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#### **Review Article**

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# Assessment of Ground water Quality with Special Focus on Fluoride Contamination and its Remediation Methods in India: A Comprehensive Review

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#### Introduction

The largest freshwater resource on Earth is ground water. It plays a pivotal role for industrial, agricultural, domestic and ecological needs of humans (Wada, 2016; Barya *et al.*, 2022; Thakur *et al.*, 2023). Only 3% of potable water is available on earth and 97% of water is found in ocean which is saline in nature.

ABSTRACT

The ground water quality varies in chemical constituents and their concentration depending on geological condition of that particular area. Due to exponential population growth, expansion of agricultural activities, urbanization and increased industrialization ground water

High fluoride levels beyond the permissible World Health Organization (WHO) limit of 1.5mg/l has been observed in various states of India. The present study gathered all the information for fluoride concentration in ground water from secondary data source or from various literatures in India and different part of the globe. Fluoride in drinking water is both positive and negative effects on health. The paper presents the current information on fluoride in environment and its effects on human health and existing methods for defluoridation.

quality is depleting rapidly (EPA, 1997; Cronin *et al.*, 2000; Thakur *et al.*, 2024). All over the world, poor drinking water is mainly accounted for 80% of the human diseases (WHO, 1984).

About 1 billion people world-wide do not have access to safe drinking water. According to revised Sustainable Development Goals the  $6^{th}$  one is clean water and sanitation.

Access to safe water is the most basic human need for health and well-being. Various Ground water quality threats i.e. faecal contaminants, salinity, arsenic, fluoride, pesticides, iron, manganese, and others but this special issue deals with fluoride related ground water quality challenges. Fluoride present in drinking water is completely absorbed whereas fluoride present in food is not absorbed (Barya *et al.*, 2022).

During the last decade, several newly affected regions have been reported all over the world, around 100 countries show impact of fluoride adulteration in underground water. One of important factor of fluoride in ground water is interaction of water with rocks. Several rocks have fluoride containing minerals like Calcium fluorophosphates  $(Ca_5(PO_4)_3F)$  also known as apatite, biotite group of black micahornblende а  $((Ca,Na)_2(Mg,Fe,Al)_5(Al,Si)_8O_{22}(OH)_2)$ and Fluorite (CaF<sub>2</sub>). The eroding of these rocks and gradual infilteration of rain water in these rocks rises fluoride in underground water. Other than these natural sources some other anthropogenic sources are also responsible for fluoride contamination in ground water. Agricultural fertilizers containing Phosphate, coal combustion, pesticides, Industrial aerosols and sewage sledges are major reason for high concentration of fluoride in water and soil (Frencken, 1992). There are also some minor factors which affect fluoride concentration in ground water such as temperature, pH of water and soil, sorption capacity of soil, depth of well, etc (EPA, 1997). High fluoride in ground water has been reported from 24 states in India (CGWB, 2023) they are AP, Telangana, Assam, Bihar, Chhattisgarh, Delhi, Gujarat, Haryana, HP, Jharkhand, Karnataka, Kerala, MP, UP, Maharashtra, Punjab. Rajasthan, Tamil Nadu, West Bengal, Uttarakhand, Odisha, Manipur, Meghalaya & Nagaland. Fluorine is the most electronegative element and has the high reactivity, therefore, it does not occur in elemental form rather it occurs in ionic form. In aquatic environments with pH less than 5, it has the tendency of forming complexes with metal ions, while at higher pH values it tends to exists as a single fluoride ion (Cronin et al., 2000; Thakur et al., 2024; Frencken, 1992). Although, the World Health Organization (WHO) has set the fluoride guideline limit of 1.5mg/l in drinking water (Brunt et al., 2004). The main objective of this work is to put together the information on fluoride levels in India.

#### **Geogenic Sources of Fluoride**

As fluorspar it is found in sedimentary rocks and as in Cryolite in igneous rocks. These fluoride minerals are nearly insoluble in water. Hence fluorides will be present in ground water only when conditions favour their solution (WHO, 1984).

#### **Biological Factors**

About 95% of the fluoride in the body is deposited in hard tissues and it continues to be deposited in calcified structures even after other bone constituents (Ca, P, Mg,  $CO_3$  and citrate) have reached steady state. Most important factor in deciding to what extent fluorine is incorporated into skeleton is Age. The uptake almost ceases in dental enamel after the age of about 30 years.

Whereas up to age of 55 years fluoride ingested with water goes on accumulating in bones. Long term consumption of fluoride rich water can interfere with carbohydrates, lipids, protein, vitamins, enzyme and mineral metabolism.

# Factors responsible for release of fluoride in ground water

#### pН

It is noticed that as the alkaline nature of water increases concentration of fluoride increases in underground water. The positive relationship between pH and bicarbonate creates an alkaline environment. It is the major governing mechanism for discharge of fluoride from the rock source (Nagendra Rao, 2003).

#### **Evaporation or evapotranspiration**

Evaporation increases the ion concentration which causes precipitation of some major minerals (United States Environmental Protection Agency, 2009) and decreases the concentration of calcium. Reduction of calcium favors increase in dissolved fluoride concentration in underground water (Sreedevi *et al.*, 2006).

#### **CO**<sub>2</sub>

Carbon dioxide is also important factor to control fluoride concentration in ground water. When carbon dioxide rich air comes in contact with rainwater, it releases of hydrogen ions is the reason for the weathering of the silicate rocks. The increase in the Na+/Ca<sup>2+</sup> ratio activates the fluoride concentration in ground water (Handa, 1975; Barnes *et al.*, 1988; Thakur *et al.*, 2021).

#### Ion Exchange

In case of fluorapatite rocks exchange of fluoride and phosphate ions takes place which forms Calcium Deca fluoride (Barnes *et al.*, 1988). Calcium Deca fluoride compound is very hard and brickle which is not suitable for skeletal structure (Thakur *et al.*, 2021).

 $Ca_5(PO_4)_3F + 9F-Ca_5F_{10}+3PO_4^{3-}$ 

#### **Health Effects of Fluoride**

#### Fluoride content in food

The fluoride concentration in food depends on two important factors. Firstly, on soil and secondly on water used for irrigation purpose. These two factors vary on climatic condition of village to village and district to district. The level of fluoride in vegetables, cereals and pulses ranges between 1.5-1.78 mg/kg (Ramteke *et al.*, 2007). About fruits, oil, nuts, mutton, condiments, fish also contain fluoride. Black salt which is used as taste maker in various food products specially pani-puri also contains rich amount of fluoride. Millions of Indians consume Pan, Tobacco, Gutka (Pan masala), Supari. On an average 2-4 sachet of pan masala consumed by average adductors which contains 0.34-1.12mg of fluoride (Thole, 2013).

#### **Fluoride in Beverages**

Among the beverages tea is the highly fluorinated one (Singh *et al.*, 1993). Almost two-third of the fluoride is soluble in water. Ooze of fluoride in tea increases with its contact time with water while boiling (Thakur *et al.*, 2022). Milk tea is always better than black tea as milk reduces concentration of fluoride. Aerated drinks, Lipton ice and coconut water also contains fluoride but it is less as compared to tea.

#### **Fluoride in Toothpastes**

The permissible limit of fluoride concentration in toothpaste, gel and paste is up to 1000mg/l as per the Bureau of Indian Standards.

#### **Remedial methods**

This section is divided in three categories.

#### **Dietary Habits**

Simple method to avoid fluorosis in spite of fluoride rich drinking water is to increase intake of calcium rich food. Best calcium rich source is curd. Just by taking one bowl of curd daily helps in preventing fluorosis. It is also observed that cell functions deteriorate due to fluoride intake which results in cell death. Vitamin C, a natural antioxidant overpowers oxidative stress in cells and protect from fluoride attack (Shreyas J. Kashyap *et al.*, 2021; Fung *et al.*, 1999; Gulati *et al.*, 1993).

#### Fluoride free Drinking water

Surface water is fluoride free and Rain water harvesting is also a good method.

#### **Several Defluoridation techniques**

Nalgonda technique is first method for fluoride removal from ground water using activated alumina. But treatment has a drawback of post management of excess of aluminum left in treated water. The alternative high performance defluoridation method is ion-exchange (Melike Ekran et al., 2021). It uses ion-exchange resin for fluoride removal. Electrocoagulation and electrodialysis are also techniques used for defluoridation (Thakur et al., 2020; Alkurdi et al., 2019; Mameri et al., 1998; Haldar and Gupta, 2020; APHA, 1995). Providing fluoride free water to fluoride rich areas is a very tough job in a developing country like India. Millions of individuals in India are affected by fluorosis. Majorly fluoride absorbed by the body occurs through fluoride rich water rather than fluoride rich food. Usage of surface water and rain water harvesting to provide fluoride free water is not feasible. Several defluoridation methods exist but they have their own drawback.

#### Table.1 Fluoride containing minerals (Ramadan and Hilmi, 2014)

Mineral	<b>Chemical Formula</b>	% Fluorine
Sellaite	$MgF_2$	61%
Villianmite	NaF	55%
Fluorite	CaF <sub>2</sub>	49%
Cryolite	Na <sub>3</sub> AlF <sub>6</sub>	45%
Fluorapatite	$Ca_3(PO_4)_3F$	3-4%

**Table.2** Different analytical water quality parameters used for testing of quality of water and their sources of occurrence and potential health effects with USEPA guidelines (Sonila Awasti *et al.*, 2002).

Sr. No.	Parameters	Source of occurrences	Potential health effect	
1	Colour	Due to presence of dissolved salts		
2	Turbidity	Soil runoff	As the level of turbidity increases possibility of bacterial diseases increases.	
3	Odour	Because of biological deprivation	unpleasant smell	
4	Electrical Conductivity	Due to presence of different dissolved solids	Corrosive nature of water increases due different dissolved solids.	
5	рН	It depends on dissolved solids and gases.	unpleasant taste; corrosive action of water increases.	
6	Total Hardness	It is caused by presence of high conc. Of alkaline earth ions especially calcium (Ca <sup>2+</sup> ) and magnesium (Mg <sup>2+</sup> ) ions in a water supply.	It prevents the formation of foam.	
7	Total alkalinity	It measures the dissolved CO <sub>2</sub> gases	Embrittlement of steel boiler. Boiled rice turns yellowish.	
8	TDS	Indicates the presence of minerals, organic material and salts in water	Unacceptable taste; gastro-intestinal irritation; corrosion.	
9	Chloride	Disinfectant	Irritation in eyes and nose; stomach upset; Corrosive nature of water rises.	
10	Nitrate	Excessive use of fertilizer; leaking from septic tanks and sewage leakages; decomposition of rocks.	Blue-baby syndrome also known as methemoglobinemia.	
11	Dissolved oxygen	Amount of oxygen present in water	It corrodes water pipe lines, boilers, etc. Lower value of dissolved oxygen makes difficult survival of aquatic animals.	
12	Biochemical Demand (B.O.D)	Contamination due to organic matter in water	It increases dissolved oxygen.	
13	Sulphate	Dissolved Sulphates of calcium, magnesium and iron in water.	Unpleasant taste; Causes gastro-intestinal problems. Calcium sulphate scale.	
14	Fluoride	Concentration of fluorine depends on several factors like type of rock, contact time and anthropogenic sources.	Fluoride concentration causes several effects like dental fluorosis, skeletal fluorosis etc.	

## Table.3 Showing adverse effect of fluoride intake (Adam, 2001)

1	Dental Fluorosis – In this deformation of enamel, mottling and brittleness of teeth is seen.		
2	Kidney Stones – It is a distinctive function of skeletal fluorosis		
3	Reproductive Toxicity- Drinking water having high concentration of fluoride shows declined birth rate.		
4	Gastrointestinal - High concentration of fluoride in digestive tract causes vomiting, nausea, diharrea and		
	abdominal pains.		
5	Neurotoxicity- High fluoride concentration causes neuron degeneration and low IQ in children is reported in		
	some of research papers.		
6	Skeletal Fluorosis- High concentration of fluorine results in bone deformation.		

# **Table.4** Shows more than 1.5mg/l concentration of fluoride in groundwater in various states in India(Shakir Ali *et al.*, 2019)

S. No.	States	Span of fluoride concentration	
		(mg/l)	
1	Andhra Pradesh	1.8-8.4	
2	Assam	1.45-7.8	
3	Bihar	1.7-2.8	
4	Chhattisgarh	1.5-2.7	
5	Delhi	1.5-6.1	
6	Gujarat	1.6-6.8	
7	Haryana	1.5-17	
8	Jammu and	2.0-4.21	
	Kashmir		
9	Karnataka	1.5-4.4	
10	Kerala	2.5-5.7	
11	Maharashtra	1.5-4.01	
12	Madhya Pradesh	1.5-10.7	
13	Orissa	1.52-5.7	
14	West Bengal	1.5-9.1	
15	Rajasthan	1.54-11.3	
16	Uttar Pradesh	1.5-3.1	
17	Tamil Nadu	1.5-3.8	
18	Punjab	0.44-6.0	

### Table.5 Different Fluoride Standards in India

S. No	Institution	Limit (mg/l)
1	World health organization WHO	1.5
2	United states Environmental Protection Agency US EPA	4
3	Bureau of Indian Standards BIS	1





Figure.2 Distribution of fluoride in India



Map showing Fluoride concentration in India (Rafique *et al.*, 2015), Punjab, Rajasthan, M.P, Chhattisgarh shows high concentration i.e range above 3mg/l.Maharashtra, Telangana, Goa, Karnataka, A.P, Gujarat, Bihar, U.P, Delhi, Haryana showing 1.5-3mg/l of range.

#### **Author Contribution**

Dipti G. Iyer: Investigation, formal analysis, writing original draft. Vaishali P. Meshram: Validation, methodology, writing—reviewing. Pravin U. Meshram:—Formal analysis, writing—review and editing.

#### **Data Availability**

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

#### Declarations

Ethical Approval: Not applicable.

Consent to Participate: Not applicable.

Consent to Publish: Not applicable.

**Conflict of Interest:** The authors declare no competing interests.

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