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Occurrence of Fluoride in the Drinking Water Sources from Gad River Basin, Maharashtra

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Abstract: High fluoride content in the groundwater is reported from parts of the Gad River Basin, Sindhudurg district, coastal Maharashtra, India. The fluoride content of up to 5 mg/l has been found in the groundwater in laterite, basalt and the Precambrian basement (gneiss) aquifers in the region. The presence of high fluoride in groundwater well above the permissible levels for consumption poses a serious health threat to the rural populace in the region. The presence of tourmaline bearing pegmatites in the Precambrian basement is considered as a potential fluorine source. Deep circulation of fluoride rich groundwater between the latetritised basalts and the underlying crystalline basement could be responsible for the occurrence of fluoride in both the shallow and deeper aquifers of the region.

Keywords: Fluoride, Groundwater, Drinking water quality, Health hazard, Maharashtra.

INTRODUCTION

The problem of excess fluoride (>1.5 mg/l in drinking water, Bureau of Indian Standards, 1991) in groundwater is endemic in 17 States of India. But its occurrence and distribution are non-uniform in different States – from one affected district in Jammu and Kashmir to all 32 districts affected in Rajasthan (UNICEF, 2000). The States of Andhra Pradesh, Gujarat and Rajasthan have become synonymous with the term 'fluorosis'. The sources for the occurrence of fluoride-rich waters in these states are traced to the presence of fluorite bearing rocks. Fluorosis is caused by ingestion of excess fluoride through drinking water, food, drugs and industrial wastes. Clinically, fluorosis appears in three forms – dental fluorosis, skeletal fluorosis and non-skeletal (soft-tissue) fluorosis, of which the former two types are irreversible. Rural populations that are dependent on groundwater for drinking water supply are the worst affected. Malnourished children, pregnant women, lactating mothers and poorer sections of the rural society are highly vulnerable to fluorosis.

Of the 6.9% of the Indian rural population at risk from fluorosis in the year 1999, 0.2% (0.14 million) are from the state of Maharashtra (UNICEF, 2000). In Maharashtra, high fluoride occurs mainly in the groundwater from Chandrapur, Nanded, Nagpur and Akola districts and is attributed to the pre-Deccan Trap rocks exposed in these districts (e.g. Kulkarni, 1998, Duraiswami, 2007, 2008). Of these occurrences, the occurrence of fluoride from Chandrapur has received considerable attention (Deshmukh et al. 1995). The occurrence of fluoride in groundwater from the drinking water sources in the Sindhudurg district of Maharashtra has been poorly reported in literature. The Precambrian rocks exposed in the district are considered to be the northwest extension of the Dharwar craton which is known for occurrences of fluoride (Suma Latha et al. 1999). Considering the direct health hazard and the social implications, compilation of information regarding the status of fluoride in this part of the State became imperative. The present paper is a preliminary note on the fluoride problem in this part of Maharashtra and discusses some low cost mitigatory measures to deal with the menace.

GAD RIVER BASIN

The Gad River Basin (~980 km^2) in the Sindhudurg district of Maharashtra constitutes a major westerly flowing drainage in the coastal belt of southern Konkan. The major towns within the basin include Phonda, Kankauli and Kasal (Fig.1). The climate across the basin is sub-tropical and is characterized by high monsoon rainfall (avg. 3642 mm), moderate temperatures (26-32°C) and high humidity (75-80%). The diurnal variation in temperature is less than 7°C between May to December and increases up to 10°C from January to March. The southwest monsoon

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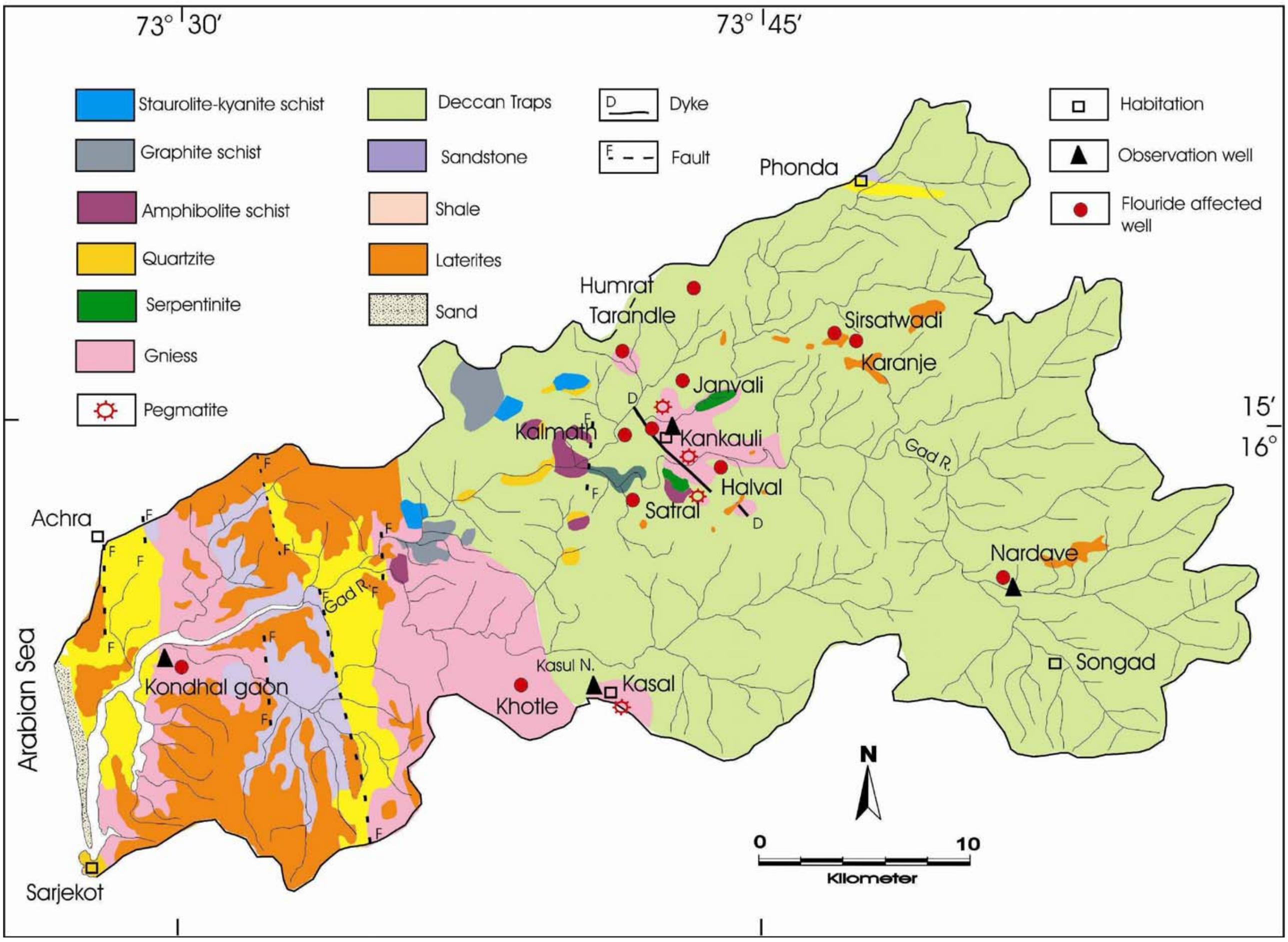

Fig.1. Photogeological map of the Gad River basin showing the fluoride-affected localities.

commences in the first week of June and continues till mid-October, but as much as 35% to 40% of the annual rainfall is received during the month of July, resulting in high run-off. Based on the Potential evapotranspiration and Moisture index, the climate of the region is classified as humid and tropical (Paranjpe, 2000).

The Gad River basin epitomizes all the physiographic characters of the Konkan coastal belt. The basin is bound on the west by the Arabian Sea and on the east by the mighty *Sahyadri* ranges. The coastline bounding the basin is characterized by the presence of the famous Achra beach – India's second longest beach (~12 km) after the Marina beach in Tamil Nadu. The coastline is followed by a broad coastal plain averaging 30 to 35 km inland above which stand out residual plateaus and hill ranges. The Gad River and its tributaries – the Janavali River and Kasul nala are the chief agents of fluvial erosion in the basin. The Gad River originates in the foothills of the *Sahyadri* ranges southeast of Songad and initially flows in the northwest direction. Near Kanedi bazar it changes direction and flows in an east-west direction until it meets the Janavali River near Kasral. The Janavali River has its source somewhere near the crest of the *Sahyadri* ranges, northeast of Phonda and flows in the general NE-SW direction. In its lower reaches, west of Marda, the Gad River suddenly swirls to attain a narrow linear almost NE-SW course, which ultimately culminates in to the Kalavli creek before discharging into the Arabian Sea near Sarjekot. The Gad River and its tributaries are rain fed and therefore the volume of water fluctuates considerably throughout the year.

GEOLOGY

A variety of rock types are exposed in the Gad River basin that span in age from the Precambrian to Recent. The Precambrian crystalline rocks equivalent to the Sargur Supergroup, and rocks belonging to the Peninsular gneissic complex are exposed at low altitudes along the major rivers (Fig. 1). Metamorphic rocks exposed towards the center of the basin, especially around Kankauli include pelitic rocks (staurolite-kyanite schist, mica-garnet schist, graphite schist), psammatic rocks (ferruginous quartzite, micaceous quartzite, dumorterite quartzite), calc-metasediments (crystalline limestone, talc schist), amphibolites, metaconglomerate and serpentinised rocks. Amphibolites comprise of a wide range of rock types like hornblende-quartz-biotite schist and hornblende schist resulting from metamorphism of basic igneous rocks and actinolite-tremolite schist from the metamorphism of impure calcareous sediments (Koregave, 1980). Highly serpentinised rocks and talc-tremolite schist are exposed within the gneiss near Vagda and Janavali represent the ultrabasic rocks in the Gad River basin. The serpentinised rock contains layers and lenses of chromite.

The gneissic rocks in the Gad River basin are exposed as inliers around Kankauli and in the southwestern part. The foliation in the gneiss is generally in the N-S and NW-SE direction with 50° to 70° dips towards E-NE. The contacts of the gneisses with the metasediments belonging to the Sargur Supergroup are generally gradational, implying that the gneisses are much younger than the metasediments (Koregave, 1980). A large number of pegmatites of varying shapes and size are present in the gneiss. The pegmatites are coarse grained, macro-block, veined bodies genetically related to the granitic residue. These are usually veined, lens-like and are often irregular, branched. The pegmatites also show pinching and swelling and exhibit typical graphic textures due to regular intergrowth of quartz and feldspars. Two generations of pegmatites are observed in the area. The older set is emplaced as thin veins along the foliation of the gneiss and the younger set that is thicker (~1 m wide) cut across the foliation. The mineral composition of the pegmatites corresponds to that of the surrounding gneissic rocks and is of the quartz-feldspar (orthoclase/ microcline)-biotite±tourmaline type. Large number of parallely disposed (trend: 160°-340°) thick quartz veins and pegmatites intrude the amphibolites south of Vagda. The quartz veins stand out as ridges and exhibit pinching and branching at places. In this area, two types of pegmatites are found viz. quartz-tourmaline-muscovite pegmatite and quartz-garnet-muscovite pegmatite. The common occurrence of tourmaline in the pegmatites (Type III of Fersman, 1940) from the Gad River basin indicates widespread boron pneumatolysis.

The conglomerates, sandstones and shales belonging to the Kaladgi Supergroup are exposed in the lower reaches of the Gad River basin. The sediments rest unconformably over the Peninsular gneissic complex and are in turn overlain by laterites and/or lava flows belonging to the Deccan Trap. The sediments show a general NNW-SSE strike direction and dip by 20°-35° towards WSW. The westerly dips of these sediments may be due to the influence of the West Coast fault system (Sarkar, 1986). The Deccan Traps and laterites cover large portions of the Gad River basin. The lava flows are confined to the middle and upper reaches of the basin. The flows are of simple variety and belong to the Ambenali Formation of the Wai Subgroup (Subbarao and Hooper, 1988). A NNW-SSE trending dolerite dyke is exposed near Kankauli and is a feeder to the lava flow exposed in the railway-cutting southwest of the town (Karmalkar et al. 1998). Laterites derived from weathering of basalts, metamorphic, and sedimentary rocks cover large tracts of the Gad River basin (Fig. 1).

HYDROGEOCHEMISTRY

Groundwater occurs under phreatic condition in the various aquifers in the Gad River basin. The aquifers can be broadly classified into three main types viz. gneissic, basaltic, and lateritic aquifers. The depth of dug wells tapping these aquifers varies from 4 to 20 m. The bore wells are usually shallow (< 60 m) and most of these are sunk with a view to provide safe, potable drinking water. There are in all 5-observation wells (Kandalgaon, Kasal, Kankauli, Kasuli and Nardave) in the Gad River basin, set up by GSDA to monitor the water levels in the different types of aquifers. The water level record from 1989 to 2009 show a declining trend in the pre- and post-monsoon seasons for the lateritic (Kandalgaon and Nardave) and basaltic (Kasuli) aquifers (Fig. 2). However, the post-monsoon water levels from the observation wells at Kasal and Kankauli i.e. gneissic aquifers show rising post-monsoon trends during this period. The declining water level trends in the basaltic and lateritic aquifers could be attributed to their lower water retention capacity due to their jointed and porous nature leading to high transmissivity. Another interesting observation is seen from long term hydrographs (Fig. 2) viz. for the same amount of rainfall, the water levels in different aquifers behaves differently. Thus a large degree of fluctuations is seen in the lateritic aquifers in comparison to the gneissic aquifer.

Groundwater samples were collected in one litre poly-ethylene bottles and the physio-chemical characteristics were determined according to the analytical procedure (APHA, AWWA and WPCF, 1981). Fluoride was determined in the samples by the SPADNS spectro-photometric method. Although a large number of water samples from different drinking water sources within the Gad River basin were collected and analysed, only those samples that contain fluoride above the permissible limits (Bureau of Indian Standards, 1991) are considered for the

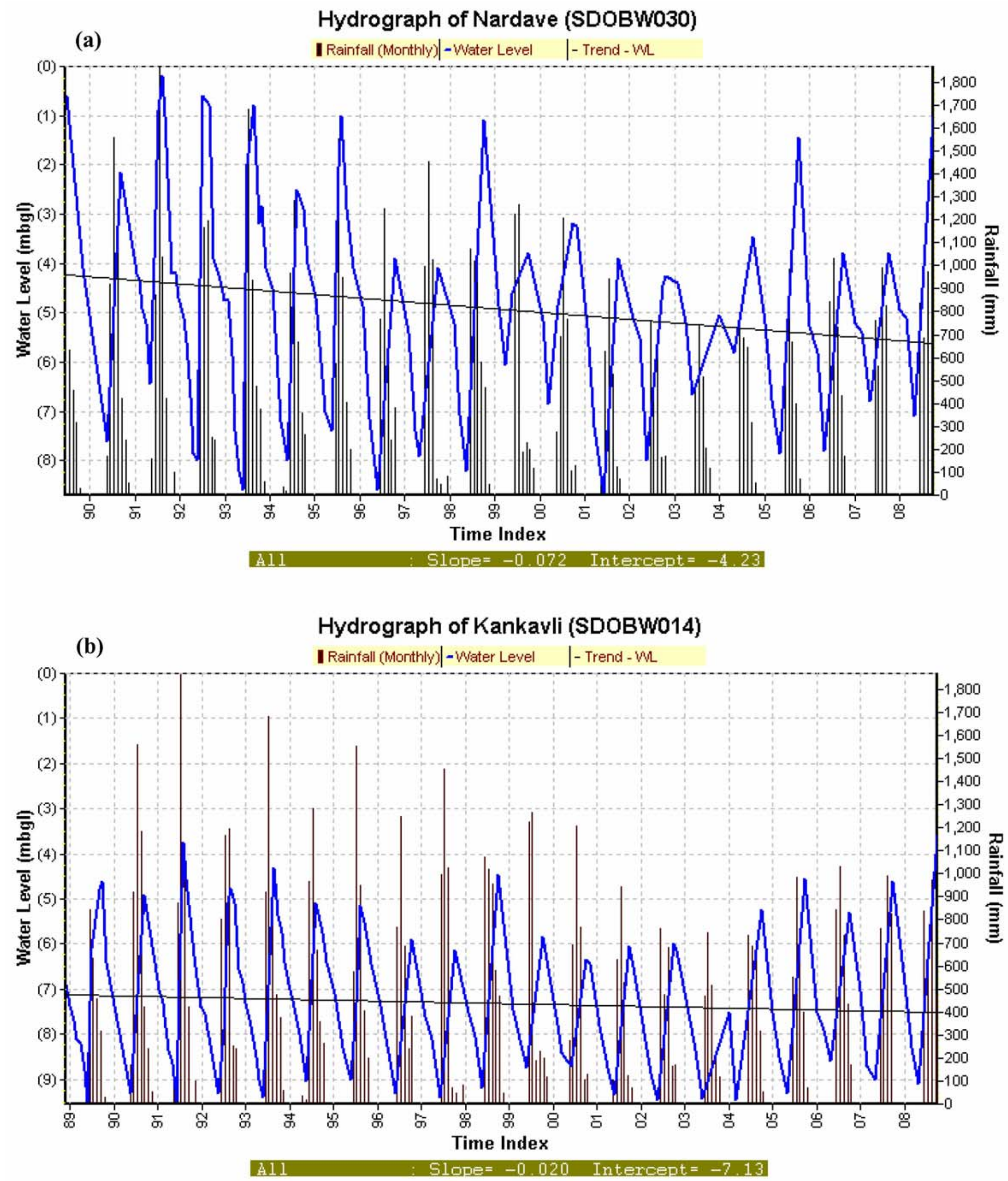

Fig.2. Hydrographs of groundwater levels in **(a)** lateritic and **(b)** granite gneiss aquifer.

present communication and are given in Table 1. The groundwater from the study area is alkaline in nature (pH: 7.0 to 8.9) with the electrical conductivity ranging from 145 to 557 micromhos/cm. A positive correlation (Fig. 3) is seen between the pH of groundwater and the fluoride content (correlation coefficient, r: 1.6529) indicating that the pH, and hence alkalinity, influences the fluoride content in the groundwater. This is because alkaline waters dissolve fluoride-bearing minerals by precipitating calcium magnesium carbonates simultaneously. Positive correlation is also seen between sodium and fluoride (*r*: 0.0207). A weak negative correlation is seen between fluoride and calcium (*r*: - 0.0367) and total hardness (*r*: -0.008) in contrast to a pronounced negative correlation with magnesium (*r*: -0.14). Negative correlation of calcium and magnesium vis-à-vis fluoride is expected due to the low solubility of the fluorides in natural groundwater (Handa, 1975a; Hem, 1991). However, in case of the Gad River basin, the weathering of plagioclase and augite from the basaltic flows has led to a steady supply of calcium to the groundwaters leading to a subdued negative correlation with fluorine.

Chemical analyses of groundwaters from the Gad River basin were classified graphically (Piper, 1994) using

Table 1. Geochemical analyses (mg/l) of select groundwater samples from Gad River basin

Village	Deulwadi	Halwal		Humrat	Janavli		Kalmath	Kankavli
Location	Deulwadi	Rly. Line	Rly. Line	Garage	BKG Rd.	School	Gavdewadi	Gangowadi
Source	BW	BW	BW	BW	BW	BW	BW	BW
Sample no.	1	2	3	4	5	6	7	8
Aquifer	Basalt	Gneiss	Gneiss	Basalt	Basalt	Laterite	Basalt	Laterite
pH	8.0	8.0	7.5	8.1	8.0	7.3	7.5	7.7
E. C.	557	218	260	534	220	145	485	426
TDS	356	140	166	842	141	93	310	273
TH	144	105	160	64	76	160	64	140
TALK	224	171	256	148	112	67	92	183
Ca	27	30	62	211	14	34	26	34
Mg	18	7	1	3	16	18	0	13
Na	46	31	31	45	17	8	67	19
K	5	0	0	0	3	0	0	4
CO_3	0	0	0	16	0	0	16	2
HCO_3	224	171	256	132	112	67	76	181
Cl	58	18	11	32	38	107	30	50
SO_4	16	3	2	113	5	5	95	5
NO_3	0	7	6	0	0	0	0	4
Fe	0.3	0.2	0.2	0.2	0.3	1.8	0.3	0.1
F	2.4	5.0	5.0	4.9	2.4	1.6	2.5	1.9

Table 1. Contd...

Village	Karanje						Nardave	Satral	Khotle	Tondavali
Location	Karanje	Karwabhe	School	Shirsatwadi	Shirsatwadi	Shirsatwadi	TS22	Varchawadi	Kondwadi	Tondavali
Source	BW	BW	BW	BW	BW	BW	DW	BW	BW	BW
Sample no.	9	10	11	12	13	14	15	16	17	18
Aquifer	Basalt	Basalt	Laterite	Laterite	Laterite	Laterite	Laterite	Basalt	Basalt	Gneiss
pH	7.9	7.8	8.9	8.0	8.0	7.8	7.9	7.4	7.3	7.6
E.C.	425	163	172	393	172	481	460	138	174	240
TDS	273	104	110	252	110	308	294	88	111	154
TH	24	92	20	155	20	20	20	0	112	135
TALK	108	156	64	171	55	92	140	84	108	195
Ca	8	18	8	42	8	8	6	22	38	44
Mg	1	12	0	12	0	5	1	6	4	6
Na	106	8	51	16	100	99	91	0	28	33
K	0	0	0	4	0	4	3	0	2	0
CO_3	8	16	9	0	0	0	0	8	16	0
HCO_3	100	140	55	171	55	92	140	76	92	195
Cl	34	140	4	36	4	36	11	36	54	14
SO_4	34	20	66	8	56	54	62	2	5	25
NO_3	4	9	0	0	0	2	0	0	0	16
Fe	0.2	0.2	0.4	3.5	0.2	0.1	0.3	0.6	3.0	0.2
F	4.0	2.0	4.8	1.6	5.6	3.2	3.2	1.6	3.0	2.0

percentages of milliequivalents of the major cations and anions (Fig. 4). Calcium-magnesium-bicarbonate is the most common groundwater quality type in the basin, especially in the Trap-dominated areas. The groundwater samples from Janavali and Karwhabe show the predominance of magnesium cation and this can be attributed to the ultrabasic (sepentinite) formation exposed in the vicinity. In samples obtained from or near proximity of the gneissic aquifers, the sodium-bicarbonate/sulphate groundwater quality predominates and could be attributed to the high incidence of alkali elements in the gneissic basement. Similarly, sodium cation is also predominant in some samples (Nardave, Shirsatwadi, Karanje) obtained from abstraction structures that tap the basaltic aquifers well within the mainland. Alternatively, the sodium could be added to the groundwater either due to leaching during the process of lateritisation or

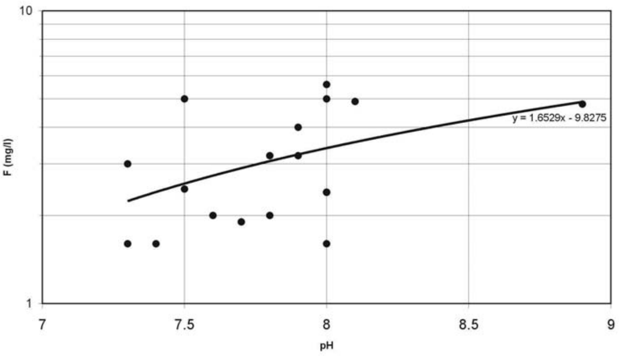

Fig.3. Positive correlation between pH and fluoride in groundwater from the Gad River basin.

through saline aerosols from the Arabian Sea. Whatever the process, it is clear that calcium cations are replaced by sodium in the groundwater by base-exchange process. Thus, the calcium-magnesium-bicarbonate water is modified to sodium-bicarbonate and sodium-sulphate water types by ion exchange.

DISCUSSION

The interplay between weathering of host lithotype, residence time of groundwater and ion exchange results in the complex distribution of fluoride in the groundwater of the Gad River basin. During weathering, in humid, sub-tropical climates the geochemical fate of fluorine is controlled by a series of intricate adsorption-desorption and dissolution-precipitation processes (Deshmukh et al. 1995).

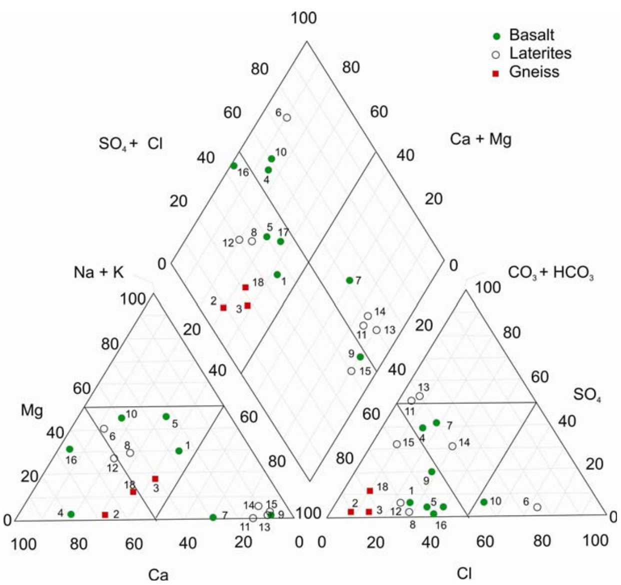

Fig.4. Trilinear diagram (Piper, 1994) used to classify chemical types of groundwater samples from Gad River basin.

Fluorine is reported from several pegmatitic minerals like biotite (0.22 to 3.05 wt%, Seraphim, 1951), muscovite (0.04 to 2.06 wt%, Deer et al. 1966; Nemec, 1969), tourmaline (0.07 to 1.27 wt%, Deer et al. 1966; Nemec, 1969) and apatite (2.37 to3.36 wt%, Shmakin and Shiryayeva, 1968). Unlike fluorine in apatite, which is relatively stable during weathering, fluorine from micas is highly susceptible and is rapidly leached out (Deshmukh et al. 1995). Therefore enrichment of fluoride in groundwater is common where Precambrian rocks are intruded by pegmatites. Precambrian gneisses and tourmaline-mica pegmatites are commonly present in the Gad River basin. The occurrence of fluoride in groundwater close to the known pegmatite occurrences around Kankauli suggests that the minerals within the pegmatite (tourmaline) could be the source of fluoride. In the Gad River basin the chemical composition of groundwater can be used as a proxy indicator of potential fluoride problem (Frencken, 1992). High fluoride groundwaters are usually associated with sodium-bicarbonate water types that have high pH (above 7) and are relatively low in calcium and magnesium. Thus, fluoride could occur within the basin in gneissic areas that are intruded by tourmaline-mica pegmatites and where sodium bicarbonate waters occur. Hence, detailed mapping and mineralogical studies of the pegmatites from the basin could help identify the probability of fluoride-affected habitations.

High fluoride concentration can also build up in groundwaters with long residence time in the aquifers. In case of the Gad River basin the fluoride appears to be concentrated in deep borewells (Table 1) rather than the shallow dug wells. The deeper crystalline rocks such as those exposed in the basin have low primary porosity with groundwater occurring in joints and fractures. In such an environment, the groundwater movement is restricted to the joints/fractures and there is very slow movement through the primary porosity. This increases the residence time of groundwater that results in the increase in fluoride concentration. Discontinuance of existing fluoride affected borewells and tapping drinking water reserves from the shallow dug wells could be effective in preventing the ill effects of fluorosis.

There is a certain seasonality of fluoride content in the groundwater regime that is pronounced in the humid tropical climate of the Konkan region. This is primarily because in pre-monsoon the mobility of fluoride is inhibited due to the high availability of calcium while in post-monsoon the mobility is greatly enhanced due to low calcium availability and higher ionic strength of the groundwater (Handa, 1975a, b; 1977). Rooftop rainwater harvesting and artificial recharge of existing borewells appear to provide viable method to dilute the fluoride content (Chakraborti, 1995).

Occurrence of fluoride in groundwaters has drawn worldwide attention as its deficiency (< 0.6 mg/l) causes dental caries and excess (>1.5 mg/l) causes skeletal fluorosis (Dissanayake, 1991). The presence of fluoride from the deeper aquifers of the Gad River basin provides ample evidence to suggest that geology plays a vital role in the occurrence of fluoride in the drinking water sources. Considering this, the present account of occurrence of fluoride in the groundwater from this part of the State should be followed by detailed geohydrological, medical, nutritional and socio-economic surveys for studying the effects of fluorosis amongst the rural habitants. The groundwater sources from the present study contain fluoride to the tune of 1.5 to 5.6 mg/l. As per these values for fluoride, the occurrence of dental fluorisis is by far the most common manifestation (Dissanayake, 1991). Children below the age of 7 are particularly vulnerable considering the fact that fluoride has a greater impact on growing teeth. Hence, it is important to undertake a survey of oral hygiene, especially in primary schools located within the Gad River basin so that the impact of long-term use of fluoride water can be evaluated.

Suitable low cost removal methods such as the Nalgona technique (Navlakhe and Bulusu, 1989) should be introduced in the area. In this technique, a combination of alum (or aluminium chloride) and lime (or sodium alunimate), together with bleaching powder, are added to high-fluoride water, stirred and left to settle. Fluoride is subsequently removed by flocculation, sedimentation and filtration. Other precipitation methods include the use of gypsum, dolomite or calcium chloride. The most common ion-exchange removal method is based on activated carbon, activated alumina, ion-exchange resins, plant carbon and clay minerals. The other low cost method to tackle fluoride is adoption of roof top rainwater harvesting (Viswanath, 2004). Other highly efficient methods of removal include electrodialysis and reverse osmosis, but high cost and O&M are problematic (Solsona, 1985). Adsorption/ion exchange by plant carbon (Solsona, 1985; Heidweiller, 1990) and other mitigatory measures (Susheela, 1993) can also be considered.

Although removal techniques are recommended, it is diet and nutrition than can control the damaging effect of fluorosis. Calcium and vitamin C deficiencies are recognized as important exacerbating factors. Hence, diet rich in calcium and vitamin C needs to be supplemented in fluoride endemic habitations. Thus, good nutritional and Ca-rich dietary habits of people residing in the Sindhudurg district has resulted in low manifestation of fluorosis despite fluoride occurrence in groundwater, while the vice versa is true in case of people residing in Chandrapur district. Hence the best way of mitigating fluorosis is to tackle and change dietary habits of vulnerable people from the lower income group. This will be a small step in tackling the menace of the ‘crippling giant’.

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