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REVIEW ARTICLE

FLUORIDES IN HEALTH AND DISEASE- A REVIEW

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ABSTRACT:
Fluoride has played an essential role in oral health. The problem of Fluorosis is world wide affecting many countries and in India especially Rajasthan which is the largest state of India. The cause of problem is high fluoride content of water more than 1 ppm and chronic long term ingestion as stated by the different studies around the world and World Health Organization (WHO). Fluorosis does not only affect oral cavity but has more severe and generalised effect on the body resulting in hard tissue and soft tissue irreversible damage in form of dental fluorosis, increased chances of caries and periodontal diseases, skeletal and non-skeletal fluorosis and acute toxicity which is lethal. The present paper discusses the impact of these fluorides in health and disease. Authors concluded that being from medical professionals it is our duty to work in preventing this endemic disease by educating our patients and people in the affected areas.
Key words: Fluorides, Fluorosis, Oral Health.

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INTRODUCTION

The word fluoride is derived from the Latin term 'Fluore' meaning 'to flow'. Fluorine belongs to Halogen group at room temperature fluorine is a pale green gas. It is the most electronegative and reactive of all elements and thus in nature rarely found in its elemental state. This element was isolated in 1886 by Nobel 'Laureate Henri Moissan' and it combines directly with most of metals and indirectly with few of metals to form Fluoride. Fluoride is ubiquitous in nature and is present in rocks, soil, water, plant food and even in air.[1,2] Fluoride is seventeenth in order of frequency of occurrence of elements representing about 0.06 to 0.09 % of the earth crust and in soil it occur in form of Fluorspar, Fluoroapatite and Cryolite. Fluoride has immense public health importance but also act as 'Double Edged Sword', lack of fluoride in drinking water leads to dental caries while excess results in fluorosis. According to 1984 guidance by WHO fluoride is an effective agent in preventing dental caries if taken in optimal amount. But a signal optimal level for daily intake cannot be agreed because the nutritional state of individuals varies greatly influences the rate of at which the fluoride is absorbed by the body. A diet poor in calcium for e.g. increases the body's retention of fluorine. [1] Water is the major source of fluoride intake. The 1984 WHO guidance suggests that in areas with a warm climate the optimal fluoride concentration in drinking water should be bellow 1 mg/ litre (1 ppm or parts per million) while in cooler climate it could go up to 1.2 mg/litre. The WHO guidance value for fluoride in water is not universal. In India for e.g. lower its upper permissible upper limit from 1.5 -1.0 ppm in 1998.[3]

FLUORIDE [4]

The relationship between fluoride and dental caries was first noted in the early part of 20th century when it was observed that residents of central area of USA developed brown stains on their teeth. Fluorosis is caused by excessive intake of fluoride. The dental effects of fluoride develop much earlier than the skeletal effects in people exposed to large amount of fluoride .The clinical dental fluorosis is characterized by staining and pitting of the teeth. In more severe cases all the enamel would be damaged.

Chronic high level of fluoride can lead to skeletal fluorosis. In skeletal fluorosis, fluoride accumulates in bone progressively over many years. The early

symptoms of skeletal fluorosis include stiffness and joint pain. In severe cases, bone structure may change and ligaments may calcify, which results in impairment of muscle and pain. Fluorosis can also result in kidney and liver problem in large dosage of fluorides and persistent intake of small amount over many years can adversely influences soft tissue as well. The most important manifestations are gastro-intestinal, respiratory and cardio-vascular symptoms, as well as allergic skin lesions. [5]

Endemic fluoride, as the effect of chronic fluoride intoxication as a universally known and continues to be a problem of public health magnitude all over the world.

FLUORIDE IONS[1, 5]

Fluorine, a gaseous element is a halogen which being most electronegative and reactive of all elements does not occur in free form in nature. This element was isolated in 1886 by Nobel 'Laureate Henri Moissan' and it combines directly with most elements and indirectly with few to form fluorides. Fluorides are ubiquitous in nature and are present in rocks, soil, water, plants, foods and even air.

Hydrogen fluoride (HF) is a colorless, pungent liquid or gas that is highly soluble in organic solvents and in water in which it forms hydrofluoric acid. Calcium fluoride (CaF_2) is a colorless solid that is relatively insoluble in water and dilute acids and bases. Sodium fluoride (NaF) is a colorless to white solid that is moderately soluble in water. Sulfur hexafluoride (SF_6) is a colorless, odorless and inert gas that is slightly soluble in water and readily soluble in ethanol and bases.

The most common procedure used to quantify free fluoride anion is the fluoride ion-selective electrode. Micro diffusion techniques are considered to be the most accurate methods of sample preparation (i.e. liberation of free ionic fluoride from organic and inorganic complexes).

Chemical Properties of Fluorine

- It is element of Halogen group with molecular weight 19 and atomic number 9.
- Fluorine is the most electro negative of all elements
- Fluorine exists as a diatomic molecule with remarkably low dissociation energy (38 K cal/mole). As a result it is highly reactive and has strong affinity to combine with other elements to produce compounds known as fluoride.

ENVIRONMENTAL FLUORIDE [1]

Fluorides are released into the environment naturally through the weathering and dissolution of minerals, in emissions from volcanoes and in marine aerosols. Fluorides are also released into the environment via coal combustion and process waters and waste from various industrial processes, including steel manufacture, primary aluminium, copper and nickel production, phosphate ore processing, phosphate fertilizer production and use, glass, brick and ceramic manufacturing, glue and adhesive production. The use of fluoride-containing pesticides as well as the controlled fluoridation of drinking-water supplies also contributes to the release of fluoride from anthropogenic sources. Based on available data, phosphate ore production and use as well as aluminium manufacture are the major industrial sources of fluoride release into the environment.

SOURCES OF FLUORIDE [6,7]

1. **Foods:**
   Nearly all foods contain small quantities of fluoride and the total daily intake through any average human diet is small except in endemic regions. In certain endemic regions of India the fluoride content of vegetables and food may be very high .The contribution of food to the total daily intake of fluoride varies from region to region.

2. **Water and beverages:** [5, 8,9]
   In case of natural waters, the variation in the fluoride content from region to region is dependent upon such factors as the source of water, type of geological formation and the amount of rainfall. Surface water generally has low fluoride while ground water may have high concentrations of fluoride as has been found in many parts of the world. The highest fluoride concentration of 28.9 ppm was reported from India. The fluoride content of seawater varies from 0.8 to 1.4 ppm which explains why the fluoride content of diet rises when sea foods are consumed. Among beverages tea has an exceptionally high fluoride content which varies in different brands from 122-260 ppm or more. Each cup of tea may supply 0.3-0.5 mg of fluoride. Bottled beverages, which are increasingly being consumed around the world, have a variable and some have high content of fluoride and should be considered as additional sources of fluoride.

METABOLISM OF FLUORIDE

Biological effects of fluoride intoxication are related to the total amount of fluoride ingested whatever the source be it food, water or air.

Absorption of fluorides [1, 9]

Soluble inorganic fluorides ingested through water and foods are almost completely absorbed and also those inhaled from the respiratory tract. But absorption of less soluble inorganic and organic fluorides varies from 60-80%. Fluorides are absorbed from the gastro-intestinal tract by a process of simple diffusion without any mechanism of active transport being involved. Various dietary components apparently influence the absorption of fluoride from the gut. It has been noticed that salts of calcium, magnesium and aluminium when added to diet reduce the quantum of fluoride absorption on account of the formation of their less soluble compounds. This is the reason why waters with high calcium and magnesium content check the incidence of fluorosis as indicated by epidemiological studies. Therefore, it is to be expected that all other factors being equal the incidence of skeletal fluorosis would be less where the calcium and magnesium content of drinking water is high. It is

noteworthy that administration of magnesium salts (serpentine and magnesium hydroxide) to patients suffering from fluorosis and experimental animals has increased the faecal and urinary excretion of fluorides. Similarly, increased absorption of fluoride from gastrointestinal tract ensues from the addition of substances like phosphates, sulphates and molybdenum to the diet and these can increase fluoride toxicity.

Distribution of fluoride [1, 8, 10]

About 96-99% of the fluoride retained in the body combines with mineralized bones, since fluoride is the most exclusive bone seeking element on account of its affinity for calcium phosphate .But it has been noticed that there is no significant retention of it in the body if very small quantities of fluorides are ingested. In fact, there was no discernible retention of fluoride when up to 4-5 mg was ingested daily. But when more than 5 mg were ingested about half of it appeared to have been retained by the skeleton and rest excreted through urine. Observations show that after absorption from the gut fluoride enters the circulation, the plasma fluoride accounting for the three-fourths of the total amount of fluoride found in the whole blood and cells for the rest. Fluoride in plasma exists in free ionic and bound forms, the later form bound to the serum albumin forming about 85% of the total amount fluoride in plasma.

Plasma fluoride in normal individuals in non-fluoridated areas ranges from 0.14-0.19 ppm and is higher in fluorotic patients. Newer methods which only measure ionic component of plasma fluoride levels are lower and range between 0.004 - 0.008 ppm when drinking water contained traces of fluoride and varied from 0.1- 0.02 ppm when water was fluoridated. Plasma fluoride concentrations tend to increase slowly over the years. It is seen that plasma levels of fluoride do not fluctuate widely despite a wide variation of fluoride levels in drinking water presumably because of the action of some regulatory mechanisms, which have not yet been clearly identified. The sequestration of fluoride into the skeleton, urinary excretion and loss sustained through sweat help in regulation of plasma fluoride. The levels of fluoride in most soft tissues of the body are lower than 1 ppm but are higher than those of plasma. The fluoride content of brain is 0.4 - 0.68 ppm and the concentration in C.S.F is 0.1 ppm, which is lower than plasma.

The uptake of fluoride by the skeleton is very rapid and depends upon the vascularity and the rate of its growth. The fluoride uptake of young bones is faster than that of mature bones. The fluoride is incorporated more readily in the active, growing and cancellous areas of bone, than in the compact regions. It has been observed that skeletal fluoride concentration increases almost proportionately to the amount of fluoride ingested and the duration of its ingestion. The amount of fluoride present in various bones of same skeleton differs from bone to bone with pelvis, vertebrae registering higher fluoride content than limb bones. Even in the limb bones amount of fluoride deposited in them depends upon the activity of muscles attached to them. In caged monkeys fluoride content of upper limb bones is more than the lower limb bones. It is this increase in the fluoride content of skeleton that provides the most reliable clue to excessive fluoride intake. The other indicators such as urine and soft tissue levels which manifest wide fluctuations cannot be relied upon. Once incorporated into the hard tissues the fluoride is retrievable though with difficulty and entails an extremely slow process of osteoclastic resorption spread over many years.

REQUIREMENT AND DEFICIENCY

Total daily fluoride intake [1, 4, 10]

The fluoride contents from all the sources determine the human intake of fluoride. In majority of endemic areas around the world the main contribution is from water and only in few areas of India and China significant amounts come from foods and rarely the polluted air is the culprit. The estimated range of safe and adequate intake of fluorides for adults is 1.5 to 4.0 mg per day and it is less for children and those with renal disease. The daily intake of fluoride in endemic regions varies from 10 to 35 mg and can be even higher in summer months.

REQUIREMENT OF FLUORIDE [1,2,11]

- Fluoride consumption in optimal amounts in the water supply imparted protection against the development of dental caries without staining the teeth (Dean 1938).
- Another benefit of fluorides is that the incidence of osteoporosis seems to occur less frequently in regions with high fluoride content in water than in those in which the inhabitants consumed little fluoride.
- Fluoride is an essential substance it can prevent or reduce dental decay and strengthen bones, thus preventing bone fractures in older people.
- Fluoride increases resistance or reduction in enamel solubility of enamel by promoting the precipitation of hydroxyapatite and phosphate mineral. When hydroxyapatite is exposed to a low fluoride concentration (1 ppm) a layer of fluoroapatite forms on the hydroxyapatite crystals. This thin layer governs the rate of dissolution.
- Fluoride increases the rate of mineralization of hypo-mineralized areas of newly erupted teeth which are often prone to caries.
- It causes remineralization of incipient lesions by deposition of minerals into previously damaged areas of tooth with resulting reduced enamel solubility.
- It interferes with micro organisms in two ways at higher concentration it is bactericidal and at lower concentration it is bacteriostatic.
- Fluoride acts as an inhibitor of demineralization from organic acids (acetic or lactic acid).

RECOMMENDED DAILY ALLOWANCE OF FLUORIDE [11]

The recommended maximum daily allowance depends on the age:-

AGE	FEMALE	MALE
0-6 months	0.01 mg	**0.01 mg**
6-12 months	0.5 mg	**0.5 mg**
1-3 years	0.7 mg	**0.7 mg**
4-8 years	1.1 mg	**1.1 mg**
9-13 years	2.0 mg	**2.0 mg**
14-18 years	2.9 mg	**3.2 mg**
>19 years	**3.1 mg**	**3.8 mg**

DEFICIENCY OF FLUORIDE [1, 2]

- Development of dental caries and incidence of osteoporosis and fracture seems to occur in regions with low fluoride content in water than in those in which the inhabitants consumed optimal amount of fluoride.
- Although, the importance of this element to normal mineralization of hard tissues and formation of caries resistant enamel has been recognized, there has been as yet no conclusive evidence proving that it is an essential element for human health (McClure 1970).
- Indeed, fluoride deficiency syndrome is yet to be described. This may be due to the fact that human body requirement of this micronutrient must be small, which is met with naturally through food and water.

FLUOROSIS [5, 12]

Fluoride intoxication presents an extraordinary degree of uniformity in its clinical manifestations. It occurs in humans as dental and skeletal fluorosis. They are separated by a prolonged relatively symptom free interval during which the skeleton does not stop accumulating fluoride. In its advanced stages, skeletal fluorosis causes crippling deformities and neurological complications. The effects of fluoride intoxication are related to the total amount of fluoride ingested, although earlier only water was taken into account presumably because the supply of fluoride by food was deemed negligible. The safest minimum daily intake of fluoride is not known, earlier reports suggested that any daily ingestion of over 28 mg of fluoride would be harmful, subsequent studies cited 20 mg as the maximum safe limit. But in endemic areas where the presence of certain local factors like nutritional status etc. and prolonged exposure tend to aggravate the fluoride toxicity the safe level of fluoride intake may be even lower. Cases of endemic skeletal fluorosis in India revealed an average fluoride intake of 9.88 mg and it is held that any intake of more than 8 mg would be harmful, so it is suggested that 3.2 mg as the highest level of fluoride intake which could be deemed safe. It is true that in persons with normal functioning kidneys there is wide margin of safety.

The severity of fluorosis has a definite relationship with the following factors:

- Fluoride concentration in drinking water,
- Period of exposure,
- Climatic factors (for example Temperature),
- Fluoride ingestion through other sources,
- Nutritional status,
- Chemical constituent of drinking water other than fluoride and
- Occupation

Concentration of fluoride in drinking water and its effects on human health

Fluoride Concentration in (mg/l)	Effect
Nil	**Limited growth and fertility**
< 0.5	**Dental caries**
0.5 - 1.5	**Promotes dental health, prevents tooth decay**
1.5 - 4.0	**Dental fluorosis (mottling and pitting of teeth)**
4.0 - 10.0	**Dental fluorosis, skeletal fluorosis (pain in neck bones and back)**
> 10.00	**Crippling fluorosis**

Dental fluorosis which is characterized by discolored, blackened, mottled or chalky-white teeth is a clear indication of overexposure to fluoride during childhood when the teeth were developing. These effects are not apparent if the teeth were already fully grown prior to the fluoride overexposure, therefore the fact that an adult may show no signs of dental fluorosis does not necessarily mean that his or her fluoride intake is within the safety limit. Chronic intake of excessive fluoride can lead to the severe and permanent bone and joint deformations of skeletal fluorosis. Early symptoms include sporadic pain and stiffness of joints, headache, stomach-ache and muscle weakness can also be warning signs. The next stage is osteo-sclerosis (hardening and calcifying of the bones) and finally the spine, major joints, muscles and nervous system are damaged.

Whether dental or skeletal fluorosis is irreversible and no treatment exists. The only remedy is prevention by keeping fluoride intake within safe limits.

DENTAL FLUOROSIS [2, 5, 8, 10]

Excessive ingestion of fluoride during the early childhood years may damage the tooth forming cells, leading to a defect in the enamel known as dental fluorosis. The teeth impacted by fluorosis have visible discoloration, ranging from white chalky spots to brown and black stains.

Teeth with fluorosis also have increased porosity of the enamel. In the milder forms the porosity remains to the sub-surface enamel, whereas in more advanced stage the porosity affects the surface enamel as well resulting in extensive pits. [13]

Both sexes are equally affected. It is the permanent teeth that are affected and they lose their normal creamy white translucent color and become rough, opaque and chalky white. Pitting and chipping are other marks of fluorosis. Brown or black pigment gets deposited on the defective enamel and once established tends to remain there permanently. Incidence of dental fluorosis in endemic areas exhibits a linear relationship to the fluoride content of water but it may also vary with other factors. Dental fluorosis does not obviously occur when there has been no exposure to fluoride in the first decade of life.

FLUOROSIS CRITERIA FOR DEAN'S INDEX [1, 3]

Score	Criteria
0	**Normal** - The enamel represents the usual translucent semi vitriform type of structure. The surface is smooth, glossy, and usually of a pale creamy white color.
0.5	**Questionable** - The enamel discloses slight aberrations from the translucency of normal enamel, ranging from a few white flecks to occasional white spots. This classification is utilized in those instances where a definite diagnosis of the mildest form of fluorosis is not warranted and a classification of "normal" is not justified.
1.0	**Very mild** - Small opaque, paper white areas scattered irregularly over the tooth but not involving as much as 25% of the tooth surface. Frequently included in this classification are teeth showing no more than about 1-2 mm of white opacity at the tip of the summit of the cusps of the bicuspids or second molars.
2.0	**Mild -** The white opaque areas in the enamel of the teeth are more extensive but do not involve as much as 50% of the tooth.
3.0	**Moderate -** All enamel surfaces of the teeth are affected, and the surfaces subject to attrition show wear. Brown stain is frequently a disfiguring feature.
4.0	**Severe** - Includes teeth formerly classified as "moderately severe and severe." All enamel surfaces are affected and hypoplasia is so marked that the general form of the tooth may be affected. The major diagnostic sign of this classification is discrete or confluent pitting. Brown stains are widespread and teeth often present a corroded-like appearance.

SKELETAL FLUOROSIS[2, 10]

Pre-skeletal stage: The duration of this stagemay vary with the amount of fluoride daily ingested. It ranges from 10 to 30 years or even longer in endemic areas and from 10 to 15 years or longer in cases of industrial fluorosis. It is usually free of any signs or symptoms in its early stages in endemic regions. The persons concerned may occasionally complain of pains in the small joints of the limbs and back, which are often mistaken for rheumatoid arthritis or ankylosing spondylitis. However, various reports from Europe and America suggest that there would be symptoms corresponding to gastrointestinal, musculoskeletal, respiratory and visceral systems during this stage. The majority of these visceral symptoms may be due to allergy to fluoride in susceptible individuals or the effect of fluoride on the various target organs and these are nonspecific.

Skeletal fluorosis: Early in the development of fluorotic changes in the skeleton the patients often complain of a vague discomfort and paraesthesia in the limbs and the trunk. Pain and stiffness in the back appear next, especially in the lumbar region followed by dorsal and cervical spines. Restriction of the spine movements is the earliest clinical sign of fluorosis. The involvement of the ribs gradually reduces the movement of the chest during breathing, which finally becomes mainly abdominal. When that happens the chest assumes a barrel shape, with the increasing immobilization of the joints due to contractures, flexion deformities may develop at hips, knees and other joints which make the patient bedridden. Bony exostosis may also appear over the limb bones especially around the knee, the elbow and on the surface of tibia and ulna. Despite the fact that the entire bone structure has become affected the mental faculties remain unimpaired till the last stage is reached.

The stage at which skeletal fluorosis becomes crippling usually occurs between 30 and 50 years of age in the endemic regions. Newcomers to a hyper endemic region may sometimes develop symptoms of skeletal involvement within 4 years of their arrival. Men suffer more than women from severe affects of the disease presumably because their work is usually more strenuous than that of women.

Neurological manifestation of fluorosis

The neurological squeal in skeletal fluorosis manifesting usually as radiculo- myelopathy arise principally because of the mechanical compression of the spinal cord and nerve roots brought about by osteophytosis and sclerosis of the vertebral column. However, it is only in later stages owing to pressure on the radicular vessels in the intervertebral foramina that vascular complications may supervene, but the neural toxicity attributable to fluorides is yet to be established. Neurological complications arise at a late stage of the disease in about a tenth of the total number of skeletal fluorosis cases. The largest number of cases with neurological manifestations was reported from two endemic belts of Punjab, Harayana, Rajasthan and adjacent Uttar Pradesh in northern India and from Andhra Pradesh in southern India, but there have been few reports of fluorosis with neurological complications from countries other than India.

Cranial lesions: Nerve inthe skull is not much affected in fluorosis and basal cranial nerve foraminae are not usually encroached upon except at advanced stages of the disease. Of the cranial nerves, the most frequently affected in a quarter of the cases investigated has been the eighth nerve. In all such cases calvarial changes caused by fluorosis are discernible. A progressive high frequency perceptive deafness is observed, moreover the bone conduction is affected more than air conduction. Nevertheless total deafness rarely occurs. It is perhaps the compression of the nerve in the sclerosed and narrowed auditory canal that accounts for the deafness in fluorosis.

Peripheral neuropathies: Exostosis, which mainly develops around the knee, elbow and ankle, may press upon the median, ulnar or lateral popliteal nerves. Pain and paraesthesia followed by weakness in the limbs may be caused by such bony growth.

Acute toxicity

Acute oral exposure to fluoride may produce effects including nausea, vomiting, abdominal pain, diarrhoea, fatigue, drowsiness, coma, convulsions, cardiac arrest and death. Severe tissue damage, respiratory effects, cardiac arrest and deaths have been noted individuals exposed accidentally to hydrofluoric acid through dermal contact. The lethal dose of sodium fluoride to the average adult has been estimated to be between 5 and 10 g (32–64 mg fluoride/kg body weight), an acute dose of 5 mg fluoride/kg body weight has been considered to be the maximum that might lead to adverse health effects. Death due to acute fluoride poisoning resulting from improperly fluoridated drinking-water, the individual was estimated to have ingested approximately 17.9 mg fluoride/kg body weight prior to death. The toxicity of fluoride is dependent upon the type or species of the compound ingested. Generally, the more soluble salts of inorganic fluorides (e.g. sodium fluoride) are more toxic that those that are either weakly soluble or insoluble (e.g. calcium fluoride). [3]

Gastrointestinal effects produced following the acute ingestion of toxic amounts of fluoride likely arise from the corrosive action of hydrofluoric acid, which is produced within the acidic environment of the stomach. Damage to the gastric mucosa (e.g. hemorrhage, loss of epithelium) has also been observed in human volunteers administered acidulated phosphate fluoride gels or sodium fluoride solutions. Some individuals may be unusually hypersensitive to stannous fluoride and manifested by ulcerations in the oral cavity after topical treatment.

Cardiac arrest following accidental exposure to high levels of fluoride has been attributed to the development of hypocalcaemia or hyperkalaemia. The acute effects of fluoride upon the central nervous system may be due to fluoride induced hypocalcaemia and the inhibition of cellular enzymes.

Respiratory effects (e.g. hemorrhage, pulmonary oedema, tracheobronchitis and shortness of breath) have been observed in individuals following inhalation of hydrogen fluoride.

FLUORIDE IN INDIA

FLUORIDE IN INDIA [5, 8, 14, 15]

In India 62 million people including 6 million children are affected with fluoride related health diseases. Excess fluoride in ground water is reported from 17 States which include Andhra Pradesh, Rajasthan, Karnataka, Maharashtra, Tamil Nadu, Madhya Pradesh, Uttar Pradesh, Jammu and Kashmir, Punjab, Kerala, Orissa, Bihar, and Gujarat. Of which Uttar Pradesh, Rajasthan, Gujarat, Andhra Pradesh, and Tamil Nadu are severely affected. As per water technology mission 25 million people residing in 8700 villages in India are affected by fluorosis. Concentration level of fluoride in India reported from groundwater varies from 0.5 to 50 mg/l. Fluoride concentration has significant positive relationship with pH and negative relationship with hardness and magnesium.

Districts known to be endemic for fluoride in various States of India [16]

States	Districts	Range of fluoride concentration mg/liter
Assam	Karbianglong and Nagaon	**0.2–18.1**
Andhra Pradesh	All districts except Aliabad, Nizamabad, West Godhavari, Visakhapatnam, Vijzianagaram and Srikakulam	**0.11–20.0**
Bihar	Palamu, Daltonganj, Gridh, Gaya, Rohtas, Gopalganj, Paschim and Champaran	**0.6–8.0**
Delhi	Palamu, Daltonganj, Gridh, Gaya, Rohtas, Gopalganj, Paschim and Champaran	**0.4–10.0**
Gujarat	All districts except Dang	**1.58–31.0**
Harayana	Faridabad, Karnal, Sonipat, Jind, Gurgaon, Rewari, Mohindergarh, Rohtak, Kurukshetra and Kaithal	**1.58–31.0**
Jammu and Kashmir	Doda	**0.05–4.21**
Karnataka	Dharwad, Gadag, Bellary, Belgam, Raichur, Bijapur, Gulbarga, Chitradurga, Tumkur, ChikmagaluManya, Bangalore and Mysore	**0.2–18.0**
Kerala	Palghat, Allepy, Vamanapuram and Alappuzha	**0.2–2.5**
Maharashtra	Chandrapur, Bhandara, Nagpur, Jalgaon, Bulduna, Amravati, Akola, Yavatmal, Nanded and Sholapur	**0.11–10.2**
Madhya Pradesh	Shivpuri, Jabua, Mandla, Dindori, Chhindwara, Dhar, Vidhisha, Seoni, Sehore, Raisen and Bhopal	**0.08–4.2**
Orrissa	Phulbani, Koraput and Dhenkanal	**0.6–5.7**
Punjab	Mansa, Faridcot, Bhatinda, Muktsar, Moga, Sangrur, Ferozpur, Ludhiana, Amritsar, Patila, Ropar, Jallandhar and Fatehgarh sahib	**0.44–6.0**
Rajasthan	All the 32 districts	**0.2–37.0**
Tamilnadu	Salem, Periyar, Dharampuri, Coimbatore, Tiruchirapalli, Vellore, Madurai and Virudunagar	**1.5–5.0**
Uttar Pradesh	Unnao, Agra, Meerut, Mathura, Aligarh, Raibareli and Allahabad	**0.12–8.9**
West Bengal	**Birbhum, Bhardaman and Bankura**	**1.5–13.0**

DEFLUORIDATION PROCESSES

Defluoridation is removal of excess fluoride from water, several methods have been suggested for removing excessive fluoride in water.

The de-fluoridation methods are divided into three basic types depending upon the mode of action and they are:[1, 9]

A. Based on some kind of chemical reaction with fluoride
B. Based on adsorption process
C. Based on ion-exchange process

A) Based on chemical reaction [17]

Nalgonda technique[1]

The Nalgonda technique (named after the village in India where the method was pioneered) employs flocculation principle. Nalgonda technique is a combination of several unit operations and the process involves rapid mixing, chemical interaction, flocculation, sedimentation, filtration, disinfection and sludge concentration to recover waters and aluminium salts. Alum (hydrated aluminium salts) a coagulant commonly used for water treatment is used to flocculate fluoride ions in the water. Since the process is best carried out under alkaline conditions lime is added. For the disinfection purpose bleaching powder is added after through stirring the chemical elements coagulate into floccus and settle down in the bottom.

The reaction occurs through the following equations:-

$2\ Al2\ (SO4)3.\ 18H2\ O + NaF + 9Na2CO3 \rightarrow [5Al\ (OH)\ 3.Al\ (OH)\ 2F] + 9Na2SO4+NaHCO3 + 8\ CO2 + 45\ H2O$

$3Al2\ (SO4)3.\ 18H2\ O + NaF + 17NaHCO3 \rightarrow [5Al\ (OH)\ 3.Al\ (OH)\ 2F] + 9Na2SO4+\ 17\ CO2 + 18\ H2O$

- Economical-annual cost of defluoridation (1991 basis) of water at 40 lpcd works out to Rs. 20/- for domestic treatment and Rs. 85/- for community treatment using fill and draw system based on 5000 population for water with 5 mg/l and 400 mg/l alkalinity which requires 600 mg/l alum dose.
- Provides defluoridated water of uniform acceptable quality.

Lime

It has been observed that while giving lime treatment to waters containing magnesium salts in sufficient quantity fluoride are absorbed on magnesium hydroxide floccus and it results in fluoride removal.

B) Based on adsorption process [2, 17]

These include several bone formulations synthetic Tri-calcium phosphate and Hydroxy-apatite and a variety of adsorbent materials. Some of them are provided below.

Bone Charcoal

The bone is processed by burning in air and palavering it to fine powders. The fluoride removal capacity of the product is 1000 mg/l.

Processed bone

Bone contains calcium phosphate and has a great affinity for fluoride. The bone is degreased, dried and powdered. The powder can be used as a contact bed for removal of fluoride in water. The exhausted bed is regenerated with sodium hydroxide solution.

Tricalcium phosphate

Tricalcium phosphates natural or prepared synthetically by reacting milk of lime and phosphoric acid have been used for defluoridation.

Activated Carbons

Activated Carbons prepared from paddy husk have a high defluoridation capacity. Similarly activated carbon prepared from cotton waste, coffee waste and coconut waste were tried for defluoridation but all these materials are academic interest only.

Activated Magnesia

Activated magnesia is a good adsorbent among several substances and activated magnesia has better fluoride removal capacities at high temperature.

Tamarind Gel

The concentration of fluoride from solution of sodium fluoride of 10 mg/l could be brought down to 2 mg/l by the addition of tamarind gel alone and to 0.05 mg/l by the addition of small quantity of chloride with the tamarind gel.

Serpentine

It is the name which applies to the material containing one or both the minerals Chrysolite and Antigorite. The medium could not be regenerated and had to be discarded after the use. The yellow variety was found to have better fluoride removal capacity than the green mineral.

Activated alumina

Defluoridation of water by activated alumina is the method of choice in developed countries its affinity for fluoride is very high. It is a porous material with the surface comprised largely of active sites. It is prepared by dehydration of Al (OH) 3 in the temperature of 300 – 600 ^{0}C. Activated alumina is regarded as an excellent material for fluoride removal. However, pH and alkalinity are the factors which affect the adsorption capacity. The exhausted material can be regenerated by washing with alkali followed by acid and finally with distilled water.

Plant materials

The plant materials such as barks of *Moringaolifera* and *Emblicaofficinalis*, the roots of *Vetiveriazizanoides* and the leaves of *Cyanodontactylon* were found to be good defluoridation agents.

Burnt clay

Burnt clay obtained from brick Chula prepared by burning a mixture of 70 % clay and 30% red soil was observed to possess commendable defluoridation property. Clay with particle size 500 microns exhibited maximum defluoridation efficiency. At low pH range the

defluoridation efficiency was more compared to that at high pH range.

C) Based on Ion exchange mechanism [9, 17]

Anion exchange resins

These are found to remove fluorides either by hydroxyl cycle or chloride cycle along with other anions. Polystyrene anion exchange resin and basic quaternary ammonium type are used to remove fluoride along with other anions. Poly anion exchange resin, Tulsion A - 27, Deacodite FF (IP), Lawatit MIH – 59 and Amberlite IRA 400 are the few examples.

Cation exchange resin

Cation exchange resins impregnable with alum solution have been found to act as defluoridating agents, "Avaram bark" based Cation exchange resin works effectively in removing fluoride from water and also Defluoron-1 and 2, Carnion, Wasoresin -14 and polystyrene .

DOMESTIC DEFLUORIDATIONMETHODS

ACTIVATED ALUMINA

Application of domestic defluoridation plant based on activated alumina has been launched by UNICEF in rural India. The unit basically consists of two chambers the upper chamber is fitted with a simple flow control device (removable circular ring) at the bottom. The average flow is 10 litres/ hour the main component of this unit is a PVC casket containing 3 Kg of activated alumina (AA) giving a bed depth of 17 cm. A perforated plate of either stainless steel or tin metal is placed on the top of AA bed to facilitate uniform distribution of raw water. The top chamber is covered with a lid.

Household level AA filter

Lower chamber is used for collection of treated water it is fitted with a tap to draw the treated water and about 10 liters of water can be collected in 1 hour. Exhausted AA can be regenerated by dip-regeneration method. In this method, the casket containing exhausted AA is placed in a plastic bucket containing 8 litres of 1% NaOH for 4 hours. The casket is then transferred to a bucket containing water and is rinsed by occasional lifting. The casket is then placed in a plastic bucket containing 8 litres of 0.20 % H_2SO_4 for 4 hours. Subsequent wash with raw water till the pH raises above 7.0 makes the AA bed ready for the next defluoridation cycle.

BRICK PIECES COLUMN

Brick pieces of 15 to 20 mm sizes packed in a 225 mm diameter PVC pipe for a height of 925 mm and a 20 mm diameter PVC pipe with a funnel at top is placed inside to pour fluoride rich water. The raw water is allowed to enter unit at bottom and moves upward.

The raw water is allowed to enter the unit at bottom and moves upward. This unit has a capacity of 16 litres and a household for drinking and cooking could use this quantity. The cost of filter unit is about Rs. 600, aluminium oxide present in the soil used for brick manufacture. During burning operation in the kiln, it gets activated. Replacement of filter media is required once in three months if fluoride content in raw water is 2.50 mg/l.

MUD POTS

Collection and storing of water in mud pots is an ancient method. Red soil and clay are used to prepare the mud pots.

Here also the burning (heat treatment) is done as in the case of brick production, hence the mud pot also will act as an adsorbent media. The fluoride removal capacity will vary with respect to the alumina content present in the soils used for pot production. So, people can be educated to keep the benefit of mud pot a low cost vessel. The cost of a pot is around Rs 15.

PRECIPITATION [1, 17]

Precipitation methods are based on the addition of chemicals (coagulants and coagulant aids) and the subsequent formation of insoluble fluoride precipitates. Fluoride removal is accomplished with solids separation from liquid. Various chemicals involved in the precipitation process are lime, alum, alum and lime (Nalgonda Technique), poly aluminium chloride, poly aluminium hydroxyl-sulphate and polyelectrolyte.

CONCLUSION

The problem of Fluorosis is world wide affecting many countries and in India especially Rajasthan which is the largest state of India. The cause of problem is high fluoride content of water more than 1 ppm and chronic long term ingestion as stated by the different studies around the world and World Health Organization (WHO). Fluorosis does not only affect oral cavity but has more severe and generalised effect on the body resulting in hard tissue and soft tissue irreversible damage in form of dental fluorosis, increased chances of caries and periodontal diseases, skeletal and non-skeletal fluorosis and acute toxicity which is lethal .

In Rajasthan all the 32 Districts are affected with high fluoride levels in water. 11909 villages and 11388 other habitations are having fluoride level in their 8-*87\][poiuground water in concentration of over 1.5 mg/l. Similarly 4250 villages and 3449 habitations are having fluoride concentration more than 3.0 mg/l, thus the fluoride affected villages and habitations constitutes of nearly 24.79% of the total number of villages and habitations.

Analyzing the epidemiological studies and data of Rajasthan from the year 1991, 1991-93, 1997-98 and the last and recent in 2001, it is seen that the number of fluoride effected villages/habitations are found to be increasing from "1991 to 2001".

The mostly badly affected areas in the state are the rural areas where lack of education, poverty, limited sources of water and cost of defluoridation are important factors in these areas.

PHED, UNICEF, and Non-Governmental Organizations (NGO's) are working in various fields. And from being medicinal professionals it is our duty to work in preventing this endemic disease by educating our patients and people in affected areas.

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