Research Journal of Recent Sciences ISSN 2277-2502
Vol. **7(4),** 36-47, April **(2018)** *Res. J. Recent Sci.*

Review Paper

Fluoride contamination in groundwater, soil and cultivated foodstuffs of India and its associated health risks: A review

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Available online at: www.isca.in, www.isca.me
Received 1st August 2017, revised 21st February 2018, accepted 3rd March 2018

Abstract

Around 200 million people of 29 countries including India are severely affected due to fluoride pollution. Ingestion of fluoride beyond the World Health Organization recommended maximum permissible level (1.5 mg/l) is associated with dental and skeletal fluorosis and other toxic responses while lacking of fluoride intake is associated with dental caries. Minerals like apatite, fluorite/ fluorspar, topaz and mica get weathered naturally and provide fluoride to soils. In site-specific cases some industries (mainly phosphorous fertilizer plants; steel, aluminum, zinc, smelting industries; glass and ceramic industries, etc) are also responsible for fluoride contamination of soils. Irrigation with fluoride-contaminated water transfers fluoride to crops, vegetables and fruits. This bioaccumulation contributes further fluoride to the food chain in addendum to the drinking water route, and thus it is causing larger risk to the already fluoride-contamination affected population. Moreover, this new avenue of fluoride highly endangers the most susceptible infants and children towards dental fluorosis. The 'Nalgonda' and 'activated alumina' processes are the most commonly used defluoridation techniques of drinking water. But, a suitable, efficient, user-friendly and cost-effective technique for defluoridation is yet to be developed. Equal emphases are to be given on creation of awareness in people regarding fluorosis and restriction of usage of fluoride-contaminated irrigation water.

Keywords: Fluorosis, fluoride pollution, fluoride bioaccumulation, fluoride in food chain.

Introduction

Moderate to high fluoride pollution in groundwater is reported as one of the major environmental issues of Algeria, Brazil, Canada, China, Ethiopia, Ghana, India, Iran, Italy, Japan, Jordan, Kenya, Korea, Malawi, Mexico, Norway, Pakistan, Sri Lanka, Thailand, Turkey, and the US[1,2] threatening an estimated 200 million people[3]. Fluoride can be found in different environmental components originated from mineral sources, atmospheric sources, and geothermal sources. After its dissolution from weathered rock it becomes mobile, and it enters soil, groundwater, cultivated crops, fruits and vegetables depending on the prevailing geological agents. The natural dissolution of fluorite, apatite, and topaz from bedrocks leads to rich content of fluoride in groundwater[4]. Moreover, different anthropogenic influences like usage of phosphate fertilizers and insecticides, discharge of sewage and sludge, overuses of groundwater in agriculture, etc., increase groundwater contamination with fluoride[5,6]. Fluoride in groundwater is mostly of geological origin. Intake of fluoride in excess amounts, mostly via drinking water (other sources are food, industrial gas and excessive use of toothpaste), causes 'fluorosis' effecting the teeth and bones[7]. Mild fluoride intake induces dental effects while long-term exposure to high concentration of fluoride results severe skeletal and other adverse illness. The associated human health risks from fluoride can be broadly categorized as: dental effects, skeletal effects, reproductive effects, developmental effects, renal effects, neurological effects, endocrine effects, and carcinogenic effects. Interestingly consumption of fluoride at modest level prevents dental caries. Thus, the most vital step to control fluorosis is to ensure permissible fluoride concentration in drinking water. The World Health Organization (WHO) has recommended 1.5 mg/l as the maximum permissible limit for fluoride in drinking water[1].

Endemic fluorosis has been reported from 20 (out of total 29) Indian states, spreading over 65% of the total rural habitations of the country[8]. More than 65 million Indians including 6 million children are at risk due to the presence of fluoride beyond the desirable 1.5 mg/l level in drinking water[8,9]. High concentration of fluoride in groundwater has been mainly reported from different regions of Assam, Andhra Pradesh, Bihar, Chhattisgarh, Gujarat, Karnataka, Madhya Pradesh, Maharashtra, Odisha, Rajasthan, Telangana, Uttar Pradesh and West Bengal[3,4,10-20]. The present review focuses on the assimilation of relevant data and information on past and present status of fluoride contamination in India, mobility of fluoride in ecosystems, and the related human health issues.

Fluoride contamination in water - source, essentiality vs. toxicity

Fluoride is present in almost all groundwater, but the presence of leachable fraction of the total fluoride depends widely on rock varities[21]. The types of minerals, residence time, and climate primarily control dissolution of fluoride from bedrock[22]. The pH, hardness, ionic strength, and other water quality parameters also have important roles in site-specific cases influencing mineral solubility, complexation, and sorption/exchange reactions[23]. Generally fluoride-bearing minerals are sparingly soluble with the exception of villiaumite[24]. Microbial presence accelerates dissolution of fluorapatite by partitioning phosphorus from the solid phase[25]. Similar is the case of fast fluorite dissolution in sodium-bicarbonate waters[23] while the release of fluoride from clay minerals depends strongly on pH[24]. The solubility of fluorite was mostly found to control the presence of fluoride in groundwater[26]. Hence there is no linear correlation between the groundwater fluoride concentration and the fluoride-bearing mineral percentages. The role of climate on fluoride contaminations in groundwater was found to be highly dependent on the recharge of groundwater through rainfall and subsequent groundwater flow[26,27]. Humid tropical regions with high rainfall have lower probability for groundwater fluoride contaminations due to leaching and subsequent dilution of soluble ions including fluorides. On the opposite side arid environments normally have high fluoride contents due to low to modest recharge of groundwater. Low rainfall leads to enhanced opportunity for water-mineral reactions which result into high salinity contents in the arid environment, which further facilitate mineral dissolution[28]. Temperature also regulates the solubility of fluorine-bearing minerals. Both high pH and low Ca and Mg contents in groundwater mostly influence fast leaching and subsequent high concentrations of fluoride in groundwater[29].

Drinking water has been categorized as the most influencing factor for entry of fluoride in the human food chain[30]. According to Wood[31] fluoride consumption is good for human health when present at 0.5–1 mg/l in water. This permitted range of fluoride helps in the production and maintenance of healthy teeth and bones. Later, the United States Environment Protection Agency (USEPA) revised the recommended range of fluoride as 0.7–1.2 mg/l[32]. Thus, any water sample with fluoride contents < 0.7 or > 1.5 mg/l should not to be utilized as a source of potable water. Fluoride may be added at low levels during treatment of drinking water in places which are devoid of adequate quantity of fluoride to aid in dental and skeletal health of the residing population, mainly children. Again, high fluoride consumption causes chronic dental and skeletal fluorosis, which is manifested by mottling of teeth in mild cases, softening of bones and neurological damages in severe cases[33-35]. Consumption of fluoride > 2 mg/l was reported to cause dense and brittle bone, and also dental problems[11]. However, the United States Public Health Service (USPHS) has advocated following a range of allowable concentrations for fluoride in drinking water for different regions depending on prevailing climatic conditions[36]. The USPHS argued that the amount of water intake and accordingly the amount of fluoride ingested being influenced primarily by the regional air temperature.

Fluoride contamination in soil–sources and significances

Fluoride is the 13th most abundant element of the earth's crust representing ~0.3 g/kg of the earth's crust. It is mainly present as NaF or HF, which can be found in minerals fluorspar, fluorapatite, topaz and cryolite. In most of the rocks fluorine is present normally in the range of 100–1300 mg/kg[37], but in soils fluoride contents generally vary in the range 20–500 mg/kg[26]. Exceptionally higher presence of fluoride (> 1000 mg/kg) was observed in soils derived from rocks with high fluorine contents[38] or in cultivated soils affected by anthropogenic inputs, like application of phosphate fertilizers[39], intrusion of sewage sludge, and industrial contamination[38]. Thus, a soil may be called as fluoride contaminated when fluoride content exceeds ~500 mg/kg. According to Bhat et al.[40] fluoride contamination of soil is primarily due to the application of phosphorous fertilizers containing $< 1\%$ to $> 1.5\%$ fluorine. The sorption capacity of soil (which varies with soil pH), types of sorbents present in soil, and soil salinity largely regulate the mobility of fluoride in soil[38,41]. In comparison to sandy soils, presence of higher clay and oxyhydroxide contents in fine-grained soils helps to retain more fluoride. Edmunds and Smedley[26] suggested that evapotranspiration increases the salinity of soil solutions which leads to higher content of fluoride ultimately reaching groundwater. The primary anthropogenic sources of fluoride pollution in soil are different chemical industries (like phosphorous fertilizer plants, steel, aluminum, zinc, smelting industries, etc), glass and ceramic industries, and power plants. The soil of the areas in vicinity of the above said industrial units contain medium to high content of fluoride[40].

Irrigation and application of fertilizers normally increase the sodicity of soils which ultimately result into high fluoride content in soils[16]. Concentration of fluoride in deep tube wells was detected to be significantly correlated with agricultural field soils[19]. In a fluoride affected area if fluoride-contaminated groundwater is used for irrigation then there is prominent possibility of fluoride bioaccumulation into irrigated crops. This would further endanger human health due to augmented fluoride consumption through cultivated crops, fruits and vegetables in addition to drinking water pathway. Soils polluted with fluoride negatively impact health via direct soil contact, through inhalation of vaporized soil contaminants, via intake of contaminated foods, and through intake of contaminated groundwater[40,42,43]. Fluorine can penetrate deep soil layers of $<$ 80 cm, and when present in high concentrations it can oppress biological activities operating in soil medium[44].

Flow of fluoride from irrigation water to cultivated crops

Plants via xylematic flow transport fluoride to different organs, mainly the leaves[45]. Bioconcentration of fluoride in plants at various levels was shown by different researchers[46-49]. The bioaccumulation of fluoride had been established to cause chronic toxicity in grazing animals and humans[50]. The chronic toxicity may finally damage bones and lead to tooth wear. Leafy vegetables are susceptible to air borne fluoride and shows wide variations in the fluoride level cultivated in different areas. According to Gupta and Banerjee[51] the higher fluoride contents in leafy vegetables were due to the enhanced metabolism and/or photosynthesis rate in leafy shoots as compared to seeds/grains or other storage organs (tubers). In another study leafy vegetables (like radish, spinach, cabbage and cauliflower) were reported to bioconcentrate fluoride more preferably (BCF>1) indicating higher rate of photosynthesis in leafy vegetables associated with higher intake of irrigation water[52]. Cereals were usually found to accumulate < 1 mg/kg fluoride as fluoride mostly gets accumulated in the outer layer of the grain and in the embryo[53]. In many studies especially in areas adjacent to industries spinach has been reported to be unusually enriched in fluoride[49,54]. Tea is one of the most popular refreshment drinks of many countries of the world including India. But, at the same time tea had been established to be one of the most fluoride-enriched conventional beverages as ~67% of the total fluoride in leaves gets solubilised in the drink[55]. Fluoride-contaminated irrigation water (7.4−14 mg/l) had been shown to transfer fluoride (mg/kg) in the cultivated crops of Rajasthan[56] (spinach: 26, methi: 19, etc) (Gautam et al. 2010). Lower translocation of fluoride in edible parts was observed for the grain-yielding crop plant (mustard), tubers (potato), and fruiting vegetables (tomato, brinjal). Leafy vegetables like spinach, coriander leaves, and marsilea were shown to have higher translocation[51].

High fluoride levels produce different negative impacts on plants like inhibition of germination, malformations of ultrastructures, reduction of photosynthetic capacity, alteration in permeability of membranes, bringing down productivity as well as biomass, and production of other physiological and biochemical disorders[56]. Fluoride significantly affects some physiological processes which result into chlorosis, leaf necrosis, leaf tip burn, change in biochemical ratio of plant body, etc[57,58]. Fluoride was observed to produce toxic effects on chlorophyll pigment and on secondary metabolites like sugar, ascorbic acid, amino acids and proteins[57,59]. Table-1 summarizes concentrations of fluoride in different environmental components as well as in cultivated crops and vegetables.

Entry of fluoride into the human food chain and its consequences

Drinking of fluoride-contaminated water is proved to be the most influencing factor for entrance of fluoride in human beings. Moreover, the flow of fluoride from contaminated irrigation water to the cultivated crops may ultimately enter the human food chain through intake of rice, wheat, pulses, vegetables and fruits. The dose of fluoride exposure through drinking of fluoride-contaminated deep tube well water of the Bankura and Purulia districts (West Bengal) was evaluated by Samal et al.[19] for infants, children and adults. The results showed that for infants the doses of fluoride exposure were 0.02−0.53 mg/kg-day. The standard dose of fluoride exposure is 0.05 mg/kg-day. This is the Agency for Toxic Substances and Disease Registry (ATSDR) recommended the minimum risk level (MRL) value[60]. In children and adults of the studied area the doses of fluoride exposure were observed in the ranges 0.01–0.24 and 0.01–0.14 mg/kg-day, respectively. The estimated values in infants, children and adults were evaluated to be around 11, 5 and 3 times higher than the ATSDR's MRL value[19]. Bhattacharya et al.[61] on the basis of the obtained results predicted that infants and children of the studied area would be very much affected by dental fluorosis and thus suggested immediate mitigation of the situation.

In a similar study at Unnao district (Uttar Pradesh, India) the toxicological risk from fluoride exposure by different age groups through intake of cereal crops and vegetables was estimated[62]. The results showed that the cumulative estimated daily intake (EDI) of fluoride (mg/kg-day) in contaminated areas were 0.065–0.082 for 3−14 year children, 0.026–0.032 for 15−18 years and 0.031–0.039 for 19−70 years, respectively. Thus, the cumulative EDI in children (3–14 years) were found to be higher in comparison to the other age groups. As children are more prone to dental fluorosis, the obtained results were of deep concern. The same was further reinforced with the evaluated higher hazard index (HI) values for this age group (HI > 1). The authors also reported that the dose of fluoride exposure from rice and wheat in all the age groups was more (≥ 95%) than the dose of fluoride exposure due to the intake of locally grown vegetables.

The average daily intake of fluoride via all possible exposure pathways contributing to risk of fluorosis in infants and children was estimated by Erdal and Buchanan[63]. The authors considered intake of fluoride through drinking water, beverages, cow's milk, foods, and fluoride supplements, consumption of infant formula, inadvertent swallowing of toothpaste (by children only) and incidental ingestion of soil (by children only) as the possible exposure pathways. The results showed that drinking water and infant formula was the two major contributors (52% and 39%, respectively) towards the cumulative daily fluoride intake for infants. Similarly, for children toothpaste (57%), drinking water (22%), and food (9%) were found to be the major contributor for cumulative EDI. According to the literature for the reasonable maximum exposure (RME) scenario intake of fluoridated drinking water and consumption of infant formula for infants and incidental ingestion of toothpaste during brushing by children resulted the HI values >1. The findings raised concerns that a segment of the infants and children living in the US might be exposed to amounts of fluoride higher than required for prevention of dental caries. Additionally the children might increase their fluoride intake by using mouth rinses, gels, and specially flavored toothpastes. As tea is a good accumulator of fluoride children who consume tea frequently are hypothesized to have higher intake of fluoride[64].

Table 1: Fluoride levels in water, soil, crops and vegetables grown in fluoride-contaminated areas of the world.

Location	Fluoride in water (mg/l)	Fluoride in soil (mg/kg)	Fluoride in crops and vegetables (mg/kg)	Reference
India (Bankura and Purulia, West Bengal)	0.08−1.3	55–399	13−63	Bhattacharya et al.[61]
India (Purulia, West Bengal)	0.01−1.7	69−417	-	Bhattacharya[20]
India (Bankura and Purulia, West Bengal)	0.08−1.3	55–399	-	Samal et al.[19]
India (Udaipur, Rajasthan)	0.36–0.71	-	101−189	Bhat et al.[40]
India (Vadodara, Gujarat)	0.8–3.3	1.3−9.2	-	Parikh et al.[104]
India (West Bengal)	0.15−1.8	-	-	Datta et al.[105]
India (Newai Tehsil, Rajasthan)	0.3–9.8	50−180	8−98	Saini et al.[49]
India (Dausa, Rajasthan)	5.1−15	-	1.1−14	Yadav et al.[106]
India (Birbhum, West Bengal)	0.58−10	-	0.4–4.2	Pal et al.[52]
India (Birbhum, West Bengal)	3.2–3.8	-	-	Mondal et al.[107]
India (Unnao, Uttar Pradesh)	-	1−4	1.1−55	Jha et al.[62]
India (Birbhum, West Bengal)	-	-	4−27	Gupta and Banerjee[51]
Iran (Isfahan)	0.01−0.4	6.4−265	1−3.6	Chavoshi et al.[108]
Nigeria (Zaria)	0.08−0.19	0.09−0.14	0.01−0.06	Paul et al.[109]
China (Liaohe River Basin)	0.7–4.5	-	0.51−3	Zheng and Sun[110]
India (Nagaur, Rajasthan)	0.92−15	-	1.9−26	Gautam et al.[56]
India (North 24 Parganas, West Bengal)	0.01−1.2	-	-	Kundu and Mandal[59]
India (Nadia, West Bengal)	0.01−1.2	-	-	Kundu and Mandal[15]
India (Murshidabad, West Bengal)	0.02–1.2	-	-	Kundu and Mandal[111]
India (Birbhum district, West Bengal)	0.62–4.1	140–144	12–13	Gupta and Banerjee[112]
Iran	0.99–2.5	-	-	Dobaradaran et al.[113]
China	-	267–552	0.26–4.6	Lan et al.[114]
Pakistan (Lahore and Kasur, Punjab)	2.5–21	-	-	Farooqi et al.[115]
India (Birbhum, West Bengal)	0.01–2	-	-	Gupta et al.[116]
Indonesia	5.5−14	-	-	Heikens et al.[117]
Croatia	-	11−12	6−12	Kalinic et al.[47]
Poland	-	-	11−33	Kusa et al.[118]
Pakistan	8–14	-	-	Shah and Danishwar[119]
Brazil	0.1–2.3	-	-	Whitford et al.[72]
Japan	0.01−7.8	-	-	Ish and Suckling[120]

Health risk assessment to fluoride exposure via drinking water and staple food in Dayyer (Iran) were reported by Keshavarz et al.[65]. The authors found seasonal variation in the daily fluoride intake (mg/kg-day) from drinking water: 0.07–0.08 in spring to 0.09–0.1 in summer for adults and children, respectively. During summer and spring the total estimated oral intake of fluoride for children were 0.12 and 0.15 mg/kg-day, respectively, and the values for adults were 0.1 and 0.11 mg/kg-day, respectively. The results established that drinking water of the region was the major factor of entry of fluoride in the studied population. Potential health risk of fluorosis was concluded by estimating the HI values (>1). Greater concentration of mean fluoride (13–63 mg/kg dry weight) in rice, pulse and vegetable samples was reported to be 9.5–16% contributor to the cumulative estimated daily intake (EDI) of fluoride[61]. The authors also evaluated higher HI for children of their study area (3.2 and 8.7 for CTE and RME scenarios, respectively).

Impacts of fluoride on human health

The impact of fluoride on human health was first identified at the end of 19^{th} century. Significant fluoride content was reported in bones and teeth of human beings[26]. People living in some specific areas of the US were found to develop brown dental stains at the beginning of the 20^{th} century which was afterwards proved to have positive correlation with the fluoride contamination in drinking water of the effected regions[66]. Interestingly at the same time it was established that optimal intake of fluoride could prevent the developing of dental caries without any dental staining[67]. Later in 1940s municipal water treatment plants started to fluoridate drinking water optimally (0.7–1.2 mg/l) relating with the average air temperature of those region[68,69].

Absorption of fluoride in the body: Fluoride is electronegative in nature and it can form strong complexes with Al, B, Be, Fe(III), Si, Na, U and V, although these complexes are usually absent in natural water bodies[22]. The magnesium-fluoride complex is dominant in potable water[26]. Epidemiology suggested that the intake of fluorosilicates and the intake of NaF or fluoride from natural waters produce dissimilar biological impacts[70].

The absorption depends on various dietary factors e.g. amendment of Ca, Mg and Al salts in diet incorporates a fraction of fluoride into some compounds with low solubility which are removed from the body through excretory process. Likewise the addition of phosphates, sulphates and Mo to diet increase fluoride assimilation from the GI tract. HPO complex comprises of >90% of the total fluoride. Formation of this complex is favoured by the gastric acid within the stomach at pH 2[71]. After its absorption from the stomach as well as from the small intestine (by diffusion process) HPO complex dissociates within mucosa (which is less acidic) to release fluoride[72]. Almost 50% of the absorbed fluoride is quickly integrated in the development of bones and teeth, and the remaining fluoride is eliminated from body via urination[73]. Nearly all the fluoride accumulation in body is thus diagnosed in bones and teeth. The fluoride accumulation process in the skeleton is the most effective in children, and the rate of accumulation gets reduced with lifetime[72]. The process mostly continues up to the age 55[74]. Fluoride incorporated into hard tissues is recoverable, but is a very slow procedure (‘*osteoclastic resorption*’)[71].

Beneficial effects of fluoride: By the year 1930 it was established that controlled intake of fluoride could prevent development of dental caries and, also responsible for stronger bone growths[26,71,74]. The sharpest decline in dental caries was found for fluoride concentrations 0.7–1.2 mg/l, and negligible further gain was observed for presence of fluoride beyond the above range[71]. Medicinal research and epidemiology showed that bone mineralization could improve by intake of fluoride with suitable Ca and Vitamin D doses, but it could not essentially cut down the count of fractures. The fluoride content was observed to follow a ‘U’-shaped curve with the number of bone fractures, and the maximum potential was observed at ~1 mg/l[68]. Very high fluoride contamination of drinking water (≥ 4 mg/l) was noticed to increase fracture cases[75]. Thus, further clinical research and epidemiological studies are required to analyze the affectivity of fluoride in the treatment of osteoporosis, and also to maximize the benefits from fluoride with minimum risks[76].

Adverse effects of fluoride: The acute effects of fluoride toxicity from accidental overdoses or due to the ingestion of NaF and dental products are vomiting, hemoptysis, cramping of the arms and legs, bronchospasm, cardiac arrest, ventricular fibrillation, fixed and dilated pupils, hyperkalemia, hypocalcemia, and sometimes death[77,78]. The chronic effects due to long term fluoride exposure in human beings are genetic mutations, birth defects, hypersensitivity reactions, allergic illnesses, repetitive bone injury, Alzheimer’s disease, etc.

Dental effects of fluoride: The most characteristic feature of dental fluorosis is mottled enamel. With the development of the enamel mineralization gets increased and matrix proteins get removed. Fluoride exposure during this enamel development process disrupts enamel mineralization. This ultimately results in anomalously large gaps in its crystalline structure, excessive retention of enamel proteins, and increased porosity[79]. The appearance of white horizontal striations on the tooth surface or opaque patches of chalky white discolorations are the symptoms of dental fluorosis at mild level[74,80]. In case of severe dental fluorosis, the opaque patches become stained yellow to brown or even black, and eventually the increased tooth porosity leads to structural damages, such as pitting or chipping[74].

Skeletal effects of fluoride: Skeletal fluorosis is the increased bone mass and density, accompanied by a range of skeletal and joint symptoms. The threshold level of fluoride intake to produce skeletal fluorosis varies widely on quality of water, quantity of water consumption, and other dietary factors. The stages of development of skeletal fluorosis are well established but, the mechanisms are not well understood yet[26]. The symptoms at the beginning are pain and stiffness in the backbone, hip region, and joints, accompanied by increased bone density ('*osteosclerosis*'). The steady increase of stiffness makes the entire spine as one continuous column of bone ('*poker back*'). Calcification and ossification of various ligaments of the spine may happen with time. In ultimate stages skeletal fluorosis produces neurological defects, muscle wasting, paralysis, crippling deformities of the spine and major joints, and compression of the spinal cord.

Reproductive effects of fluoride: The reproductive effects of fluoride on humans are not well studied yet. Freni[81] studied the influence of higher fluoride contents in drinking water (> 3 mg/l) on the decrease in birth rates of the US population. The results of other studies suggested that high fluoride ingestion had negative reproductive effects on males. The effects include morphology and mobility of sperm or the levels of testosterone, follicle-stimulating hormones and inhibin B[82].

Developmental effects of fluoride: Some studies have shown that the fluoride concentrations in maternal blood plasma is positively correlated with the umbilical cord blood plasma, suggesting passive diffusion of fluoride from mother to fetus through the placenta[83,84]. Adverse developmental outcomes were found when very high doses of fluoride were applied in clinical experiments on animals; but in case of human beings the results were inconclusive[71]. Possibility of fluoride induced prevalence of Down's syndrome, especially for children born to mothers of age < 30 was shown by Whiting et al[85]. The occurrence of '*spina bifida occulta*' was reported to be exceptionally high in areas with high fluoride contamination[86].

Neurological effects of fluoride: The effects of dietary fluoride ingestion on the intelligence of children were reported[87,88]. The results showed that the children consuming higher quantity of fluoride (> 2 mg/l) scored badly on IQ tests than by the children who were exposed to lower levels of fluoride (< 1 mg/l). The IQ and the urinary fluoride contents of school going children in India were detected to be significantly inversely related[89]. Spittle[90] et al. concluded that the threshold fluoride dose for neurotoxicity response in children was in the range 2–4 mg/l. Fluoride may induce different biochemical changes in proteins and enzymes which may interfere with the normal brain functioning and cause impaired cognition and memory[91].

Gastrointestinal effects of fluoride: Variety of gastrointestinal effects like nausea, vomiting, diarrhea and abdominal pain are induced by acute fluoride toxicity. The symptoms are common in fluoride-contaminated areas with poor nutrition by the population[92,93]. Clinical studies on animals revealed that fluoride is able to stimulate stomach acid secretion, reduce blood flow away from the stomach lining, and can kill gastrointestinal tract epithelium cells[71]. According to Doull et al.[71] when fluoride content in water is < 4 mg/l, the gastrointestinal hypersensitive people (< 1% of the population) would show gastrointestinal indications.

Renal effects of fluoride: Excess fluoride from different organs is excreted through the renal system[77], and thus it is at higher risk of fluoride toxicity than most of the soft tissues of the body. The chronic ingestion of fluoride was shown to have non-carcinogenic effects on the kidneys[71]. Juuti and Heinonen[94] observed that residents living in areas with groundwater fluoride level > 1.5 mg/l experienced higher hospitalization for '*urolithiasis*'. Singh et al.[95] by studying a fluoride-contaminated area of India (fluoride concentrations in drinking water: 3.5–4.9 mg/l) reported that patients with clear symptons of skeletal fluorosis were 4.6 times more likely to develop kidney stones.

Carcinogenic effects of fluoride: Linkage between fluoride content in drinking water and the prevalence of uterine and colon cancer were reported by some researchers[96,97]. Increased '*osteosarcoma*' (bone cancer) and '*osteoma*' (noncancerous bone tumors) in animals were also shown[71]. Evidence of association between fluoride exposure and the incidence of kidney and bladder cancer was reported by some studies[97,98]. Carcinogenic impacts of fluoride is to be evaluated separately for each type of cancer as epidemiology for the identification of the carcinogenic potential of chronic fluoride toxicity is dependent on many cancer as well as potential causal factors[99-101].

Management and mitigations of fluoride contamination

The fluoride contamination has spread in many parts of India causing both health and social problems. Thus, mitigation and management of this burning issue are urgently needed. Prevention of fluorosis is the only appropriate measure as it is irreversible. It can be done by using various intervention steps. To prevent or minimize toxic responses of fluoride provision has to be made to supply alternate safe drinking water, to remove fluoride beyond the maximum permissible limit from drinking water, and to improve the overall nutritional status of people at risk. The simplest remedial option for fluoride poisoning is the use of safe surface water sources or consumption of groundwater with fluoride level < 1.5 mg/l. The other effective measures of minimizing fluoride are defluoridation of water via flocculation and adsorption processes, creation of awareness in people about fluorosis, providing health education, etc. The presently used defluoridation processes have their own merits and demerits in terms of their use, effectiveness and costing. Some defluoridation techniques are discussed below:

Nalgonda process: It involves direct addition of lime in water to maintain the pH and also direct addition of a known quantity of alum in water depending on the fluoride content of raw water. Therefore, it is difficult to standardize the alum dose as it is different for each source of water. The cumbersome technique is unsuitable for operation by laymen, thus has limited rural applications. Further, this technique can only be utilized for water sources with fluoride content < 10 mg/l. High content of free residual aluminum (2–7 mg/l) which is beyond the IS 10500 recommended maximum permissible limit[35] (0.2 mg/l) is usually present in output water. Moreover, when this treated water is boiled in aluminum utensils it further aggravates the free aluminum content in water. The taste of the treated water is also generally not acceptable. As aluminum is a neurotoxin and the presence of aluminum as low as 0.08 mg/l in drinking water was found to cause Alzheimer's disease.

Activated alumina process: Reduction of fluoride concentration from ~10 to < 1 mg/l is easily possible using activated alumina filters. The removal of fluoride is proportional to the contact time of water with the alumina filter media. Thus, more the contact time more efficient will be the fluoride removal in the treated water. Efficiency of this process was also observed to increase with acidic water (low pH) and water having low temperature. Ideal pH of this technique is 5.5, which allows up to 95% fluoride removal. But, the reactivation of filter material (treatment of filter bed by acid and alkali) is cumbersome, and it can be done only by trained personnel, who are generally unavailable in most of the fluoride affected villages of India. This procedure also results in moderately high (0.1–0.3 mg/l) residual aluminum in output water. Overall it is an expensive defluoridation process.

Other defluoridation techniques: Venkatramanan et al.[102] developed a technique for separating fluoride by using paddy husk carbon impregnated with alum. The process involved autoclaving paddy husk carbon by 1% NaOH and soaking overnight in 1% alum solution. Fluoride can be reduced from water by precipitation using $CaSO_4$ or $Fe_2(SO_4)_3$ solution. But elimination of excess Ca^{2+} is difficult and if iron is taken regularly, it will have adverse effect to body because it does not have any metabolic exit. Sarkar and Banerjee[103] had developed a simple, low cost and techno-feasible method for removal of fluoride from drinking water. The locally available natural material '*laterite*' (or red soil) was used as the adsorbent. Processes like electrodialysis, reverse osmosis, etc. require special equipments, especially trained persons to operate, and need constant maintenance. These processes are very expensive also.

A suitable, efficient, user-friendly and cost-effective technique for defluoridation is yet to be developed. The process should be such that it would be suitable for any fluoride concentration, pH, TDS, alkalinity and temperature of input water, and can be handled by rural population.

Conclusion

The past and present scenario of fluoride pollution in water, soil, crops, and vegetables are reviewed in this article. Fluoride exposure to humans through intake of drinking water and other dietary routes is one of the alarming global problems. Irrespective of some beneficial effects fluoride is responsible for dental and skeletal fluorosis, and other adverse reproductive, developmental, neurological, renal, carcinogenic, endocrine and gastrointestinal effects. For improving the status of fluoride contamination on long-term basis an efficient fluorosis risk management plan should involve identification of all potential exposure pathways, mapping of affected zones, supplying of defluoridated drinking water, restriction in usage of contaminated water for irrigation, health education, improvement of the nutritional status of populations at risk, encouragement for utilizing surface water, harvesting of rainwater, etc. Provision for adequate nutrition of people comprising of calcium, iodized salt, vitamin C and antioxidants in the fluoride-contaminated areas should be arranged by the government to protect the residents from early and severe fluorosis, which is an irreversible and untreatable disease.

Acknowledgement

Piyal Bhattacharya is thankful to the Science and Engineering Research Board (DST-SERB), Government of India for funding this investigation (Sanction Number: YSS/2015/000454). The authors declare that they have no actual or potential competing financial interests.

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