

## **Incidence and effects of fluoride in Indian natural ecosystem: A review**

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### **ABSTRACT**

*Fluoride contamination in drinking water due to natural and anthropogenic behavior has been documented as one of the major problems worldwide impressive a serious threat to human health. Fluoride in drinking water has an intense effect to teeth and bones. The WHO and BIS has been decided fluoride concentration up to 1-1.5 mg/L as a permissible limit for drinking. concentration of fluoride in the level of 1.5-4mg/L result in dental fluorosis whereas with prolonged consumption at still higher fluoride concentrations (4-10mg/L) dental fluorosis leads to skeletal fluorosis. High fluoride concentrations in ground water occur widely in many parts of world. This review article is aimed at providing precise information about fluoride effects and contamination in soil and water Indian states such as Andhra Pradesh, Rajasthan, Haryana, Uttarpradesh, Madhya Pradesh, Gujarat, Maharashtra, Tamil Nadu and Karnataka are reviewed.*

**Keywords:** Fluoride, Dental Fluorosis, Skeletal Fluorosis, Indian natural ecosystem, Incidence

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### **INTRODUCTION**

Over all 200 million people worldwide rely on water source contaminated with high fluoride. The probability of occurrence of high fluoride concentration in ground and surface water was detected in varies countries include India, China, Argentina, Mexico and in several African countries and Pakistan, Italy, Iran, Bangladesh, Newzeland, Ethiopia, UK were fluoride contaminated countries. The dental fluorosis and skeletal fluorosis are endemic in number of countries, they are U.S.A, Morocco, Algeria, Libya, Egypt, Jorden, Turkey, Franfrac, kenya, Tanzania, South Africa, Australia, Japan, Thailand, Canada, Saudi Arabia, Persian Gulf, Srilanka, Syria. The above said countries are most prominent fluorosis countries in worldwide [1]. Ground water is a valuable natural gift that is very important for human health, socio-economic development and functioning of ecosystems [2,3]. Ground water will become contaminated by naturally or manmade activities, municipal activities, commercial, industrial and agricultural activities can all contaminate ground water quality [4,5]. The more dependence on groundwater to convene ever-increasing demands of domestic, agriculture, and industry sectors has resulted in over utilization of groundwater resources in several states such as Gujarat, Rajasthan, Punjab, Haryana, Uttar Pradesh, Tamil Nadu, among others [6].

In India, 65 million peoples at risk in dental fluorosis and skeletal fluorosis, in India many states are endemic fluorosis, Andhra Pradesh, Tamilnadu, Karnataka, Gujarat, Rajasthan, Punjab, Haryana, Bihar and Kerala [7]. These states are contributing dental fluorosis, which drinks above 1.5 mg/l of fluoride of drinking water. In Tamilnadu,

Salem, Erode, Dharmapuri, Coimbatore, Thiruchirapalli, Vellore, Madurai, Viruthunagar and Krishnagiri are having fluoride contamination and people risk with dental and skeletal fluorosis. According to WHO the permissible limit of fluoride concentration in drinking water is 1.5 mg/l. In India, most of the populations dependent on groundwater source for drinking water supply [8].

The main source of fluoride contaminant in drinking water, are water additives and fluoride discharge from fertilizer and aluminum factories [9]. The excessive fluoride intake, in consequence to the inadequate use (or) swallowing of fluoride containing toothpaste, is responsible for the development of dental fluorosis. Children up to 5 years old swallow around 30% of the amount of toothpaste used every time they brush their teeth [10]. The use of fluoride is considered an important factor in the prevention and management of dental caries, inhibiting demineralization and stimulating remineralization, due to the widespread of many other fluoride sources a decline in dental caries and increase in the prevalence of dental fluorosis have been documented in communities with and without fluorinated drinking water [11]. The linkage between fluoride and human health was first postulated during the late nineteenth century, when chemists recognized the variable fluoride contents of bones and teeth in human [12]. Skeletal fluorosis is due the threshold level of fluoride. Skeletal fluorosis is characterized by increased bone mass and density [13]. Diagnostic method of fluorosis, the adequate diagnosis of fluorosis requires inspection of dry and clean dental surfaces, under a good light source. The clinical appearance of mild dental fluorosis is characterized by bilateral, diffuse, opaque and white striation that runs horizontally across the enamel. Nowadays, the differential diagnosis between fluorosis and non-fluorosis induced opacities needs to establish differences between symmetrical and asymmetrical and /or discrete patterns of opaque defects controlling the fluoride intake is the best preservative measures for dental fluorosis. Bleaching and enamel micro abrasion techniques are conservatives and provide highly satisfactory results [14,15,16].

The wide range of defluoridation techniques have been developed to bring down the excess fluoride concentration below permissible limit. They are physical, chemical and biological method for defluoridation. Defluoridation techniques include precipitation, electro coagulation, electro dialysis, membrane process and adsorption. Precipitation method is commonly used economical method, which involves addition of salts. Nalgonda technique developed at our institute is based on precipitation with alum and lime and is one of the mostly widely used techniques for fluoride removal. Although membrane methods have successfully reduced fluoride concentration to acceptable level, surface adsorption retains a major place in defluoridation research and practice because it's general great accessibility and low cost [17].

Biosorption is an emerging technique for water treatment utilizing abundantly available biomaterials. Number of biosorbents has been developed for fluoride removal. Among various biosorbents, chitin and chitosan derivatives have gained wide attention as effective biosorbents due their low cost [18]. Besides chitosan, some other biosorbents such as algal and fungal biomass also has been examined fluoride removal [19]. Agricultural waste materials being economic and eco-friendly due to their unique chemical composition, renewable and low cost are viable option for defluoridation [20].

## 2. Sources of fluoride

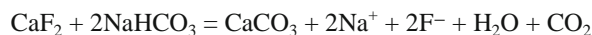
Fluorine is the thirteenth most abundant element in the Earth's crust. It rarely occurs as the element but normally is found as the fluoride ion or as a number of inorganic and organic fluorides. It occurs in varying concentrations in rocks, soil, water, air, plants and animals both naturally and as a consequence of human activity such as agricultural or industrial processes. Human exposure may be through any or all of these sources. This review is restricted to consideration of human exposure to fluoride through diet and the gastrointestinal tract (GIT), although exposure may also occur through inhalation of aerosols or dust particulates (e.g. cryolite,  $\text{Na}_3\text{AlF}_6$ ) in the workplace or through volatile anesthetics (eg. Halothane,  $\text{CH}_3\text{CHBrCl}_3$ ) used in certain types of surgery.

### 2.1. Fluoride distribution in water

Water is an essential natural resource for sustaining life and is among nature's most valuable gifts. Once viewed as an infinite and bountiful resource, today, water often defines the limits of human, social, and economic development for a region. The main source of freshwater for sustaining life on earth is groundwater. Unfortunately, groundwater is either being increasingly depleted for irrigation of crops, industrial, or other uses, or is becoming contaminated by various pollutants. The presence of fluoride as a contaminant of groundwater has become a worldwide problem,

because it is commonly found in groundwater sources. The problem of high fluoride content in groundwater resources is important, because of both toxicological and geo-environmental concerns. The chief source of fluoride in groundwater is fluoride-bearing minerals that exist in rocks and soils. The weathering and aqueous leaching processes that occur in soils play an important role in determining the amounts of fluoride that reaches groundwater.

The various factors that govern the release of fluoride into water from fluoride-bearing minerals are (i) the chemical composition of the water, (ii) the presence and accessibility of fluoride minerals to water, and (iii) the contact time between the source mineral and water [21]. Overall water quality (e.g., pH, hardness, and ionic strength) also plays an important role by influencing mineral solubility, complexation and sorption/exchange reactions [22].



The above equation clearly shows the processes that could control negative (Between fluoride and calcium) and positive relationships (between fluoride and Bicarbonate) when both are in contact with each other. Water samples in which Fluoride levels exceed 5 mg/l are oversaturated with regard to fluorite. Once fluorite reaches equilibrium, calcite is removed by precipitation, which allows the fluoride concentration to increase [23].

In groundwater, the natural concentration of fluoride depends on the geologic, chemical, and physical characteristics of the aquifers, porosity and the acidity of the soils and rocks, the temperature, the action of other chemical elements, and the depth of the wells. In natural water, the fluoride forms strong complexes with Al, and therefore, fluorine chemistry is largely regulated by Al concentration and pH level. Below pH 5, fluoride is almost entirely complexes with Al, predominantly with the  $\text{AlF}^{2+}$  complex, and consequently the concentration of free fluoride is reduced to low levels. As the pH increases, the Al–OH complexes dominate over the Al–F complexes, and the free fluoride level increases. Fluoride occurs at some level in almost all groundwater, but the concentration found in most potable waters is less than 1 mg/l [24].

It has been postulated that fluoride-bearing minerals are normally only sparingly water soluble, with the exception of villiumite, and these minerals release fluoride to water slowly. The rate of fluorite dissolution may be faster in sodium bicarbonate-containing waters, and the release of fluoride from clay minerals depends strongly on the pH level. The maximum concentration of fluoride in groundwater is usually controlled by the solubility of fluorite [25]. Once the solubility limit for fluorite ( $\text{CaF}_2$ ) is reached; an inverse relationship will exist between fluoride and calcium concentrations. Earlier studies have revealed that there is a close association between high fluoride content and soft, alkaline (i.e., sodium bicarbonate) groundwater that is depleted of calcium [26]. Igneous rocks that have been formed from highly evolved magmas are a rich source of fluorine bearing minerals. The plagioclase composition of igneous rocks is typically high in albite, the sodium-rich end-member [27]. As a result, the groundwater in contact with these rocks is often soft and calcium deficient, which allows for higher fluoride concentrations when equilibrium with fluorite is, attained [28].

## 2.2. Fluoride distribution in soil

Whereas the fluorine content of most rocks ranges from 100 to 1,300 mg/kg soil concentrations typically vary between 20 and 500 mg/kg.<sup>12</sup> However, much higher concentrations (1,000 g/kg) can occur in soils that are derived from rocks with high fluorine contents or in soils affected by anthropogenic inputs, such as phosphate fertilizers [29]. Most of the fluorine found in soils occurs within minerals or is adsorbed to clays and oxy-hydroxides, with only a few percent or less dissolved in the soil solution. Fluoride mobility in soil is highly dependent on the soil's sorption capacity, which varies with pH, the types of sorbents present, and soil salinity [30].

Although aluminum smelters, and therefore fluoride emissions, exist in the temperate regions, there is a lack of studies dealing with the effects of addition of fluoride on the chemistry of temperate soils. Moreover the published studies refer to forest soils, whereas the behavior of managed soils under this circumstance has not been investigated. However, in highly F-polluted soil, as the soil becomes more acidic or alkaline, the risk of zootoxic concentrations of F in shoots of plants would increase [31]. The San Cipri'an Aluminium Smelter-Alumina Refinery Complex, located on the north coast of Galicia, NW Spain, since 1978, emits fluoride to the atmosphere, resulting in increased concentrations of fluorine in soils and vegetation in the immediate surroundings [32]. Stated that the soils in the vicinity of the smelter have a high fluoride sorption capacity. The fluoride sorption may bring about changes

in the soil composition. Understanding those changes is relevant for the management of the soils close to the smelter [33].

### **3. Health effect of fluoride in human beings**

Fluoride consumption is often regarded as a double-edged sword. When ingestion of fluoride in inadequate quantities (less than 0.5 ppm), F causes health problems (e.g., dental caries, lack of formation of dental enamel, and deficiency of mineralization of bones), especially among children (WHO, 1996). In contrast, if fluoride is consumed or used in excess (more than 1.0 ppm), it can cause health problems in the young and old. The various forms of fluorosis that may arise from excessive intake of fluoride through drinking water. If fluoride is consumed in more than 4.0 ppm, it can promote the dental fluorosis in children. If fluoride is consumed in more than 10.0 ppm, it can promote dental fluorosis, skeletal fluorosis and crippling skeletal fluorosis, possibly cancer [34].

#### **3.1. Dental fluorosis**

Fluorosis is a preventable disease of teeth and bones that afflicts millions of people worldwide. It is caused primarily by the prolonged ingestion of fluoride-rich drinking water, which is most often groundwater that has percolated through and leached volcanic and sedimentary deposits [35,36,37]. Dental fluorosis is an accumulation of fluoride in teeth and is caused by ingestion of fluoride during the period of tooth development, i.e. prior to tooth eruption [38,39]. The fluoride becomes incorporated into the crystal lattice structure of the enamel and causes hypomineralization which increases the porosity of the enamel [40,41].

Fluoride at excessive consumption levels causes the enamel to lose its luster. In its mild form, dental fluorosis is characterized by the appearance of white, opaque areas on the tooth surface, and in severe form, it is manifested by the appearance of yellowish brown to black stains and severe pitting of the teeth. This discoloration may be in the form of spots or horizontal streaks. Normally, the degree of dental fluorosis depends on the amount of fluoride exposure up to the age of 8–10. This is true because fluoride stains only the developing teeth while they are being formed in the jawbones and are still under the gums. The effect of dental fluorosis may not be apparent if the teeth are already fully grown prior to excessive fluoride exposure. The amount of fluoride absorbed by the body depends on a number of complex variables to do with the health and condition of the individual [42,43]. Mild forms of dental fluorosis are evidenced by the appearance of white horizontal striations on the teeth surface or opaque patches of chalky white discoloration [44].

#### **3.2. Skeletal fluorosis**

Skeletal fluorosis is characterized by increased bone mass and density, accompanied by a range in skeletal and joint symptoms. In early stages, the Symptoms include pain and stiffness in the backbone, hip region, and joints, accompanied by increased bone density (osteosclerosis). The stiffness increases steadily until the entire spine becomes one continuous column of bone, a condition known as ‘‘poker back’’. As this condition progresses, various ligaments of the spine can also become calcified and ossified. In its most advanced stages, fluorosis produces neurological defects, muscle wasting, paralysis, crippling deformities of the spine and major joints, and compression of the spinal cord. The threshold level of fluoride ingestion needed to cause skeletal fluorosis varies depending on water Intake, water quality, and other dietary factors [45].

Skeletal fluorosis affects both children and adults. It does not easily manifest itself until the disease attains an advanced stage. Fluoride is mainly deposited in the joints of the neck, knee, pelvic, and shoulder bones, and once it takes place, it makes movement or walking difficult. The symptoms of skeletal fluorosis are similar to those of spondylitis or arthritis. Early symptoms include sporadic pain, back stiffness, burning-like sensation, pricking and tingling in the limbs, muscle weakness, chronic fatigue, and abnormal calcium deposits in bones and ligaments. At an advanced stage, osteoporosis in long bones and bony outgrowths may occur. A rare bone cancer, osteosarcoma, may result, and finally, the spine, major joints, muscles, and the nervous system may sustain damage. Crippling skeletal fluorosis is the advanced and severe form of skeletal fluorosis. The prevalence of high levels of fluoride intake over the long term, accompanied by malnutrition, strenuous manual labor, and impaired renal function, leads to severe skeletal fluorosis (Reddy 1985). Some cases of skeletal fluorosis have been documented in the Unites States [46].

The concentration of fluoride in the bone also varies with age, sex, type, and specific part of bone, and is believed to reflect an individual’s long-term exposure to fluoride. It was observed that approximately 99% of the fluoride in the

body is found in bones and teeth [47] (though the amount of fluoride in teeth is very small compared to bones), with the remainder distributed in highly vascularized soft tissues and blood. Besides skeletal and dental fluorosis, excessive consumption of fluoride may lead to many other disease manifestations: neurological manifestations, depression, gastrointestinal problems, urinary tract malfunctioning, nausea, abdominal pain, tingling sensation in fingers and toes, muscle fiber degeneration, low hemoglobin levels, deformities in RBCs, excessive thirst, headache, skin rashes, nervousness, reduced immunity, repeated abortions or still births, male sterility, reduced intelligence etc. Also found a significantly elevated risk of hip fractures in residents living in countries with fluoridated water [48].

### 3.3. Renal effects

The renal system is responsible for excreting most of the body's excess fluoride and is exposed to higher concentrations of fluoride than are other organs [49]. This suggests that it might be at higher risk of fluoride toxicity than most soft tissues. The chronic ingestion of fluoride can have non-carcinogenic effects on the kidney, and both pertain to the incidence of kidney stones [50]. More than 18,700 people living in a region of India where fluoride concentrations in the drinking water ranged from 3.5 to 4.9 mg/l and found that patients with clear signs of skeletal fluorosis were 4.6 times more likely to develop kidney stones.

### 4. Effect of fluoride on animals

Although the literature on fluorosis in cattle is extensive, information about fluorosis in horses is almost nonexistent. In 1974, US National Academy of Science report on effect of fluoride in animals. Most of horses in Justus Farm in Pagosa Springs, Colorado, affected by fluorosis due to consumption artificially fluorinated water, concentration up to 1.3 to 3.4 ppm. Fluorosis affected horses symptoms were dental fluorosis, crooked legs, hyperostosis and enostosis, hoof deformities and reduced bone resorption [51]. In Rajasthan dungarpur district natural occurrence of fluorosis was observed in a survey of domesticated dromedary camels. Among these eight camels were affected with mild to severe dental fluorosis [52]. Osteo- dental and non skeletal fluorosis was observed in domesticated cattle living in Chani village, Bikaner district of Rajasthan. Fluoride in drinking water of this village varies between 1.5 and 2.5 ppm [53]. In Rajasthan herbivores animals were affected by dental and skeletal fluorosis due to excess of high fluoride concentration in water [54]. The highest fluoride concentration found in invertebrates detritivores, compared to invertebrate herbivores and carnivores [55].

### 5. Fluoride contamination in India

Occurrence of fluoride in Indian groundwater's in several states of Andhra Pradesh, Rajasthan, Haryana, Uttarpradesh, Madhya Pradesh, Gujarat, Maharashtra, Tamilnadu and Karnataka in India was well documented [56]. In India, it was first detected in Nellore district of Andhra Pradesh in 1937. Approximately 62 million people including 6 million children suffer from fluorosis because of consumption of water with high fluoride contaminations [57,58].

#### 5.1. Fluoride contamination in Andhra Pradesh

Andhra Pradesh is most prominent fluoride contaminated state in India. In Andhra Pradesh, Nellore district, in the region of Udayagiri Taluk villages having high fluoride concentration in drinking water. They are Turkapalli 4.01 ppm, and Pakeerpalem 4.00 ppm, Varikunta padu 6.74 ppm, Bijjam palli 2.92 ppm, Masi peta 2.37 ppm, Singa reddy palli 2.98 ppm, Boda banda 3.47 ppm, Kolangadi palli 5.12 ppm, Gangi reddy palli 4.43 ppm, Basine palli 3.12 ppm [59]. The fluoride contamination in the South-eastern part of Ranga reddy district, in Andhra Pradesh. The endemic district of Andhra Pradesh has indicated that the fluoride rich ground water present in the wells located downstream water and close to the surface water is getting low fluoride [60]. Fluoride concentration in surface and ground water samples were determined in eight villages of Andhra Pradesh in India. Among these villages thirty-eight samples were collected and analysed for fluoride content along with  $P^H$ , electrical conductivity, total dissolved solid (TDS), total hardness, total alkalinity, chlorides, sulfates and nitrate. Fluoride concentrations in surface and ground water samples varied from 0.5 and 9.0 mg/l [61].

Ground water quality in the Varaha river basin located in the Visakhapatnam district of Andhra Pradesh has been investigated. An obtained chemical data of ground water suggest that the number of ground water samples show fluoride content greater than the safe limit. Some factors responsible for the occurrence of fluoride in the ground water are evapotranspiration, long contact time of water and agricultural fertilizers [62]. Fluoride concentrations in groundwater samples were determined in Uravakonda, Anantapur district of Andhra Pradesh. Fluoride concentrations in groundwater samples of these villages varied between 0.5 to 7.2 mg/l. Fluoride contamination in



groundwater in parts of Kudupa district, Andhra Pradesh. The fluoride concentration in groundwater of this region ranged from 0.226 to 3.52 mg/l [63]. In Andhra Pradesh, Kandukur revenue the concentration of fluoride ion in all drinking water samples has varied from 1.22 to 3.09 mg/l and 1.4 to 4.6 mg/l [64]. In Gunthakal area of Ananthapur district drinking water has fluoride level in the range of 0.18 to 2.00 mg/l [65]. Range between 1.1 to 5.8 mg/l fluoride concentration was determined in drinking water of Kommala area, Warangal district in Andhra Pradesh [66].

## 5.2. Fluoride contamination in Tamil Nadu

Tamil Nadu as a state has severe problem with fluoride contamination. In Tamilnadu, the high concentration of fluoride in groundwater is found to be in Dharmapuri, and Krishnagiri, Salem, closed followed Coimbatore, Madurai, Trichy, Dindugal and Chidambaram district. The districts having low fluoride are Thirunelveli, puthukotai, North Arcot, and Ramnad district. In Dharmapuri, region of garimangalam having the fluoride availability in groundwater varies from 0 to 2mg/l [67]. Excess fluoride more than the permissible limit of 1.5mg/l is observed at K.vetrapatti and Karukanchavadi areas (Dhinakaran, Central Ground Board, 2009) 22% of the samples are having higher concentration of fluoride (>1.5mg/l), in parts of Edapadi, Attayam patti and Jalakandapuram. This is observed the majority of the samples are characterized by higher concentration of NO<sub>2</sub>, SO<sub>4</sub>, and F than the BIS permissible limit [68].

In Cuddalore district, chidamparam taluk having higher fluoride concentration in ground water. The fluoride concentration was observed the maximum concentration of fluoride 2.6 ppm in following villages North pichavaram, Senjicherry, kanagarapattu, keezhaperambai in cuddalore district [69]. The fluoride concentration of the drinking water from various sources in the areas covered ranged from 0.47 to 6.6ppm. The mean concentration of fluoride in drinking water in Vellore district, in the region of Danakonda palli and madanancheri having 2.1 & 1.1 ppm in drinking water, and Dharmapuri in the region of Chinna Kuppam (1.8 ppm), Papireddi patty TP (1.7 ppm), Pattukoanam Patti (4.6 ppm). Krishnagiri district, in the region of Alapatti (1.9 ppm), periamuthur (2.9 ppm), Khammam palli (1.7 ppm), Salem district, in the region of Viruthasam patti (1.2 ppm), Ramreddi patti (1.1 ppm), Chittur (1.3 ppm). Erode district, in the region of Villarasam Patti (1.1 ppm), Chinnam palayam (0.6 ppm) [70].

Ambattur industrial area in chennai city has been highly contaminatated by fluoride. Ten different habitations were selected for the study and compared. Fluoride concentration in the range of 0.8 to 1.4 mg/l was finded in 10 different location of Ampatture industrial area [71]. The fluoride concentration in groundwater of the Tirupur and Coimbatore area varies between 0 to 2 mg/l with an average value of 0.9 and median was 0.6 mg/l. The concentration was higher than 1.5 mg/l in eight locations of Tirupur and Coimbatore [72]. The Blocks in the Coimbatore district, which is situated on the Western part of the study area, were found to contain 180 to 2,600 mg/kg F<sup>-</sup> (Handa, 1975). In Thoothokudi district, in the region of Ottapidaram block, having fluoride concentration was ranged from 0.936 to 4.34 mg/l with highest fluoride level at Ackkanicken Patti (4.34 mg/l), and lowest at Saminatham (0.936 mg/l).

The fluoride level lower than 1.0 mg/l was observed as 3.28% at two locations (Rajavinkovil and Saminatham), between 1 and 1.5 mg/l it was 14.75% at nine locations and at the fluoride level greater than 1.5 mg/l, it was 81.97% at fifty location. There are maximum numbers of village (21 samples) fall on the region of fluoride concentration between 1.5 to 2.0 mg/l [73]. Tirunelveli district of Tamilnadu, in the region of Sankarankovil block having higher concentration of fluoride in drinking water, out of 50 villages, only 24 villages contain water sources having fluoride within the limits. The remaining 26 villages had their drinking water sources contaminated with excessive fluoride. Among them, the samples of four villages namely Achampatty, melavayil, Kelavayil and Supplapuram have more than 3mg/l of fluoride. Among these highest concentration of fluoride was observed in supplapuram and the values were 3.84 mg/l. The lowest fluoride level (0.66 mg/l) was determined in Nainapuram [74].

In Dindigal district, certain blocks having higher incidence of fluoride (>1.2 mg/l) was reported in Dindigal, Nilakkotai, Palani, and Veda sandur blocks [75]. The high fluoride concentration in groundwater was identified in palacode region of Dharmapuri district in Tamilnadu, where it is only source of drinking water, in which fluoride was also higher concentration in the groundwater of these villages varied from 1.4 to 2.4 mg/l, causing dental fluorosis and teeth matting, among people in general and children [76]. In Salem district, some parts having fluoride contamination comparison between post-monsoon and pre-monsoon. At 4.20 mg/l was the maximum fluoride level in pre-monsoon season, and 0.36 mg/l was maximum fluoride level in post-monsoon season in groundwater.

Compare to both season variation of fluoride level, the pre-monsoon was having high concentration of fluoride in groundwater, due to the weathering and leaching of the greater availability of fluoride bearing minerals in soil [77].

The fluoride contamination was observed in Madurai district, in the region of Thirunagar, due to this, that area peoples and school going children's having dental fluorosis [78]. The fluoride concentration in underground water was determined in four Panchayats of Vellore district of Tamilnadu, where it is only source for drinking water. The prevalence of dental fluorosis and skeletal fluorosis was determined among the people of Narasingapuram panchayat of Alangayam block, Vellore district, where fluoride concentration in drinking water ranges from 2.35 to 4.59 [79]. In Tiruchirappalli district the fluoride concentrations in drinking water in the level of 0.45 to 2.09 mg/l were reported [80]. In Tamilnadu, Pappireddipatti block of Dharmapuri has fluoride content in the range of 1 to 14 mg/l [81].

### 5.3. Fluoride contamination in Gujarat

The fluoride concentration in the ground water varies in the district of north Gujarat region. It varies from 0.99 to 5.48 ppm in Sabarkantha district, 1.96 to 10.85 ppm in Patan district, 3.82 to 12.08 ppm in Mehsana district and 2.77 to 6.64 ppm in Banaskant district. Around 80% of the ground water samples from Sabarkantha district contain fluoride above the maximum permissible limit and 20% are only safe limit (0.99 to 1.25 ppm). In Patan district about 95% ground water samples contain above the maximum permissible limit, in Mehsana district also 95% of samples having higher fluoride level, in Banaskant district 92% samples having fluoride above maximum permissible level [82]. In Gujarat the number of fluoride affected habitations were increased from 2,826 in the year 1992 to 4,187 by year 2003. In ground water of Gujarat, the fluoride concentration in these villages ranged from 1.5mg/l to as a 18.90 mg/l [83].

The Kheralu taluka of mehsana district people have health risk due to the excess of fluoride present in the drinking water. This block is rich in agricultural but is highly depending on groundwater, for both irrigation and drinking water purposes. During the last century, large scale utilization of groundwater for irrigation, so that groundwater source getting low rate. As a result, Kheralu taluka region face the problems like dissolution of Fluorides and other dissolved salts in drinking water. The process of deterioration of groundwater quality continues unabated and progressively increasing proportion of the population is affected by fluorosis. In Kheralu taluka water samples are collected from 60 villages, among these villages most of the water samples have high fluoride contamination above the permissible limit [84]. Ground water quality problem have risen in many geographical parts due to natural environmental processes and manmade activities in the ecosystem. Hydro geochemical of fluoride contamination in groundwater was investigated in Mehsana district, Gujarat state. In Mehsana district some location has fluoride contamination. They are Bhatson, Bhandu, Dabhoda and Pamol [85].

### 5.4. Fluoride contamination in Maharashtra

The Occurrence of fluoride was observed in ground water of Pandharkawada area in Yavatmal district of Maharashtra. The following villages having high fluoride concentration in ground waters viz, Chikhaldara 0.48 mg/l, Mohadari 0.34 mg/l, Runjha 0.61 mg/l, Khatara 4.81 mg/l, Sonurli 3.03 mg/l, Karanji (phul pod) 2.45 mg/l, Wadhona (Bk) 5.76 mg/l, Wadhona (Kh) 5.75 mg/l, Dharna 13.41 mg/l, Sakhra 11.9 mg/l, Nilgiri 3.50 mg/l, Ganeshpur 2.84 mg/l, Wai 3.02 mg/l, Datpari 2.91 mg/l, Pimpri 0.90 mg/l, Gevrai munch 4.81 mg/l, Marathwakdi 1.0 mg/l, Dhoki 1.77 mg/l, Shushri 5.95 mg/l, Pendhari 2.88 mg/l, Tembhi 1.58 mg/l, Washa 1.77 mg/l, Arli (Bk) 0.61 mg/l, Pimpershenda 1.01 mg/l, Karegaon 0.30 mg/l [86]. Fluoride contamination was observed in Gad river basin in Maharashtra. The following blocks are having fluoride concentration in groundwater, block of Deulwadi (2.4 mg/l), Humrat (4.9 mg/l), and Kalmath (2.5 mg/l) [87]. In Maharashtra Chandrapur block groundwater having fluoride concentration above 1.5 mg/l. The water samples collected from open well and bore wells from 27 different locations. From the water sample collected are having fluoride concentration in the range of 1.0 to 3.0 ppm [88]. Assessment of fluoride concentration was carried out in Godavari River and ground water of the Nanded city in Maharashtra. In which people has health risk due to the excess of fluoride present in drinking water above permissible limit (1.5 mg/l). So peoples are affected by many diseases viz, dental and skeletal fluorosis, some cases of deaths. Fluoride was found to be in the range of 0.43mg/l to 2.0mg/l. In Nanded city municipal tap water and ground water fluoride concentration above 1.5mg/l [89].

### 5.5. Fluoride contamination in Rajasthan

In Northern Rajasthan many parts were contaminated by excess fluoride contamination in drinking water. The following areas are having high Fluoride contamination in drinking water, Fluoride range between 4.78 to 1.01 mg/l. Some parts of Rajasthan risk in endemic fluorosis, where as above 1.5 ppm of fluoride concentration in drinking water [90]. A study on distribution and health hazards by fluoride contaminate in groundwater was performed in 1,030 habitations of Bhilwara district of central Rajasthan. 1030 samples were collected and analyzed for fluoride concentration. Fluoride content in these villages varies from 0.2 to 13.0 mg/l [91].

Fluoride distribution was observed in groundwater and survey of dental fluorosis in villages of Didwana Tehsil of Nagaur district of central Rajasthan in India. Totally 54 water samples were collected and fluoride determined by Orion fluoride electrode. The fluoride concentration in the Tehsil ranges from 1.1 to 8.5 mg/l. The maximum fluoride concentration was recorded from village Didwana (8.4 mg/l) [92]. The Indira Gandhi Canal catchment area has fluoride concentration in drinking water in the range of 2.50 to 4.48 mg/l, villages of Bhakra canal catchment area fluoride concentration 1.00 to 5.75 mg/l, villages of Gang canal catchment area having fluoride concentration from 1.50 to 3.50 mg/l [93]. The Ground water quality assessment of Nawa Tehsil area ground water having high fluoride contamination between 14.62 to 24 ppm. Fluoride contamination in some villages of Nagaur district, they were Todas 3.81 ppm, lalas 5.13 ppm, Khorandi 4.17 ppm [94].

### 5.6. Fluoride contamination in West Bengal

The Assessment of potential hazards of Fluoride contamination in drinking water was reported in West Bengal of India. The following villages have high fluoride contamination in ground water viz, Mogra having Fluoride contamination in the range of (0.11 to 0.96 µg/ml), Chanditala-II (0.03- 1.12 µg/ml), Haripal (0.07-1.05 µg/ml), Tarakeswar (0.07 to 1.00 µg/ml), Dhaniakhali (0.02 to 1.00 µg/ml) [95]. In West Bengal fluoride rich groundwater was reported in Birbhum district. The occurrence of fluoride rich groundwater in Asanjola, Madhabpur and Narayanpuram villages of Birbhum district in West Bengal, in which high concentration 1.2 to 20.9 mg/l of fluoride was, detected [96]. Distribution of fluoride was investigated in groundwater of Raniganj coal field, West Bengal in India. The fluoride concentration varies from 0.20 to 1.6 mg/l [97]. Bankara district of West Bengal peoples affected by dental and skeletal fluorosis due to the high concentration of fluoride in drinking water [98]. Purulia district of Westbengal has fluoride concentration in the range of 0.07 to 2.27 mg/l [99].

### 5.7. Fluoride contamination in Uttar Pradesh

In Uttar Pradesh Makur and Unno districts having high fluoride concentration in drinking water. In Makur block, the fluoride concentration in the groundwater was found to vary between 1.05 to 13.9 mg/l. Which exceeded the maximum desirable limits of 1.0 mg/l of fluoride in the drinking water as lay down by BIS [100].

### 5.8. Fluoride contamination in Karnataka

In Karnataka Kolar and Tumkur districts, have high fluoride concentration in drinking water. The fluoride concentration in Kolar district, in the range of 0.36 to 3.34 mg/l and fluoride concentration in Tumkur district in the range of 0.78 to 5.35 mg/l [101]. Fluoride release into ground waters in Ilkal area of Bagalkot district in Karnataka water samples were collected from bore wells, dug wells, mine pit, infiltration gallery, lake, river for the measurement of fluoride. Fluoride concentration in water samples varies from 0.1 to 6.5 mg/l. Ilkal block of bagalkot district, benakandoni habitation having high fluoride concentration at 6.5 mg/l [102]. Hathiguddur in Gulbarga district has a fluoride level of 7.4 mg/l, while 5.75 mg/l was observed in Farhatabad. District bellary showed a wide range of fluoride concentration. Village Sanavasapur and Tekalakota have 7.4 mg/l, while Kurugodu and Verupayur have as low as 0.95 mg/l [103]. Ground water samples collected from 50 different locations from the Jamakhanali taluk, the fluoride concentration were varies from 0.062 to 0.061 ppm.

### 5.9. Fluoride contamination in Kerala

Kerala, as a state, has only mild problem with fluoride concentration compare to other Indian states. In Palakkad, the fluoride levels varied from the detection limit to 1.8 mg/l and fluoride concentration in Alappuzha varied from 0.3 to 1.6 mg/l [104].



**5.10. Fluoride contamination in Assam**

Fluoride concentration was reported in groundwater of small Tea gardens in Sonitpur district, Assam in India. The fluoride concentration in Tea garden water samples varied from 0.6 to 5.602 ppm [105]. 30 water samples were collected from different locations of Tinsukia district, the fluoride levels were varies from 0.0912 to 0.2283 mg/l [106]. Lukhimpur district of Assam, among six tea gardens has fluoride contamination. The fluoride level in the range of 0.39 to 1.06 mg/l [107]. In Assam Nalbari district drinking water has fluoride concentration in the range of 0.02 to 1.56 mg/l [108].

**5.11. Fluoride contamination in Chattisgarh**

Fluoride contamination was reported in groundwater of Tamnar area, Raigarh district in the state of Chattisgarh. Hence this district peoples are risk in dental fluorosis [109].

**5.12. Fluoride contamination in Haryana**

Haryana is one of the fluorosis states in India. Assessment of fluoride in spent wash of the distilleries of Haryana. The spent wash samples were collected from distilleries areas of Haryana. The samples were having fluoride concentration in the range of 1.95 to 2.32 mg/l [110]. Fluoride concentration was found to be more than permissible limits at seven locations. Highest value of 19.36 mg/l was observed at Korawal [111]. In Panipet city of Haryana has fluoride contamination in the range of 6.6 to 7.5 mg/l [112]. Dabwali town in Sirsa district of Haryana drinking water samples has fluoride concentration in the range of 0.90 to 34.50 mg/l [113]. The fluoride occurrence of in underground water was quantified in five villages of Hodal block, Faridabad district of Haryana. The fluoride concentration in Hodal blocks area in the range of 1.0 to 40.0 mg/l [114].

**5.13. Fluoride contamination in Orissa**

In India Orissa is one of the fluoride contaminated state. In Orissa, Nayagarh district was have high fluoride contamination in drinking water. In Nayagarh district many villages having fluoride contamination in the range of 0.3 to 10.1 mg/l in groundwater [115]. Delineation of fluoride contaminated groundwater around a hot spring in Nayagarh district of Orissa. The fluoride concentration varies from 0.2 to 12.7 mg/l in ground water [116]. Hydrogeochemical method was processed for controlling the high fluoride concentration in ground water in Boden block area of Orissa. The Boden block area of Orissa having fluoride concentration varies from 0.0 to 6.4 mg/l. Fluoride contamination in Angul district of Orrisa due to releases of fluoride from various industries. So that assessment of fluoride carried out. Eighteen groundwater samples were collected from different locations (open well and tube well). Fluoride content in drinking water in the range of 0.2 to 2.4 mg/l was recorded [117]. Balasore district of Odisha is one of the fluoride contamination are in India. The data show that many of peoples in this region of Odisha have either dental or skeletal fluorosis. Chakulia, Kunnarpur, Nuagun areas having fluoride contamination in the range of 0.6 to 5.83 mg/l [118].

**5.14. Fluoride contamination in Himachalpradesh**

Himachalpradesh is one of the fluoride contamination states in India. Quality of water from hot springs in Mandi district of Tattapani habitation has brackish water with EC ranging from 1480 to 9700  $\mu\text{S}/\text{cm}$  and fluoride concentration ranging from 1.03 to 1.66 mg/l [119].

**5.15. Fluoride contamination in Jharkhand**

Fluoride contamination in groundwater sources in Palmu district, Jharkhand. Groundwater samples were collected from different parts of Palmu district. The maximum fluoride concentration (4.2 mg/l) was observed in daltongani block [120]. Chronic fluoride intoxication in the form of dental and skeletal fluorosis was surveyed in five villages of the Palamau district, in Jharkhand. Out of 238 sample of drinking water, mainly from groundwater, the majority had elevated fluoride concentrations capable of causing health risk to the community. Among these one water sample has high fluoride concentration 12 mg/l [121]. In Jharkhand State, Garhwa district water samples are analyzed for fluoride content in drinking water. From this block of Garhwa 4012 water samples were collected, among these 295 (7.4%) samples were tested in laboratory for analyzing the concentration of fluoride. Fluoride concentration in drinking water in the range of 0.018 to 5.92 mg/l. kharaundhi and untari blocks has high fluoride concentration [122].

### 5.16. Fluoride contamination in Punjab

Ministry of water resources, Government of India, thirteen states in India have been identified as fluorosis affected to the presence of natural fluoride bearing minerals in subsoil. Punjab (Bhatinda and Sangrur), is one of them. The maximum value fluoride 22.6 mg/l has been reported in kachikhanuri in Sangrur district [123]. Fluoride contamination was observed in groundwater of kalalanwala village in east Punjab. The maximum fluoride concentration was 22.8 mg/l [124]. In Punjab many Blocks has fluoride contamination above permissible limit (1.5 mg/l) viz. Amiritsar, bhatinda, faridkot, Firozepur and Gurdaspur [125].

## CONCLUSION

The brief account on sources, impacts and contamination of fluoride in Indian natural ecosystem have been presented. The sources of fluoride have been divided in two sections dealing with water and soil. Impacts of fluoride on humans and animals are discussed. Dental fluorosis, skeletal fluorosis, crippling fluorosis, renal effects are discussed with their exceeding limit fluoride in water. For that, In India populations have been facing high health risk on fluorosis. In worldwide the millions of people affected by dental fluorosis and skeletal fluorosis. Effect of fluoride on humans, animals, plants and marine ecosystem due to the release of fluoride from industries like, Coal mining, aluminium smelter and etc. Today, there are several fluoride containing dental restoratives available in the market including glass ionomers, resin modified glass ionomer cement and tooth paste. So to overcome the fluorosis in worldwide by safe drinking and avoid the discharge of industries waste to water bodies.

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