Effect of Groundwater Fluoride Concentration on Human Health: A Systematic Review

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Abstract-In studies conducted by various researchers it was observed that the groundwater is polluted by fluoride in most of the parts of World. Due to reported health related issues some workers carried out carcinogenic risk assessments and meta-analysis of fluoride exposure to humans. In this context, fluoride concentration data in groundwater across India by systematic searches conducted in various international search engines databases was collected. In some tests High water and urine fluoride levels in children indicated the chronic exposure to fluoride and suggested that the population is at high risk of developing chronic fluorosis in some regions of World. This paper highlighted the research work of various workers wherein Fluoride concentration risk factors related to human health are pointed and also discussed the steps to reduce fluoride in ground water.

Keywords: Fluoride, Carcinogenic risk assessment, meta-analysis, ground water, chronic fluorosis.

Introduction

French chemist Henri Moissan discovered Fluorine which is a common halogen element with the symbol F and atomic number 9. It ranks as the 13th most naturally occurring element on the planet Earth It is extensively dispersed in the environment, constituting more than 0.32% of the Earth's crust (WHO, 1984). Among all known elements it is one of the most reactive and electronegative elements naturally found in soil, water and food. It is most recognized for its role in preventing and reversing dental caries and building strong teeth and bones [1]. Fluoride (F–) and hydroxide ions (OH–) have almost similar ionic sizes, and both are negatively charged. Hence, F– ion can replace the OH-ion within the mineral structures during chemical reactions [2,3]. Most fluoride is absorbed in the gut and stored in bones and teeth. Unabsorbed fluoride is excreted in urine. Children absorb fluoride more efficiently than adults, as their teeth and bones are rapidly forming. Fluoride, a mineral, is naturally present in many foods and available as a dietary supplement. Fluoride protects teeth from decay by demineralization ad remineralization. Too much fluoride can lead to dental fluorosis or skeletal fluorosis which can damage bones and joints. It is also produced synthetically for use in drinking water, toothpaste, mouthwashes and various chemical products.[5]

Approximately 80% or more of orally ingested fluoride is absorbed in the gastrointestinal tract [1]. Most fluoride is absorbed in the gut and stored in bones and teeth. Unabsorbed fluoride is excreted in urine. Children absorb fluoride more efficiently than adults, as their teeth and bones are rapidly forming In adults, about 50% of absorbed fluoride is retained, and bones and teeth store about 99% of fluoride in the body [1,4]. The other 50% is excreted in urine. In young children, up to 80% of absorbed fluoride is retained because more is taken up by bones and teeth than in adults [1].

The water sources that are rich in ions like sodium (Na^+), potassium (K^+ chloride (Cl^-), and calcium ($Ca2^+$) are incline to have high concentration of fluoride as its concentration in most waters is controlled by the solubility of the main fluoride-bearing mineral that is fluorite (CaF_2). Overall, groundwater contains more fluoride than surface water due to greater contact and residence times with fluoride-bearing minerals in rock-water interactions. Fluoride

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concentrations are decidedly inconstant depending on the weathering and availability of leachable fluoride in rocks in similar lithologies or climate conditions. The studies shows that in addition to various natural sources, anthropogenetic diffusion of fluoride occurs into the water by means of the aluminium and coal industries, fertiliser use, and manufacturing processes [6]. On the basis of physical properties, fluoride is sectioned into three major types: a) hydrogen fluoride (HF), which is highly soluble in water, forming hydrofluoric acid ;b) sodium fluoride (NaF), which is moderately soluble in water and c) fluorosilicic (H_2SiF_6) acid, which is also known as hexosilicic acid, and substantially soluble in water [11]. In natural waters, fluoride occurs mainly as free fluoride ions through the complexes of Be, B, Al, and Si under specific conditions. The main factors that influence the higher concentration of fluoride in groundwater include climate, evaporation, precipitation, geology, geomorphology, and hydro geochemistry [7]. One of the factors which is responsible for the release of fluoride into groundwater is the pH, which governs the mobility of fluoride concentration. The positive correlation between pH and bicarbonate indicates an alkaline environment as the dominant controlling mechanism for the leaching of fluoride from the source rock. The correlation between the TDS and fluoride concentration, suggested a strong ionic strength and an increase in the solubility of fluoride in groundwater [8]. In some researches it was noted that there is a positive correlation between fluoride and bicarbonate and a negative correlation between calcium and fluoride concentrations in groundwater. Under alkaline condition the high concentration of fluoride is detected.with high concentrations of fluoride. Natural phenomenon like evaporation, or evapotranspiration, is an important process that increases the fluoride concentration in groundwater. [8-10]. Fluoride in deep source springs or shallow groundwater may arise from the discharge of hydrothermal groundwater from host rocks at depth, short-circuiting via fault pathways to the surface [12] Sometimes, fluoride concentrations are highly inconstant depending on the weathering and availability of leachable fluoride in rocks in similar lithologies or climate conditions [6]. Fluoride in the groundwater is derived from the weathering and subsequent leaching of fluoride-bearing minerals in the rocks and soil also from volcanoes. In the Icelandic literature of 1000 CE, there was a mention of an animal disease named ‘gaddur’ that was believed to have developed due to fluoride derived from volcanic eruptions. The presence of fluoride in water and its relationship between calcium and fluoride were first revealed by Hoag and Snee in 1930 by using a spectrograph [13]. In 1925, Fredrick McKay was the first to report the development of an unusual permanent stain of moulted enamel on the surface of teeth linked with drinking water and fluoride concentration. Fluorosis was identified as an occupational disease in humans in 1930 [13-15]. Reddy et al suggested that in the deep aquifer, continuous and long-duration water–rock interaction would enrich the fluoride concentration in groundwater.[16]

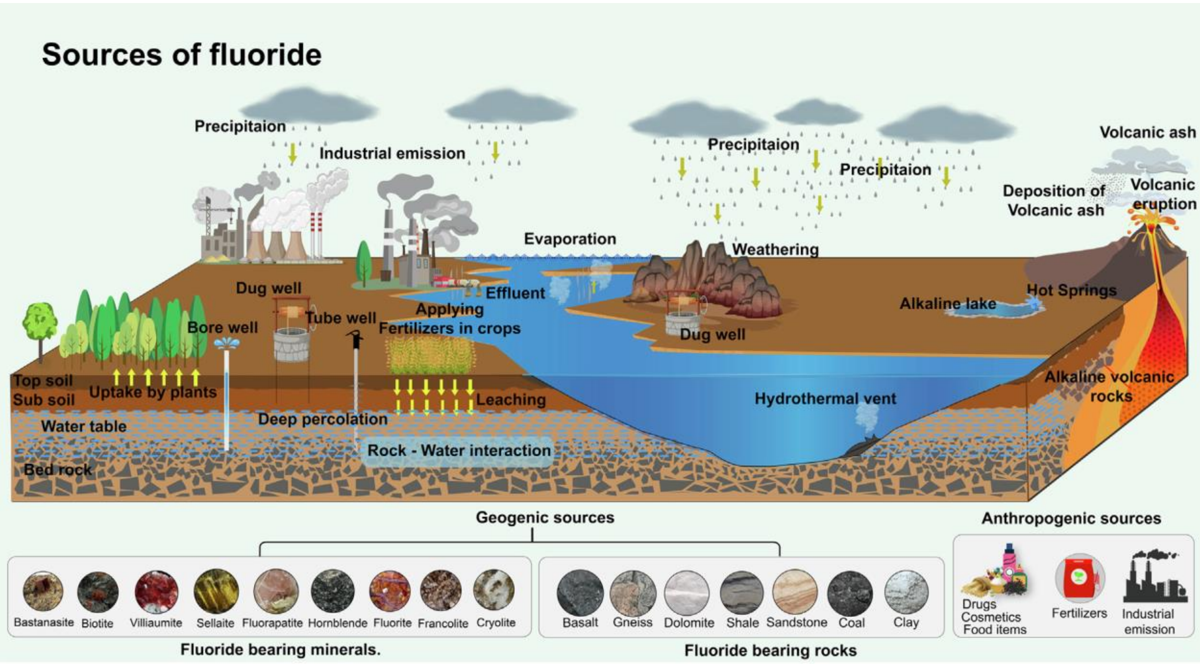

Fig 1.Various natural and anthropogenic sources of fluoride in surface and groundwater systems[17-18,35]

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Literature Survey

1.World fact files

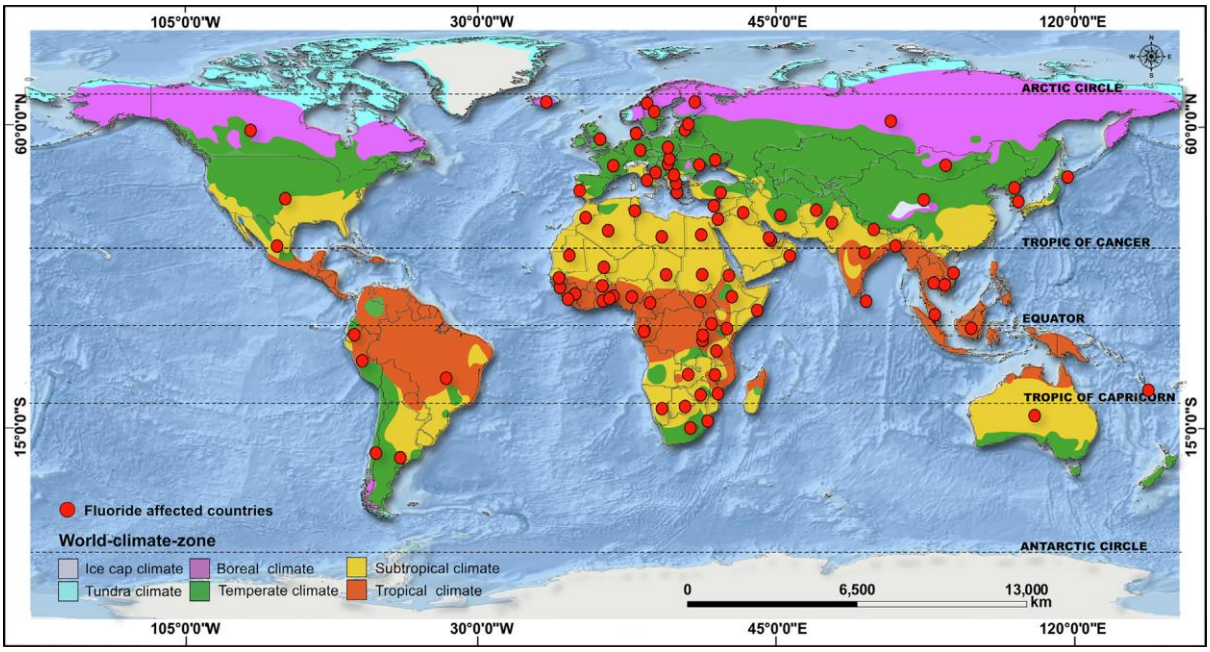

Fig.2.Fluoride-affected areas superimposed on the world climate map. (Source of climate base map: https://www.britannica.com/science/radiative-forcing) [35]

57.4 million people receive naturally occurring fluorinated water at or optimal levels in countries like Swedan, China, Sri Lankas, Zimbabwe and Gabon. The health of millions of people worldwide is negatively impacted by chronic exposure to elevated concentrations of geogenic fluoride in groundwater. Due to health effects including dental mottling and skeletal fluorosis, the World Health Organization maintains a maximum guideline of 1.5 mg/L in drinking water. Community water fluoridation is rare in Continental Europe, with 97–98% choosing not to fluoridate drinking water. The different parts of world are affected by fluoride concentration at different levels. Of the approximately 180 million people potentially affected worldwide, most reside in Asia (51–59% of total) and Africa (37–46% of total), with the latter representing 6.5% of the continent's population. Africa also contains 14 of the top 20 affected countries in terms of population at risk. We also illuminate and discuss the key globally relevant hadrochemical and environmental factors related to fluoride accumulation. Due to varying patterns of population density and groundwater usage, the implications of groundwater fluoride hazard [20]. The sternly affected countries include Sri Lanka, India, Pakistan, China, Iran, Jordan, West Bengal, Nepal, Japan, South Korea, North Korea, Libya, Ghana, Sudan, Burundi, Tanzania, Ethiopia, Algeria, Australia, and Chile. [19-22] Most of the fluoride pollution-prone zones are located in high grade metamorphic terranes or those associated with alkaline intrusions. The geothermal hot springs and volcanic regions are closely correlated with arid or semi-arid climatic conditions.[23]

Fluorine shows atmophile affinities and occurs in many common rock-forming minerals. Fluoride arises from the groundwater dissolution of fluoride-rich rocks containing minerals like amphiboles and biotite, particularly in metamorphic basement rocks, volcanic rocks, especially those of alkaline composition and clay minerals and micas [24-25] Fluoride in deep source springs or shallow groundwater may arise from the discharge of hydrothermal groundwater from multitude rocks at penetration, short-circuiting via fault pathways to the surface, and augmented concentrations of fluoride may occur in late-stage hydrothermal fluids and pegmatitic mineralization.[26]

Table1.List of countries affected by fluoride contamination in groundwater [35]

Continent	Countries
Asia (28)	Afghanistan, Bahrain, Bangladesh, Cambodia, China, India, Indonesia, Iran, Iraq, Israel, Japan, Jordan, Malaysia, Mongolia, Myanmar, Nepal, North Korea, Oman, Palestine, Pakistan, Qatar, Saudi Arabia, South Korea, Sri Lanka, Thailand, Yemen, Turkey and Vietnam
Africa (38)	Algeria, Benin, Botswana, Burkina Faso, Burundi, Cameroon, Chad, Cote D'Ivoire, Egypt, Eritrea, Ethiopia, Gabon, Ghana, Gunia, Kenya, Libya, Malawi, Mali, Mauritania, Morocco, Mozambique, Namibia, Niger, Nigeria, Rwanda, Senegal, Sierra Leone, Somalia, South Africa, South Sudan, Sudan, Swaziland, Tanzania, Togo, Tunisia, Uganda, Zambia and Zimbabwe
Europe (24)	Croatia, Cyprus, Denmark, Estonia, Finland, France, Germany, Greece, Greek, Hungary, Iceland, Italy, Latvia, Moldova, North Macedonia, Norway, Poland, Portugal, Serbia, Slovakia, Spain, Sweden, UK, and Ukraine
North America (3)	Canada, Mexico and the United States
South America (5)	Argentina, Brazil Chile, Ecuador, and Peru
Australia (2)	Australia and Vanuatu

2.Adverse Effects of Fluoride

Utmost reports divulge that a small amount of fluoride is good for dental health; however, exceeding the permissible limit causes dental fluorosis, skeletal fluorosis, crimping fluorosis, and osteosclerosis. Fluorosis was identified as an occupational disease in humans in 1930 [14]. For reducing or controlling fluoride in water sources, water fluoridation process is suggested which helps in controlling the level of fluoride in public water supplies solely to reduce tooth decay. On January 25, 1945, Grand Rapids, Michigan, became the first town in the world to use artificially fluoridated water. Singapore became the first country in Asia to start a fluoridation programme in the early 1950s [27]. The National Health and Medical Research Council of the Australian Government has also been practicing adding fluoride to water supply schemes since 1953. Other countries that adopted this practice in the early years include Canada (1945), the Netherlands (1953), and New Zealand (1954). From 1970 on, fluoridated toothpaste came on the market, although the concentration in the toothpaste was a little higher than the recommended value for artificial fluoridation of water supplies, which ranges from 0.5 mg/L to 1.0 mg/L [28]. Many researchers published their work on study of various diseases caused due to exceeded value of fluorides in various water resources. **Shrivastava et al.** pointed out that young adults suffer mostly from skeletal fluorosis which is chronic invasive systemic bone disease caused by long term excessive intake of fluoride [30].**Sabine etal** found in their studies that long term intake of excessive fluoride damages soft and hard tissues, teeth and bones intensely.[31].Thyroid hormone is also affected by high Fluoride concentration as in many studies TSH level of children in fluorosis area was found to be higher than any normal or unaffected areas. **Xiu et al** concluded in their studies [32]. Most importantly IQ level of children in area of high fluoride was lesser than in areas with low fluoride and significant difference was observed by **Feiging et al.** and prevention measures were suggested.[33]. **Chen et al** found that accumulation of excess fluoride in environment poses serious health risks to plants, animals and humans. Studies also indicated that excess fluoride concentration have negative impacts on food chain and hence ecological balance gets disturbed. [34] About 65 million people in the country are having fluorosis of different kind. A survey of literature has shown that groundwater in 21 states of the Indian Republic is facing the problem of fluoride. The major cause of fluoride in Indian sub-continent groundwater is leaching of flouride from rock minerals of earth's crust. Besides these anthropogenic activities, for example industrial discharges, aluminium smelter, brick kiln and air pollution from industries may also contribute to this problem. [36] Excessive intake of fluoride is hazardous to human beings and reducing to permissible limit can control various diseases and disorders to a great extent.

Some popular methods for Fluoride removal/reduction are discussed here.

1. Reverse Osmosis (RO): RO is a physical process in which the anions are removed by applying pressure on the feed water to direct it through the semi permeable membrane. RO works at higher pressure with more prominent rejection of dissolved solids. The membrane rejects the ions taking into account the size and electrical charge. RO membrane process is the reverse of natural osmosis as a consequence of applied hydraulic pressure to the high concentration side of the solution, it forces solvent filter through the membrane, against a pressure gradient into the lower-concentration solution. In RO, utilizing a mechanical pump, pressure is applied to a solution via one side of the semi-permeable membrane to overcome inalienable osmotic pressure. The process likewise removes soluble and particulate matter, incorporating salt from seawater in desalination [37]. This process can remove up to 90% of fluoride from water and is an effective technique for small-scale applications such as household water treatment systems. However, it requires a lot of energy and produces a large amount of wastewater. Additionally, the membranes that can be used in the reverse osmosis systems must be replaced periodically, which can be expensive.[35]. **Briao et al.** used reverse osmosis for desalination of water from the Guarani Aquifer System for drinking purpose in southern Brazil. The rejection of 100% of fluoride, 97% of total dissolved solids (TDS) and 94% of sulphate ions was achieved by RO at 2MPa pressure and 1.61 m/s of cross section flow velocity. The recovery rate of 93% of drinking water was obtained by blended water created by mixing groundwater with permeate [38]

2. Ion exchange process: Fluoride can be removed from water supplies with a strongly fundamental anion-exchange resin containing quaternary ammonium functional groups. The removal takes place according to the following reaction: The fluoride ions substitute the chloride ions of the resin. This process proceeds until every one of the sites on the resin are possessed.

Electro-coagulation is a straightforward and efficient technique to remove the flocculating agent produced by electro-oxidation of a sacrificial anode and generally made of iron or aluminium. Electrocoagulation includes electrolytic oxidation of a proper anode material. Electro-coagulation reactor is comprised of an electrolytic cell with one anode and one cathode [39]. **Yang et al.** studied the electrochemical removal of fluoride by delivering aluminium sorbent in a parallel-plate electrochemical reactor. Defluorination was done by anodic dissolution of aluminium electrodes in a dilute sodium chloride (NaCl) aqueous solution. The NaCl in the solution adequately diminished the power utilization and advanced the sorbent generation by deactivating the aluminium–water electrochemical system.

3.Adsorption Process: Adsorption procedures include the water's entry through a contact bed where fluoride is removed by ion exchange or surface chemical reaction with the solid bed matrix. As contrast with different procedures of defluorination, adsorption method is prominent because of its straightforwardness and also accessibility of extensive variety of adsorbents. Adsorption onto solid surface is straightforward, flexible and suitable procedure for treating drinking water systems, particularly for small groups. Adsorption technique is efficient and can remove ions over an extensive variety of pH to a lower leftover concentration than precipitation [40,41]. A few adsorbent materials have been attempted in the past to check their possibilities and techno-economic feasibility as defluoridating specialists.

Nalgonda technique created by NEERI is coagulation – precipitation method includes an expansion of aluminium salt, lime and bleaching powder took after by quick mixing, flocculation, sedimentation and filtration. Aluminum salt is utilized to remove fluoride from water. Lime and alum are the most usually utilized coagulants for Nalgonda technique for defluoridation of water [42].Vivek et al presented a novel cost effective defluorination method that is based on surface modification of rice husk ash (RHA) by coating aluminium hydroxide. RHA is obtained by burning rice/paddy husk which is an abundantly available and is an inexpensive raw material. The results showed excellent fluoride removal efficiency and the adsorption capacity was found to be between 9 and 10 mg/g. [43]

Conclusion

Fluoride in small portion is needed for overall growth of human. Fluoride is known to have both beneficial and detrimental effects on health, depending on the dose and duration of exposure. The optimum fluoride level in drinking water should be below 1.5 mg/L as per WHO guidelines. The groundwater fluoride concentration varies from place to place depends on many factors. As it is present in food, water and soil in different proportions it cannot it avoided. But once the concentration crosses the permissible limit it indicates various side effects. This paper discussed about various adverse effects of high fluoride concentration. There are various treatment methods to reduce and remove excess fluoride by using surface water and collection of rainwater for drinking purposes is the best method in absence of any treatment process.[36]. Fluorosis can cause a reduction in children's IQ, and this effect was found to be associated with the type of fluoride exposure. Based on the prevalence of dental fluorosis and its effect on the intelligence of children, it appears that reducing fluoride levels in drinking water and monitoring water quality are important strategies for the prevention and treatment of dental fluorosis. At the community and domestic levels, active alumina and its different materials are widely applied to remove the fluoride in the water in the ion exchange and adsorption processes [44]. There are various methods of defluoridation but according to environmental conditions, geological conditions and financial conditions pertinent method will vary. Still research is head on for affordable defluoridation method so that poor people could be benefited. There is not a universal method which is appropriate under all social, financial, economic, environmental and technical conditions. None of the methods has been implemented successfully at a large scale in many parts of the world. All available defluoridation methods do have disadvantages.

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