Fluoride in Groundwater: Causes, Implications and Mitigation Measures

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Fluoride in Groundwater: Causes, Implications and Mitigation Measures

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ABSTRACT

Groundwater is the major source for various purposes in most parts of the world. Presence of low or high concentration of certain ions is a major issue as they make the groundwater unsuitable for various purposes. Fluoride is one such ion that causes health problems in people living in more than 25 nations around the world. Fluoride concentration of atleast 0.6 mg/l is required for human consumption as it will help to have stronger teeth and bones. Consumption of water with fluoride concentration above 1.5 mg/l results in acute to chronic dental fluorosis where the tooth become coloured from yellow to brown. Skeletal fluorosis which causes weakness and bending of the bones also results due to long term consumption of water containing high fluoride. Presence of low or high concentration of fluoride in groundwater is because of natural or anthropogenic causes or a combination of both. Natural sources are associated to the geological conditions of an area. Several rocks have fluoride bearing minerals like apatite, fluorite, biotite and hornblende. The weathering of these rocks and infiltration of rainfall through it increases fluoride concentration in groundwater. Fluoride which is present in high concentration in volcanic ash is readily soluble in water and forms another natural source. Anthropogenic sources of fluoride include agricultural fertilisers and combustion of coal. Phosphate fertilisers contribute to fluoride in irrigation lands. Coal which is a potential source of fluoride is used for combustion in various industries and in brick kilns. The aerial emission of fluoride in gaseous form during these activities reaches the surface by fall out of particulate fluorides and during rainfall they percolate with the rainwater thus reaching the groundwater table. Also the improper disposal of fly ash on ground surface contributes to fluoride in groundwater. Since ingestion of high fluoride has a long term effect on human health it is essential to monitor its concentration in groundwater used for drinking periodically and take steps to bring them within the permissible range of 0.6 to 1.5 mg/l. There are several methods available for the removal of fluoride from groundwater which is insitu or exsitu. To dilute the groundwater contaminated with fluoride, artificial recharging structures can be built in suitable places which will decrease its concentration. Rainwater harvesting through existing wells also will prove effective to reduce the groundwater fluoride concentration. Exsitu methods which are conventional treatment methods like adsorption, ion exchange, reverse osmosis, electrodialysis, coagulation and precipitation etc can be practiced at community level or at households to reduce fluoride concentration before ingestion. But the choice of each method depends on the local conditions of the region such as the quality of groundwater and the source of contamination whether it is natural or anthropogenic. Fluoride contamination being a prominent and widespread problem in several parts of the world and as causes for this are mostly natural and unpreventable, educating the people and defluorinating the groundwater before consumption are essential for a healthy world.

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INTRODUCTION

It is well known that about 70% of the earth's surface is covered with water. Most of the water is in the oceans (96.5%) in the unusable form while some of them are frozen (1.74%). Lakes, swamp water and rivers hold 0.014% and soil moisture accounts for 0.001%. Water also exists in the form of vapour in the air (0.001%) and as groundwater beneath the sub surface in the aquifers (1.7%) (Gleick, 1996). World's water needs are met from surface and groundwater resources. However, use of groundwater is advantageous as it is comparatively fresh and widely distributed unlike the surface water. Threats to groundwater have been increasing everyday due to raise in population and their needs. Thus with increasing demand of groundwater for domestic, industrial and agricultural needs, the pressure on this resource has become enormous. Overexploitation and improper management has also lead to contamination of this resource. The degradation of groundwater may be due to natural or anthropogenic processes. Natural causes are inherent geological conditions while anthropogenic causes include wastewater from sewage treatment plants, discharge from industries, improper solid waste disposal, agrochemicals, runoff from agricultural fields, leakage from underground storage tanks etc. When the chemical composition of groundwater is not within the prescribed standards for drinking or irrigation or industrial water, they become unsuitable. Arsenic, fluoride, nitrate, iron, manganese, boron, most heavy metals and radionuclides are few contaminants that are of great concern if not present within permissible limits. In this chapter, the causes and implications due to presence of increased fluoride in groundwater used for drinking purpose and measures to be adopted to mitigate the problem are discussed.

PROPERTIES OF FLUORIDE

Fluoride belongs to halogen family represented as 'F' with atomic weight 18.998 and atomic number 9. It occurs as a diatomic gas in its elemental form and has a valence number 1. It is the most electronegative and the most reactive when compared to all chemical elements in the periodic table (Greenwood and Earnshaw, 1984; Gillespie et al., 1989). It has an oxidation state of -1 and occurs as both organic and inorganic compounds. It is the 13^{th} most abundant element in the earth's crust (Weinstein and Davison, 2003). Its natural abundance in the earth's crust is 0.06 to 0.09% (Fawell et al., 2006) and the average crustal abundance is 300 mg/kg (Tebutt, 1983). Fluoride does not exhibit any colour, taste or smell when dissolved in water. Hence, it is not easy to determine it through physical examination. Only chemical analysis of the groundwater samples can determine the concentration of this ion. The widely used method for the estimation of fluoride in groundwater sample is colorimetric SPANDNS (sodium 2-(parasulfophenylazo)-l,8-dihydroxy-3,6-naphthalene disulfonate) method. The other colorimetric method extensively used is the complexone method. Fluoride concentration can also be quantified using sophisticated instruments like ion chromatograph. Ion selective electrodes are available to measure fluoride concentration in water, which can be used both in the field and in laboratory. Fluoride is one of the important micronutrient in humans which is required for strong teeth and bones. In humans, about 95% of the total body fluoride is found in bones and teeth. WHO (World Health Organisation) (1984) has prescribed the range of fluoride from 0.6 to 1.5 mg/l in drinking water as suitable for human consumption. BIS (Bureau of Indian Standards)

(1992) has set a required desirable range of fluoride in drinking water to be between 0.6 and 1.2 mg/l. However, this standard suggests the maximum permissible limit can be extended up to 1.5 mg/l. This required fluoride is supplied to the human body usually through drinking water. Consumption of water with fluoride below or above the prescribed range is detrimental to human health. Hence, it is essential to monitor the groundwater quality regularly which is used directly without treatment as drinking water.

FLUORIDE IN GROUNDWATER

Groundwater is considered as the major source of drinking water in most places on earth. Usually people use groundwater for drinking and other domestic household purposes such as cooking without any physical or chemical treatment. This is not a healthy practice and may lead to number of health disorders. However, this practice cannot be avoided due to lack of treated piped water supply system in several parts of developing countries.

Groundwater with fluoride concentration above the permissible limit set by WHO i.e 1.5 mg/l have been recorded in several parts of the world. In 1984, WHO estimated that more than 260 million people living all over the world consume water with fluoride concentration above 1 mg/l (WHO, 1984). The problem of high fluoride in groundwater has been reported by several researchers in India, China, Japan, Sri Lanka, Iran, Pakistan, Turkey, Southern Algeria, Mexico, Korea, Italy, Brazil, Malawi, North Jordan, Ethiopia, Canada, Norway, Ghana, Kenya, South Carolina, Wisconsin and Ohio (Dissanayake, 1991; Gaciri and Davies, 1993; Srinivasa Rao, 1997; Banks et al., 1998; Oruc, 2003; Rukah and Alsokhny, 2004; Kim and Jeong, 2005; Tekle-Haimanot et al., 2006; Valenzuela-Va´squez et al., 2006; Zheng et al., 2006; Chae et al., 2007; Farooqi et al., 2007; Mirlean and Roisenberg, 2007; Msonda et al., 2007; Vivona et al., 2007; Davraz et al., 2008; Messaïtfa, 2008; Moghaddam and Fijani, 2008; Oruc, 2008; Suthar et al., 2008; Desbarats, 2009; Li et al., 2009; Karthikeyan et al., 2010; Keshavarzi et al., 2010; Kim et al., 2010; Looie and Moore, 2010; Naseem et al., 2010; Reddy et al., 2010a; Yidana et al., 2010; Young et al., 2010; Brindha et al., 2011). The other possible sources of intake of fluoride apart from drinking water are through food, beverages and dental products like tooth paste.

Most of the people affected by high fluoride concentration in groundwater live in the tropical countries where the per capita consumption of water is more because of the prevailing climate. In places like Ghana, people consume 3 to 4 liters of water which is higher than the WHO estimate of 2 l/adult/day (Apambire et al., 1997). The risk of fluorosis is higher in these places. However, incidence of fluorosis in people living in other parts of the world has also been reported. The intensity of fluorosis problem is very serious in the two heavily populated countries of the world namely India and China (Ayoob and Gupta, 2006). In most cases, fluoride in groundwater is contributed by the host rocks which are naturally rich in fluoride. Because of rock water interaction, long residence time and evapotranspiration, the concentration of fluoride increase. Most of the studies indicate the increase in fluoride composition in groundwater with increase in depth from ground surface (Hudak and Sanmanee, 2003; Edmunds and Smedley, 2005; Kim and Joeng, 2005; Valenzuela-Va´squez et al., 2006). But this is not always common (Apambire et al., 1997; Reddy et al., 2010a). The geochemistry of high fluoride groundwater are often associated with neutral to alkaline pH, low calcium concentration and high sodium and bicarbonate concentrations (Handa, 1975; Ramamohana Rao et al., 1993; Kundu et al., 2001; Smedley et al., 2002; Edmunds and Smedley, 2005; Chae et al., 2007). In some cases, there is also occurrence of high nitrate where there is high fluoride in groundwater (Handa, 1975; Reddy et al., 2010b). Saxena and Ahmed (2001) put forth that alkaline conditions with pH ranging between 7.6 and 8.6 are favorable for dissolution of fluorite mineral from the host rocks. Sodium bicarbonate type waters are typical of high fluoride waters (Handa, 1975; Srinivasa Rao, 1997; Chae et al., 2007). Fluoride exhibits a positive relationship with sodium and bicarbonate while it extends an inverse relationship with calcium. Handa (1975) observed that groundwater is generally undersaturated with respect to fluorite but in some cases they are saturated or over saturated (Srinivasa Rao, 1997; Smedley et al., 2002; Chae et al., 2007). There are also cases where both calcite and fluorite saturation occurs in groundwater with high fluoride. Overall, the natural concentration of fluoride in groundwater depends on the geological, chemical and physical characteristics of the aquifer, the porosity and acidity of the soil and rocks, the surrounding temperature, the action of other chemical elements, depth of the aquifer and intensity of weathering (Feenstra et al., 2007).

CAUSES FOR FLUORIDE

The possible causes and sources through which fluoride exists in the environment are schematically shown in Figure 1.

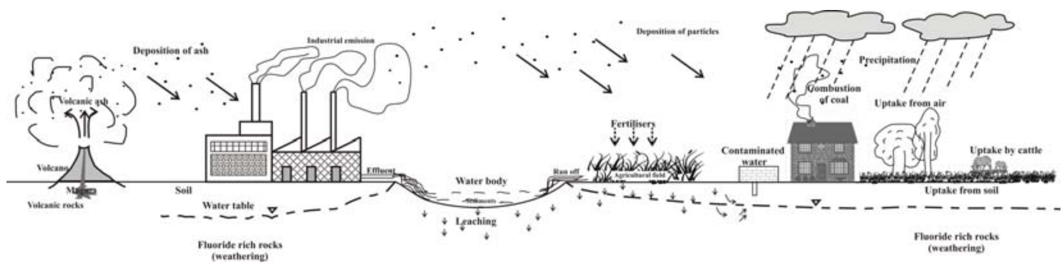

Figure 1. Possible casues for fluoride in groundwater

Aquifer material

Most of the fluoride in groundwater is naturally present due to weathering of rocks rich in fluoride. Water with high concentration of fluoride is mostly found in sediments of marine origin and at the foot of mountainous areas (WHO, 2001; Fawell et al., 2006). Known fluoride belts on land include: from Syria through Jordan, Egypt, Libya, Algeria, Sudan and Kenya, from Turkey through Iraq, Iran, Afghanistan, India, northern Thailand and China. There are also same kind of belts in the America and Japan (WHO, 2001). Fluorite occurs in igneous and sedimentary rocks. Deer et al., (1983) reported that the occurrence of fluoride in both these rock types is almost similar. Fluoride occurs as sellaite [MgF_2], fluorite or fluorspar [CaF_2], cryolite [Na_3AlF_6], fluorapatite [$3Ca_3(PO_4)_2$ $Ca(F,Cl_2)$], apatite [$CaF_2.3Ca_3(PO_4)$], topaz [$Al_2SiO_4(F,OH)_2$], fluormica (phlogopite) [$KMg_3(Si_3Al)O_{10}(F,OH)_2$], biotite [$K(Mg,Fe)_3$ $AlSi_3O_{10}(F,OH)_2$], epidote [$Ca_2Al_2(Fe^{3+};Al)(SiO_4)(Si_2O_7)O(OH)$], amphibole such as tremolite [$Ca_2Mg_5Si_8O_{22}(OH)_2$] and hornblende [$Ca_2(Mg,Fe,Al)_5(Al,Si)_8O_{22}(OH)_2$], mica, clays, villuanite and phosphorite (Matthess, 1982; Pickering, 1985; Hem, 1986; Handa, 1988; Haidouti, 1991; Gaumat et al., 1992; Gaciri and Davies, 1993; Datta et al., 1996; Apambire et al., 1997; Kundu et al., 2001;

Mohapatra et al., 2009). Fluoride minerals such as fluorite and cryolite are not readily soluble in water under normal pressure and temperature. But under alkaline conditions and range of specific conductivity between 750 and 1750 µS/cm, dissolution rate of fluorite minerals increase (Saxena and Ahmed, 2001). Granitic rocks which are a typical source of fluoride rich rocks contain fluoride ranging between 500 and 1400 mg/kg (Koritnig, 1978; Krauskopf and Bird, 1995), which is much higher than any other rock type. The world average content of fluoride in granitic rocks is 810 mg/kg (Wedepohl, 1969). The weathering of these rocks results in increased fluoride content in groundwater. Longer residence time in aquifers with fractured fluoride rich rocks enhance fluoride levels in the groundwater. Naseem et al. (2010) put forth that granitic rock and clay in Pakistan contained average fluoride of 1939 and 710 mg/kg respectively. Granite and granitic gneisses in Nalgonda, India contain fluoride rich minerals such as fluorite (0 to 3.3%), biotite (0.1 to 1.7%) and hornblende (0.1 to 1.1%) (Ramamohana Rao et al., 1993). Mondal et al. (2009) reported from the same area that the rocks contain 460 to 1706 mg/kg of fluoride. Laboratory studies conducted by Chae et al. (2006) showed that leaching of fluoride from granitic rocks contributed 6 to 10 mg/l of fluoride to water.

Volcanic ash

Volcanic rocks are often enriched in fluoride. Hydrogen fluorine is one of the most soluble gases in magmas and comes out partially during eruptive activity (D'Alessandro, 2006). The aerial emission of fluoride in the form of volcanic ash during volcanic eruption reaches the surface by fall out of particulate fluorides and during rainfall. This fluoride from the soil surface will easily reach the groundwater zone along with percolating rainwater. Volcanic eruptions are common in Iceland and fluorosis poisoning in livestock and humans was identified long ago in 1978 from the Laki eruption (Fridriksson, 1983; Steingrímsson, 1998). The fluoride content in ash from Hekla eruption in 2010 was 23-35 mg/kg (Matvælastofnun, 2010). Volcanic ash is readily soluble and thus the risk of fluoride contamination in groundwater is very high. These volcanic sources have also been found to cause fluoride contamination in groundwater of Kenya (Gaciri and Davies, 1993).

Fly ash

Like volcanic ash, fly ash from the combustion of fossil fuels also account for high fluoride. More than 100 to 150 million tons of fly ash is produced worldwide annually due to the combustion of coal especially from power plants (Prasad and Mondal, 2006; Piekos and Paslawska, 1998). Inappropriate disposal of this fly ash will result in the leaching of fluoride to groundwater. Churchill et al. (1948) reported that coal contains 40 to 295 mg of fluoride/kg. But the fluoride content of coal depends on the type of coal being burnt. Brick kilns which use coal for burning also account as a source for fluoride pollution (Jha et al., 2008).

Fertilisers

Phosphate containing fertilisers add up to the fluoride content in soil and groundwater (Motalane and Strydom, 2004; Farooqi et al., 2007). It is evident that superphosphate (2750 mg of F/kg), potash (10 mg of F /kg) and NPK (Nitrogen Phosphorous Potassium) (1675 mg of F /kg) which are phosphatic fertilisers contain remarkable amount of fluoride (Srinivasa Rao, 1997). Also, fluoride concentration in irrigation water accounts to be 0.34 mg/l. In agricultural areas successive irrigation had lead to the increase in fluoride concentration in groundwater (Young et al., 2010). The amount of water soluble fluoride in the soil near a phosphate fertiliser plant in Brazil was 10 mg/kg. Datta et al. (1996) put forth that if an agriculture field of 1 ha receives 10 cm of irrigation water containing 10 mg/l of fluoride, then the soil can obtain 10 kg of fluoride. This is considered as a potential threat for increase in fluoride concentration in soil and groundwater.

Apart from these, industrial processes such as aluminium smelting (Haidouti, 1991), cement production and ceramic firing (WHO, 2002) also lead to release of fluoride into the environment.

HEALTH IMPLICATIONS

Intake of fluoride higher than the optimum level is the main reason for dental and skeletal fluorosis. Depending upon the dosage and the period of exposure fluorosis may be acute to chronic. Ayoob and Gupta (2006) quoted that around 200 million people, from among 25 nations all over the world are under the dreadful fate of fluorosis of which India and China, the two most populous countries of the world, are the worst affected. In India 62 million people including 6 million children are estimated to have serious health problems due to consumption of fluoride contaminated water (Andezhath and Gosh, 2000). Endemic fluorosis is prevalent in 29 provinces of China, municipalities and autonomous regions (Wang and Huang, 1995). Dental and skeletal fluorosis was predominant in China due to the indoor burning of coal to make brick tea or for heating purposes (Ando et al., 1998; Watanabe et al., 2000; Ando et al., 2001). Dissanayake (1991) stated that dental fluorosis was predominant in the dry zone where fluoride concentration was high and dental caries was prevalent in wet zones where low fluoride occurs in groundwater of Sri Lanka. Of the 10 million people living in the Ethiopian Rift Valley, more than 8 million are exposed to elevated concentration of fluoride (Rango et al., 2010). The health outcome by consuming fluoride at different concentration was given by Dissanayake (1991) i.e. when fluoride concentration in drinking water is below 0.5 mg/l it causes dental carries; fluoride between 0.5 to 1.5 mg/l results in optimum dental health; 1.5 to 4 mg/l causes dental fluorosis; 4 to 10 mg/l induces dental and skeletal fluorosis while fluoride above 10 mg/l results in crippling fluorosis. However, fluorosis results not only due to the presence of high concentration fluoride in drinking water but also depend on other sources such as the dietary habits which enhance the incidence of fluorosis.

Dental fluorosis

Tooth enamel is principally made up of hydroxyapatite (87%) which is crystalline calcium phosphate (Brudevold and Soremark, 1967). Fluoride which is more stable than hydroxyapatite displaces the hydroxide ions from hydroxyapatite to form fluoroapatite. On prolonged continuation of this process the teeth become hard and brittle. This is called dental fluorosis. Dental fluorosis in the initial stages results in the tooth becoming coloured from yellow to brown to black. Depending upon the severity, it may be only discolouration of the teeth or formation of pits in the teeth. The colouration on the teeth may be in the form of spots or as streaks. Usually these streaks on the teeth are horizontal. Children who are exposed to excess fluoride from childhood show symptoms of fluorosis very often than compared to adults. Hence the fluoride problem in an area may not be decided on the fact that the adults have good teeth with no symptoms of discolouration. Though the main source for dental fluorosis is fluoride ingestion through drinking water, it can also be ingested through toothpastes containing fluoride. It is common for children to swallow toothpastes which has to be avoided to prevent fluorosis. A significant relationship between fluoride intake by water and the prevalence of dental fluorosis has been reported by several researchers (Heller et al., 1997; Viswanathan et al., 2009; Mandinic et al., 2009; Viswanathan et al., 2010).

Skeletal fluorosis

Exposure to very high fluoride over a prolonged period of time results in acute to chronic skeletal fluorosis. It was stated in 1993 that crippling skeletal fluorosis might occur in people who have ingested 10 to 20 mg of fluoride per day for over 10 to 20 years (National Research Council, 1993). India and China has been largely affected by crippling skeletal fluorosis with 2.7 million people being affected in China. Of the 32 states in India, 17 have been identified as endemic areas with 6 million people affected by skeletal fluorosis. Apart from ingestion of fluoride through drinking water, skeletal fluorosis also may be caused due to indoor use of coal as fuel and by air borne fluoride. Ingestion of fluoride through inhalation in factories and industries is one of the occupational health problems. Skeletal fluorosis does not only affect humans but also animals fed with fluoride rich water and fodder. Fluorosis is also now associated with heavy consumption of tea (Cao et al., 1996; Watanabe et al., 2000; Whyte et al., 2008; Joshi et al., 2010). Early stages of skeletal fluorosis start with pain in bones and joints, muscle weakness, sporadic pain, stiffness of joints and chronic fatigue. During later stages, calcification of the bones takes place, osteoporosis in long bones, and symptoms of osteosclerosis where the bones become denser and develop abnormal crystalline structure. In the advanced stage the bones and joints become completely weak and moving them is difficult. The vertebrae in the spine fuse together and the patient is left crippled which is the final stage. Skeletal fluorosis is usually not recognized until the disease reaches an advanced stage.

Other effects

Other health disorders that occur due to consumption of high fluoride in drinking water to be muscle fibre degeneration, low haemoglobin levels, deformities in RBCs, excessive thirst, headache, skin rashes, nervousness, neurological manifestations, depression, gastrointestinal problems, urinary tract malfunctioning, nausea, abdominal pain, tingling sensation in fingers and toes, reduced immunity, repeated abortions or still births, male sterility, etc (Meenakshi and Maheshwari, 2006). As fluoride is excreted in urine through the kidneys, they affect the effective functioning of the kidneys. They facilitate in the formation of kidney stones. Li et al. (1988) reported that fluoride might have genotoxical effects. Several studies also reported these effects on humans and animals (Sheth et al., 1994; Joseph and Gadhia, 2000; Tripathi et al., 2009). Consumption of drinking water with high fluoride by children may affect their intelligence. Tang et al. (2008) who studied this phenomenon however could not come out with a mechanism by which the IQ of children is lowered. Guan et al. (1999) suggested that when phospholipids and ubiquinone contents gets altered in the brain of rats affected by chronic fluorosis, changes in their membrane lipids may be the cause of this problem. Several other studies carried also comply with this fact (Trivedi et al., 2007; Ge et al., 2010). The presence of excessive fluoride in groundwater has its impact not only on humans but also on soil fertility and plant and animal growth.

STATUS OF GROUNDWATER FLUORIDE OCCURRENCE IN THE WORLD

Dean (1933) reported the prevalence of fluorosis in Arizona, Arkansas, California, Colorado, Idaho, Illinois, Iowa, Kansas, Minnesota, Nevada, New Mexico, North Carolina, North Dakota, Oklahoma, Oregon, South Carolina, South Dakota, Texas, Utah and Virginia. In 1980's the United States incorporated several recommendations to reduce fluoride ingestion. But Kumar et al. (1998) found that the prevalence of dental fluorosis has not declined even more than ten years after the recommendations. High concentration of fluoride (>3.5 mg/l) was noticed in South Carolina where 40% of the people depend on groundwater for their needs (South Carolina Ambient Groundwater Quality Report, 2003). The groundwater of Lake Saint-Martin area, Manitoba represented geochemical signature of groundwater originating from a deep regional aquifer, which are upwelling in a previously unrecognized discharge zone created by structural uplift associated with the impact event (Desbarats, 2009). It was further found that Na–HCO_3–SO_4 groundwater type with low chloride and less depleted ^{18}O concentrations represented modern recharge displacing groundwater of Na-mixed anion type with high chloride and highly depleted ^{18}O compositions representing discharge from Winnipeg formation in Manitoba. The volcanic ash deposits in Texas which was the reason behind high fluoride in groundwater up to 6.27 mg/l showed better correlation with well depth (Hudak and Sanmanee, 2003). Emission from phosphate fertiliser production factory which was the cause of high fluoride in groundwater in southern Brazil was found to be influenced by vegetation cover i.e. places with grasslands had higher fluoride than those with eucalyptus plantation (Mirlean and Roisenberg, 2007).

In Ireland, from 1964 fluoridated water was supplied to the public in order to meet the fluoride requirement of the human body. The Department of health, Social Services and Public Safety of Ireland put forth that 71% of the population are exposed to this fluoridated water in 2007 (Oral Health Strategy for Northern Ireland, 2007). In 1984, the National Survey of Children's Dental Health in Ireland examined people until 15 years and found that they had fluorosis from mild to questionable level but higher levels were absent (Whelton et al., 2004). Federation Dentaire International stated that most Germany had low fluoride water (Federation Dentaire International, 1990). In Belgium, drinking water contains fluoride below 0.3 mg/l. However, cases of dental fluorosis have been reported by Carvalho et al. (1998) in Belgium.

Schwartz and Friedrich (1973) assessed the concentration of fluoride in spring and stream waters to determine occurrences of fluorite in Osor district, Spain. Groundwater in Norway had high fluoride up to 8.26 mg/l due to fluoride rich rocks. Fluoride concentrations in groundwater were high in areas of granite while it was low in anorthosites. It was observed that dental fluorosis was detected in parts of Norway which are related to the bedrock source (Banks et al., 1998). Czarnowski et al. (1996) studied the groundwater quality in Poland with respect to fluoride and found that they were well below limits in most places. However, fluoride concentration of 1.38 mg/l was detected around a phosphate industry waste disposal site. Over 90% of the population in Estonia consumed water with fluoride below 1.5 mg/l. Even then fluoride concentrations of about 7 mg/l occur naturally in western Estonia which is due to Silurian-Ordovician aquifer system (Indermitte et al., 2009). The risk of getting affected by dental fluorosis in this region is 4.4 times higher in category of exposure ranging between 1.5 and 2.0 mg/l. Alumina production plants had increased the fluoride concentration in Greece. Studies by Haidouti (1991) showed that total soil fluoride collected from near the alumina production plant at depths of 0-5, 5-15 and 15-30 cm decreased with distance from the emission source and reached background levels at about 20 km. Also, total soil fluoride decreased with depth at high impact areas and it was vice versa at low impact areas. This high fluoride in soil may leach through the unsaturated zone during precipitation and increase fluoride in groundwater.

Oruc (2008) stated that in Turkey endemic fluorosis related to high fluoride ranging from 1.5 to 4 mg/l in drinking water was observed since 1955. Public drinking water supply system in Isparta, Turkey draws water from lakes and springs from 1962. Springs discharged from volcanic rocks, Golcuk pyroclastic and Miocene clastic rocks contained fluoride between 3.71 and 5.62, 1.81 and 3.95, 0.39 and 1.02 mg/l respectively. This resulted in elevated fluoride concentration in groundwater which resulted in dental fluorosis in south west Turkey (Davraz et al., 2008). North Jordan has very low fluoride concentration (Rukah and Alsokhny, 2004) where it becomes necessary to provide fluorinated water to meet the daily requirement of fluoride by the human body. It is found that the fluoride concentration follows the regional topography in that area. In Saudi Arabia 34% of the water needs are supplied by groundwater. The fluoride concentration in groundwater that is supplied to the water treatment plants before reaching the public contains 0.63 to 1.6 mg/l of fluoride (Alabduláaly, 1997). Thus fluoride concentration is well within limits in drinking water in Saudi Arabia. Moghaddam and Fijani (2008) found that groundwater occurring almost everywhere in basaltic rocks in north western Iran contain fluoride beyond the suitable range.

Dental decay was found increasing in southern Algeria. Messaïtfa (2008) reported the concentration of fluoride up to 2.3 mg/l in groundwater in Algeria. About 70% of the fluoride intake for the people of this region is through groundwater used for drinking. Apart from these, dates and tea contribute to 10% and 20% of fluoride intake respectively. Thus the daily intake of fluoride ingested by an adult exceeds the threshold limit of 0.05 to 0.07 mg of fluoride/kg/day (Burt, 1992). Fluoride contents in some rivers (12-26 mg/l), springs (15-63 mg/l) and alkaline ponds and lakes (60-690 mg/l) were found to be very high in Tanzania (Nanyaro et al, 1984). Weathering of fluoride rich nephelinitie and carbonatitic rocks and soils and gaseous emanations through mineral springs were the probable contributing sources. Gaciri and Davies (1993) noticed that in natural waters of Kenya, fluoride concentration was greater in lake water than groundwater and springs which was greater than river water. Evaporation would have been a major cause to increase the concentration of fluoride in lakes of this region.

Groundwater studies on fluoride in South Korea show that the concentration of fluoride depends on the residence time (Kim and Jeong, 2005) due to geogenic source of fluoride (Chae et al., 2007; Kim et al., 2010). People living in Ikeno district of Japan were accidentally exposed to drinking water containing 7.8 mg/l fluoride for 12 years (Ishii and Suckling, 1991). After the realization of the problem, they were substituted with water containing 0.2 mg/l of fluoride. Because of this the children in this area developed dental fluorosis. In Japan, as there are number of volcanoes, there is probability of fluoride contamination through volcanic ash. Ash from volcanic explosion of Sakurajima volcano, Japan was found to contain average fluoride concentration of 788.1 mg/kg (Nogami et al., 2006). This forms a potential source of threat to groundwater around this area. Abdelgawad et al. (2009) found that weathering and alteration of granitic rocks was the factor affecting oversaturation of fluoride ion in groundwater in Mizunami area, Japan.

The incidence of mottled enamel was first reported in 1930 by Anderson and Stevenson (1930) in China. About 26 million people in China suffer from dental fluorosis due to high fluoride consumption through water. Also 16.5 million people suffer from dental fluorosis resulting from coal smoke pollution (Liang et al., 1997). It was estimated that more than 30 million people suffer from chronic fluorosis. Fluoride problems in China occur through drinking water, indoor coal combustion and brick tea. In Taiyuan basin of China, Guo et al. (2007) noticed that in recharge area interactions between groundwater and fluoride containing minerals were the sources for high fluoride whereas in discharge area evaporation and mixing of karst water contributed to high fluoride. Migration and enrichment of fluoride in North west China was studied by Genxu and Guodong (2001). This study concluded that on the basis of spatial distribution the causes for fluoride can be differentiated as dissolution-runoff, evaporation-runoff and leaching-evaporative enrichment depending on hydrogeochemical zones.

Fertiliser containing leachable fluoride ranging from 53 to 255 mg/kg and coal containing fluoride ranging from 5 to 20 mg/kg were reported to pollute groundwater with high fluoride in east Punjab, Pakistan by Farooqi et al. (2007) where 2 million people are at risk of being exposed to high fluoride. The granitic rocks with average fluoride concentration of 1939 mg/kg in Nagar Parkar area, Pakistan, contain fluoride in kaolin deposits between 468 and 1722 mg/kg and secondary kaolin deposits have 270 mg/kg which are the source of fluoride up to 7.85 mg/l in groundwater in this area (Naseem et al., 2010). Studies on fluoride in groundwater in Sri Lanka carried out by Dissanayake (1991) and Young et al. (2010) shows that the condition has not changed even after about two decades with fluoride above 4 mg/l in groundwater. It was found that high fluoride areas lie within low plains and low fluoride areas were usually highlands. This was because the contact time with the geological material was longer in the plains and there exists slow groundwater movement compared to highlands (Dharmagunawardhane and Dissanayake, 1993). A detailed description on the concentration of fluoride in groundwater and its sources in various regions of the world based on literature are given in Table 1 and Figure 2.

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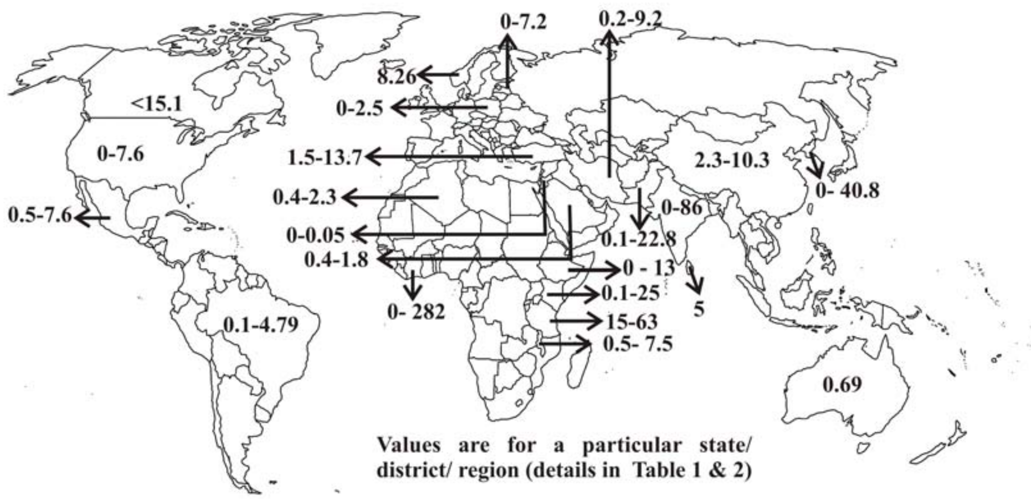

Figure 2. Occurrence of fluoride in groundwater in various parts of the world based on literature

Table 1. Concentration of fluoride in groundwater and its sources in various parts of the world based on literature

Country (in aphabetical order)	Source	General range of fluoride concentration in groundwater	Reference
Algeria	Fluorinated minerals	0.4 to 2.3 mg/l	Messaïtfa, (2008)
Australia	Atmospheric	Up to 0.69 mg/l	Petrides and Cartwright, (2006)
Brazil	Phosphate fertilizer production emission	0.1 to 4.79 mg/l	Mirlean and Roisenberg, (2007)
Canada	Fluoride rich rock	Up to15.1 mg/l	Desbarats, (2009)
China, Taiyuan basin	Dissolution of the fluorine minerals and evaporation	>2 mg/l	Li et al., (2009)
China	Fluorine rich minerals and rocks	2.5 to 10.3 mg/l	Genxu and Guodong, (2001)
China, Mongolia	Fluorite from Holocene sediments	2.3 to 9.8 mg/l	Zheng et al., (2006)
China, Taiyuan basin	Limestone	Up to 6.20 mg/l	Guo et al., (2007)
Estonia	Silurian-Ordovician carbonaceous aquifer	0.01 to 7.2 mg/l	Indermitte et al., (2009)
Ethiopia	Geochemical characteristics	0.01 to 13 mg/l	Tekle-Haimanot et al., (2006)
Ghana, Keta basin	Mineral weathering	0 to 282.29 mg/l	Yidana et al., (2010)
Ghana	Fluorine enriched Bongo coarse grained hornblende granite and syenite suite	0.11 to 4.60 mg/l	Apambire et al., (1997)
Iran	Dolomite and limestone along with gypsum	0.7 to 6.6 mg/l	Looie and Moore, (2010)
Iran, Isfahan	Amphibole and mica group minerals in metamorphic and granitic rocks	0.2 to 9.2 mg/l	arzi et al., (2010)
Iran, Maku	Basaltic rocks	0.30 to 5.96 mg/l	Moghaddam and Fijani, (2008)
Jordan	Fluorite and calcite solubility	0.009 to 0.055 mg/l	Rukah and Alsokhny, (2004)
Kenya	Volcanic activity and chemical weathering	0.1 to 25 mg/l	Gaciri and Davies, (1993)
Korea, Gimcheon	Pegmatite	Up to 2.15 mg/l	Kim et al., (2010)
Korea	Granitic rocks	> 5 mg/l	Kim and Jeong, (2005)

Korea	Geological	0 to 40.8 mg/l	Chae et al., (2007)
Malawi	Geological, chemical and physical characteristics of the aquifer	1.65 to 7.5 mg/l	Sajidu et al., (2008)
Malawi, Lilongwe	Weathering of rocks containing biotite, dissolution of hornblende, fluorite and amphibole	0.5 to 6.98 mg/l	Msonda et al., (2007)
Mexico, Sonora	Deep regional flows, heating processes and fluorite dissolutions in granitic rocks	0.53 to 7.59 mg/l	Valenzuela-Va´squez et al., (2006)
Mexico, San Luis Potosí basin	Fluorite	2.10 to 3.65 mg/l	Carrillo-Rivera et al., (2002)
Norway	Lithological	Up to 8.26 mg/l	Banks et al., (1998)
Pakistan, Punjab	Phosphate fertilizers and coal combustion	0.11 to 22.8 mg/l	Farooqi et al., (2007)
Pakistan, Thar desert	Granitic rocks	1.13 to 7.85 mg/l	Naseem et al., (2010)
Poland	Anthropogenic	0.3 to 2.45 mg/l	Czarnowski et al., (1996)
Saudi Arabia	--	0.42 to 1.8 mg/l	Alabduláaly, (1997)
Sri Lanka	Granitic gneiss	0.01 to 4.34 mg/l	Young et al., (2010)
Sri Lanka	Fluoride in rocks	> 5 mg/l	Dissanayake, (1991)
Tanzania	Fluoride rich nephelinitie and carbonatitic rocks	15 to 63 mg/l	Nanyaro et al., (1984)
Turkey	Fluorite in limestone	1.5 to 13.7 mg/l	Oruc, (2008)
USA, Wisconsin	Felsic igneous and equivalent metamorphic rocks	0.01 to 7.60 mg/l	Ozsvath, (2006)
USA, Texas	Volcanic ash deposits	0.3 to 6.27 mg/l	Hudak and Sanmanee, (2003)
USA, South Carolina	Cryptocrystalline fluoroapatite	> 3.5 mg/l	South Carolina Ambient Groundwater Quality Report, (2003)
USA, Utah	Fluorite rich rocks	0.01 to 0.6 mg/l	Mayo and Loucks, (1995)
USA, Ohio	Bedrock	0.2 to 2.8 mg/l	Deering et al., (1983)

India

Of the 85 million tons of fluoride deposits on the earth's crust, 12 million are found in India (Teotia and Teotia, 1994). Hence it is natural that fluoride contamination is widespread, intensive and alarming in India. Endemic fluorosis is prevalent in India since 1937 (Shortt et al., 1937). It has been estimated that the total population consuming drinking water containing elevated levels of fluoride is over 66 million (FRRDF, 1999). Different parts of India where elevated concentration fluoride in groundwater as reported in literature are shown in Figure 3 and a detailed list of concentration of fluoride in groundwater and their sources in different places are given in Table 2.

Some regions in north western and southern India are heavily affected with fluorosis (Agarwal et al., 1997; Yadav et al., 1999). About 50% of the groundwater in Delhi exceeds the maximum permissible limit for fluoride in drinking water (Datta et al., 1996). Jacks et al. (2005) observed that high fluoride in groundwater in many parts of India was due to evapotranspiration of groundwater with residual alkalinity. Fluoride content was higher in deeper aquifers of Maharashtra (Madhnure et al., 2007) which was due to long residence time than shallow groundwater.

The rocks in southern India are rich with fluoride which forms the major reason for fluoride contamination in groundwater. It is a well established fact that groundwater in Nalgonda district, Andhra Pradesh, has high fluoride due to the inherent fluoride rich granitic rocks. The granitic rocks in Nalgonda district contain fluoride from 325 to 3200 mg/kg with a mean of 1440 mg/kg. The mean fluoride content in Hyderabad granites is 910 mg/kg (Ramamohana Rao et al., 1993). The Nalgonda granties contain much higher fluoride than the world average fluoride concentration of 810 mg/kg (Wedepohl, 1969). In Kurmapalli watershed, rocks are enriched in fluoride from 460 to 1706 mg/kg (Mondal et al., 2009). Calcretes act as a sink for fluoride. Co-precipitation and/or adsorption of fluoride by calcrete deposits in Wailapalli watershed had resulted in high fluoride in groundwater (Reddy et al., 2010a)

Brindha et al. (2011) found that when groundwater fluctuation was within 4.5 m below ground level, fluoride concentration was high when the water level was low and the fluoride concentration decreases with the rise in water table. This was due to direct evaporation of groundwater from these wells. If groundwater fluctuation was beyond 4.5 m below ground level the concentration of fluoride measured in groundwater after the monsoonal rains were higher than the preceding months. This was because evaporation resulted in the precipitation of fluoride rich salts on the soil which reached the groundwater along with percolating rainwater. The fluoride rich rocks form the main source for high fluoride groundwater in India. Also agriculture is intensively practiced in most parts of India. Hence it is possible that the fertilisers also add up to the sources of fluoride contamination in groundwater. Thus treatment of groundwater especially for fluoride before using it for drinking purpose is very essential in India.

Brindha, K. and Elango, L. (2011) Fluoride in Groundwater: Causes, Implications and Mitigation Measures. In: Monroy, S.D. (Ed.), Fluoride Properties, Applications and Environmental Management, 111-136.
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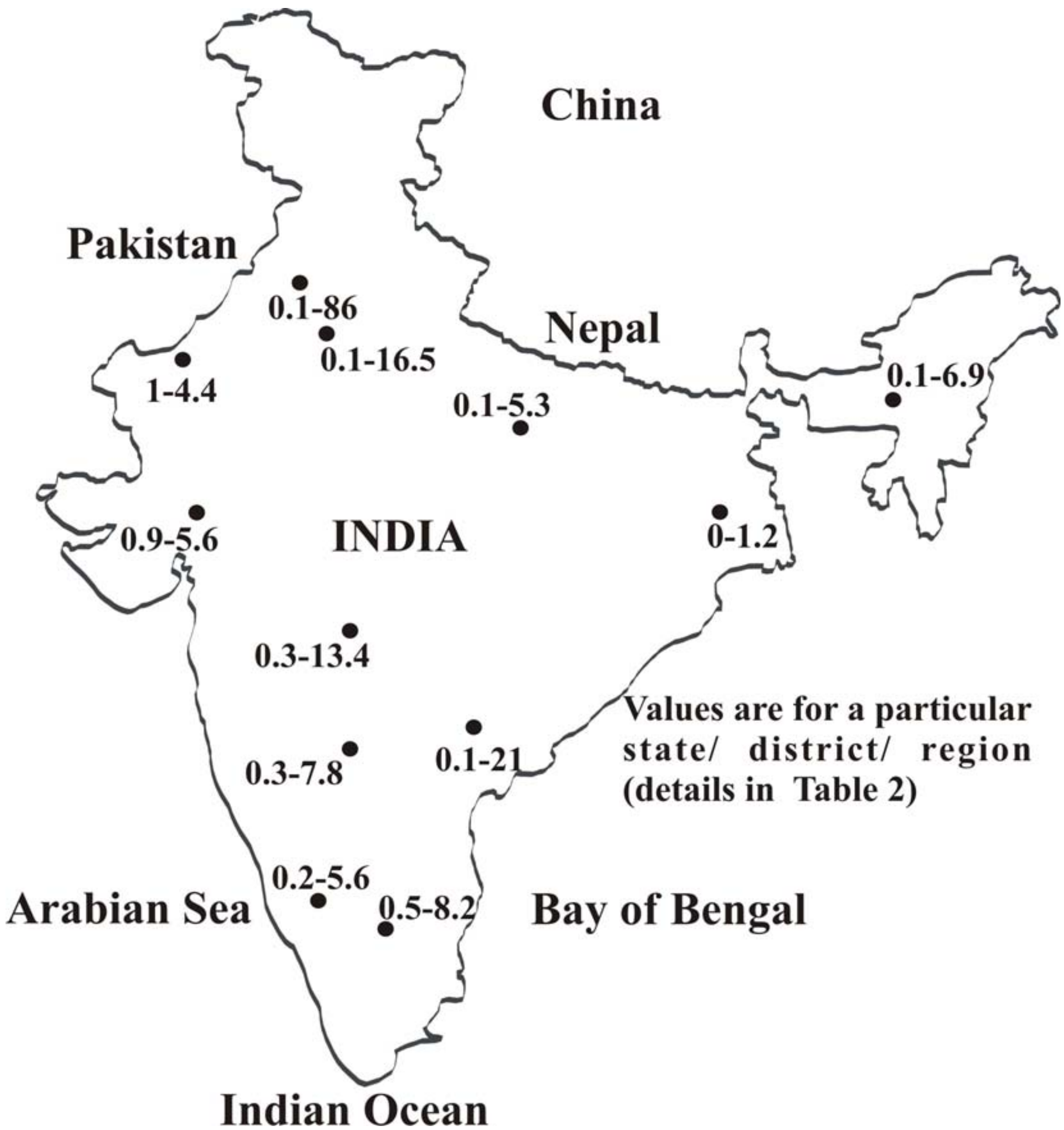

Figure 3. Range of fluoride in groundwater in India based on literature

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Table 2. Concentration of fluoride in groundwater in India and its sources based on literature

State, district/place (in aphabetical order)	Source	General range of fluoride concentration in groundwater	Reference
Andhra Pradesh, Kurmapalli watershed	Fluoride rich rocks	Up to 21.0 mg/l	Mondal et al., (2009)
Andhra Pradesh, Nalgonda	Fluoride rich grantic rocks	0.4 to 20 mg/1	Ramamohana Rao et al., (1993)
Andhra Pradesh, part of Nalgonda district	Fluoride rich granitic rocks	0.1 to 8.8 mg/l	Brindha et al., (2011)
Andhra Pradesh, Vamsadhara river basin	Pyroxene amphibolites and pegmatites	Up to 3.4 mg/1	Srinivasa Rao, (1997)
Andhra Pradesh, Visakhapatnam	Granitic rocks	0.6 to 2.1 mg/l	Subba Rao, (2009)
Andhra Pradesh, Wailapally watershed	Hornblende, biotite, apatite, fluorite and fluoride rich calcretes	0.5 to 7.6 mg/l	Reddy et al., (2010a)
Andhra Pradesh, Wailapally watershed	Fluorite bearing rocks	0.97 to 5.83 mg/l	Reddy at el., (2010b)
Andhra Pradhesh and Jharkhand	Coal ash	0.1 to >4 mg/l	Prasad and Mondal, (2006)
Assam, Guwahati	Granite	0.18 to 6.88 mg/l	Das et al., (2003)
Delhi	Irrigation water and brick industries	0.1-16.5 mg/l	Datta et al., (1996)
Gujarat, Mehsana	Granite, gneiss and pegmatite	0.94 to 2.81 mg/l	Salve et al., (2008)
Gujarat, Mehsana	Calcite and dissolution of dolomite	1.5 to 5.6 mg /l	Dhiman and Keshari, (2006)
Haryana, Bhiwani	Rock	0.14 to 86 mg/l	Garg et al., (2009)
Karnataka, Bellary	Apatite, hornblende and biotite	0.33 to 7.8 mg/l	Wodeyar and Sreenivasan, (1996)
Keral, Palghat	Hornblende and biotite gneiss	0.2 to 5.75 mg/l	Shaji et al., (2007)
Maharashtra, Yavatmal	Amphibole, biotite and fluoroapatite	0.30 to 13.41 mg/l	Madhnure et al., (2007)
Rajasthan, Hanumangarh	Fluoride bearing host rocks	1.01 to 4.42 mg/l	Suthar et al., (2008)
Tamil Nadu, Erode	Host rocks and weathering of fluorite	0.5 and 8.2 mg/l	Karthikeyan et al., (2010)
Uttar Pradesh, Kanpur	--	0.14 to 5.34mg/l	Sankararamakrishnan et al., (2008)
West Bengal, Hooghly	Super phosphate fertilizer	0.01to 1.18 mg/l	Kundu and Mandal, (2009)

MITIGATION MEASURES

Everybody needs clean water. When high fluoride in the drinking water source has been identified, it is better to avoid that source and look for other sources. But this is not a long lasting solution. Insitu and exsitu methods are available to treat groundwater with high fluoride and bring it to the usable form.

Insitu-treatment methods

Insitu method aims at directly diluting the concentration of fluoride (in groundwater) in the aquifer. This can be achieved by artificial recharge. Construction of check dams in Anantapur district, India has helped widely to reduce fluoride concentration in groundwater (Bhagavan and Raghu, 2005). Rainfall recharge also called as rainwater harvesting can be adopted using percolation tanks and recharge pits which may prove helpful. Recharge of rainwater after filtration through the existing wells can also be planned to improve the groundwater quality.

Exsitu-treatment methods

Numerous exsitu methods are available for defluoridation of water either at household or community level. Adsorption method involves the passage of water through a contact bed where fluoride is adsorbed on the matrix. Activated charcoal and activated alumina are the widely used adsorbents (Chidambaram et al., 2003; Chauhan et al., 2007). Brick, bone char, fly ash, serpentine, red mud, waste mud, rice husk, kaolinite, bentonite, charfines, ceramic etc. are some of the other absorbents capable of effectively removing fluoride from groundwater (Srimurali et al., 1998; Çengeloğlu et al., 2002; Chidambaram et al., 2003; Yadav et al., 2006; Sarkar et al., 2006; Castillo et al., 2007; Ma et al., 2008; Kemer et al., 2009; Tor et al., 2009; Ma et al., 2010; Ganvir and Das, 2010; Chena et al., 2010). The effective removal of fluoride by these absorbents depends on the initial concentration of fluoride, pH, contact time, type of absorbent and its size.

In ion exchange process, when water passes through a column containing ion exchange resin, the fluoride ions replace calcium ions in the resin. Once the resin is saturated with fluoride ions, it is backwashed with solution containing chloride such as sodium chloride. The chloride ions thus again replaces the fluoride ions in the resin and is ready for reuse. But the backwash is rich in fluoride and hence care should be taken in disposing this solution. Similarly in precipitation methods, the disposal of sludge with concentrated fluoride is a great problem. Precipitation involves addition of chemicals such as calcium which results in the precipitation of fluoride as fluorite. Aluminum salts are also used for this process. Nalgonda technique which is a well known technique uses alum, lime and bleaching powder followed by rapid mixing, flocculation, sedimentation and filtration. This was developed in India by National Environmental Engineering Research Institute to serve at community and household levels. The resulting sludge from this process contains high amount of aluminium and fluoride, the disposal of which is yet another problem. These above mentioned ex-situ methods are simple and cost effective.

Membrane processes is also an ex-situ technique which includes methods called reverse osmosis and electrodialysis. These are advanced techniques which require high cost input. Both these methods use a semipermeable membrane which removes dissolved solutes from the water when they pass through them. But the negative point is that even the ions which are essential for the human body are also removed. The difference between these techniques is that reverse osmosis works on pressure while electrodialysis works on direct potential. Also reverse osmosis can be practiced at household level whereas electrodialysis involves huge set up and is even more expensive. All these methods have their own advantages and disadvantages. Hence it is necessary to evaluate the prevailing local conditions and cost effectiveness before choosing a particular defluorination method for an area.

Apart from all these it is essential to create environmental awareness among public regarding the ill effects of high fluoride. Reduction in the use of fertilisers, especially phosphatic fertilisers is important. It is better to adopt organic farming in places of fluoride threat. In countries with high temperature, it is advisable to reduce evapotranspiration by increasing vegetation cover. This will prevent the deposition of fluoride salts on the unsaturated zone which will subsequently reach the groundwater during rainfall. Other way of combating fluorosis is to modify the dietary intake of the people. Food with more calcium and vitamin C can prevent fluorosis to a certain extent. Usage of coal for combustion indoors should be avoided and the resultant fly ash obtained from combustion of fossil fuel in industries has to be disposed cautiously.

SUMMARY AND CONCLUSION

It is evident from studies by several researchers worldwide that fluoride in groundwater has been a potential problem to human society. The main source of fluoride in groundwater is the rocks which are rich in fluoride. Fluoride occurs in sellaite, fluorite, cryolite, fluorapatite, apatite, fluormica, biotite, amphibole and several other rocks. Weathering of these rocks and prolonged residence time leads to high fluoride groundwater. Low calcium, high sodium and high bicarbonate are typical of high fluoride groundwater. Volcanic ash and combustion of coal are the next major source for fluoride. The other sources for fluoride are infiltration of agricultural runoff containing chemical fertilisers, improper disposal of liquid waste from industries, alumina smelting, cement production and ceramic and brick firing. Some amount of fluoride is essential for the human body for healthy teeth and bones. But when they are present above the recommended limit of WHO and BIS i.e. 1.5 mg/l it results in mild dental fluorosis to crippling skeletal fluorosis as the quantity and period of exposure increases. Dental fluorosis is more prevalent in children than in adults. Skeletal fluorosis occurs when an individual is exposed to fluoride of above 10 mg almost every day over a period of one or two decades. Apart from fluorosis there are also several health disorders due to ingestion of drinking water with high fluoride. To remediate the groundwater with high fluoride, defluorination techniques are adopted. They include adsorption, ion exchange, coagulation and precipitation, reverse osmosis and electrodialysis. Of these, reverse osmosis has been considered as the best available technology. Onsite treatment includes artificial recharge methods such as rain water harvesting, constructing check dams, percolation ponds, facilitating recharge of rain water through existing wells etc. Adopting a particular method depends on the initial fluoride concentration, source and cost effectiveness in an area.

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