FLUORIDE CONTAMINATION ISSUES OF GROUNDWATER IN RAJASTHAN: A REVIEW

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ABSTRACT

This paper depict the major groundwater contamination that is fluoride, it is the common widespread pollutant worldwide. This paper gives idea about distribution, sources and defluoridation of this pollutant. Groundwater exploitation and contamination is common issue in this state. Fluoride is the major issue in almost all the district of the state while common problem. Throughout the world fluorosis is one of the common issue related to drinking water and human health, around 200 million people with 6 million children globally suffered from fluorosis.Pure potable water is still a challenge for world, yet one billion people suffer for safe drinking water and likewise global attention seek towards resolving the groundwater quality problem mainly in developing countries. Rajasthan is the largest state with huge number of population but having only 1% of India's total water resources. Fluorine is chemically reactive and it is the most electronegative univalent gaseous of halogen family and it is the 9th element of the periodic table. It is found in very small quantity in water, plant, animal and air. China and India are two most common countries suffer from endemic fluorosis whereas in India, Rajasthan is having the dreadful condition with all the 33 district having fluoride contamination. This review paper contains the present status of fluoride of all the 33 district of Rajasthan and its hostile effect.

Keywords: Groundwater, Fluorosis and Defluoridation

Introduction:

India is the agrarian country and 68% of workforce directly or indirectly depends on agriculture (WHO 2015). Many regions in the country are under the grip of economy and environmental stock, and most groundwater is being used for agriculture due to unavailability of surface water. In India 50% of urban areas and 80% of household needs in rural region fulfil by groundwater which is under threat due to existence of various contamination like arsenic, fluoride, nitrate, iron and salinity

(Ayoob S & Gupta A K, 2006). Rajasthan is a state in north India, having 10.4% of total geographical area of India. It covers large geographical area of the India. The major problem of the world's groundwater from past few years is arsenic and fluoride. Fluoride is one of the most important elements for maintaining human health and life. It is the most electronegative elements. When it is present in small quantity it normalizes bones and help in formation of dental enamel (Chouhan and Flora, 2010). It is the lightest member of halogen family and most electronegative among all the chemical elements. Nearly about 200 million people in worldwide including 6 million children are suffers from serious health risk because of intake of fluoride in their drinking water (IWMI). Fluoride above the permissible limit (1.5 mg/l) reveals detrimental effect on teeth and bones and the disease caused by it is called fluorosis and also non skeletal diseases is caused such as anaemia, fatigue and ache, pain in joints, polydipsia, muscles weakness and low haemoglobin. Fluoride enters in the human body mainly through drinking water and also from food that contain fluoride, crops that grow in fluoride bearing soil and some other. Fluorosis is endemic in many parts of the world including India and in India Tamil Nadu, Jharkhand, Rajasthan, Uttar Pradesh, Andhra Pradesh, Gujarat, Punjab and Haryana. Fluoride contamination is important toxicological and geo-environmental issues in India. Fluoride is mainly contributed due to geogenic process but now a days it also deposited in the environment due to anthropogenic activities i.e. phosphate based fertiliser used in agriculture land (Handa 1975). Fluorite, fluorspar, fluorapatite these are the principle sources of fluoride and found in granite, granite gneisses and pegmatite (Rama Rao 1982). Fluoride concentration less than 0.8mg/l shows dental caries. Therefore it is necessary to maintain fluoride concentration between 0.6 to 1.2 mg/l in drinking water (WHO 1984). If the limit is below 0.6 mg/l the water resources need to be rejected. The maximum and minimum fluoride concentration which is prescribed by the Indian Council of Medical Research, are 1.0 to 1.5 mg/l respectively. Fluoride containing compounds are used in various industries and in other sources such as ceramic industries, fertilizer industries, anhydrous hydrogen fluoride is used in production of refrigerant, herbicides, plastic, electrical component, aluminium, pharmaceuticals and in many others (Siddiqui 1970). Volcanoes are the one of the major source for hydrogen fluoride while sodium fluoride has been used as a fungicides, insecticides and rodenticides. Groundwater contamination due to fluoride minerals was

first reported in Prakasham district of Andhra Pradesh in 1937 (Short et al 1937). In India fluoride is contributed in groundwater mainly due to geogenic process and geological sources. Rock water interaction is the process from which fluoride bearing minerals dissociated from sources and added to groundwater through dissolution (Gupta et al. 2006).

Method:

Search and available sources strategy:

In this review published article and literature is systematically searched and collected from official sites until 2018. Also published article in various national and international institutes is used as reference study with having professional criteria in mind.

Sources of fluoride:

Fluoride is basically geogenic and present 0.06 to 0.09% in earth's crust and the amount of fluoride is influenced by local & regional geological condition of any area. Major fluoride bearing rocks are quartzites, basalts, pegmatite, hornblende, biotite, muscovite, fluoraptite, fluormica, fluorite, cryolite, delorites. Fluoride in groundwater varying from geological settings and types of rocks present there. Besides these rocks alkali rocks and hydrothermal solution also contribute in fluoride pollution whereas clay mineral is the main sources for fluorine in soil (McGown & Suttie 1977). Igneous rock and volcanic rocks have fluorine accumulation from 100 ppm to < 1000 ppm (Frencken et al. 1992). Generally fluoride aggregate during magmatic crystallization and differentiation process and the remaining magma enhanced in fluorine. Groundwater from crystalline rocks mainly (alkaline) granites deficient in calcium are especially prone to high fluoride accumulation and these types of rocks are found mainly in Precambrian basement areas. Sedimentary rocks have fluorine accumulation from 200 ppm up to 1000 ppm i.e. limestone & shales (Frencken et al. 1992). In carbonate sedimentary rocks the fluorine is exist as fluorite. Clastic sediments have greater fluorine absorption as the fluorine is determined in micas and illites in the clay fractions. High accumulation may also be creating in sedimentary phosphate bed or volcanic ash layer. Metamorphic rocks have a fluorine concentration from 100 ppm up to more than 5000 ppm (Dissanayake et al. 1986). When fluoride rich minerals that is present in rocks and soils, came in contact with highly alkaline water they release fluoride in water by hydrolysis process replacing hydroxyl ion. Due to weathering of rocks the calcium-magnesium or carbonate deliberation that is mainly form in arid and semi-arid areas

seem to be good sink fluoride ion (Hussain J. et al. 2004). Fluorosis demonstrated mainly in three ways: skeletal fluorosis, dental fluorosis and non-skeletal fluorosis. Intake of fluoride above 1.5 mg/l in drinking water cause dental fluorosis that is determined by mottling & pitting of teeth, chipping of enamel, staining, yellowish to brownish colour of teeth (Madhnure et al. 2007). Due to the electronegativity character of fluoride it is attracted to positively charged calcium in bones and teeth (Rao Subba. 2003).

These are the following reasons responsible for high fluoride contamination in groundwater:

Rock-water interaction:

It is the main reason for high fluoride in arid and semi-arid reasons with presence of some fluoride bearing rocks and rocks geochemistry. Physiological condition of rocks like decomposition, dissociation & dissolution of rocks for long time contribute to the fluoride contamination. Solubility of fluoride in groundwater differ from one rock type to other

Contact time:

The ultimate concentration of fluoride in groundwater for the most part depends on reaction times with geological formation minerals.High fluoride concentra tions are often engineered up in groundwater's that have long residence times within the aquifers. Such groundwater is sometimes relatedto deep g eological formation systems and slow groundwater movement Shallow aquifers that contain recently infiltrated rain sometimes have low fluoride (Kanduti Domen et al., 2016). Exceptions will occur in shallow aquifers settled in active volcanic areas full of hydrothermal alteration. Below such conditions, the solubility of fluorspar will increase with increasing temperature and fluoride could also be superimposed by dissolution of HF gas.

Climate:

Arid regions are intent to high fluoride concentrations. Here, groundwater flow is slow and the contact times with rocks are finally high. The fluoride contents of water might increase throughout evaporation process if resolution remains in equilibrium with calcite and pH is bigger than hardness (Das S. et al. 2000). Dissolution of evaporation salts deposited in arid zone is also a vital supply of fluoride. Fluoride increase is a smaller amount pronounced in wet tropics, thanks to high rain inputs and their diluting impact on the groundwater chemical composition.

Chemical characteristics of groundwater:

Mainly fluoride in groundwater related to Na-HCO3 type of water and comparatively little calcium and magnesium

concentrations. This type of water have basically high pH values which is more than 7.

Anthropogenic sources:

Anthropogenic sources also contribute a good amount of fluoride in the environment from food materials, drinking/beverage materials, canned fruit/food, milk and other products also vegetable that grow in the soil which enriched with fluoride minerals shows a high amount of fluoride which is intake by humans. Industries as well as fertilizers based company especially phosphate based contribute fluoride in the environment. Artificially high fluoride soil happens by contamination through the application of fertiliser, sewage sludge and pesticides (Kanduti Domen et al. 2016). Movement of F in soil is complex and principal factors that control the amount of F in soil solution are clay minerals, absorption Ca & P in soils and pH of soil also absorption rate is high when pH of soil is 6 to 7. The amount of fluoride vary in different food items like milk has very low F concentration while tea leaves contain a good amount of F. Factors that control F content in groundwater is ratio of soluble & insoluble F in source rocks, contact time of water with rock, rainfall pattern, soil temperature & oxidation-reduction process.

Fluoride contamination in Rajasthan:

Rajasthan is the driest state of India and support only 1% of water resources and also receives very less rainfall. Due to absence of surface water groundwater exploited on regular basis for drinking and other common uses. Rajasthan supports 15 rivers out of which only 2 are perennial i.e. Chambal & Mahi (Munoth et al 2015). Total area covered by this state is 3, 42,239 sq.km and total blocks / districts of the area is 236 / 32 respectively (Ministry of water resources, Government of India). The state is divided into three hydrogeological unit's i.e. unconsolidated sediments, semi-consolidated sediments and consolidated rocks. This state has high number of fluoride affected areas with average concentration of 16 mg/l due to presence of fluoride bearing rocks and minerals in the area. Ajmer, Alwar, Banaswara, Barmer, Bharatpur, Bhilwara, Bikaner, Jaipur, Dausa, Rajasamand, Hanumangarh, Pali, Nagur, Sirohi, Ganganagar these are the major fluoride affected area of the state. Fluoride deposits due to weathering of rocks, arid climate, presence of various fluoride bearing rocks & minerals, velocity of flowing water, temperature, pH, due to concentration of major ions in water (Kala Jayshri Prof Sharma Gunawat et al., 2014). Geology and geographical setup of the area downfall the water quality and it receive only 5mm to 20 mm rainfall annually. According to the study of Rajasthan voluntary health association in 1994 total

2433 village with almost 2.6 million people affected by fluoride problem.

Effect of fluoride on human health: Skeletal and dental fluorosis:

Fluorine is chemically reactive electronegative univalent gaseous halogen found in small amount in the water, air, plants and animals. Fluoride is important element for the formation and maintenance of bones and teeth. In the earlier phase of 1900s people living in the U.S. residential area suffered from brown stains on their teeth this is directly related to fluoride in drinking water and as research continued researchers reveals that optimal amount of fluoride in drinking water is necessary for proper maintenance of teeth and bones. Human health disorder caused by chronic exposure of high concentration of fluoride responsible for fluorosis. Fluorosis is one of the serious problem related to drinking water globally 200 million people with 6 million children suffered from endemic fluorosis (). Initially fluorosis attack on skeletal tissue and people having bone deformities, pain and proximal muscles weakness. Mottled teeth, discolouration, blackened or chalky white are the clear indication high exposure of fluoride in human body which is called dental fluorosis while skeletal fluorosis increase bone mass and density and fluoride get deposited in all the joints which makes difficult in walking and movement. Others problem that is also related to fluorosis are low haemoglobin, nausea, gastrointestinal problems, skin rashes, neurological manifestation, kidney failure (Meenakshi et al. 2006). Dietary intake of fluoride in different food items are shown given below in the table.

Table 1. Concentration of fluoride in different food items

Types of food	Concentration (ppm) 1mg/l = 1ppm
Black tea	3-5
Cucumber, onion, celery	0.01
Apple	0.03
Yoghurt	0.12
Shellfish product	2-3
Wine	1-2
Green tea	1.2
Eggs	0.01
Tomato	0.02
Peach, strawberry	0.04
Boiled or ram beef	0.22
Chicken meat	0.15
Radish	0.06
Boiled pasta	0.07

Source: United State Agriculture Department (USDA) 2005.

Fluoride level in the food items also depends on the materials used in it such as cookware used to prepare food like Teflon. About 90% of the fluoride consumed accumulated in gastrointestinal tract and

remaining 10% is excreted through faces. In pregnant women uptake of fluoride concentration in placenta is depend on the accumulation of fluoride in their bloodstream (). It is noted IQ of the children living in high fluoride area are mostly low. Fluoride mainly enter into human body through consumption of water and then food.

Neurotoxical effect of fluoride:

Various study has been conducted on animal and human for the study of fluoride content in the environment. IQ level of children in China deficits due to fluoride exposed more than 2.5 – 4 mg/l in drinking water. Fluoride caused neurotoxicity research performed in laboratory on animal reveals that it affects learning and memory of children. An animal study on rodent reported change in the behaviour of it due to fluoride treatment but the researcher didn't find any considerable change in magnitude. Some molecular, cellular and anatomical change in nervous system were seen which is due to certain physiological and environmental circumstances.

Effect of fluoride on reproductive health:

Now a day's problem of infertility is common and a study by Freni C. Stan reveals that with increasing fluoride concentration decreases birth rate. To calculate fluoride rate sociodemographic covariables were used from which total annual fertility rate were observed which decreased gradually.

Tremendous effect of fluoride on human health due to toothpaste:

All the previous research proves that use of fluoride based toothpaste before the age of 6 increase the risk of fluorosis. Children below the age group of 6 years swallow can swallow the up to 25 to 35% of toothpaste during their each brushing because their swallowing reflex is not fully developed (Baxter PM 1980). All the possibility that strengthen the risk of fluorosis in children due to toothpaste is basically the children begin brushing before the age of 2 years while they are also not supervised some children swallow about 0.5 mg of fluoride in each brushing (Levy SM 1993). According to Mascarenhas AK & Burt BA 1998 risk of fluorosis were high in children who begun their brushing before the age of 2 years and also male were more severely suffer from fluorosis risk than females because fluorosis is prevalent in later developing teeth like premolars and second as teeth in males develop later than females. A crude risk to the child who use toothpaste before the age of 6 years.

Methods of fluoride removal:

Fluoride concentration in water varies fluorite solubility in calcium poor aquifers and high fluorite concentration mainly found in groundwater because it is mostly derived through. It is naturally present in soil, water, plant and animal in very small quantity. There are various methods used for defluoridation: - Ion exchange and coagulation, membrane technologies, nalgonda technique, biosorbent and many more which are not yet established.

Nalgonda method:

Nalgonda is also one of the common method for fluoride removal lime and alum are the most commonly used coagulant but there are some limitations with this method, it need daily addition of chemical, it is very less effective for the water having high TDS & hardness and produce large amount of sludge. When alum is added to water two reaction are formed first one is alum reacts with alkalinity to supply insoluble aluminium hydroxide and in second one alum reacts with fluoride ion in water. Best fluoride is adept at pH 5.5 to 7.5 (Razbe N. et al. 2013). Nalgonda technique evolved by NEERI, Nagpur it is a simple and economical method used for defluoridation. It involves bleaching powder, aluminium salt and lime by rapid mixing, flocculation, sedimentation and filtration. Aluminium salt is only responsible for elimination of fluoride the dosages of aluminium salt increase with increase in fluoride and alkalinity in the water. The dose of lime is $1/20^{th}$ by the dose of aluminium salt, lime responsible for making dense floc for rapid settling and bleaching powder is added to the raw water at 3 mg/l rate for disinfection (Padmashri J.P. 2001). This process is suitable for small amount of water only. It is easy to use because of no handling of caustic acids and alkalies, simple in design, construction, operation and maintenance, no regeneration of media. While there are some demerits of Nalgonda technique it need high dose of (Al (OH) 3 and use of aluminium sulfate increase sulphate ion concentration greatly in water which causes catharatic impacts in humans (Dahi E. 1996). This is only useful for small population such as 200 people and take about 2-3 hours. Also an important disadvantage of this method is gives high residual aluminium concentration i.e. 2-7 mg/l whereas allowable limit is 0.2 mg/l.

Membrane process:

Membrane process achieved great importance from few decades in industrial viewpoints. It purify wastewater and desalinize sea water, its probability to remove fluoride ion and other detrimental chemical constituents from water is

influencing. In membrane separation process there are two or more than two mixture are taken, is partially apart from each other with the help of semi-permeable membrane from which species move faster than other. The membrane must not be disintegrate or dissolve (singh kalpana et al 2013). The semi-permeable membrane or barrier should be porous polymeric film or non-porous, ceramic film or metal material or also liquid, gas or solid whereas most popular membrane separation process are donnan dialysis, electrodialysis, reverse osmosis and nanotechnology.

Donnan Dialysis: This is similar to ion exchange membrane process while different from electromembrane process which shows that it does not need electrical current but relatively a difference in chemical potential, it is also called diffusion dialysis (S. Sanghratna et al 2015). It use diluted solution with several anion-exchange membrane

Electrodialysis: Simply it is the removal of ionic component from aqueous and electrolytic feed solution with the use of ion selective membrane process with the help of electric force. It is just like reverse osmosis including electric current instead of pressure to separate ionic contamination of water. In this method multiple electrodialysis cells are used and arranged into a configuration with alternating anion and cations exchange membrane, for experimental procedure of this method two detachable chamber made of Teflon were used. Initially SB 6407 anion exchange membrane were placed between these two chambers (Ergun Erdem et al 2008).

Reverse Osmosis: Reverse Osmosis gives highly pure water. S. Sanghratna and Arfin Tanvir studied that RO is basically a membrane based process where anions are removed by applying pressure on feed water. Feed water passes under high pressure through semi-permeable membrane, where water seeps the small pores of the membrane and is brought as a filtered water. While a solution is alienated from pure solvent by semi-permeable and the pressure applied on the solution is more than the osmotic pressure, the solvent start flowing from the solution towards the pure solvent, this is purely known as reverse osmosis (Singh Kalpana et al., 2012). The reverse osmosis process is very costly but it is very effective in fluoride removal too. This process is basically used for desalination and it is membrane based process. Reuse of membrane-based seawater desalination and wastewater proves as excellent way to decrease water scarcity and augment water shortage (S. lee et al., 2010). Whereas revesre osmosis and electrodialysis are most appropriate for brackish water desalination while RO

serves distillation process also (Kalogirou., 2005). RO process is very efficient but it is not suitable for rural areas as well as it makes water acidic and need pH improvement. RO technique remove important minerals from water which is used for fitting development so remineralization of water is necessary.

Nanotechnology: According to the researchers it is concluded that nanofilteration and reverse osmosis effectively reduce fluoride up to 95% to 76% respectively. Nanotechnology is the latter innovation in terms of membrane process. NF delivers slightly larger pores as compare to RO and readily allow the entry of solute and solvent that means less energy, lower pressure requirement and elimination of solute very low and flow rate very fast (Arfin Tanvir, 2015). This method is basically for portable water and removes harmful contaminates like pesticides, chloride and nitrate. Nanotechnology is advance as compare to RO and lots of experimental and theoretical study is being carried out to mark clear view on the mechanism of solute retention (Mohapatra M. et al., 2009). According to Kalplana Singh et al., Nanofilteration membrane NF90 most appropriate for fluoride removal.

Adsorption technique:

This method is quite possible in recent year because of its simplicity, low cost, high accessibility and gives highly purified water. Adsorbent of fluoride over solid adsorbent occurs mailnly on three basic steps (Stanic 2014):

a. Transport of fluoride ion to the external surface of the adsorbent from the bulk solution across the boundary layer surrounding the adsorbent particle, which is called external mass transfer.
b. Adsorbent of fluoride ion on to particles surface.
c. The adsorbed fluoride ion probably exchanged with the structural element inside adsorbent particle depending on the chemistry of solids or adsorbed fluoride ions transferred to the internal surface for porous materials.

Higher adsorption capacity of fluoride shown by various metal oxides and hydroxide, especially prepared in nano-form. Hydrous titanium dioxide (TiO_2) has been found extremely powerful for removal of fluoride (Fan et al., 2003). There are lots of adsorbent materials but it is difficult to compare all because of its variation in data and parameters like pH, particle size, temperature, presence of different ions, initial concentration of fluoride, ionic

strength etc.). Whereas it was concluded that pH is the dominant factor of in fluoride concentration, it has been observed that adsorption of fluoride is high where pH is acidic or closer to neutral and concentration decrease if pH increase. Second important factor is the type of ion present and its concentration pin treated water. Mirna et al,. 2014 conclude that metal oxides and hydroxides various titanium, iron and aluminium oxides and hydroxides were commonly tested and indicated the maximum adsorption capability over wide range of pH as well as great selectivity for fluoride removal.

Conclusion:

A brief review on fluoride presented in this article. It demonstrate from above literature that fluoride content in water in Rajasthan mainly due to geogenic (rock and mineral) effect and some contribution due to anthropogenic activity also. Fluoride does not bring any change in colour, odour or taste and it acts as invisible poison. Fluoride is one of the important beneficial micronutrient for human body. According to WHO 0.6 mg/l/day of fluoride intake are enough for beneficial effect while 2 mg/l/day in fluoride rich area. For the valuable effect of fluoride people must be very alert while consuming fluoride in edible products. It can be toxic when consume in very high concentration otherwise not. Variety of process available for the defluoridation. Simplest and traditional method is adding lime and bleaching powder to the contaminated water. There are huge number of defluoridation method available some of them are actual while others are cost effective or ineffective. Broadly researchers discuss about membrane process, adsorption, and precipitation, electrochemical. Adsorption is conventional method which include various adsorbent such as carbon based material commonly use adsorbent belong to carbonaceous group and activated carbon, clay and soils, alumina based material and many more. Membrane peocess include reverse osmosis, nanofilteration, elctrodialysis and donan dialysis. Groundwater of the area highly affected with fluoride and nitrate, a thorough study is necessary before the use of it for domestic and drinking purpose.

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