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Fluoride in Environmental Compartments – A Comprehensive Review of Literature

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Abstract

Fluoride occurs naturally within many types of rock that has health importance. However, excessive fluoride contamination in drinking water due to natural and anthropogenic activities has been reported as one of the major health related problems worldwide, with the developing world being no exception. In view of this a comprehensive literature review was carried out to determine the current body of knowledge pertaining to fluoride related research work with focus on the environment and human health related aspects. The literature published in the standard journals was used for the purpose of this study and it (literature) was reviewed by following the principles of deductive reasoning. The literature shows that In India, several states are affected with excess fluoride in groundwater and pose a serious health threat to the inhabitants of these regions. Hence, it is important that specific region specific studies should be carried out to generate more and more data, which can help in delineating meaningful and efficient strategies for mitigating fluoride related problems. Also, the literature shows that there is scope for developing the region specific technologies for addressing the fluoride contamination in water in general and groundwater in particular.

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Introduction

Fluoride is a chemical that occurs naturally within many types of rock. Fluoride contamination in drinking water due to natural and anthropogenic activities has been recognized as one of the major problems worldwide imposing a serious threat to human health. Hydrochemical characteristics of groundwater, the lithological features of the aquifer sediments and fluoride in rocks are widely reported as the points of genesis of high fluoride groundwater (> 1.5 mg/L), which often makes that area an endemic fluorosis area. Furthermore, it has been reported by Kravchenko et al., (2014) that elevated level of fluoride (F^-) in drinking water is a well-recognized risk factor of dental fluorosis in humans.

Through various intermediate steps, but also directly, fluoride passes into the natural water system and components of the food chain. Ingestion by man is mainly through drinking water and other beverages. However, the correct knowledge of fluoride in the water, especially the groundwater is very little, which makes large population vulnerable to fluoride related ailments. Most of the fluoride found in groundwater is naturally occurring from the breakdown of rocks and soils or weathering and deposition of atmospheric volcanic particles. Fluoride can also come from i) runoff and infiltration of chemical fertilizers in agricultural areas, ii) septic and sewage treatment system discharges in communities with fluoridated water supplies, iii) liquid waste from industrial sources.

It has been reported that at low concentrations fluoride can reduce the risk of dental cavities. However, exposure to higher amounts of fluoride can cause dental fluorosis, which is a cause of concern. In its mildest form this results in discolouration of teeth, while severe dental fluorosis includes pitting and alteration of tooth enamel. Even higher intakes of fluoride taken over a long period of time can result in changes to bone, a condition known as skeletal fluorosis, which can cause joint pain, restriction of mobility, and possibly increase the risk of some bone fractures. In view of the environmental and human health importance, this study was carried out to comprehensively review the literature pertaining to the fluoride related studies. In addition to above, evolution of fluoride related research focusing on the prevalence of fluoride in the groundwater and its consequences on human health were given more attention.

Material and Methods

A literature review was carried out to identify the previous research efforts and directions related to our focal area. The objective was to identify the research gaps and highlight research motivations. To the extent possible, care was taken to reproduce the original terminology used by the authors, to preserve the originality of the views. Furthermore, the literature was reviewed in a chronological order (of publication), which indicated the evolutionary pattern in the selected area of research i.e. fluoride pollution. The literature review addressed various issues encompassing the field of groundwater pollution vis-à-vis fluoride. In this study care was taken to review literature published in standard journals i.e. journals with science citation index. The review of the literature is as follows

Discussion

Pickering (1985) has reviewed the factors which promote retention or release of soluble fluoride species by soils with the aim of identifying key processes. The topics considered include fluoride levels in soils, mobility as demonstrated in leaching studies, and fluoride adsorption by soils. Author stated that the mobility of fluoride tends to be a function of soil type, system pH and F levels, with retention being favoured in acidic sediments containing clays and poorly ordered hydrous oxides of aluminium. Furthermore, Dissanayakeab (1991) had stated that the presence or excess of very low concentrations of fluoride in the groundwater has been a major factor in the dental epidemiology. The author stated that the high fluoride content in the groundwater (sometimes in excess of 5 mg/l) was related to high frequency of dental fluorosis, while very low fluoride content of water results in dental caries.

Warnakulasuriya et al., (1992) examined the prevalence of dental caries and dental fluorosis 14-yr-old children living in four geographic areas of Sri Lanka with water F levels of 0.09-8.O ppm. Based on the study results, authors stated that the their data are comparable with findings from other tropical countries, e.g. Kenya and Senegal, and reaffirm that WHO guidelines for the upper limit of F in drinking water may be unsuitable for developing countries with a hot, dry climate. Gaciri and Davies (1993) have stated that the acquisition of considerable additional data on the hydro geochemical behavior of fluoride in natural waters of Kenya has been made possible by extensive surface-water and groundwater sampling campaigns as well as by improvements in analytical techniques.

Reimann et al., (1996) in their study analysed hard rock groundwater samples collected from private drinking water wells in the environs of Oslo and Bergen for their radon and fluoride contents. Their results indicated that contrary to popular perception, concentrations of several elements (e.g. Ba, F, Fe, Mn, Na, Rn) exceeded drinking water action levels in a significant number of cases. Authors advocated a continuous program for assessment of the economic and toxicological impacts of such contaminants. In view of the high fluoride content in the groundwater, Zevenbergen et al., (1996) presented a simple defluoridation method that used Kenyan soil derived from volcanic ash (e.g. Ando soils or soils with 'andic' properties) as a fluoride sorbent. These results were extended to possible technical application using a one dimensional solute transport model. Based on the results authors concluded that the use of Ando soils appeared to be an economical and efficient method for defluoridation of drinking water on a small scale in rural areas.

Datta et al., (1996) had reported that the groundwater was used extensively in the Delhi area for both irrigation and raw water requirement. Fluoride contamination in groundwater was therefore a matter of concern for the planners and managers of water resources. Authors discussed stable isotope (18O) and fluoride signatures in groundwater, to characterise the sources and controlling processes of fluoride contamination. Their study indicated that almost 50% of the area was affected by fluoride contamination beyond the maximum permissible limit and the wide range (0.10–16.5 ppm) in fluoride concentration suggested contributions from both point and non-point sources. Very high fluoride levels in groundwater were mostly found in the vicinity of brick kilns. Authors stated that the process of adsorption and dispersion of fluoride species in the soil as well as lateral mixing of groundwater along specific flow-paths control the groundwater fluoride and 18O composition.

Apambire et al., (1997) had stated that the oncentrations of groundwater fluoride in excess of the World Health Organization (WHO) maximum guideline value (1.5mg/l) in the Bongo area of Ghana have been known since 1978. However, the effect of fluoride on people ingesting the water showed that 62% of the total population of school children in the study area had dental fluorosis. Upon investigation, authors stated that the source of groundwater fluoride within the study region was dissolution of the mineral fluorite and dissolution of and anion exchange with micaceous minerals and their clay products. However, authors suspect that a much higher population is susceptible to developing dental and skeletal fluorosis than originally suspected. The purpose of this study by Hudaka (1999) was to compile, map, and evaluate regional patterns of fluoride concentrations in Texas groundwater. Author reported that several factors contribute to elevated fluoride levels in Texas aquifers, including seepage from nearby saline formations, sparse recharge and dilution, and native mineral constituents of the aquifers. Results of this study suggest that: (1) regional geology influences fluoride concentrations, (2) statewide, the pattern is not random, (3) fluoride levels are generally higher in the western part of the state, and (4) regions which warrant further monitoring include west- central and north- central Texas.

Latha (1999) in her study showed that occurrence of fluoride is highly sporadic and localized in eastern and southeastern Karnataka and the concentration of fluoride varied from 1 to 7.4 mg/l. The geological strata near the wells influence the fluoride content in phreatic groundwater. Effective and cheap methods of defluoridation are few and hence author advocated the need to use biological defluoridation, which may be the best alternative to the conventional methods. Chakraborti et al., (2000) in their study examined 2063 people from 8 villages in the Bagpani area of Karbi-Anglong district of Assam, India for dental and skeletal fluorosis. 646 (31.3%) were identified with dental fluorosis and 36 (1.74%) with skeletal fluorosis. A positive correlation was also observed of fluoride with calcium and sodium in drinking water; no positive correlation with potassium was observed. Analysis of groundwater from villages of Karbi-Anglong and Nagaon districts of Assam showed that of a total of 75 samples analyzed, 43 samples had fluoride levels above 1.0 mg/litre.

Riveraa et al., (2002) had stated that significant amounts of fluoride are found in the abstracted groundwater of San Luis Potosí. This groundwater withdrawal induces a cold, low-fluoride flow as well as deeper thermal fluoride-rich flow in various proportions. Flow mixing takes place depending on the abstraction regime, local hydrogeology, and borehole construction design and operation. Ekanayake and Hoek (2002) conducted a study to assess caries and developmental defects of enamel in relation to fluoride levels in drinking water and the association between caries experience and the severity of diffuse opacities in children living in Uda Walawe, an area with varying concentrations of fluoride in drinking water in Sri Lanka. The prevalence of enamel defects increased significantly with the increase in the fluoride level in drinking water. In conclusion, the relationship that was observed in this study between fluoride levels in drinking water, diffuse opacities and caries suggests that the appropriate level of fluoride in drinking water for arid areas of Sri Lanka.

Sujatha (2003) had stated that the fluoride level in groundwater is controlled by the distribution of $Ca2+$ and $SO42-$, ionic strength and the presence of complex ions in its composition. In the study area, situated in the Ranga Reddy district, Andhra Pradesh, India, the concentrations of fluoride in the groundwater varied from 0.7 to 4.80 mg/l and from 0.4 to 4.20 mg/l during the pre-and post-monsoon seasons respectively. From the correlation coefficient studies, it was observed that fluoride was inversely related with $Ca2+$ and positively related with $HCO3-$, whereas the correlation coefficient between fluoride and other ions was very poor during both seasons. Rao and Devadas (2003) studied groundwater samples from Anantapur District, Andhra Pradesh, India for fluoride ($F-$) along with other chemical parameters. The results suggest that the main sources of $F-$ in groundwater in the district are the country rocks, in which fluorine is strongly absorbed in soils consisting of clay minerals. A strong positive correlation between $F-$ and lithogenic sodium reflects weathering activity was also recorded.

Hoeka et al., (2003) described the association between source of drinking water and other potential risk factors with dental fluorosis. Authors stated that from the point of view of prevention of dental fluorosis, drinking water from surface sources or from shallow wells located close to surface water would be preferable. Péreza et al., (2003) studied reproductive parameters in a population exposed to fluoride at doses of 3–27 mg/day (high-fluoride-exposed group—HFEG). The results of this study indicate that a fluoride exposure of 3–27 mg/day induces a subclinical reproductive effect that can be explained by a fluoride-induced toxic effect in both Sertoli cells and gonadotrophs. Shomar et al., (2003) determined the fluoride levels in water, soil and tea, and identified the major fluoride minerals in soil that supply water with fluoride ions and found a high positive correlation between fluoride concentrations in groundwater and occurrence of dental fluorosis. Saxena and Shakeel (2003) conducted a geochemical study of groundwater from 58 selected fluoride-rich areas in different parts of India that includes eight states indicates that: 1. These groundwaters are alkaline in pH (7.4–8.8) and their electrical conductivity varies from 530–2,680 µS/cm and fluoride concentration from 1.7–6.1 mg/l. Presence of fluoride-bearing minerals in the host rocks and their interaction with water is considered to be the main cause for fluoride in groundwater. 2. The decomposition, dissociation and dissolution are the main chemical processes for the occurrence of fluoride in groundwater. 3. This study indicated that 85% groundwater samples have EC: 1,000–2,000 µS/cm, pH: 7.5–8.5, and HCO3/Ca (epm ratio): 0.8–2.3. 4. The Ca and HCO3 contents of groundwater samples have shown good correlation with fluoride

Abu and Khaled (2004) reported that the groundwater fluoride concentration shows a positive relation to pH and HCO3, whereas Cl, Mg, Ca, and Na initially increase and then decrease with increasing fluoride in the water. Authors reported that the chemistry of the groundwater was controlled by the fluorite and calcite solubility. Reimanna and Banksb (2004) focused on the inorganic chemical quality of natural groundwater. Possible health effects, the problems of setting meaningful action levels or maximum admissible concentrations for drinking water, and potential shortcomings in current legislation are discussed and expressed a need for setting action levels based on transparency, toxicological risk assessment, completeness, and identifiable responsibility was suggested by the authors on the basis of empirical data.

In addition to above studies, Meenakshi et al., (2004) determined fluoride concentration in underground water in four villages of Jind district of Haryana state (India) where it is the only source of drinking water. The study results indicated considerable variations among the analyzed samples with respect to their chemical composition with majority of the samples did not comply with Indian as well as WHO standards for most of the water quality parameters measured. The fluoride concentration in the underground water of these villages varied from 0.3 to 6.9 mg/l, causing dental fluorosis among people especially children of these villages and overall water quality was unsatisfactory for drinking purposes without any prior treatment in majority of studied locations. Water quality data set from the alluvial region in the Gangetic plain in northern India, which is known for high fluoride levels in soil and groundwater, has been analysed by chemometric techniques by Singha et al., (2005). Study showed that the groundwater samples are dominated by variables having origin both in natural and anthropogenic sources in the region, whereas, variables of industrial origin dominate the surface water samples. It suggested that the groundwater sources are contaminated with various industrial contaminants in the region. Furthermore, the stable isotope analysis results by Kima and Jeongb (2005) provide substantial information for the relative ages of groundwaters revealing that the F-rich groundwaters are deeply circulating paleoground waters and occur along the faults due to upward flow along the fault plane indicating that rock chemistry, groundwater age, well depth, and geologic structure are the important factors controlling the occurrence of high F groundwaters.

Jacksa et al., (2005) has stated that India has an increasing incidence of fluorosis, dental and skeletal, with some 62 million people at risk. High fluoride groundwaters are present especially in the hard rock areas south of the Ganges valley and in the arid north-western part of the country. The phenomenon is related to groundwater with residual alkalinity. Fluoride concentrations are governed by adsorption equilibria and by fluorite solubility. Ncube and Schutte (2005) have reported that dental fluorosis was observed in those areas of South Africa in which fluoride levels were higher than the recommended guidelines for drinking water. Ayooba and Guptaa (2006) have stated that the incidence of dental, skeletal and crippling skeletal fluorosis has been reported in India with average fluoride concentrations as low as 0.5, 0.7 and 2.8 ppm respectively. Distribution of fluoride in groundwater and its suitability assessment for drinking purpose Groundwater samples collected either from the bore-wells (forms a part of municipal water supply) or from the hand pumps (direct consumption) were analysed for fluoride in Hisar city, India by Khaiwal and Garg (2006). The study results indicated that in most of the groundwater samples the concentration of fluoride was found to be moderately higher, when compared to the WHO standard for drinking water, which may lead to associated health risks in urban population, if the groundwater is being used without proper treatment.

Maheshwari (2006) has stated that excessive fluoride concentrations have been reported in groundwaters of more than 20 developed and developing countries including India where 19 states are facing acute fluorosis problems. Based on the study results it has been concluded that the selection of treatment process should be site specific as per local needs and prevailing conditions as each technology has some limitations and no one process can serve the purpose in diverse conditions. Singh et al., (2007) determined the fluoride content in drinking water samples from Southern Haryana, India. The mean fluoride concentration in drinking water samples of Pataudi, Haily Mandi and Harsaru villages was 1.68 ± 0.35, 3.22 ± 1.18 and 1.78 ± 0.12 mg/l, respectively, which was very high. Misraa and Mishra (2007) have stated that in marginal and central alluvial plains of India, the inland salinity is continuously increasing, canal network and arid to semi-arid climatic conditions that led to excessive evapotranspiration concentrates the salt in soil and thereby escalating the groundwater salinity, which is severely affected by endemic fluorosis due to consumption of fluoride-contaminated water. Result of the study shows that there is a variation and continuous escalation in the groundwater salinity and fluoride concentration in deep and shallow aquifers.

Rafique et al., (2008) has reported that of the Thar Desert of Pakistan is severely contaminated with fluoride ion with fluoride ion concentrations ranging between 0.09 and 11.63 mg/l. Moreover, Dhar and Bhatnagar (2009) have reported that though fluoride is an essential element needed for normal development and growth of animals and extremely useful for human beings, its monitoring in the water sources should be carried out regularly. Ozsvath (2009) has stated that the relationship between environmental fluoride and human health has been studied for over 100 years by researchers from a wide variety of disciplines and most scientists believe that small amounts of fluoride in the diet can help prevent dental caries and strengthen bones, but there are a number of adverse effects that chronic ingestion at high doses can have on human health, including dental fluorosis, skeletal fluorosis, increased rates of bone fractures, decreased birth rates, increased rates of urolithiasis (kidney stones), impaired thyroid function, and lower intelligence in children. Recently, Reddy et al., (2010) have reported that high concentrations of fluoride (up to 7.6 mg/L) are a recognized feature of the Wailapally granitic aquifer of Nalgonda District, Andhra Pradesh, India where the basement rocks provide abundant sources of F in the form of amphibole, biotite, fluorite and apatite. In light of groundwater pollution Negrel et al., (2011) studied the stable isotopes of the water molecule (δ18O and δ2H) from a hard-rock aquifer in the Maheshwaram watershed, Andhra Pradesh, India and stated that in such a watershed, the effect of overpumping can be severe and the environmental effects of water abstraction and contamination are of vital importance.

Sujana and Anand (2011) explored the feasibility of utilizing bauxite for fluoride removal from synthetic and natural fluoride bearing groundwater samples of Orissa, India. the study results show that the estimated thermodynamic parameters (ΔH°, ΔS° and ΔG°) indicated that the adsorption was spontaneous and exothermic in nature. Ganvir and Das, (2011) have stated that fluoride content in groundwater that is greater than the WHO limit of 1.5 mg/L, causes dental and skeletal fluorosis. Authors reported that several fluoride removal techniques are reported in the literature amongst which the Nalgonda technique and use of activated alumina have been studied extensively. In view of this authors proposed a novel cost effective defluoridation method that is based on surface modification of rice husk ash by coating aluminum hydroxide and the study results showed excellent fluoride removal efficiency and the adsorption capacity was found to be between 9 and 10 mg/g. Currell et al., (2011) carried out chemical analysis of groundwater and sediments to investigate causes of elevated F^- (1.5–6.6 mg/L) and reported that positive correlations between F and As concentrations and Na/Ca ratios (r2 = 0.67 and 0.46, respectively) indicate that groundwater major ion chemistry plays a significant role.

Shan et al., (2012) have stated that insightful knowledge of geochemical processes controlling fluoride mobility is fundamental to understand the occurrence of elevated F in groundwater. Whereas Jayawaedana et al., (2012) have reported that soil geochemistry suggests that the meta-igneous rocks in the fluoride-rich districts may have been influenced by a fluoride-rich residual melt, whereas the fluoride-poor districts are associated with acidic meta-igneous rocks and meta-sedimentary rocks. Furthermore, Pettenati et al., (2012) in their study focused on modelling the F^- accumulation caused by irrigation return flow below rice paddy fields in the small endorheic Maheshwaram watershed (Andhra Pradesh, Southern India). their study found that the principal sources of F^- are fluorapatite dissolution and, to a lesser extent, allanite and biotite dissolution. Anthropogenic sources of F^-, such as fertilizers, are probably very limited. After simulating an entire dry-season irrigation cycle (120 days), the results are in good agreement with the observed overall increase of Cl^- in the Maheshwaram groundwater.

Chakrabortty et al., (2013) carried out a modeling and simulation study along with economic evaluation for removal of fluoride from contaminated groundwater in a flat sheet cross flow nanofiltration membrane module and concluded that such a filtration system is quite promising in purifying fluoride-contaminated groundwater at low cost. Chhabra et al., (2012) has stated that significantly ($P < 0.05$) higher plasma F concentrations were observed in animals of fluorotic region in comparison to healthy control animals, which indicates that along with humans the animals are also susceptible to adverse effects of high fluoride concentration.

Conclusions

'Water is life,' so central to human life, yet over one billion people across the world have no access to safe drinking water. Of late, there has been increasing global attention focused on resolving water quality problems especially in developing countries, as the lack of access to clean water denies the most essential of all rights, the right to life. The latest estimates suggest that around 200 million people, from among 25 nations the world over, are under the dreadful fate of fluorosis. India and China, the two most populous countries of the world, are the worst affected. India is plagued with numerous water quality problems due to prolific contaminants mainly of geogenic origin and fluoride stands first among them. The weathering of primary rocks and leaching of fluoride-containing minerals in soils yield fluoride rich groundwater in India which is generally associated with low calcium content and high bicarbonate ions. The unfettered ground water tapping exacerbates the failure of drinking water sources and accelerates the entry of fluoride into groundwater. Most of the scientific literature substantiates the benefits of low fluoride concentrations in preventing dental decay, however, the situation is not acceptable in case of high fluoride concentrations.

The literature shows that In India, several states are affected with excess fluoride in groundwater. The problem is aggravated due to the lack of appropriate and user friendly defluoridation technology. In view of this it can be concluded that the geochemical symbol plot maps can help geochemists understand factors controlling the distribution and uptake of fluoride in the different geographical regions. Hence, it appears to be important to carry out comprehensive fluoride mapping by collecting data at regular intervals. Although various technologies are being used to remove fluoride from water, the literature shows that there is ample scope for formulating different strategies for mitigating fluoride related problems.

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