



Fluoride distribution and contamination in the water, soil and plants continuum and its remedial technologies, an Indian perspective— a review[☆]

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ABSTRACT

Fluorine is an essential element required in trace amounts but gets toxic for human beings at levels more than $1.5 \text{ mg F}^- \text{ L}^{-1}$ primarily through drinking contaminated water. It is the 13th most abundant element and constitutes about 0.06–0.09% in the earth crust. It is electronegative in aqueous medium forming fluoride ion (F^-). Fluoride contamination in the environment occurs mostly due to anthropogenic and geogenic sources. Fluoride is widely distributed in all components of environment, air ($0.1\text{--}0.6 \mu\text{g L}^{-1}$) soils ($150\text{--}400 \text{ mg Kg}^{-1}$) rocks ($100\text{--}2000 \text{ mg Kg}^{-1}$), plant ($0.01\text{--}42 \text{ mg Kg}^{-1}$) and water ($1.0\text{--}38.5 \text{ mg L}^{-1}$). Human beings and animals are being exposed to F^- primarily from water ($0.2\text{--}42.0 \text{ mg L}^{-1}$) and plants ($0.77\text{--}29.5 \mu\text{g g}^{-1}$). Fluorosis, a health hazard due to F^- is a major problem in many countries across the world affecting about 200 million people globally. In India, > 62 million people in twenty states are facing problem due to F^- . The most affected states are Rajasthan (7670 habitations), Telangana (1,174 habitations) and Karnataka (1122 habitations). To mitigate this problem, there is an urgent need to understand the current status and brief knowledge of F^- geochemistry. The objective of this review is to highlight different sources of F^- that contaminate different environmental matrices including plants, the extent of contamination level in India, uptake, translocation and toxicity mechanism in plants. The review also highlights currently available mitigation methods or technologies through physio-chemical and biological means.

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1. Introduction

Fluorine, an element of halogen group has earned a dubious distinction as “double-edged sword” due to its narrow range of safe limit. Fluoride (F^-) is beneficial for human beings in small quantities ($32\text{--}64 \text{ mg Kg}^{-1} \text{ F}^- \text{ Kg}^{-1}$ body weight), but become toxic by consuming water having F^- levels above 1.5 mg L^{-1} . Fluoride in water ($1 \mu\text{g mL}^{-1}$) keeps the teeth strong and healthy by converting hydroxyapatite into fluorhydroxyapatite/fluorapatite (less soluble) and thus reduces the solubility of enamel and tooth decay (Dhar and Bhatnagar, 2009). It also enhances the resistance of teeth to acid attack from sugar and plaque bacteria. However, F^- combines with Ca^{2+} in the blood to form insoluble calcium fluoride (CaF_2) thereby resulting in hypocalcemia (Khairnar et al., 2015). The

prominent health hazard due to F^- includes fluorosis which remains a significant problem in many countries across the globe like Afghanistan, China, Japan, Iraq, Iran, Turkey, etc. including India.

The area affected with more than $1 \mu\text{g mL}^{-1} \text{ F}^-$ in water, have intake ranging between 0.02 and $0.048 \text{ mg F}^- \text{ kg}^{-1} \text{ day}^{-1}$ for adults while areas with less than $0.3 \mu\text{g mL}^{-1} \text{ F}^-$ intake through water and dietary ranges between 0.004 and $0.014 \text{ mg F}^- \text{ kg}^{-1} \text{ day}^{-1}$ (ATSDR, 2003). Even infants that feed upon milk reconstituted with fluoridated water, might ingest $0.12\text{--}0.18 \text{ mg F}^- \text{ kg}^{-1} \text{ day}^{-1}$ (WHO, 1984). According to WHO 1984 and Indian standard drinking water specification 1991, The permissible limit of F^- in drinking water was recommended as 1.5 mg L^{-1} , whereas according to Kumar and Puri (2012) highest desirable limit is 1.0 mg L^{-1} . Other Indian agencies like Central Pollution Control Board (CPCB) and Central Public Health and Environmental Engineering Organization (CPHEEO) also recommends permissible limit i.e., 1.5 mg L^{-1} while Indian Council of Medical Research (ICMR) has declared $1.0\text{--}2.0 \text{ mg L}^{-1}$. According to Aoba and Fejerskov (2002) intake of F^- about 1 mg day^{-1} by an adult is harmless but above than 5 mg

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day⁻¹ could cause chronic F⁻ poisoning like dental or skeletal fluorosis.

Major sources of F⁻ are mostly natural, i.e., F⁻ containing mineral rock like fluor spar, rock phosphate, cryolite, apatite, mica, etc. High F⁻ concentrations occur in large and extensive geographical belts associated with a) sediments of marine origin in mountainous areas, b) volcanic rocks and c) granitic and gneissic rocks (Murray, 1986). Fluoride dissolved from these rocks contaminated groundwater (Edmunds and Smedley, 1996). Human beings including cattle are exposed to excess F⁻, primarily through drinking water and also through vegetables and cereals grown on contaminated soils. Thus, there is an urgent need to develop cheaper and sustainable decontamination techniques of F⁻ for drinking water. In recent times, rather than physicochemical methods, biological agents including plants and microorganism are being investigated for defluoridation and many have been found to be efficient and economic (Agarwal and Chauhan, 2015; Merugu et al., 2012). Among all the environmental components, studies on the levels of F⁻ in different plants particularly the edible crops are scantily reported in the literature. Economically viable and readily available plants like *Prosopis juliflora*, *Acacia tortilis*, *Cassia fistula*, etc. are reported for the accumulation of F⁻ (Saini et al., 2012). Use of bio-absorbent prepared from algae, fungi, woody plant, etc. has also been investigated for the application of biological agents in defluoridation (Mohan et al., 2007).

The objective of this review is to account for F⁻ sources and its contamination in various regions of India, levels of F⁻ accumulation in different plants including crops and the mechanism of uptake/transportation manifesting into toxicity. The review also presents brief information about different available methods for defluoridation through physio-chemical and biological means. Thus, the review highlights the research gap regarding reviewing the reports of F⁻ levels in plants particularly edible plants and use of biological means for defluoridation. The review may facilitate the researchers and policymakers to focus their research and effort to find a sustainable solution.

2. Sources of fluoride

Fluoride occurs naturally in varying concentrations in rocks, soil, water, air, plants and animals and anthropogenically in soil and water. The occurrence and movement of F⁻ in various environmental compartments have been schematically presented in Fig. 1.

2.1. Natural source

Fluoride accounts for about 0.06–0.09% of the Earth's crust (Koritnig, 1951). Some of the common minerals containing F⁻ are fluor spar or fluorite (CaF₂), cryolite (Na₃AlF₆), fluorapatite (Ca₅(PO₄)₃F) villiamite (NaF) and topaz (Al₂(SiO₄)F₂). Fluoride containing rocks are considered as the reservoir of F⁻ (WHO, 1984). Common rocks containing F⁻ are: Killas (1873 mg Kg⁻¹) > schist (1703 mg Kg⁻¹) > gneiss (1563 mg Kg⁻¹) > granite (1043 mg Kg⁻¹) > silixite (982 mg Kg⁻¹) > conglomerate (963 mg Kg⁻¹) > sandstone (903 mg Kg⁻¹) (He et al., 2013). The highest content of F⁻ was reported from volcanic rock (2000 mg Kg⁻¹) (Anazawa, 2006) followed by alkaline igneous rock (1300 mg kg⁻¹) and in ultramafic rocks and limestone ~100 mg Kg⁻¹ (Hem, 1985). The weathering of the primary minerals Fluorite, releases the F⁻ to the soil and groundwater. Volcanic activities and hydro-geothermal vents also release F⁻ into the environment which contaminates the soil; release of HF containing gases contaminates the air with F⁻ (CEPA, 1996; Vithanage and Bhattacharya, 2015). The third major natural source of F⁻ is from the marine aerosol, which contributes about 20,000 Kg globally of

inorganic F⁻ annually (CEPA, 1996).

High F⁻ content in soil occurs in clay minerals (Robinson and Edington, 1946). Alternatively, during the hot, dry pre-monsoon season, high rate of evapo-transpiration causes precipitation of salts, including F⁻ salts, temporarily in the top layers of the soil which act as a semi-permanent reservoir of soluble F⁻. During monsoon season, the precipitation infiltrates the soil, resulting in high TDS and addition of F⁻ to the soil and groundwater through the leachate from soils (Umar and Alam, 2012).

The enrichment of F⁻ in the aquifers depends largely on physical characteristics of soil such as porosity, depth of the wells and the acidity of the soils and rocks, temperature, ion exchange, presence of cations/anions and other on-going chemical reactions (Annaduraia et al., 2014). Fluoride is found in ground water up to 67 mg L⁻¹ while in most surface water is < 0.1 mg L⁻¹, however, sea water has a relatively high F⁻ content of 1.2–1.4 mg L⁻¹ (IPCS, 2002). This has been attributed to erosion and run-off due to streams and rivers from mountains and highlands over the period of time.

2.2. Anthropogenic sources

The use of F⁻ bearing fertilizers like aluminum phosphate fertilizer (Borah and Saikia, 2011), fumigants and pesticides containing barium fluorosilicate (BaSiF₆), sodium silico-fluoride (Na₂SiF₆), sulfuryl fluoride (SO₂F₂), trifluralin (C₁₃H₁₆F₃N₃O₄) at large-scale over the time, is attributed for the rise of F⁻ level in soil and groundwater. Industries related to glass, aluminum, steel etc. emit particulate F⁻ like AlF₃, NaAlF₆, CaF₂. Fluorinated compounds present in the atmosphere occurs as organic compounds like fluorocarbons, perfluoro-compounds, trifluoroacetic acid or inorganic gases i.e., HF, SF₆, SiF₄, F₂, H₂SiF₄, and minerals, i.e., CaF₂, NaF, Na₂SiF₆, NaAlF₄ (Ozsvath, 2009; Shoeib et al., 2004). The mixing ratio of inorganic and organic fluorine in troposphere is 0.1–0.4 ppb and ~1 ppb, respectively while some fluorinated gases are also transported to the stratosphere (De Angelis and Legrand, 1994; Ravishankara et al., 1993) and these gaseous fluorine eventually get deposited in soil over a period of time (Ozsvath, 2009). Meteorological conditions like temperature, wind direction and speed, turbulence, and precipitation of any area also determine the level of F⁻ contamination in air and soil (Gasic et al., 2010; Scheringer, 2009). Burning of coal and deposition of fly ash cause wet and dry deposition of gaseous and particulate F⁻. Fluoride emission in air due to biomass burning was estimated up to 76 Gg F⁻ yr⁻¹ and it depends on the type of biomass burned and the geographic location where it was harvested (Jayarathne et al., 2014).

Apart from HF in atmosphere, anthropogenic F⁻ in the form of CFC has annual atmospheric accumulation rate of 9.5–17 pptv (1 pptv = 1 × 10⁻¹² mol mol⁻¹) (NAP, 1992). Besides, a person addicted to smoking could intake 236 ppb F⁻ (1 × 10⁻⁹ mol mol⁻¹) (Ayooob and Gupta, 2006). In Uttar Pradesh, India, ash from brick kiln was observed to contain F⁻, which was reported to contribute to its contamination to its nearby areas (Faruqi, 2002). Coal-fired power plants are also important sources of anthropogenic HF emissions. Coal contains F⁻ in the range of 4–40 g Kg⁻¹ (McDonald and Berkeley, 1969). In 2001, the total air emission from HF by electrical utilities is estimated to be 55.8 million tons (Ranjan and Ranjan, 2015).

Even both the natural and anthropogenic sources are responsible for the contamination of F⁻ in different component of environment (air, water and soil), natural sources also contributes significantly. Among natural sources, F⁻ concentration in soil have been reported to contain >5000 mg Kg⁻¹ due to presence of gneisses and granite rocks (Vithanage and Bhattacharya, 2015) and 788.1 mg Kg⁻¹ from ejected ash volcanic ash, whereas,

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