkot and 20.25% in Kutch (Susheela 2003). The dental fluorosis was found 77.1% in 17–22 years old population of Rajasthan, where the fluoride level was found 2.6 ppm in drinking water (Choubisa SL 2001)

11.4.3 Skeletal Effects of Fluoride

Bone illness or changes in bone structure and calcification of ligaments are symptoms of skeletal fluorosis, and it is caused by excessive intake or accumulation of fluoride through drinking water and diet of prevalent fluoride zone (Krishnamachari, 1986). The chronic ingestion of excessive fluoride deposited in the joint of the knee, shoulder bones, neck and pelvic creates difficulty in movements and walking. According to the reports of the WHO 1970 and 2002, accumulation of fluoride in bone depends on sex, bone types and age. Fluoride accumulation capacity of cancellous bone is high as compared to cortical bones. Ninety-nine percent of total fluoride deposits in teeth and bone of human and remaining deposits in blood and vascularised soft tissue (Husdan et al. 1976; Kaminsky et al. 1990; WHO 2002). The skeletal fluorosis was developed and 43 percent people affected in Anantapur district of Andhra Pradesh, where the fluoride level was ranged from 1.2 to 2.1 mg/L (Reddy and Prasad 2003). Long-time exposure to high level of fluoride (more than 5 mg/L) in drinking water can cause *crippling skeletal fluorosis*, and it is marked by:

- Kyphosis: unusually enlarged convexity in the bend of thoracic spine
- Flexion deformity: flexion contracture or bend of the knee
- Scoliosis: sideway curvature of the spine or vertebral column
- Paraplegia: paralysis or de-activation of muscles function in the lower body
- Quadriplegia: paralysis or permanent loss of the torso and four limbs (Ayoob and Gupta 2006)

11.5 Amelioration Techniques

11.5.1 Adsorption

The fluoride-contaminated water passes through a packed column with fluoride adsorption media like coated silica gel, activated coconut shell, activated alumina, calcite, natural clay soil, red mud, activated saw dust, bone charcoal, activated carbon, ground shell, fly ash, serpentine, magnesia, coffee husk and rice husk

which are used for removal of fluoride. This method is easily prepared and suitable for household and community purposes.

11.5.1.1 Activated Alumina

Activated alumina is an attractive adsorbent with high fluoride removal capacity as compared to another adsorbent media due to highly porous aluminium oxide that exhibit more surface area. The cationic networks spread out over the crystal of alumina provide positive charge which are attracted to anionic species. Hardness and fluoride concentration of water both affect the removal efficiency of activated alumina. The process is pH specific, hydroxide and silicate act as competitor in more than pH 7, while alumina dissolves at pH below 5 (Bishop and Sansoucy 1978). In Rajasthan, India, a microfilter bucket (5 kg alumina pin down) was provided to the people in fluoride-affected area by Sarit Sanshtan Udaypur with the help of the UNICEF.

Merit

- ~90% fluoride removal efficiency.
- This process is cost-effective.

Demerit

- The processes need pretreatment.
- Specific removal efficiency at pH 5–6.
- Alumina bed fouls in high TDS.
- Regeneration is required after 5 months.

11.5.1.2 Activated Carbon

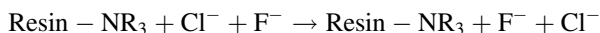
Removal of fluoride by activated carbon was used, and it was investigated in 1934 (Mckee and Johnston 1999). Activated carbon give good removal capacity under pH 3. So, the process needs to maintain pH.

11.5.1.3 Brick Pieces Column

The bricks are utilized as adsorbent of fluoride for removal of fluoride because they contain AlO_2 . The brick particles is activated through burning or heating process then the activated brick particle is packed into adsorption colum. The fluoride contaminated water passing through the adsorption colum filled with activated brick particle which adsorb fluoride from raw water.

11.5.2 Ion-Exchange Resin

Many synthetic chemicals (polyanion, Amberlite IRA 400, and XE-75) were used as a cation or anions transferable resins mostly in hydroxide and chloride forms. Ammonium functional groups with chloride were used as ion-exchange resins. It is a strong-base resin for ion exchange between chloride and fluoride. The resin is cleaned after using by backwashing of supersaturated salt of sodium chloride (Meenakshi, and Maheshwari 2006).



Several materials such as calcium, alumina, magnesium, activated carbon, sulphonated carbon and lime were used for fluoride removal as ion-exchange resin removed from 5 mg/L to 1.5 mg/L (Mohan Rao and Bhaskaran 1988).

Merit

- Fluoride removal capacity up to 90–95%

Demerit

- pH low and elevated level of chloride in treated water.
- Elevated level of alkalinity; sulphate and carbonate reduced the removal efficiency.
- Disposal of resin is a major problem.

11.5.3 Coagulation-Precipitation

Coagulation-precipitation is a process for the removal of soluble fluoride in the form of insoluble salt by adding of chemicals called coagulant aids. In this technique polyaluminium (chloride and hydroxy sulphate), brushite and aluminium salt are commonly required raw materials.

In India, the Department of Chemistry, IIT Jodhpur, developed gravity-based water filtration systems followed by precipitation method through natural materials such as neem, Ayurvedic waste, sesame, seed powder of gaur, etc. (Choudhary et al. 2014).

11.5.3.1 Nalgonda Technique

This is the most famous technique used for amelioration of F^- from groundwater in India. This technique was primarily started in Nalgonda village of Andhra Pradesh. The lime and alum, both are added with rapid mixing of water in a tank, for better flocculation and sedimentation of contaminants. The bleaching powder is added for the disinfection of filtered water. The operation may be conducted on both large and

small scale, and it started in various countries, i.e. Tanzania, Kenya and Senegal (NEERI 1978). NEERI Nagpur also developed hand pump attached technique under Rajiv Gandhi National Drinking Water Mission. Lime and alum (hydrate aluminium salts) are the most commonly used coagulants, and lime precipitate to fluoride can cause rising of pH up to 11–12. According to Potgeiter (1990), when alum was added in water, it formed aluminium hydroxide due to reaction with alkalinity and settled down after reaction with fluoride as a form of insoluble calcium fluoride.

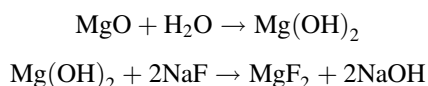
Merit

- Nalgonda technique is the most effective technique for fluoride removal (NEERI, 1978).
- Technique worked well in the range between 1.5 and 20 mg/L.
- TDS and hardness should be below TDS 1500 and 600 mg/L.

Demerit

- Aluminium dissolved in treated water can cause problem.
- Maintenance required 3000 rupees for 10,000 L capacity of plant per day.
- Larger amount of sludge became a problem for drying and disposal.
- It is not an automatic process; it required manpower.

According to Rao and Mamatha 2004, Nalgonda technique is modified by Indian Institute of Science, Bangalore, by using sodium bisulphate, lime and magnesium oxide. Fluoride was removed in the form of magnesium fluoride salt.



Magnesium oxide rises pH up to 10 to 11 in treated water, and it is sharply reduced between 6.5 and 8 by dissolving in sodium bisulphate (0.15–0.2 g/L). When the concentration of HCO_3^- is higher than 200 mg/L, then it creates problem in the working of sodium bisulphate. The problem is solved by adding 0.3 mg and 0.8 mg of lime and magnesium oxide (MgO).

11.5.4 Membrane Techniques/Physical Methods

Reverse osmosis (RO) and nanofiltration both remove fluoride by their membrane. Nanofilter allows only microscopic size and removes all dissolved solid having larger size by low pressure. The reverse osmosis removes dissolved ions by semipermeable membrane and its works on opposite of osmosis process. The high pressure applies on a semipermeable membrane which not allow to larger particles passing from it. Most of the researcher found 90% removal capacity of fluoride. The main factor that affects the process is characteristic of raw water, recovery, selection of membrane,

water rejection and size of plants. Babra et al. (1997) found in a study that efficiency of RO plants decreases with boosting of plants capacity. Meenakshi et al. (2004) found the following merits and demerits:

Merit

- This technique is extremely successful for fluoride removal and also reduces dissolved solids, pesticides, microorganisms, inorganic and organic pollutants, etc.
- Little maintenance and quick process.
- Without limitation of pH and temperature does not affect removal efficiency.

Demerit

- pH of treated water have low level.
- It rejects all essential mineral.
- It rejects high volume of reject water.

11.6 Preventive Measures

The following measures should be adapted for prevention and control of excessive intake of fluoride:

11.6.1 Awareness

To aware the inhabitant is a first step for prevention of negative health impact of fluoride contamination, spreading awareness about safe limit of fluoride in drinking water and their incurable effect on health through poster, interpersonal communication or group discussion in school and villages, wall painting, newspaper or news channel, etc.

11.6.2 Rainwater Harvesting and Surface Water Resource: Alternative Water Source

The water harvesting technology is a better way to supply less fluoride-containing drinking water to inhabitants. Generally groundwater has high fluoride content as compared to surface water which may be used after conventional treatment of surface water.

11.6.3 *Changing the Dietary Habits*

Amelioration technique shall not remove the fluoride level under safe limit; then the following prevention can be adopted through dietary habit of inhabitants:

Calcium Take calcium-rich diet in fluorosis area such as sesame seeds, curd, jiggery, green leafy vegetables, drumstick, etc. High intake of calcium reduced the absorption efficiency of fluoride.

Vitamin C It was reported that vitamins C and E protect against oxidative stress and damage of endometrial in rats in fluoride intoxication (Guney et al. 2007). Some edible items have high vitamin C contents such as orange, sprouted pulses, lemon, coriander and tomato while vitamin E in nuts, green vegetables, dried beans, white grain pulses or cereals, etc.

Antioxidants Items such as ginger, papaya, green leafy vegetables, carrot, garlic, pumpkin, etc. show antagonistic effect; thus they play prophylactic function to avoid fluorosis (Kumar et al. 2014).

Toothpaste In fluorite belt area, 4–6-year-old children can use toothpaste having low fluoride concentration for dentifrices (Villena 2000). Some dental specialist suggested to use toothpastes having low concentration of fluoride (500–550 ppm) as compared to standard concentration (1.0–1.1 ppm) of fluoride (Negri and Cury, 2002; Stookey et al. 2004).

11.7 Fluorosis Management in India

In 2008, for controlling of the troubling of fluorosis, the Planning Commission of India starts the NPPCF programme in 11th 5-year plan during 2007–2012. This programme has been handled by various NGOs and governmental bodies such as MDWS (Ministry of Drinking Water and Sanitation), RGNDWM (Rajiv Gandhi National Drinking Water Mission), MHFW (Ministry of Health and Family Welfare) and the UNICEF (United Nation International Children's Emergency Fund). Baseline data was collected by the DDWS (Department of Drinking Water Supply), Central Government, in 196 districts in 19 states of India and fund released to 100 districts in four phase, according to collected baseline data (Dharmshaktu 2013).

All districts were provided rapid management, community diagnosis, capacity building, medical and laboratory manpower, surgery, education and communication materials, etc. Nellore, Jamnagar, Nagpur, Nayagarh, Ujjain districts of India were provided fund under NPPCF programme in Ist Phase during 2008–2009, and another 15 districts were provided fund during 2009–2010 in IInd Phase. Third and fourth Phase provided fund to 40–40 districts during 2010–2011 (Dharmshaktu 2013; MHFW 2017).

11.8 Conclusion

Gradually, fluoride and fluorosis illness becomes a prevalent disease because of environmental fluoride present in the rocky surface of the underground aquifer. The underground fluoride ion becomes subsurface when groundwater is exploited for various uses. Once it contaminates land, the whole environment and flora and fauna get affected due to accumulation of fluoride. The magnitude of adverse effects of fluoride could be brought down through proper management of environmental pollution and changing the food habits. The government should give enough emphasis in time to control the fluoride-related illness before it becomes epidemic. Since there is no specific antidote for this illness, prevention is better than cure.

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