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Fluoride in Drinking Water ·Its Effects and Removal

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Abstract. Occurrence of fluoride in water, its metabolism, excretion, effect o ingestion in human and cattle system and methods of fluoridation and defluoridation have been discussed. The presence of fluoride in waters occurring in India, with special reference to Rajasthan desert has been reviewed. Based on the survey and physico-chemical analyses of about 2,700 water samples of Rajasthan, distribution of fluoride in this area has been discussed with reference to drinking water standards. A water resources map showing concentrations of fluoride in four arid districts of Rajasthan is also presented.

Introduction

The Government of India, during the International Drinking Water Supply and Sanitation Decade, (1981-1990), has undertaken to supply safe drinking water for the country's urban and rural population. It is reported[1] by WHO in 1981 that approximately three out of five persons in the developing countries do not have access to safe drinking water. The urban areas are better served, 75 per cent of the population having some form of water supply through house connections or stand pipes while only 29 per cent have equivalent water supply in rural areas.

Fluoride has been reported by health organisations[2-5] as a health affecting substance. It has been recommended as an essential substance in water for building healthy teeth at levels within 1 mg/l concentrations. The maximum permissible limit of fluoride in water has been laid down as 1.5 mg/l by WHO and ICMR and 0.7 to 1.7 mg/l by USPHS. The effects of fluoride in drinking water have been studied in detail. Consumers exhibiting positive symptoms after taking such water have attracted the attention of the medical profession. The research work[6-11] which led ultimately to the introduction of fluoridation, began as an investigation into the cause of mottled teeth in certain communities in USA. Much work has been reported by Dean[12,13] and Elvove[14] on the relation between mottled teeth (dental fluorosis) and the fluoride level in water.

In India, a disease in humans similar to mottled enamel was first reported by Vishwanathan[15] to be prevalent in Madras Presidency in 1933. Mahajan[16] reported a similar disease in cattle in certain parts of old Hyderabad State. However, Schortt[17] was the first to identify the disease as 'fluorosis'. Subsequent to these findings, cases of fluorosis have been reported from several other parts of the country.

The arid zone in India occupies nearly 12 per cent of the total area sustaining a human population of over 19 million and livestock over 23 million, The Thar desert, extending 4.46 lakh sq. km. in India and Pakistan territories, is covered with wind blown sand and sand dunes. Rajasthan desert covers an area of 1.96 lakh sq.km. Defence Laboratory, Jodhpur (DLJ) is monitoring the water quality of this area since 1960 and our studies have shown that the ground waters are generally brackish. Further, fluoride and nitrate are present throughout. The national objective is to provide safe and adequate water supply of proper quality to all the 1,53,000 problem villages by 1990. A problem village has been roughly defined as one which has no water source of its own and the nearest one is 1.6 km. or more away, Most of the villages of arid zone of Western Rajasthan fall under this category. Construction of well or tubewell costs about Rs. 50,000 in south-west portion and about Rs. 1.5 lakh in other parts. A water supply scheme at village level would cost about Rs. 5 lakhs.

In this paper, the chemistry of fluoride, its occurrence in water, metabolism, excretion, effect of ingestion in human systems and methods of fluoridation and defluoridation have been discussed. Occurrence of fluoride in Indian waters and detailed studies on ground waters of Rajasthan with special reference to desert waters have also been reviewed and some comments offered on permissible limit of fluoride in water. Scope for further investigations in the area is indicated.

2. Occurrence

2.1 *Concentration of Fluoride in Water*

Surface waters generally do not exceed 0.3 mg/l of fluoride unless they are polluted from other sources[18,19]. Extremely high fluoride concentrations (greater than 1000 mg/l) in surface waters, however, have been found where fluoride-rich volcanic rocks are common[20,21]. The average concentration of fluoride in sea water[22] is about 1.35 mg/l. Values ranging from 1 to hundreds of mg/l have been reported in geothermal fluids[23-25]. But fluoride in surface waters of peninsular India covering a few important rivers, streams and springs have been found by Deshmukh[26] to go up to 3 mg/l in Godavari bed, Andhra Pradesh, 3.5 mg/l in Chitravati and Tambraparni beds, Tamil Nadu and 12 mg/l in Kongal, Nalgonda, Andhra Pradesh.

Many workers have reported fluoride bearing waters in Andhra Pradesh[27-29], Punjab and Haryana[30,31], Maharashtra[32], Tamil Nadu[26], Karnataka[33], Madhya pradesh[34], Gujarat[35], Uttar Pradesh[36], Bihar[37] and Kerala and Lakshadweep[38]. Rao[38]

Table 1. Fluoride concentration in different states of India

Region	Range of Fluoride in Ground Waters in Different Places (mg/l) 0—1.5	1.5—4.0	4.0—8.0	More than 8.0
		3	4	
Andhra Pradesh	Anantapur, Chitoor, Medak	Anantapur, Hyderabad, Krishna, Kurnool, Nalgonda, Prakasam	Anantapur, Karimnagar, Krishna	
Maharashtra	Pachora, Patas	—	—	—
Tamil Nadu	—	Coimbatore, Dharampuri Madurai, North Arcot, Salem, Tiruchirapalli		— — —
Karnataka		Kolar, Tumkur Chikmangalur, Chitradurg		
Madhya Pradesh	Mehdipur, Tikri, Anjeri, Waghtachoti	Gorakhar Village, Chikli Village, Bijasni, Borepani	Chandora Village, Betul	
Gujarat	—	Mehasana, Surendra Nagar, Ahmedabad, Banaskantha	South-Eastern & Western parts of Mehasana Dist., Southern part of Kutch Dist., Western part of Jamnagar Dist.	Naliya in Kutch Dist.
Sourashtra		Bhavnagar, Jamnagar, Rajkot Dist.	—	
Kerala	Quilon, Alleppey Ernakulam, Trichur			
Lakshadweep	Kavaratti, Minicoy, Aminidivi			

(*Contd.*)

	2	3	4	5
Rajasthan	Major part of State	Parts of Ganganagar, Churu, Nagaur, Alwar, Bharatpur, Jaipur, Tonk, Jalore, Sirohi, Pali, Barmer, Jaisalmer, Bikaner and Jodhpur.	Parts of Jhunjhunu, Sirohi, Pali, Jalore, Churu, Jaipur, Nagaur, Barmer, Jaisalmer, Bikaner and Jodhpur;	Parts of Ganganagar Nagaur. Sirohi, Jalore, Pali, Jodhpur, Bikaner Jaipur, Bhilwara, Udaipur, Bharatpur Barmer and Jaisalmer.
Punjab		—	Faridkot	Bhatinda, Ferozepur and Sangrur.
Haryana	—	—	—	Parts of Haryana
Uttar Pradesh	—	— —	Unnao, Muradnagar, Modinagar, Muzaffarnagar, Meerut.	Raebareli.

reviewed the above results and categorised the areas into four groups showing the concentrations in the range of 0-1.5, 1.5-4.0, 4.0-8.0 and over 8.0 mg/l. Data are presented in Table 1.

2.2 *Distribution of fluoride in ground water of Rajasthan*

Out of 27 districts of Rajasthan 16 districts (Table 2) have fluoride bearing waters in concentration more than the permissible limit of 1.5 mg/l laid down by WHO[39]. Saxena[40] reported that in Rajasthan 6,000 villages (out of 33,000) have water with fluoride concentration exceeding the permissible limit and that dental and skeletal fluorosis is wide-spread in these villages[41,42]. During the last decade, Gopal and co-authors[43-46] conducted extensive survey of Rajasthan desert and reported the incidence of fluoride in ground waters of arid areas based on physico-chemical, toxicological and epidemeological studies. A number of workers[47-52] have also reported fluoride toxicity in ground waters of Rajasthan. Analyses of about 2700 water samples by the authors and other workers indicate that the fluoride concentration in some pockets is alarming and needs immediate attention.

Table 2. Maximum concentration of fluoride in 16 districts of Rajasthan (mg/l)

Districts	No. of samples	Fluoride concn.
Jaipur	120	28.1
Bhilwara	118	24.0
Udaipur	166	21.6
Bikaner	60	20.0
Bharatpur	100	18.4
Nagaur	110	12.3
Barmer	320	11.5
Jodhpur	450	11.2
Jaisalmer	387	8.0
Churu	444	30.0
Jalore	165	14.2
Pali	236	18.3
Sirohi	—	8.0
Ganganagar		8.0
Tonk		4.0
Alwar		4.0

As part of our regular programme, survey and assessment of water quality supplied to services have been carried out by undertaking detailed studies[45,46] of about 1300 water samples from four arid districts of Barmer, Jaisalmer, Bikaner and Jodhpur.

Some relevant parameters - pH, TDS, alkalinity, and fluoride are given in Table 3. Percentage frequency of ground waters of this area having different fluoride ranges is presented in Table 4. A water resources map (Fig. 1) showing distribution of fluoride in waters of these arid districts is also discussed. The map indicates the areas of high fluoride (> 10 mg/1), medium fluoride (4 – 10 mg/1) and low fluoride levels (2 – 4 mg/1), and areas containing potable water (F < 2 mg/1).

Table 3. Physico-chemical characteristics of ground waters of Barmer, Jaisalmer, Bikaner and Jodhpur districts of Rajasthan

Details	Range			
	Barmer	Jaisalmer	Bikaner	Jodhpur
pH	7.2–8.7	7.0–8.9	6.6–9.0	7.2–8 5
TDS (mg/1)	200–26140	160–29568	430–29140	104–11914
Alkalinity (as $CaCo_3$ in mg/1)	Traces–2837	128–1956	139–3301	210–3920
Fluoride (mg/1)	0.2–10.9	Traces–8.0	Traces–20.0	Traces–11.2

Table 4. Percentage frequency of ground waters of Barmer, Jaisalmer, Bikaner and Jodhpur having different, fluoride ranges

Details Fluoride range (mg/l)	Barmer	Jaisalmer	Bikaner	Jodhpur
0–1.5	56	61	17	55
1.5– 2	11	15	6	21
2–10	30	24	55	21
Above 10	3	Nil	22	3

3. Drinking Water Standards for Fluoride in Water

The first presentation of water quality standard was made in 1914 by USPHS. The fluoride concentration laid down by USPHS (1962)[53], WHO (1963)[54], and ICM*R* (1962)[55] have been amended (Table 5). The Ministry of Health, Govt. of India[56] has prescribed 1.0 and 2.0 mg/l as permissible and excessive limits for fluoride in drinking water. The USPHS drinking water standards of 1962 get a mandatory limit of fluoride based on the annual average of maximum daily ambient temperature as given in Table 6. The fluoride-temperature relationship is based on the premise that children drink more water in warm climates, and therefore, the fluoride content in the water supply should be reduced to prevent excessive total fluoride consumption[57–59].

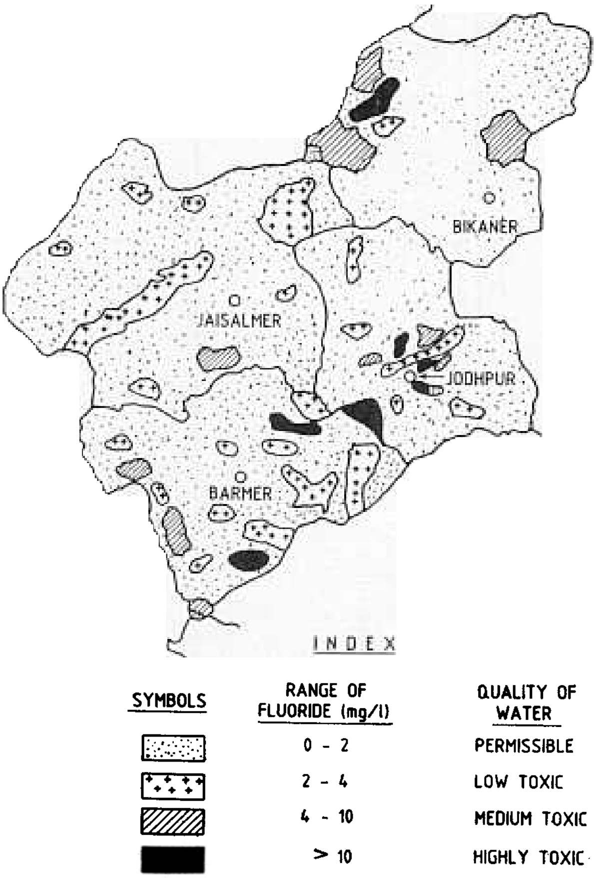

Figure 1. Distribution of fluoride in ground waters of four arid districts of Rajasthan.

Among the many methods suggested for determination of fluoride ion in water given in the official British and American Compilation of Methods, the calorimetric and electrode methods are the most satisfactory at the present time[60].

4. Metabolism

The role of fluoride in animal or human metabolism is not known with certainity. The element is metabolised from both electrovalent and covalent compounds. Fluorine in small quantities is essential for the formation of caries-resistant dental enamel and for the normal process of mineralisation in hard tissues[61]. Fluoride inhibits several enzyme systems, diminishes tissue respiration and anaerobic glycolysis. Low fluoride concentrations stabilise the skeletal system by increasing the size of the apatite crystals and reducing their solubility. The great affinity of fluorine for calcium phosphate is perhaps the most important from the physiological

Table 5. Drinking water standards

Chemical Constituents	U.S.P.H.S.		W.H.O			I.C.M.R.			
	Year	Maximum Allowable Limit (mg/l)	Year	Maximum concentration (mg/l)	Maximum Allowable concentration (mg/l)	Year	Permissible limit (mg/l)	Excessive limit (mg/l)	Highest Desirable limit (mg/l)
Fluoride	1962	0.7—1.2	1963		1.5	1962	1.0	2.0	—
(*F*)			1971	0.8—1.0	1.0—1.5	1975	—	1.5	1.0

Table 6. USPHS drinking water standard : recommended limit of fluoride concentration

Annual average of maximum daily air temperature		Recommended control limits of fluoride (mg/l)			Approval
°F	°C	Lower	Optimum	Upper	Limit
50.0—53.7	10.0—12.1	0.9	1.2	1.7	1 8
53.8—58.3	12.2—14.6	0.8	1.1	1.5	1.7
58.4—63.8	14.7—17.7	0.8	1.0	1.3	1.5
63.9—70.6	17.8—21.4	0.7	0.9	1.2	1.4
70.7—79.2	21.5—26.2	0.7	0.8	1.0	1.2
79.3—90.5	26.3—32.5	0.6	0.7	0.8	1.1

Sources : Drinking Water standards and guidelines, Water Supply Division, EPA, 1974.

point of view; it results in its accumulation in all tissues exhibiting physiological or morphological calcification. About 95 per cent of the fluoride in the body is deposited in hard tissues and it continues to be deposited in calcified structures even after other bone constituents (Ca,P,Mg,CO_3 and citrate) have reached a steady state. A pattern broadly similar to that in bone is followed in the fluoride concentrations in teeth. However, the uptake almost ceases in dental enamel after the age of about 30 years.

4.1 *Effects of Fluoride Ingestion in Human and Cattle*

The chronic toxic effects (fluorosis) of excessive intake of fluoride are usually observed as skeletal abnormalities or damage. Many detailed medical investigations have confirmed that there is no adverse effect on the body from continuous long-term consumption of 1 mg/l of fluoride in drinking water[62]. Mc Clure[63] has computed for children of various age groups the total daily intake of fluorides from food and water and established the following facts concerning the mottling of teeth[64].

4.1.1 Mottled enamel can be produced only during the period of calcification of the teeth. In other words, after about 12 years, mottled enamel cannot be produced whatsoever the level of fluoride in the water.

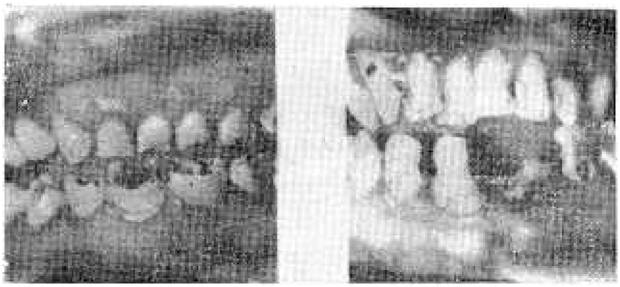

Figure 2. Fluorosis of teeth

4.1.2 Once such lesions are formed they cannot be repaired, either during the calcification or thereafter.

4.1.3 Fluoride appears to be the only agent that is ordinarily a part of the diet which has an effect on enamel production.

4.1.4 After calcification is completed the structure of the enamel remains unaltered despite changes in diet.

It has been established that physiology of human skeletan is not adversely affected by fluoride up to a level of 8 mg/l in drinking water[65]. Ingestion of 20-30 mg/day or more through water over a period of 10-20 years results in crippling fluorosis and severe osteosclerosis. Calcification of certain ligaments, rendering movements of joints difficult[66], is usually associated with atleast 10 mg/l of fluoride in drinking water. A WHO report[67] and studies of Smith andH edge[68] have also related concentration of fluoride to the biological effects. The above studies report the effects on thyroid at 50 mg/l, growth retardation at 100 mg/l and kidney changes at 120 mg/l concentration of fluoride. The bone structure was found to be blurred and it becomes a diffuse structureless shadow with uneven contours. These changes were marked in the spine and ribs. The acute lethal dose for man is between 2.5 and 5.0 gm depending upon the solubility of the compound and susceptibility of the man.

Effects of fluoride on animals are analogous to those on human beings. Studies in Australia have shown the development of exostosis of the long bone and mandibles in mature sheep, as well as the abnormal development and wear of teeth erupted by young sheep exposed to excessive fluoride[69].

4.2 *Excretion of Fluoride*

Fluorides are excreted through urine, faeces, sweat, and other body fluids. Urine of an average adult living in fluoridated water supply areas usually accounts for the principal excretion of fluorides eliminating 5.0 mg of fluoride daily[70]. Fluorides appear rapidily in urine after ingestion and generally the level reflects the daily intake. Depending upon the diet, the faecal excretion accounts for 10-30 per cent of the daily intake. Sweating may account for as much as 50 per cent of the total fluorides excreted in hot climate[65,71].

4.3 *Fluoride and Dental Caries*

Fluoride is the only halogen which combines with hydroxyapatite to form fluoroapatite structure in the tooth enamel making the tooth stronger and stable to acid. This chemical crystalline change occurs after 5-6 years of age and then exists for the life of the tooth[72,73]. Fluoride supplements become necessary for children living in areas having unfluoridated drinking water. Oral fluoride supplements are given in the form of tablets, drops, lozenges, fluoridated salt and multivitamin combinations. Flouride supplements during pregnancy have no conclusive beneficial effects since the crossover through the placental barrier of fiuoride is minimal. Topical fluorides including gels and toothpastes do not compare well with ingested flourides in caries prevention. However 30-40 per cent reduction in dental caries has been shown by use of such preparations.

Toothpastes contain 0.4 per cent stannous fluoride, 0.22 per cent sodium fluoride or 0.76 per cent monofluorophosphate, the last being the one more popularly used in our country. It has been shown that constant use of toothpaste reduces caries incidence by 15-30 per cent. Like the fluoride toothpastes, mouthrinses and mouthwashes containing 0.2 per cent sodium fluoride cause toxic symptoms.

Optimum concentration of fluoride intake from water for any temperature can be calculated by a generalised formula[74].

$$\text{Optimum } F = \frac{0.34}{E} \text{ mg/l}$$

where $E = (-0.038 + 0.0062\, t)$, is the estimated daily average water consumption for children of ten years of age in terms of ounces of water per pound of body weight and t is the annual mean of daily maximum temperature (°F).

5. Fluoridation

Fluoridation[75,76] is done in drinking waters in a number of European and non-European countries. Opposition to fluoridation has also been raised from time to time. During the last three decades hundreds of studies have been undertaken to correlate the concentration of waterborne fluoride and the incidence of dental caries. Health authorities throughout the world have endorsed that fluoride in drinking water prevents dental caries and controlled fluoridation is an acceptable public health measure. Recent studies[76] have indicated that fluoride also benefits older people in reducing prevalence of osteoporosis and hardening of arteries.

5.1 Chemicals Used

The three most commonly used fluoride compounds[77] in water treatment are sodium fluoride, sodium silicofluoride and fluosilicic acid, also known as hydrofluosilicic, hexafluosilicic or silico-fluoric acid. Besides these chemicals for fluoridation of water, others are hydrofluoric acid (HF), fluospar (CaF_2), ammonium fluosilicate [$(NH_4)_2 SiF_6$], magnesium fluosilicate ($MgSiF_6 . 6H_2O$) and stannous fluoride. Out of these only stannous fluoride is used in America owing to insufficient evidence about the long term effects of the other reagents. Ammonium and magnesium fluosilicate pose the danger of overdosing. Fluospar is insoluble as such but soluble in the presence of $Al_2(SO_4)_3$ and therefore its use is restricted. HF is highly corrosive and difficult to handle. Personal safety[78] in handling fluoride chemicals, limitation on dose of chemicals, provision of proper feeders for powder or liquid forms of chemicals and regular medical examination of workers etc. should be carried out.

6. Defluoridation

A variety of methods for fluoride removal are known[79–82]. They can be divided into two categories, viz precipitation and adsorption.

6.1 *Precipitation methods*

Methods involve the addition of chemicals and the formation of fluoride precipitate or co-precipitates. The chemicals employed include lime, magnesium compounds and aluminium sulphate. Lime (CaO) is the cheapest chemical employed for the removal of fluoride from water.[81] However, large amounts of fluoride are removed by reaction with calcium oxide followed by reaction with susperphosphate and calcium oxide or aluminium sulphate for the removal of residual fluoride. Based on this principle a number of workers[83–86] studied the use of lime or sodium aluminate, iron salts, alum and polyelectrolytes for coagulation. The well-known Nalgonda-technique[87] involves the addition of lime powder and bleaching powder (for disinfection) first and after thorough mixing with water, the alum solution is poured and stirred for 10 minutes. The contents are allowed to settle for one hour and the clear water is withdrawn. When magnesium hydroxide, magnesia and calcined magnesite were used along with lime, the fluoride removal capacity was found[83,88–91] to be better. Empirically the amount of fluoride removed is given by $0.07F\sqrt{Mg}$, where F represents fluoride initially present in mg, and Mg the magnesium removed in the form of flocks (mg).

The drawbacks of the precipitation method include the necessity for additional reagents, higher transportation and treatment costs, and the large volume of sludge produced. Filter alum is not economically feasible in some cases because of dosage requirements[79].

6.2 *Adsorption methods*

Adsorption methods[79,82] utilise the passage of the fluoride containing water through a contact bed. Fluoride is removed by ion exchange resins[32] and adsorbents like zeolites, phosphatic compounds with hydroxyapatite[83,92–94] activated alumina[83,95], activated carbon[96], Defluoron-1[32,97], Defluoron-2[98,99], and serpentine[100].

There are no known commercial anion exchange resins which are selective for fluoride only. Polystyrene anion exchange resins in general and strongly basic quarternary ammonium type resins in particular are known to remove fluorides from water along with other anions[101–102]. Cation exchange resins impregnable with alum solution have been found to act as defluoridating agents[103]. Venkatraman *et al*[104] reported that 'Avaram Bark' based cation exchange resins are effective in removal of fluoride from water. Active carbon prepared from spent coffee husk[105], coconut shell[106], saw-dust[107] and their sulphonated compounds with alum have also been found to remove fluoride efficiently.

Water with high fluoride (> 10 mg/l) concentration can economically be treated by split treatment using lime and alum in two stages[61]. Adsorption methods are usually appropriate for relatively low concentration of fluoride (10-20 mg/l) after the removal of fluoride by precipitation method to the 10-20 mg/l level.[108]

7. Fluoride Levels in Waters of Rajasthan Desert and Some Comments on Standards

Detailed studies on 1317 water samples of arid districts of Jaisalmer, Barmer, Bikaner and Jodhpur (Table 3) have shown that about 9 per cent waters are potable considering 500 mg/l total dissolved solids limit laid down by WHO and ICMR. It is observed that TDS varied between around 100 and 30,000 mg/l. The waters are generally brackish and alkaline having pH between 6.6 and 9.0. It is found that alkalinity due to carbonate is practically nil and very few samples were found to contain the maximum value of 190 mg/l. The alkalinity due to bicarbonate is found to be as high as 3920 mg/l in Jodhpur district.

All the waters have been found to contain fluoride and a maximum value of 20 mg/l was found in Bikaner. Rao[109] observed that natural waters having toxic amounts of fluorides are usually associated with alkalinity (as $CaCO_3$) in the range of 400-1000 mg/l. The arid waters of Rajasthan in general have shown[110] similar characteristics of excessive fluoride with high salinity and alkalinity. Optimum alkalinity level in the four arid districts have been found to vary from 1956 to 3920 mg/l. It is seen from Table 4 that about 47 per cent waters fall within the permissible limit of 1.5 mg/l of fluoride. For fluoride level exceeding 10 mg/l the frequency of samples is about 7 per cent. Saxena[40] and Gopal[43-46] *et al* have shown the occurrence of dental and bone fluorosis in the areas having excessive fluoride concentration. Continuous withdrawal of groundwater and depletion of water sources particularly in summer months have been shown to further increase the fluoride concentration. The above findings are in agreement[52,109] with recent surveys undertaken by Rajasthan Ground Water Department, Public Health Engineering Department, Rajasthan and Zonal Laboratory, National Environmental Engineering Institute, Jaipur.

Gopal *et al*[43-46,110-113] have reviewed the last 20 years work undertaken on the detailed survey and physico-chemical, toxicological, microbiological and desalination studies of brackish waters of Thar desert. It has been found that local population of the arid-areas is consuming for generations waters having TDS, 2000-3000 mg/l and fluoride up to 2 mg/l without any apparent deleterious effects. It is also gathered from Public Health Engineers, that a number of waterworks in Rajastan and Gujarat are supplying waters having fluoride up to 2 mg/l. Ministry of Health, Govt. of India[61] has also prescribed 2 mg F/l as the excessive limit in drinking water. In view of the scarcity of potable waters in arid districts of Rajasthan based on our studies and drinking water practices, this laboratory has recommened the maximum allowable limit of fluoride and TDS be extended up to 2 mg/l and 2000 mg/l respectively for potable water of these areas. With such relaxed limits of fluoride, 60 per cent of waters of these arid districts may be considered fit for consumption.

8. Conclusions

i) Fluoride bearing waters have been reported in Andhra Pradesh, Maharashtra, Punjab, Haryana, Tamil Nadu, Karnataka, Madhya Pradesh, Gujarat, Uttar Pradesh, Kerala and Lakshadweep. Some parts of a few states are fluorosis endemic areas. It is found that fluoride is widely distributed in waters Rajasthan. Dental and bone fluorosis are wide spread in 6000 villages of Rajasthan.

ii) Sedimentary rocks of limestone and sandstone formations having fiuorspar (CaF_2) are the main contributors of fluoride in ground waters of arid region. Quality of water of Thar desert is far from satisfactory. The ground waters of western Rajasthan are generally brackish having high salinity up to 10,000 mg/l (as $NaCl$) and high amounts of Sodium. Alkalinity due to carbonate is practically nil but bicarbonate (pH up to 9) is present in high concentrations (as high as 3920 mg/l). Residual sodium carbonate is high in fluoride bearing ground waters than the maximum limit of 2.5 mg/l.

iii) In general there exists a good linear positive relationship between F and HCO_3 ions and negative relationship between F and $Ca + Mg$ ions in high fluoride bearing ground waters. Most of these high F waters due to very high Na fall under high to very high alkali hazard waters.

iv) Methods of removal of fluoride have been enumerated in order to control excessive intake of F.

v) Based on detailed studies this laboratory has recommended[111,112] relaxation in the limit for F from 1.5 to 2 mg/l and TDS from 1500 to 2000 mg/l for desert waters. Water resources maps showing quality and quantity of water of arid areas have been prepared and found useful by local population, survey parties, armed forces and BSF deployed in this area.

9. Proposals for future studies

i) Water resources maps showing pockets of allowable concentrations of F and problem areas are required to be prepared district wise. The consumers should be educated on the beneficial and ill effects of this health affecting ion.

ii) Defluoridation schemes should be undertaken at Central and State Government levels particularly in areas where water is scarce and highly fluoridated.

iii) Detailed studies are required to be undertaken to establish correlation between the alkalinity and incidence of fluorosis as found in this study.

iv) The important weak hydrogen bonds formed between amides in biological systems are distrupted[114] by fluoride in the formation of much stronger bonds between *F* and amides. These raise the fluoride blood level and interfere in the healthy operations of living systems. The study needs immediate attention.

v) Fluoride blood levels in cattle and *F* content fixed in foodstuffs grown in fluoride bearing waters have not been studied. Rajasthan and Gujarat, the *F* prone states are one of the major suppliers of meat, milk and milk-products. The biochemical studies for fixation of *F* in these consumable products from *F* bearing waters and fodders etc. are required to be conducted country-wide to control the excessive F intake, which is being received from a growing number of sources [115,116].

vi) Not ignoring the beneficial effects of fluoridation up to 1.5 mg/l in drinking water in deficient areas indiscriminate use of *F* containing dental products like dentalgels and toothpastes, tablets and mouthwashes should be controlled immediately in endemic areas.

vii) Monitoring of water quality should be continued for *F* also using simple and rapid tests incorporated in field testing kits as undertaken by this laboratory with one such kit developed recently[113].

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