Available online <u>www.jocpr.com</u>

Journal of Chemical and Pharmaceutical Research, 2015, 7(11):292-305



Review Article

ISSN: 0975-7384 CODEN(USA): JCPRC5

Groundwater pollution and adverse effects on health by fluoride ions

M. Suneetha¹, B. Syama Sundar¹ and K. Ravindhranath^{*2}

¹Department of Chemistry, Acharya Nagarjuna University, Guntur, A.P., India ²Dept. of Chemistry, KL University, Vaddeswaram, Guntur Dist., A.P., India

ABSTRACT

Water is one of the most vital components for all forms of life and is indispensable to the maintenance of life on the earth. Water may be contaminated by natural sources or by industrial effluents. One of such contaminants is fluoride as it has a profound effect on teeth and bones when the concentration in drinking waters exceeds the permitted limits. Concentrations in the range of 1.0-1.5 mg/lit strengthens the enamel and in the range of 1.5-4.0 mg/lit results dental fluorosis whereas with prolonged exposure to higher concentrations (4-10 mg/lit) results skeletal fluorosis in children as well as adults. The fluoride concentrations in groundwater more than 30 mg/lit occur widely in many parts of the world such as India, Pakistan, West Africa, Thailand, China, Sri Lanka, Southern Africa, United States of America, Argentina, and Ethiopia. This article reviews the present scenario of groundwater pollution and a number of adverse effects on health caused by fluoride ions.

Key words: Groundwater pollution, fluoride ions, fluorosis

INTRODUCTION

Water Pollution:

Safe drinking water is indispensable life sustaining essential needs of living creatures. This wonderful gift of nature is being contaminated by the human beings in the advent of modernization, industrialization, urbanization. The other sources of pollution are improper disposal of sewage, agricultural runoff, urban storm water runoff and chemical wastes dumped by industries (Figure 1). These pollutants are detrimental to human health and also to the microorganisms living in the water bodies and to the biota and plants.

The World Health Organization estimates that safe drinking water could prevent 1.4 million child deaths from diarrhea each year [1]. It has been suggested that water pollution is single largest leading worldwide cause of diseases and deaths and that it accounts for the deaths of more than 14,000 people daily and 11,000 of them are children under the age of five years [2-3] and 1.8 million people die every year from diarrheal diseases including cholera [4].

The use of contaminated surface water sources such as lakes, ponds, rivers and rainwater sources contribute to the transmission of water-borne diseases, including cholera, dysentery and typhoid. And also the increasing population and necessities have led to the deterioration of surface and sub surface water [5]. However, around 300 million people still live in absolute poverty in both urban and rural areas, and often lack access to clean drinking water and basic sanitation; nearly half of the population is illiterate, not at all aware of the waterborne diseases affecting their

health. The World Health Organization and UNICEF proposed the large-scale use of tube wells for drinking water, anticipating that groundwater would be relatively free of the contaminants plaguing surface water.



Figure 1: Various sources of groundwater contamination

Groundwater has long been regarded as the pure form of water compared to surface water because of the purification of the former in the soil column through anaerobic decomposition, filtration and ion exchange. This is one of the reasons for the excessive consumption of groundwater as the drinking water in both urban and rural areas all over the world [6-7]. In India about 80% of the rural population and 50% of the urban population use groundwater for domestic purposes [8]. Unfortunately, it has been reported that more than 33% of the country's groundwater resources are unfit for consumption [9] as they are contaminated with a wide variety of pollutants and the reasons for which is mostly geological in nature. In many parts of India, the groundwater pollution is attributed mainly to the presence of excessive fluoride, arsenic and nitrate. Amongst the most notable of these contaminants is fluoride [10]. So, fluoride is a one of the main groundwater contaminants in India and high fluoride levels in drinking water posing severe threat to human health.

Classification of Water Pollutants:

The sources and causes of water contamination are numerous, and are as diverse as human activities [11]. The major sources of water pollution can be classified as: Municipal, Industrial and Agricultural. The effects of water pollution are varied and they depend on what chemicals are dumped and in what locations. Industries discharge a variety of pollutants in their waste water including anionic and cationic contaminates, heavy metals, organic toxins, dissolved inorganic compounds, oils, nutrients, solids and dyes.

Organic Pollutants:

These are mainly resulting from human activities and to a lesser degree even due to natural processes. These natural processes usually take place near the surface in the humus-containing soil, but may also be present in deeper layers where peat, lignite, coal, or even shallow oil deposits are present and in contact with ground water. Through metabolism, decay, dissolution, advection, and other processes, organic compounds are formed and become part of the groundwater system. Humic acids, hydrocarbons and pectins are amongst the natural organic substances occurring in ground water.

Inorganic Pollutants:

This category of water pollutants consists of metal compounds, finely divided metals, inorganic salts, trace elements, mineral acids, metallic complexes, organo-metallic compounds and poly phosphatic detergents etc. The presence of various hazardous contaminants like fluoride, nitrate, pesticides, arsenic, other heavy metals, etc. in under groundwater has been reported from different parts of India [12-17]. The distribution of these constituents largely depends on the type of geological formations in contact with the groundwater flowing through. Fluoride is a potential pollutant in India and it is mainly of natural origin found in groundwater and it leaches into the groundwater due disintegration and dissolution of igneous and metamorphic rocks containing minerals such as

amphiboles and micas [18]. Further, the effluents from chemical, metallurgical industries and coalmines also contribute to the fluoride contamination in surface waters.

Occurrence of Fluoride:

Fluorine is a ubiquitous and the 13^{th} most abundant naturally occurring element in the Earth's crust, one of the most reactive and electronegative of all the elements [19]. Fluorides account for 0.06-0.08% of the Earth's crust, but their average abundance is low (300 mg kg⁻¹) [20] and the content of fluorine in the lithosphere varies between 100 and 1500 g/ton. Fluorine cannot be found in nature in its elemental state, because of its great reactivity, but it exists as fluoride mineral complexes. Fluoride ions have the same charge and nearly the same radius as hydroxide ions and may replace each other in mineral structures [19].

Fluorides are found in a wide variety of minerals, including fluorspar [CaF₂], sellaite [MgF₂], rock phosphate/fluoroapatite [Ca₅ (PO₄)₃ F], cryolite [Na₃AlF₆], mica, hornblende and others [21]. Fluorite (CaF₂) is a common bearer of fluoride and is found in granite, granite gneisses and pegmatite [22-23] and as cryolite in igneous rocks. Fluoride also associated with mono valent cations such as NaF and KF which are water soluble, while the one formed with divalent cations such as CaF₂ and PbF₂ is generally insoluble in water. Thermal waters with high pH are also rich in fluoride [24]. Rock phosphates are converted into phosphate fertilizers by the removal of up to 4.2 per cent fluoride [21].

Fluoride pollution in the environment occurs through two different channels which are natural and anthropogenic sources [25]. The majority of fluoride in the Earth's surface is derived from rock minerals whereas other sources such as air, seawater and anthropogenic activities constitute a relatively small proportion [26-27]. Fluorine and its compounds are used as raw materials in industry which extensively generate the toxic waste wastes containing fluorine/fluoride [28]. Elemental fluorine is necessary in the preparation of many fluoride compounds, which play an important role in semiconductors, phosphate fertilizers, pesticides, production of high purity graphite, electrolysis of alumina and in nuclear applications. Burning of coals often contains fluoride as an impurity and it is being leached down to the groundwater [22, 29-31]. The amount of fluoride present naturally in groundwater is governed by climate, pH, composition of the host rock and hydrogeology [32-33]. During weathering of alkali, igneous and sedimentary rocks and circulation of water in fluoride bearing rocks and soils, fluorine can be reached out and dissolved in groundwater [34-42].

Drinking-water is typically the largest single contributor to daily fluoride intake [21]. The fluoride content of air, water and food determine the intake of fluoride and depend mainly on the geographical areas. Food seems to be the source of 80-85% of fluoride intake; the intake from drinking water is 0.03-0.68 mg/day and from tooth pastes 0.2-0.3mg [43]. Based on the previous discussion, it follows that total daily fluoride exposure can vary markedly from one region to another.

Fluoride has dual significance, i.e., beneficial effects on teeth at low concentrations in drinking-water but on excessive exposure to fluoride in drinking-water can give rise to a number of adverse effects. In India, concentrations of fluoride in drinking water in different parts of the country vary from 0.5 to 50 mg/lit and the extent of fluoride contamination in groundwater varies from 1.0 to 48 mg/lit [44]. The permissible limits of fluoride concentration in drinking water prescribed by various organizations [44-45] are shown in Table 1.

S. No.	Name of Organization	Permissible limit of fluoride ion (mg/lit)
1	World Health Organization (International standard for drinking water)	0.6-1.5
2	US Public Health Standards	0.8
3	The committee on public health engineering manual and Code of practice, Government of India	1.0
4	Indian Council of Medical Research (ICMR)	1.0
5	Bureau of Indian Standards (BIS)	0.6-1.5

Table 1: Permissible	limit of fluoride in drinl	ing water prescribed	by various Organizations

Thus the requirement of fluoride content varies among countries and depends on the geography and the age of people involved [46]. However, from several studies, a rough estimate of total daily fluoride exposure in a temperate climate is approximately 0.6 mg per adult per day in an area in which no fluoride is added to the drinking-water and 2 mg per adult per day in a fluoridated area [47].

Fluoride Distribution in Water:

Fluoride is found in all natural waters at varied concentrations. Seawater contains about 1mg/lit while rivers and lakes contain concentrations of less than 0.5 mg/lit [48]. In ground waters, the presence of fluoride can occur depending on the nature of the rocks and the occurrence of fluoride-bearing minerals. Waters with high fluoride concentrations occur in large and extensive geographical belts associated with sediments of marine origin in mountainous areas, volcanic rocks and granitic and gneissic rocks [48]. Fluoride concentrations increase in ground waters in which cation exchange of sodium for calcium occurs [24]. The high concentrations in groundwater are a result of dissolution of fluorite, apatite and topaz from the local bedrock. A total of 60-70 million people in India as a whole may be considered to be suffered from fluoride contamination [49].

Fluoride Distribution Atmosphere:

Fluorides are widely distributed in the atmosphere due to dust, industrial production and use of phosphate fertilizers, coal ash from the burning of fluoride-containing coal and volcanic activity. However, air is responsible for only a small fraction of total fluoride exposure [50]. In non-industrial areas, the fluoride concentration in air is $0.05-1.90\mu$ g m⁻³[21]. High levels of atmospheric fluoride occur in areas of Morocco and China [51-52] and more than 10 million people in China are reported to suffer from fluorosis due to the burning of high fluoride coal [53].

Fluoride Contamination in Worldwide:

A large number of fluoride contaminated areas are founded throughout the world where ground waters contain exceeded levels of fluoride (Figure 2). In 1984, WHO estimated that more than 260 million people living all over the world consume water with fluoride concentration above 1mg/lit. There are 14 countries in Africa, 8 countries in Asia, and 6 countries in the America that all having water considered unsafe by the World Health Organization [54].

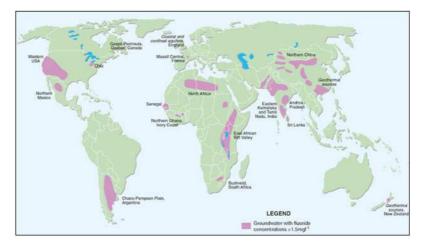


Figure 2: Regions of the World with significant levels of fluoride in the Groundwater

High groundwater fluoride concentrations associated with igneous and metamorphic rocks such as granites and gneisses have been reported from India, Pakistan, West Africa, Thailand, China, Sri Lanka, and Southern Africa. In some countries like India, Argentina, and Ethiopia, the fluoride contamination of groundwater has been significant [55-57].

The fluoride concentrations in groundwater more than 30 mg/lit occur widely in the United States of America, Africa and Asia [58-67]. In Kenya, a detailed survey of fluoride in groundwater has been undertaken by Nair, K.R. et al., [68]. High fluoride concentrations is found in United Republic of Tanzania, where 30 per cent of waters used for drinking exceeded 1.5 mg/lit fluoride [69] with concentrations in the Rift Valley even up to 45 mg/lit. In Sri Lanka, Dissanayake [70] found concentrations of up to 10 mg/lit in ground waters in the Dry Zone, associated with dental and possibly skeletal fluorosis. More than 23 countries in the world, including India, have problems with fluoride in the drinking water [71] and more than 260 million people in worldwide consume drinking water with a fluoride content of >1.0 mg/lit [47] and the majority of these people live in tropical countries where the problem is exacerbated by the need to drink more water because of the heat.

Fluoride Contamination in India:

In worldwide, India is most severely fluoride affected country. Of the 85 million tons of fluoride deposits on the earth's crust, 12 million are found in India [72]. Fluoride epidemic has been reported mostly in granite and gneissic geological formation of different states in India [73-75]. Some regions in north western and southern India are heavily affected with fluorosis [67, 76]. The rocks in southern India are rich with fluoride which forms the major reason for fluoride contamination in groundwater [77]. The disease as fluorosis in human beings is first reported in Nellore district of composite state of Andhra Pradesh even in 1937 [78]. The granites in the district of Nalgonda, composite state of Andhra Pradesh contain much higher fluoride than the world average fluoride concentration of 810 mg/kg [79]. The highest concentration observed in India is 48 mg/lit in Rewari District of Haryana [49]. In India alone, more than 62 million people including 6 million children below 14 years of age are affected with dental and skeletal fluorosis [49, 80-83]. In India in 2002, 17 states have been identified as endemic for fluorosis [84] and now the problem exist in more than 17 states and severely affected states are Rajasthan, composite state of Andhra Pradesh, Madhya Pradesh, Punjab, Haryana, Gujarat, Tamil Nadu and Uttar Pradesh [73, 85-86].

According to the Department of Drinking Water Supply, out of 593 districts from which data is available, water in 203 districts has shown high fluoride [80]. Different states of India where elevated concentrations of fluoride in groundwater as reported in literature are shown in Figure 3 and a detailed list of concentration of fluoride in groundwater in different states of India are given in Table No's 2 and 3. As seen from the Figure 3, almost all states in India have districts where groundwater contains excess levels of fluoride.

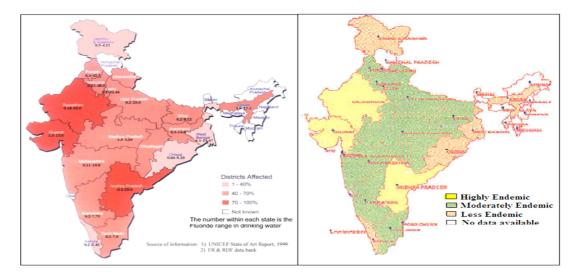


Figure 3: Map of India showing endemic states for fluorosis

Fluoride Contamination in composite state of Andhra Pradesh:

Groundwater is the major drinking water source in the villages of composite state of Andhra Pradesh which is the most prominent fluoride contaminated state in India. Several areas of Andhra Pradesh have fluoride concentration greater than the permissible limit by WHO and particularly the fluoride affected districts are mainly Cuddapah, Guntur, Nalgonda, Prakasam, Nellore, Anantapur and Rangareddy and concentrations of fluoride in groundwater in different districts of composite state of Andhra Pradesh are given in Table 3.

It is a well established fact that groundwater in Nalgonda district, composite state of 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 Nalgonda granties contain much higher fluoride than the world average fluoride concentration of 810 mg/kg [119]. In Kurmapalli watershed, rocks are enriched in fluoride from 460 to 1706 mg/kg [120]. The mean fluoride content in Hyderabad granites is 910 mg/kg [121].

Table 2: Concentration of fluoride in groundwater in different states of India where elevated concentrations of fluoride as reported in literature apart from composite state of Andhra Pradesh

States	Districts/Places	Range of fluoride conc. (mg/lit)	Ref.
	Karbi Anglong, Nagaon	0.2-18.10	[87]
Assam	Guwahati	0.18-6.88	[88]
	Sonitpur	0.17-5.60	[89]
Bihar	Palamu, Daltongani, Gridh, Gaya, Rohtas, Gopalgani, Paschim Champaran	0.60-8.0	[80]
D 11:	Kanjhwala, Najafgarh,	0.10-16.5	100.001
Delhi	Alipur city	0.40-10.0	[80,90]
	All districts except Dang	1.58-31.0	[80]
Gujarat	Mehsana	0.94-2.81	[91]
•	Mehsana	1.50-5.60	[92]
	Rcwari, Faridabad, Karnal, Sirsa, Sonipat, Gurgaon, Kaithal, Mohindergarh, Kurukshetra,	0.17-24.7	[80]
	Jind	0.20-2.00	[93]
Haryana	Bhiwani	0.14-86.0	[94]
	Rohtak	0.40-4.80	[95]
J & K	Doda,	0.05-1.21	[80]
	Garhwa	0.52-7.62	[96]
Jharkhand	Palamu	0.10-12.0	[97]
Karnataka	Dharwad, Gadag, , Belgam, Raichur, Bijapur, Gulbarga, Chitradurga,Tumkur,Chikmagalur, Mandya, Banglore, Mysore	0.20-18.0	[80,98]
	Bellary	0.33-7.8	[99]
77 1	Allepy, Vamanapuram	0.20-2.50	[80]
Kerala	Palghat	0.50-5.75	[100]
	Chandrapur, Bhandara, Nagpur, Jalgaon, Bulduna, Amravati, Akola, Nanded, Sholapur	0.11-10.2	[80]
Maharashtra	Yavatmal	0.3-13.41	[101]
Madhya	Jabua, Mandla, Dindori, Chhindwara, Dhar, Vidhisha, Seoni, Sehore, Raisen ,Bhopal	0.08-4.20	[80]
Pradesh	Shivpuri	1.65-3.91	[102]
0.1	Phulbani, Koraput, Dhenkanal	0.60-5.70	[80]
Orrissa	Balasore	0.60-5.83	[103]
Punjab	Mansa, Faridcot, Bhatinda, Mukstar, Moga, Sangrur, Fcrozpur, Ludhiana, Amritsar, Patila, Ropar, Jallandhar, Fatehgarh sahib	0.44-6.00	[80]
	All the 32 districts	0.20-37.0	[80]
	Hanumangarh	1.01-4.42	[104]
	Ajmer	0.12-16.9	[105]
Delether	Central Rajasthan	1.50-5.91	[106]
Rajasthan	Deoli Tehsil	0.30-9.60	[107]
	Dungarpur	1.50-4.40	[108]
	Jaipur	0.10-12.5	[109]
	Malpura Tehsil, Tonk	0.08-11.3	[31]
	Salem, Periyar, Dharampuri, Coimbatore, Tiruchirapalli, Vellore, Madurai, Virudunagar	1.50-5.00	[80]
	Erode	0.50-8.20	[110]
	Dindigul	2.47-5.26	[111]
T	Ottapidaram	0.94-4.34	[112]
Tamilnadu	Tirunelveli	0.73-3.02	[113]
	Rameswaram Area	1.50-2.50	[114]
	Salem	0.10-2.80 0.40-4.00	[115]
	Unnao, Agra, Meerut, Mathura, Aligarh, Raibareli, Allahabad	0.12-8.90	[80]
Uttar Pradesh	Kanpur	0.14-5.34	[116]
	Mathura	3.40-4.60	[117]
	Birbhum, Bhardaman, Bankura, Purulia	1.50-13.0	[80]
West Bengal	Hooghly	0.01-1.18	[118]

Studies on the quality of groundwater of Kakinada town [122] indicated that fluoride concentration is within permissible limits, but it has been found that fluoride content is increased with decrease in salinity. The high fluoride concentration in groundwater is attributed to the presence of fluorite and apatite minerals in the porphyritic granites of the region. The use of phosphate fertilizers, which are being leached down to the main groundwater body by return irrigation flows, might also be one of the contributing factors for high fluoride concentration.

State	Districts/Places	Range of fluoride conc. (mg/lit)	Ref.
	Podili, Darsi and Kanigiri Talukas of the erstwhile Nellore District	1.60-8.60	[123]
	Udayagiri Taluk villages of Nellore	2.37-6.74	[124]
	Wailpalli watershed, Nalgonda	0.97-5.83	[125]
	Parts of Nalgonda	0.10-8.80	[126]
	Nalgonda District	0.40-20.0	[121,127
	Sivannagudem area, Nalgonda	1.50-10.0	[128]
	Kurampalli watershed, Nalgonda	Up to 21.0	[120]
	Vamsadhara river basin	Up to 3.40	[129]
	Visakhapatnam	0.35-8.35	[130]
	Varaha River Basin, Visakhapatnam	0.60-2.10	[131]
	Rural parts of Guntur	0.60-2.30	[132]
	Uravakonda village, Anathapur	0.50-7.20	[133]
Composite state of Andhra Pradesh	Gunthakal area, Anathapur	0.18-2.00	[134]
	Ralla village, Anathapur	0.50 - 5.80	[135]
	Vempalli and Vemula mandals, Kadapa	0.226-3.52	[136]
	Badvel mandal, Kadapa	0.245-4.22	[137]
	Kandukur revenue division, Prakasam	1.22-3.09 1.44-4.55	[138]
	Marripudi mandal, Prakasam	0.50-9.00	[139-140
	Kurnool District	1.50-6.50	[141]
	Madanapalle sub-division, Chittoor	>2.50	[142]
	Gampalagudum mandal, Krishna	1.60-4.00	[143]
	Kommala area, Warangal District	1.10-5.80	[144]
	Ranga Reddy District	0.70-4.80	[145-146
	Hyderabad (MCH)	0.34-3.12	[147]

Table 3: Fluoride in groundwater as per reported in literatures for composite state of Andhra Pradesh

HEALTH IMPACTS OF FLUORIDE

Fluorosis

Fluoride in drinking water has a profound effect on teeth and bones. Fluoride in drinking water has been considered as an essential micro-nutrient for human body at low concentrations in drinking-water [148-149], but excessive exposure to fluoride in drinking-water, or in combination with exposure to fluoride from other sources, can give rise to a number of adverse effects. Excessive fluoride exposure may cause irreversible demineralization of bone and tooth tissues, a condition known as fluorosis. Hence, it is necessary for the controlled addition of fluoride to drinking water supply and is effective for preventing tooth decay. Even though helpful for dental health in small dosage, continuously exposure to fluoride in large amounts through food, water and air interferes with bone formation.

Higher fluoride concentration makes the teeth and the bones denser, harder and more brittle. In the teeth this causes mottling and embitterment, a condition known as dental fluorosis. The disease earlier called mottled enamel is first reported by Viswanathan [150] to be prevalent in human beings in Madras Presidency in 1933. Mahajan [151] reported a similar disease in cattle in certain parts of old Hyderabad state. However, Shortt [78] is the first to identify the disease as fluorosis in human beings in Nellore district of composite state of Andhra Pradesh. With prolonged exposure to higher fluoride concentrations, dental fluorosis progresses to skeletal fluorosis [152]. This range from mild dental fluorosis to crippling skeletal fluorosis as the level and period of exposure increases.

The World Health Organization permits maximum fluoride concentration in drinking water up to 1.5 mg/lit [47] at which fluorosis is minimum and this is being followed in most of the nations and is also the Australian recommended limit [153-154]. The effect of fluoride on human health by the prolonged use of drinking water with fluoride content [24,152] is presented in Table 4.

Fluoride is thus considered beneficial in drinking water at levels of about 0.7 mg/lit but once it exceeds 1.5 mg/lit causes adverse effects including severe dental fluorosis, skeletal fluorosis and crippling fluorosis [24, 81, 83, 155-157] and also other adverse affects on human health have been reviewed [158-159]. Both national and international groups [50, 160] have comprehensively reviewed available data on the metabolism and health effects of fluoride in both laboratory animals and humans.

Fluoride ion concentration, (mg/lit)	Health outcome
<0.5	Dental caries
0.5-1.5	Optimum dental health
1.5-4.0	Dental fluorosis
4.0–10	Dental and skeletal fluorosis
>10.0	Crippling fluorosis

Table 4: Health effects on prolonged use of fluoridated drinking water

Dental Fluorosis:

Dental fluorosis is defined as yellowish or brownish striations or mottling of the enamel and it is a cosmetic effect, associated pitting of the teeth in severe forms and is first reported in 1888 in Mexico in a family from Durango who attributed their black teeth for fluorosis [48]. The mottling of teeth is one of the earliest and most easily recognized symptoms [161]. They lose their normal creamy white translucent color and become rough, opaque and chalky white. Various stages of dental fluorosis are as shown in Figure 4.

An early stage of dental fluorosis is dental mottling, which is characterized by opaque white patches on teeth. In advanced stages of dental fluorosis, teeth display brown to black staining followed by pitting of teeth surfaces. Dental fluorosis produces considerable added dental costs (tooth deterioration) and significant physiological stress for affected population. Dental fluorosis is endemic in 14 states and 1, 50,000 villages in India. The problems are most pronounced in the states of Bihar, composite state of Andhra Pradesh, Gujarat, Madhya Pradesh, Punjab, Rajasthan, Tamil Nadu and Uttar Pradesh [162].

High levels of fluoride present in concentrations up to 10 mg/lit are associated with dental fluorosis while low levels of fluoride, less than 0.1 mg/lit, are associated with high levels of dental decay [24]. The margin between the beneficial effects of fluoride and the occurrence of dental fluorosis is small and public health programmes seek to retain a suitable balance between the two [160].



Figure 4: Various stages of Dental fluorosis

Dental fluorosis has been studied in several parts of South Africa [163-165] and in West Africa in Senegal, especially for the endemic region of Fatick. In China, 38 million people are reported to suffer from dental fluorosis [166] and its affect is felt at several locations of the USA [167] and in many other countries around the world.

Skeletal Fluorosis:

Skeletal fluorosis is characterized by deformation of bone structure and affects children as well as adults. Fluoride mainly gets deposited in the joints of neck, knee, pelvic and shoulder bones and makes it difficult to move or walk. The symptoms of skeletal fluorosis are similar to spondylitis or arthritis [168]. Continuous exposure to elevated concentrations of fluoride associated with skeletal fluorosis [160]. Fluoride concentration in the range of 4.0 to 10.0 mg/lit causes skeletal fluorosis [24]. It does not easily manifest until the disease attains an advanced stage. Vertebrae may fuse together and eventually the victim may be crippled.

Crippling skeletal fluorosis, which is associated with the higher levels of exposure (>10.0) [47], can result from osteosclerosis, ligamentous and tendinous calcification and extreme bone deformity and finally spine, major joints, muscles and nervous system get damaged [169-170]. The stage at which skeletal fluorosis becomes crippling usually occurs between 30 and 50 years of age in endemic regions. The factors which govern the development of skeletal fluorosis are (i) the prevalence of high levels of fluoride intake, (ii) continual exposure to fluoride, (iii) strenuous manual labour, (iv) poor nutrition, (vi) impaired renal function due to disease [171]. Figure 5 shows the victims of skeletal fluorosis.

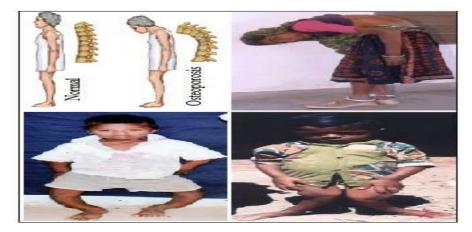


Figure 5: Victims of Skeletal fluorosis

Endemic skeletal fluorosis is occurring severely in several parts of the world, including India, China and Africa. In China, 1.7 million people suffer from the more severe skeletal fluorosis [166] and in India; around one million people suffer from serious and incapacitating skeletal fluorosis [172].

Other Health Effects:

A number of studies have been carried out to examine other possible adverse affects as a consequence of exposure to fluoride, either from drinking-water or as a consequence of occupation. Studies of occupationally exposed populations, have reported lung and bladder cancer and from cancers in other sites [48]. Besides skeletal and dental fluorosis, excessive consumption of fluoride may lead to infertility in women, brain and hepatit damage, skin rashes, nervousness, Alzheimer syndrome [173-178], depression, gastrointestinal problems, urinary tract malfunctioning, nausea, abdominal pain, tingling sensation in fingers and toes, reduced immunity, repeated abortions or still births, male sterility, mental disorders in children, reductions in the IQs of children [179], osteosclerosis, structural changes in DNA [180], dwarfishness, low haemoglobin levels, deformities in RBCs, excessive thirst, lesions of the endocrine glands, thyroid, liver and other organs and even death in extreme cases [181-184]. The majority of fluoride is excreted via the kidneys [50], hence those with impaired renal function might be at greater risk of fluoride toxicity. It is also responsible for alterations in the functional mechanisms of systems such as liver, respiratory, digestive, reproductive, excretory, central nervous and destruction of about 60 enzymes [185].

It had been reported that the concentration of fluoride ions in ground waters of many places exceeds the permissible values [76]. Due to high toxicity of fluoride to mankind, it is imperative and significant for removing excessive fluorides from fluoride-contaminated drinking water to make it safe for human consumption.

CONCLUSION

A brief review on groundwater pollution with respect to fluoride ions and its various health effects on human beings have been discussed. The World Health Organization has specified the tolerance limit of fluoride content of drinking water as 1.5 mg/L. Fluoride contamination in groundwater is a world-wide issue. The occurrence of high fluoride concentrations in groundwater and risk of fluorosis by human consumption of fluoride rich water is a problem faced by many countries, notably India, Pakistan, West Africa, Thailand, China, Sri Lanka, Southern Africa, United States of America, Argentina, and Ethiopia. Presence of excessive levels of fluoride is the cause of fluorosis and other related disorders in human beings. There is no treatment to the disease and prevention is the only solution. The

primary preventive measure is to drink of fluoride-free water. This can be achieved by the removal of fluoride from fluoride polluted waters using de-fluoridation techniques. Thus we can conclude that some cost effective and efficient technologies are required to remove excess fluoride in drinking water.

REFERENCES

[1] WHO, World Health Organization, Safe Water and Global Health, Geneva, 2008.

[2] H. Pink Daniel, Investing in Tomorrow's Liquid Gold, 2006.

[3] W. Larry, World Water Day: A Billion People Worldwide Lack Safe Drinking Water, 2006.

[4] WBCSD, World Business Council for Sustainable Development, Water Facts and Trends, 2009.

[5] T.S. Dhiviyaa Pranavam, T. Venkatesa Rao, L. Punithavathi, S. Karunanithi, A. Bhaskaran, *Indian J. Sci. Technol.*, **2011**, 4 (1), 19-21.

[6] D.P. Gupta, Sunita, J.P. Saharan, Researcher, 2009, 1(2), 1-5.

- [7] N. Kannan, J. Sabu, World Academy of Sci. Eng. and Technol., 2009, 52, 475-493.
- [8] D. Chakraborti, Bhaskar Das, M. T. Murrill, Environ. Sci. Technol., 2011, 45, 27-33.
- [9] The Times of India. Ground water in 33% of India undrinkable. March 12, 2010.

[10] UNESCO, Trace Elements in Water and Public Health, Paris, France, 2004.

[11] A. Zaporozec, J.C. Miller, Groundwater Pollution. UNESCO, Paris, France, 2000, 24.

[12] Akoijam, Chem. Environ. Research, 1997, 6, 301.

[13] C.N. Mulligan, R.N. Yong, B.F. Gibbs, Eng. Geol., 2001, 60 (1-4), 193-200.

- [14] F.H. Charles, C.H. Swartz, A.B.M. Badruzzaman, K.B. Nicole, W. Yu, A. Ali, J. Jay, R. Beckie, V.
- Niedan, D. Brabander, Comptes Rendus Geosci., 2005, 337(1/2), 285-296.

[15] A.K. Singh, Curr. Sci., 2006, 91(5), 1-7.

[16] S. Suthar, P. Bishnoi, S. Singh, P.K. Mutiyar, A.K. Nema, N.S. Patil, J. Hazard Mater., 2009, 171(1-3), 189-99.

[17] A. Agrawal, R. S. Pandey, B. Sharma, J. Water Resource and Protection, 2010, 2, 432-448.

[18] J. D. Hem, Study and Interpretation of the Chemical Characteristics of Natural Water (2nd ed.), U.S. Geological Survey, Washington, DC, USA, **1970**, 363.

[19] J.D. Hem, Study and Interpretation of the Chemical Characteristics of Natural Water (3rd ed.), U.S. Geological Survey, Washington, DC, USA, **1989**, 263.

[20] T.H.Y. Tebbutt, Relationship between natural water quality and health, Paris, France, 1983.

- [21] J.J. Murray, Appropriate Use of Fluorides for Human Health, WHO, Geneva, 1986.
- [22] A.N. Deshmukh, K.C. Shah, A. Sriram, Gondwana Geological Magazine, 1995, 9, 21-29.
- [23] S. Rao, Environ. Monitoring & Assessment, 2009, 152, 47-60.

[24] W.M. Edmunds, P.L. Smedley, Geological Society Special Publication, 1996, 113, 91-105.

[25] Y. Cengeloglu, E. Kir, M. Ersoz, Sep. and Purifi. Technol., 2002, 28(1) 81-86.

[26] R. Fuge, Environmental Geochemistry and Health, 1988, 10(2), 51-61.

[27] P. Lahermo, H. Sandstrom, E. Malisa, J. of Geochemical Exploration, 1991, 41, 65-79.

[28] G. D. Puente, J.J. Pis, J.A. Menhdez, P. Grange, J. Anal. Appl. Pyrolysis, 1997, 43, 125-138.

[29] F.A. Smith, H.C. Hodge, Critical Reviews in Environmental Control, 1979, 8(2), 241-245.

[30] M.A. Anderson, L.W. Zelazny, P.M. Bertsch, Crop Sci. Soci. Amer. J., 1991, 55(1), 71-75.

[31] G.S. Tailor, C.P.S. Chandel, Nature and Science, 2010, 8(11), 20-26.

[32] S. Gupta, S. Banerjee, R. Saha, J.K. Datta, N. Mondal, Fluoride, 2006, 39(4), 318-320.

[33] N.J. Raju, S. Dey, K. Das, Current Science, 2009, 96 (7), 979-985.

[34] B.K. Handa, Groundwater, 1975, 3(3), 275-281.

- [35] N.S. Rao, *Hydrological Sciences J.*, **1997**, 42, 877-892.
- [36] W. Genxu, C. Guodong, J. of the Arid Environment, 2001, 49, 601-614.
- [37] V. K. Saxena, S. Ahmed, Environ. Geol., 2001, 40, 1084-1087.

[38] W.M. Edmunds, P.L. Smedley, Fluoride in natural waters, In: Selinus, O. (Ed.), Essentials of Medical Geology, Elsevier Academic Press, London, **2005**, 301-329.

[39] G. Jacks, P. Bhattacharya, V. Chaudhary, K.P. Singh, Appl. Geochem., 2005, 20, 221-228.

[40] G.T. Chae, S.T. Yun, M.J. Kwon, S.Y. Kim, B. Mayer, *Geochem. J.*, **2006**, 40, 95-102.

[41] P.D. Sreedevi, S. Ahmed, B. Made, E. Ledoux, Gandolfi, Environ. Geol., 2006, 50(1), 1-11.

[42] Q. Guo, Y. Wang, T. Ma, R. Ma, J. of Geochemical Exploration, 2007, 93, 1-12.

[43] WHO, World Health Organization, Fluoride in Drinking-water, Background document for development of WHO Guidelines for Drinking-water Quality, Geneva, Switzerland, **2004**.

[44] WRSD, Water Resources Systems Division, National Institute of Hydrology, Jalvigyan Bhawan, Roorkee, India.

[45] S. Gopalkrishnan, S. Narsimha, Senipandian, Ind. J. of Envi. Protec., 1991, 11(2), 118-123.

[46] WHO, World Health Organization, Guidelines for drinking water quality (Drinking water quality control in small community supplies, Geneva, Switzerland, **1985**, 8,121.

[47] WHO, World Health Organization, Guidelines for drinking water quality, Health criteria and other supporting informations, Geneva, Switzerland, **1984**

[48] J. Fawell, K. Bailey, J. Chilton, E. Dahi, L. Fewtrell, Y. Magara, World Health Organization, Fluoride in Drinking-water, Published by IWA Publishing, London, UK, **2006**.

[49] UNICEF, State of the art report on the extent of fluoride in drinking water and the resulting endemicity in India, Report by Fluorosis Research & Rural Development Foundation for UNICEF, New Delhi, **1999.**

[50] USNRC, Health Effects of Ingested Fluoride. National Research Council, National Academy Press, Washington, DC, 1993.

[51] Y. Haikel, J.C. Voegel, R. M. Frank, Archives of Oral Biology, 1986, 31(5), 279-286.

[52] Y. Haikel, P. M. Cahen, J.C. Turlot, R.M. Frank, J. of Denta. Resear., 1989, 68(8), 1238-41.

[53] S.L. Gu, J. Rongli, C. Shouren, Biomed. and Environ. Sciences, 1990, 3(4), 384-390.

[54] R. Tekle-Haimanot, A. F. Bushera, Y. Mekonnen, Fluoride Levels in Water and Endemic Fluorosis in Ethiopian Rift Valley, 1st International Workshop on Fluorosis Prevention and Defluoridation of Water, Tanzania, The International Society for Fluoride Research, **1995**.

[55] J.D. Paoloni, C.E. Fioerentino, M.E. Sequeira, Environ. Toxicol., 2003, 18, 317-320.

[56] B. Nayak, M. M. Roy, B. Das, A. Pal, M. K. Sengupta, S. P. De, D. Chakraborti, *Clin. Toxicol.*, **2009**, 47 (4), 292-295.

[57] UNICEF, Fluoride contamination and treatment in Ethiopian Rift Valley, 2009.

[58] S.J. Gaciri, T.C. Davies, J. Hydrol., 1992, 143, 395-412.

[59] W. Czarnowski, K. Wrzesniowska, J. Krechniak, Sci. of Total Environ., 1996, 191, 177-84.

[60] G. Moges, F. Zewge, M. Socher, J. Afr. Earth Sci., 1996, 21, 479-482.

[61] W.B. Apambire, D.R. Boyle, F.A. Michel, Environ. Geol., 1997, 33(1), 13-24.

[62] N. Azbar, A. Turkman, Water Sci. and Technol., 2000, 42, 403-407.

[63] T. Chernet, Y. Trafi, V. Valles, Water Res., 2002, 35, 2819-2832.

[64] H. Mjengera, G. Mkongo, Appropriate defluoridation technology for use in fluorotic areas in Tanzania, 3rd Water Net Symposium Water Demand Management for Sustainable Development, **2002.**

[65] W.K.N. Moturi, M.P. Tole, T.C. Davies, Environ. Geochem. and Health, 2002, 24, 123-130.

[66] W.Y.Wang, R.B. Li, J.A. Tan, K.L. Luo, L. Yang, H. Li, Y. Li, Fluoride, 2002, 35, 122-29.

[67] M., Agarwal, K., Rai, R. Shrivastav, S. Dass, J. Cleaner Produc., 2003, 11, 439-444.

[68] K.R., Nair, F. Manji, J.N. Gitonga, The occurrence and distribution of fluoride in groundwaters of Kenya, In: Challenges in African Hydrology and Water Resources, Proceedings of the Harare Symposium, IAHS Publ., **1984**, 144, 75-86.

[69] M. C. Latham, P. Gretch, American Journal of Public Health, 1967, 57, 651-660.

- [70] C. B. Dissanayake, Int. J. of Environmental Health Studies, 1991, 38, 137-156.
- [71] A.K., Susheela, M. Bhatnagar, R. Bahadur, *Fluoride*, **1993**, 20(9), 100-104.
- [72] S.P.S. Teotia, M. Teotia, J. of Association of Physicians of India, 1984, 32, 347-352.

[73] V. Agrawal, A.K. Vaish, P. Vaish, Current Science, 1997, 73(9), 743-746.

[74] S. Tripathy, M.K. Panigrahi, N. Kundu, Environ. Geochem. and Health, 2005, 27, 205-216.

[75] N. J., Raju, P. Ram, S. Dey, J. of the Geological Society of India, 2009, 73(2), 178-192.

[76] RGNDWM, Rajiv Gandhi National Drinking Water Mission, Prevention and control of fluorosis in India, New Delhi, Government of India, **1993**, 25.

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

[78] H. E. Shortt, C. G. Pandit, T. N. S. Raghvachari, Ind. Medical Gazette, 1937, 72, 396-400.

[79] S., Yadav, T.I., Khan, S., Gupta, A.B. Gupta, R.N. Yadava, Fluorosis in India with special reference to Rajasthan, In: Proceedings of the International Conference on Water, Environment, Ecology, Socioeconomics and Health Engineering Seoul National University, **1999**, 3-10.

[80] A.K. Susheela, Fluorosis: Indian scienario, a treatise on fluorosis. Fluorosis Research and Rural Development Foundation, New Delhi, India, *Fluoride*, **2001**, 34, 181-183.

[81] A.K. Susheela, Fluorosis Types: Skeletal Fluorosis. Treatise on Fluorosis, 3rd ed. Fluorosis Research & Rural Development Foundation of India, Delhi, **2007a**, 30-8.

[82] A.K. Susheela, Industrial Fluorosis. In: Treatise on Fluorosis. 3rd ed. Fluorosis Research & Rural Development Foundation of India, Delhi, **2007b**, 68-76.

- [83] S. Ayoob, A. K. Gupta, Criti. Reviews in Environ. Sci. and Technol., 2006, 36(6), 433-487.
- [84] S.L. Choubisa, *Fluoride*, 2001, 34, 61-70.
- [85] P. Kumaran, G.N. Bhargava, T.S. Bhakuni, Ind. J. Environ. Health, 1971, 13, 316-324.
- [86] S.P.S. Teotia, M. Teotia, D.P. Singh, R.S. Rathour, C.V. Singh, N.P.S. Tomar, M. Nath, N.P. Singh, *Fluoride*, **1984**, 17, 48-52.

[87] D. Chakraborti, C.R. Chanda, G. Samantha, U.K. Chowdhury, S.C. Mukherjee, A.B. Pal, B. Sharma, K.J. Mahanta, H.A. Ahmed, B. Sing, *Current Science*, **2000**, 78 (12), 1421-1423.

[88] B. Das, J. Talukdar, S. Sarma, B. Gohain, R.K. Dutta, H.B. Das, S.C. Das, *Current Science*, **2003**, 85(5), 657-660.

[89] J.Dutta, M. Chetia, J.P.Baruah, M. A. Kumar, Arch. of Appl. Sci. Res., 2010, 2(5), 226-238.

[90] P. S. Datta, D. L. Deb, S. K. Tyagi, J. of Contaminant Hydrology, 1996, 24, 85-96.

[91] P. R.Salve, A. Maurya, P. S. Kumbhare, D.S. Ramteke, S. R. Wate, *Bull Environ Contam Toxicol.*, 2008, 81, 289-293.

[92] S. D. Dhiman, A. K. Keshari, Hydrological Sciences Journal, 2006, 51(6), 1149-1162.

[93] A. Singh, J. S. Laura, A. Rana. Int. J. of current research, 2013, 5(4), 998-1002.

[94] V. K. Garg, S. Suthar, S. Singh, Sheoran, Garima, Meenakshi, S. Jain, *Environmental Geology*, **2009**, 58, 1329-1340.

[95] A. Kaushik, K. Kumar, I.S. Sharma, H.R. Sharma, J. Environ. Biol., 2004, 25(2), 173-180.

[96] S. C. Samad, Rakesh Kumar, Anjani Kumar, Amrit Kumar Jha, Rameswar Prasad Sah and Triyugi Nath. *Advan. in appl. Sci. research.*, **2013**, 4(5), 325-329.

[97] R. Srikanth, Fluoride, 2008, 41(3), 206-211.

[98] S. Sumalatha, S.R. Ambika, S. J. Prasad, Current Science, 1998, 76(6), 730-734.

[99] B. K. Wodeyar, G. Sreenivasan, Current Sci., 1996, 70, 71-74.

[100] E. Shaji, B. J. Viju, D.S. Thambi, Current Science, 2007, 92(2), 240-245.

[101] P. Madhnure, D. Sirsikar, N. Tiwari, B. Ranjan, D. Malpe, Curr. Sci., 2007, 92(5), 675-79.

[102] D.N.Saksena, Y.S. Narwaria, Int. J. of Environmental Sciences, 2012, 3(3), 1141-1149.

[103] K. K., Das, T. Panigrahi, R. B. Panda. J. of Environment, 2012, 1(2), 33-39.

[104] S. Suthar, V. K. Garg, S. Jangir, S. Kaur, N. Goswami, S.Singh, *Environmental Monitoring and Assessment*, 2008, 145, 1-6.

[105] C. Vikas, R.K. Kushwaha, M.K. Pandit, J. geological society of India, 2009, 73,773-784.

[106] I. Hussain, M. Arif, J. Hussain, Environ. monitoring and assessment, 2012, 184(8), 5151-8.

[107] K.S. Meena, R.K. Gunsaria, K. Meena, N. Kumar, P.L. Meena, R.R. Meena, J. of Chemical, Biological, and Physical Sciences, 2011, 1(2), 275-282.

[108] S.L. Choubisa, Biological sciences, 2012, 82(2), 325-330.

- [109] S. Saxena, U. Saxena, Int.J. of Environmental Sciences, 2013, 3(6), 2251-2260.
- [110] K. Karthikeyan, K. Nanthakumar, P.Velmurugan, S. Tamilarasi, P. L. Perumalsamy, *Environmental Monitoring Assessment*, **2010**, 160(1-4), 141-155.
- [111] M. M. Hanipha, A. Zahir Hussain, Int. Res. J. Environ. Sci., 2013, 2(1), 68-73.
- [112] V. Veeraputhiran G. Alagumuthu, Int. J. of Environ. Sci., 2010, 1(4), 558-566.

[113] G. Alagumuthu, M. Rajan, Rasayan J. Chem., 2008, 1(4), 757-765.

[114] V. Sivasankar, T. Ramachandramoorthy, J. Environ. & Analytical toxicology, 2011, 1(3).

[115] K. Srinivasamoorthy, K. Vijayaraghavan, M. Vasanthavigar, S. Sarma, S. Chidambaram, P. Anandhan, M. Rama, *Arab J. Geosciences*, **2012**, 5(1), 83-94.

[116] N. Sankararamakrishnan, A. K. Sharma, L. Iyengar, *Environ. Monitoring and Assessment*, 2008, 146, 375-382.

[117] K.S. Rawat, A.K. Mishra, V.K. Sehgal, J. of Appl. and Nat. Sci., 2012, 4(1), 117-122.

[118] M.C. Kundu, B. Mandal, Environmental Monitoring and Assessment, 2009, 152, 97-103.

[119] K. H. Wedepohl, Handbook of geochemistry, Berlin: Springer-Verlag, Vol. II-l, 1969.

[120] N. C. Mondal, R. K. Prasad, V. K. Saxena, Y. Singh, V. S. Singh, Environ. Earth Sciences, 2009, 59, 63-73.

[121] N. Ramamohana Rao, K. Suryaprakasa Rao, R. Schuiling, Environ. Geol., 1993, 21, 84-89.

- [122] K.S. Rao, et al., Indian J. Env. Prot., 1994, 14(3), 167-169.
- [123] T.N.S. Raghavachari, K. Venkataraman, Indian. J. med. Res., 1940, 28,517-532.
- [124] P. Jaganmohan, S.V. Narayana, K.R. Sambasivarao, World J. Med. Sci., 2010, 5(2), 45-48.
- [125] A. Reddy, D.Reddy, P. Rao, K. Prasad, Environ. Monit. Assess., 2010, 171(1-4), 561-577.

- [126] K. Brindha, R. Rajesh, Murugan, Elango, Enviro. Monit. Assess., 2011, 172(1-4), 481-492.
- [127] K. Somasekhara Rao, K.A. Rama Raju, K. Emmanuel, M.Kishore, *Chem. Environ. Res.*, 2007, 16(1-2), 123-131.
- [128] V. Sudharshan, B.R. Reddy, Indian J. Env. Prot., 1991, 11(3), 185-192.
- [129] N. Srinivasa Rao, Hydrological Sciences, 1997, 42(6), 877-892.
- [130] D. R. R. Sarma, S.L.N. Rao, Bull. Environ. Contam. Toxicol., 1997, 58, 241-247.
- [131] N. Subba Rao, Environ. Monitoring & Assessment, 2009, 152(1-4), 47-60.
- [132] N. Subba Rao, Hydrological sci., 2003, 48(5), 835-847.
- [133] V. Sunitha, B. Rajeswara Reddy, M. Ramakrishna Reddy, Int. J. of Research in Chemistry and Environment, 2012, 2(1), 88-96.
- [134] A. Narsimha, N. Anitha, V. Sudarshan, Advances in Appl. Sci. Reserc, 2013, 4(2), 70-76.
- [135] K. Sarala, P.R. Rao, Fluoride, 1993, 26, 177-180.
- [136] V. Sunitha, B. M. Reddy, J. Khan, M. Reddy, Asian J. Exp. Biol. Sci., 2012, 3(2), 293-297.
- [137] V. Sunitha, J. A. Khan, B. M.Reddy, Indian J. of Advan. in Chem. Sci., 2013, 2 (1), 78-82.
- [138] Y. Hanumantharao, M. Kishore, K. Ravindhranath, J. of Chemical and Pharmaceutical Research, 2012, 4(1), 601-607.
- [139] S.V. Ramanaiah, S. Venkatamohan, B. Rajkumar, P.N. Sarma, J. of Environ. Science & Engg., 2006, 48, 129-134.
- [140] K. Somasekhara Rao, M. Kishore, M. Ravi, K. Krishna Vani, Ind. J. Environ. Prot., 2005, 25(10), 919-924.
- [141] M.C.S. Reddy, Indian J. Environ. Health., 1998, 40(3), 240-252.
- [142] A. S. Manohar Rao. Assessment of fluoride in drinking water in Andhra Pradesh- A report by arghyam, Experiece in Madanapalle sub-division, Chittor District, **2011.**
- [143] V. B. M. Rao, M. S. Rao, V. Prasanthi, M. Ravi, Rasayan J. Chem., 2009, 2(2), 525-530.
- [144] V. Radhika, G.V. Praveen, Advances in applied science research, 2012, 3 (4), 2523-2528.
- [145] V. V. Kumar, C. S. Sai, P. L. Rao, C.S. Rao, J. of Fluorine Chemi., 1991, 55(3), 229-236.
- [146] D. Sujatha, B. Rajeswara Reddy, J. Environ. Geol., 2003, 44, 579-586.
- [147] S.S. Asadi, P.Vuppala, M. Reddy, Ind. Int. J. Environ. Res. Pub. Health, 2007, 4(1), 45-52.
- [148] M. Srimurali, A. Pragathi, J. A. Karthikeyan, J. Environ. Pollut., 1998, 99(2), 285-289.
- [149] M. R. Boldaji, A. H. Mahvi, S. Dobaradaran, Int. J. Enviro. Sci. Tech., 2009, 6 (4), 629-32.
- [150] G. R.Viswanathan, Annual Report Madras, Indian Council of Agricultural Research, New Delhi, Quoted from Indian Institute of Science, **1935**, 33A, 1, 1951.
- [151] Mahajan, Annual Report, VIO Hyderabad State, 3, Indian Council of Agricultural Research, New Delhi, **1934.** [152] C. B. Dissanayake, *Int. J. of Environ. Studies*, **1991**, 37, 247-258.
- [153] J. Smet, Fluoride in drinking water, In: Frencken, LE (Ed.), Endemic Fluorosis in Developing Countries-Causes, Effects and Possible Solution: Report of a Symposium Held in Delft, The Netherlands, Netherlands Organisation for Applied Scientific Research, **1990**.
- [154] NHMRC, National Health and Medical Research Council, Australian Drinking Water Guidelines, 2004.
- [155] E. A. Savinelli, A. P. Black, J. American Water Works Association, 1958, 50 (1), 33-44.
- [156] P. L. Bishop, G. Sansoucy, J. American Water Works Association, 1978, 70(10), 55.
- [157] M. Srimurali, G. Karthikeyan, Defluoridation of Water and Household Application-A Study, Proceeding of the Twelfth International Water Technology Conference, Alexandria, Egypt, **2008**, 153-163.
- [158] K. Bailey, J. Chilton, E. Dahi, M. Lennon, P. Jacksonand, J. Fawell, Fluoride in Drinking Water, World Health Organisation, IWA Publishing, London, **2006**.
- [159] V. Ganvir, K. Das, J. of Hazardous Materials, 2011, 185(2-3), 1287-1294.
- [160] IPCS, International Programme on Chemical Safety, Fluorides, Environmental Health Criteria 227, World Health Organization, Geneva, **2002.**
- [161] W. Choi, Y. Chen, J. American Water Works Association, Water Tech., 1979, 562-570.
- [162] K.S. Pillai, V.A. Stanley, J. Environ. Biol., 2002, 23, 81-87.
- [163] I. L. Carstens, A. J. Louw, E. Kruger, J. of Dent. of S.Afr., 1995, 50(9),405-411.
- [164] U. M. Chikte, A.J. Louw, I. Stander, J. of South Afri. Dent. Assoc., 2001, 56(11), 528-532.
- [165] A. J. Louw, S. R. Grobler, T. J. van Wyk Kotze, Gen. Dent., 2002, 13, 352-356.
- [166] WRI, World Resources Institute, World Resources, a Guide to the Global Environment: the Urban Environment, WRI/UNEP/UNDP/WB, Oxford University Press, **1996.**
- [167] G.V. Black, F.S. McKay, Dent. Cosmos., 1916, 58, 129-156.
- [168] C. Dinesh, Ind. J. of Environment Protection., 1998, 1(2), 81-89.
- [169] M. Teotia, S.P.S Teotia, K. P. Singh, Ind. J. Pediatr., 1994, 65, 371-381.

- [170] P. Ekambaram, P. Vanaja, Environ. Toxicol. Pharmacol., 2001, 9, 141-146.
- [171] A. Nicolay, P. Bertocchio, E. Bargas, F. Coudore, G. A. Chahin, J.P. Reynier, *Clin. Chim. Acta.*, **1999**, 281(1-2), 29-36.
- [172] A. K. Susheela, T.K. Das, International Symposium on Environmental Life Elements and Health, Beijing, **1988**, 89.
- [173] N. Chinoy, Indian J. Environ. Toxicol., 1991, 1(1), 17-32.
- [174] Y. Shivarajashankara, A. Shivashankara, P. Bhat, S. Rao, Fluoride, 2001, 34(2), 108-113.
- [175] X. Guo, G. Sun, Y. Sun, Fluoride, 2003, 36(1), 25-29.
- [176] A. Jamode, V. Sapkal, V. Jamode, J. Indian Inst. Sci., 2004, 84(5), 163-171.
- [177] A. Mahvi, M. Zazoli, M. Younecian, B. Nicpour, J. Med. Sci., 2006, 6(4), 658-661.
- [178] S.Dobaradaran, A.Mahvi, S.Dehdashti, D.Abadi, I.Tehran, Fluoride, 2008, 41(3), 220-226.
- [179] J.D. Sharma, S. Deepika, J. Parul. Fluoride, 2009, 42, 127-32.
- [180] M. M. Emamjomeh, M. Sivakumar, J. Hazard. Mater., 2006, 131,118-125.
- [181] H. Lounici, L. Addour, D. Belhocine, H. Grib, S. Nicolas, B. Bariou, N. Mameri, *Desalination*, 1997, 114, 241-251.
- [182] M., Hichour, F., Persin, J.Sandeaux, C.Gavach, Sep. & Purifi. Technol., 2000, 18, 1-11.
- [183] O. Barbier, L. A.Mendoza, L. M. D. Razo, Chem. Biolog. Interact., 2010, 188(2), 319-333.
- [184] E., Gazzano, L., Bergandi, C., Riganti, E., Aldieri, S., Doublier, C., Costamagna, A. Bosia, D. Ghigo, *Current Medicinal Chemistry*, **2010**, 17, 2431-2441.
- [185] J.D, Sharma, M.K. Sharma, P. Agrawal, Asian J. Exp. Sci., 2004, 18(1&2), 37-46.