

Geochemistry of Fluoride Bearing Groundwater in Parts of Telangana State, India

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Abstract-Hydrogeochemical investigations carried out in parts of granitic tracts of Telangana region of India revealed that the fluoride concentrations in the groundwater are much higher than the prescribed limit ($>1.5\text{mg/l}$). It is observed that fluorine bearing minerals like fluorite and apatite present in granitic rocks are the major source for the excessive fluoride in groundwater. Geology of the area and climatic conditions play vital role for the high concentrations of fluoride in groundwater. In addition, depletion of groundwater levels due to climatic changes is another factor for high incidence of fluorosis.

Keywords- Fluoride; Fluoride Contamination; Groundwater; Granitic Terrain; Fluorine Bearing Minerals; Telangana; India

I. INTRODUCTION

One of the serious health problems facing the country today is the prevalence of the disease known as fluorosis involving millions of people from the arid and semi-arid regions of the world which arises primarily due to excess fluoride in drinking water in the areas concerned. Human beings as well as livestock are seriously affected and even crippled by this dreaded disease [1]. Fluoride ingested with drinking water is almost completely absorbed by the human body. It is rapidly distributed and mainly retained in skeleton. A small portion may be retained in teeth. WHO prescribes 1.5 mg/L fluoride concentration in drinking water as the upper limit. However, prevalence of fluorosis is reported at much lower concentrations in many parts of the globe. The problem of high concentration of fluoride in groundwater resources has become one of the most important toxicological and geo-environmental issues in India [2, 3]. In most of the fluorosis endemic areas, the average summer temperature is above 27.5°C and average drinking water consumption is more than 4 liters per day [4]. The fluorosis is endemic in 17 states of India. The most seriously affected areas are Andhra Pradesh, Telangana, Punjab, Haryana, Rajasthan, Gujarat, Tamil Nadu and Uttar Pradesh [5-7]. In India, about 62 million people including 6 million children suffer from fluorosis because of consumption of water with high fluoride concentrations [8]. Most parts of Andhra Pradesh and Telangana in India have highly endemic fluorosis zones [9-16]. The fluoride is beneficial to certain extent when present in the concentration of 0.8 to 1.0 mg/L for calcification of dental enamel especially for children below 8 years, whereas causes dental fluorosis if present in excess of 1.5 mg/L and skeletal fluorosis beyond 3.0 mg/L if such water is consumed for 6 months to several years [17]. Most F- accumulation in the human body occurs through F- contaminated drinking water, substantial amounts of F- can also be ingested through crops and vegetables irrigated with F- contaminated water. In the present paper, distribution of fluoride in the groundwater occurring in the granitic terrain of parts of Telangana region of India is highlighted with an aim to identify and delineate high fluoride bearing groundwater zones.

Study area: Hydrogeochemical investigations have been carried out in five areas of Telangana region of India to understand the occurrence of fluoride in the groundwater (Fig. 1). The Telangana region of India is occupied by the Granitic rocks of Archaean age which forms part of the stable Dharwar craton of south Indian shield (Fig. 2) [18].

Chennaraopet area (Warangal district): The area is located in the central eastern part of Warangal district of Telangana, India and forms a part of the Survey of India toposheet 56 O/13. The area geographically lies between longitude $79^\circ 32' - 79^\circ 54' \text{E}$ and latitude $17^\circ 33' - 17^\circ 55' \text{N}$.

Gangadhara area (Karimnagar district): The study area is located in the central eastern part of Karimnagar district of India and forms a part of the Survey of India toposheet 56N/2, 56J/14. The area geographically lies between longitudes $78^\circ 59' - 79^\circ 30' \text{E}$ and latitudes $18^\circ 20' - 18^\circ 35' \text{N}$.

Kalwakurthy area (Mahabubnagar district): The study area falls in the Survey of India toposheet 56 L/6 and 56 L/10 and lies in between longitudes $78^\circ 24' 00'' - 78^\circ 28' 48'' \text{E}$ and latitudes $16^\circ 34' 30'' - 16^\circ 42' 00'' \text{N}$.

Devarakonda area (Nalgonda district): The study falls in between longitudes $78^\circ 45' - 79^\circ 0' \text{E}$ latitudes $16^\circ 40' - 16^\circ 45' \text{N}$ and in Survey of India Toposheet No. 56L/14.

Basara area (Adilabad district): The study area is located in the south-western part of the Adilabad district and forms a part of the Survey of India toposheet 56 F/13. The area lies between longitudes $77^\circ 54' - 77^\circ 59' \text{E}$ latitudes $18^\circ 51' - 18^\circ 59' \text{N}$.

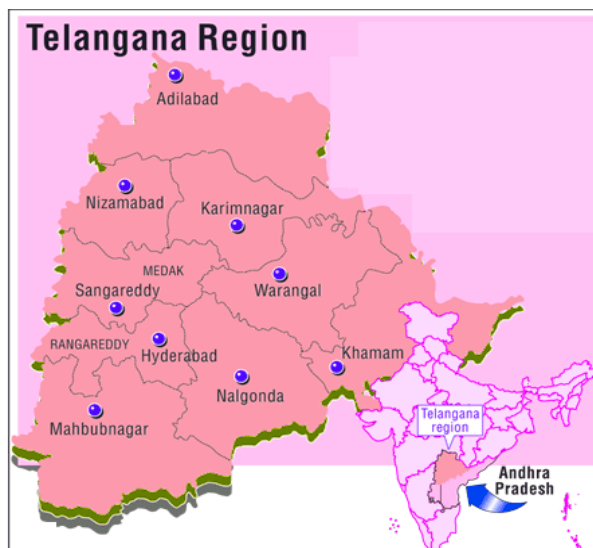


Fig. 1 Map showing Warangal, Karimnagar, Mahabubnagar, Nalgonda and Adilabad in Telangana state, India



Fig. 2a Geological map of Telangana region, India [18]

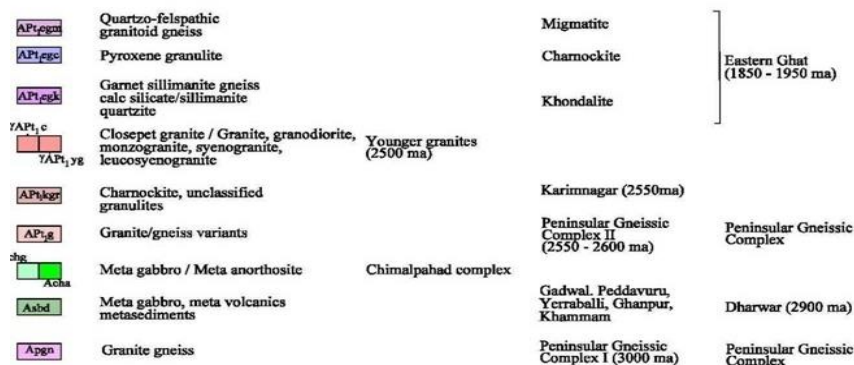


Fig. 2b Legend of the Geological map [18]

II. MATERIALS AND METHODS

Fifty seven groundwater samples from Chennaraopet area, one hundred and seven groundwater samples from Gangadhara area, fifty six groundwater samples from Kalwakurthy area, sixty groundwater samples from Devarakonda area and thirty four groundwater samples from Basara area were collected in clean two liter polythene bottles and analyzed for pH, electrical conductivity (EC), fluoride (F-) and other major ions as per standard methods [19]. Fluoride concentrations were measured

with Orion ion analyzer. The analytical results are presented in the Tables 1&2. The fluoride distribution maps are presented in Figs. 3 to 7.

III. RESULTS AND DISCUSSION

Fluoride concentration in the groundwater varies from 0.3 mg/L to 8.0 mg/L (Fig. 3) while 47% of groundwater shows excess fluoride prescribed for drinking purpose (>1.5 mg/L) in the Chennaraopet area of Warangal district. The pH in the groundwater ranges from 7.45 to 8.62 and EC varies from 92 to 5974 $\mu\text{S}/\text{cm}$. In the Gangadhara area of Karimnagar district, the fluoride concentration varies from 0.6 mg/L to 5.0 mg/L (Fig. 4) and 65% of groundwater shows excess fluoride prescribed for drinking purpose. The pH varies from 7.12 to 8.56 and EC ranges from 130 to 4332 $\mu\text{S}/\text{cm}$. Fluoride concentration in the groundwater of Kalwakurthy area of Mahabubnagar district varies from 0.16 mg/L to 3.4 mg/L (Fig. 5) while 46% of groundwater shows excess fluoride prescribed for drinking purpose. The pH varies from 7.4 to 8.8 and EC ranges from 78 to 1569 $\mu\text{S}/\text{cm}$. In the Devarakonda area of Nalgonda district, the fluoride concentration varies from 0.17 mg/L to 2.84 mg/L (Fig. 6) and 28% of groundwater shows excess fluoride prescribed for drinking purpose. The pH varies from 7.65 to 9.72 and EC ranges from 157 to 1020 $\mu\text{S}/\text{cm}$. In the Basara area of Adilabad district, the fluoride concentration varies from 0.06 mg/L to 4.33 mg/L (Fig. 7) and 20% of groundwater shows excess fluoride prescribed for drinking purpose. The pH varies from 6.47 to 8.03 and EC ranges from 235 to 2118 $\mu\text{S}/\text{cm}$. The ranges and average concentrations of other major ions are presented in Table 2.

TABLE 1 CONCENTRATION OF FLUORIDE (F-) IN THE GROUNDWATER OF DIFFERENT AREAS OF TELANGANA REGION, INDIA

Parameters	Chennaraopet (Warangal)	Gangadhara (Karimnagar)	Kalwakurthy (Mahabubnagar)	Devarakonda (Nalgonda)	Basara (Adilabad)
pH	7.45-8.62 (8)	7.12-8.56 (7.84)	7.42-8.8 (8.1)	7.65-9.72 (8.5)	6.47-8.03 (7.1)
EC*	92-5974 (1851)	130-4332 (2231)	78-1569 (824)	157-1020 (434)	235-2118 (595.2)
TDS	59-3341 (1412)	276-2641 (1458)	50-1004 (527)	100-652 (277)	150-1355 (377.8)
TH	20-815 (214.9)	75-625 (95)	75-560 (317)	72.5-450 (187)	35-850 (197.3)
Ca ²⁺	2-208.4 (22.8)	6-184 (95)	16-154 (85.2)	20-154 (45)	8-339 (58.9)
Mg ²⁺	2.4-111 (38.5)	1.2-102 (51.6)	2.9-83.8 (43.4)	2.42-48.6 (18.4)	1-29 (11.9)
Na ⁺	8-125 (61.8)	4-94 (49)	17-182 (99.5)	17-192 (80)	15-810 (146.2)
K ⁺	1-35 (4.96)	1-22 (11.5)	10-47 (28.5)	1-18 (4)	1-39 (5.7)
Cl ⁻	24.8-1118.2 (311.7)	21.3-78 (49.7)	21.3-678 (350)	32-500 (148)	42-1010 (182.1)
HCO ₃ ⁻	18.3-427 (222.6)	140-1250 (195)	48.8-220 (122)	73-226 (149)	43-183 (104.4)
NO ₃ ⁻	3-40.7 (28.9)	6-140 (73)	1.1-112 (57.3)	1.5-125 (61)	0.4-80 (22.1)
SO ₄ ²⁻	1-101.2 (34.9)	10.5-112 (61.2)	4-180 (92)	3-62 (25)	2.5-375 (56.9)

TABLE 2 RESULTS OF THE CHEMICAL ANALYSIS OF MAJOR IONS (RANGE AND AVERAGES IN MG/L) OF DIFFERENT PARTS OF TELANGANA STATE (*MS/CM)

Area	Lowest F concentration in mg/L	Highest F concentration in mg/L	Average F concentration in mg/L	% of groundwater exceeding the prescribed limit of 1.5 mg/L
Warangal district Chennaraopet area	0.3	8.0	1.65	47.0
Karimnagar district (Gangadhara area)	0.6	5.0	2.8	65.0
Mahabubnagar district (Kalwakurthy area)	0.16	3.4	1.86	46.0
Nalgonda district (Devarakonda area)	0.17	2.84	1.06	28.0
Adilabad district (Basara area)	0.06	4.33	1.13	20.0

The principal fluorine-bearing minerals of the granitic rocks present in these areas as accessories are fluorite (CaF_2), apatite ($\text{Ca}_5(\text{PO}_4)_3(\text{F}, \text{OH}, \text{Cl})$), muscovite $\{\text{K}_2\text{Al}_4[\text{Si}_6\text{Al}_2\text{O}_{20}](\text{OH}, \text{F})_4\}$, biotite $\{\text{K}_2(\text{Mg}, \text{Fe}^{2+})_{6-4}(\text{Fe}^{3+}\text{Al}, \text{Ti})_{0-2}(\text{Si}_{6-5}\text{Al}_{2-3}\text{O}_{20})\text{OH}, \text{F}_4\}$. These minerals are major source for the fluoride in the groundwater of the areas concerned. The amount of fluoride occurring naturally in groundwater is governed by climate, composition of the host rock, and hydrogeology [20]. The fluoride content is a function of many factors such as availability and solubility of fluorine bearing minerals, velocity of flowing water, temperature, pH, concentration of calcium and bicarbonate ions in water, etc. [21, 22]. Many fluorine bearing minerals occur in nature and experimental data reveals that under the conditions of normal circulation, water with pH range of 6 to 8, fluorite can

release amounts of fluoride 20 to 50 times more than other fluoride bearing minerals [23]. Another important relationship between fluoride concentration in groundwater and seasonal water level fluctuation has been reported and found that with an increase in the water level in the well, fluoride concentration of water was found to be decreasing. They attributed this phenomenon to the dilution of fluoride with fresh inflow into the system and concentration as the water slowly depleted [24].

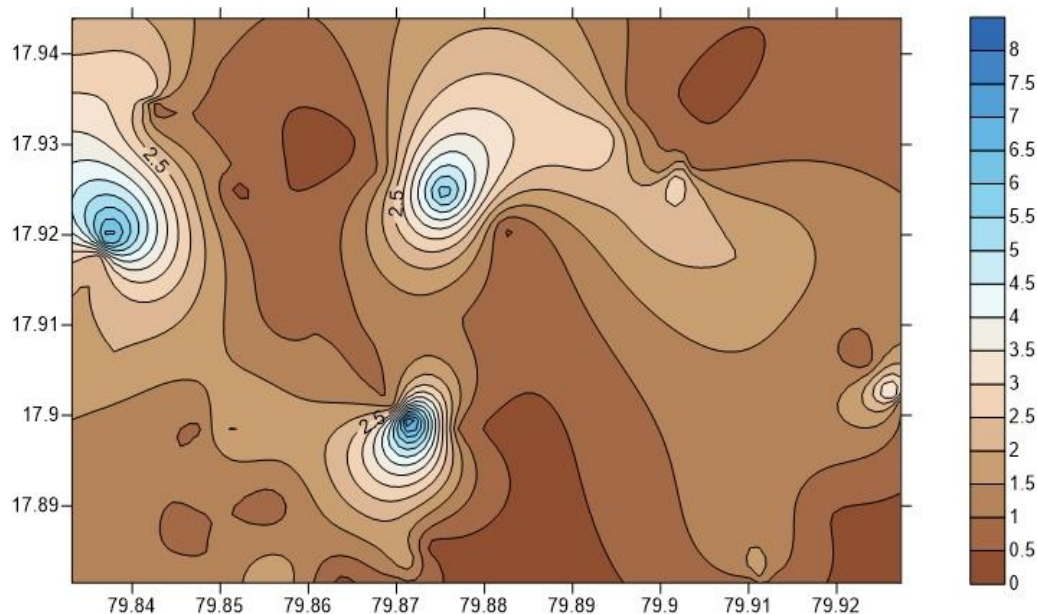


Fig. 3 Fluoride distribution map of Chennaraopet area, Warangal district, Telangana, India

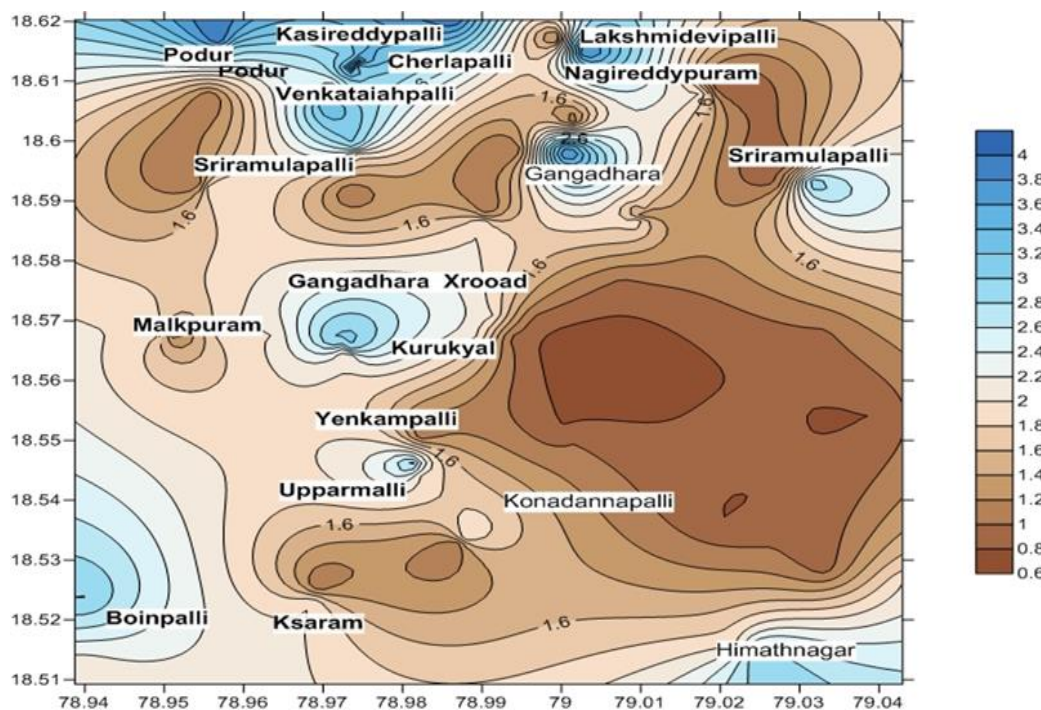


Fig. 4 Fluoride distribution map of Gangadhara area, Karimnagar district, Telangana, India

The formation of high-fluoride groundwater in India is principally governed by climate, composition of parent rock, hydrogeology and hydrogeochemistry of a basin. Endemic areas in general are hot and dry (climatically) which demands for high amount of daily water intake. Concentration of fluoride in water and amount of water intake are two important factors that resulted in high prevalence of fluorosis in the country. It is observed that area similar to India with semi-arid climate, crystalline igneous country rock and alkaline soils are most suitable for ingestion of excessive fluoride in natural waters [25]. Geology, climate, choice of water source makes Indian people vulnerable to high incidence of fluorosis. Moreover, in areas such as Telangana and Andhra Pradesh where the country rock is fluoride-rich Pink Granite (covers most part of Peninsular India) the problem is more serious in terms of endemicity. The worst hit states within India are Telangana and Andhra Pradesh where problem not only has wide distribution but highest level of fluoride in groundwater from a single area.

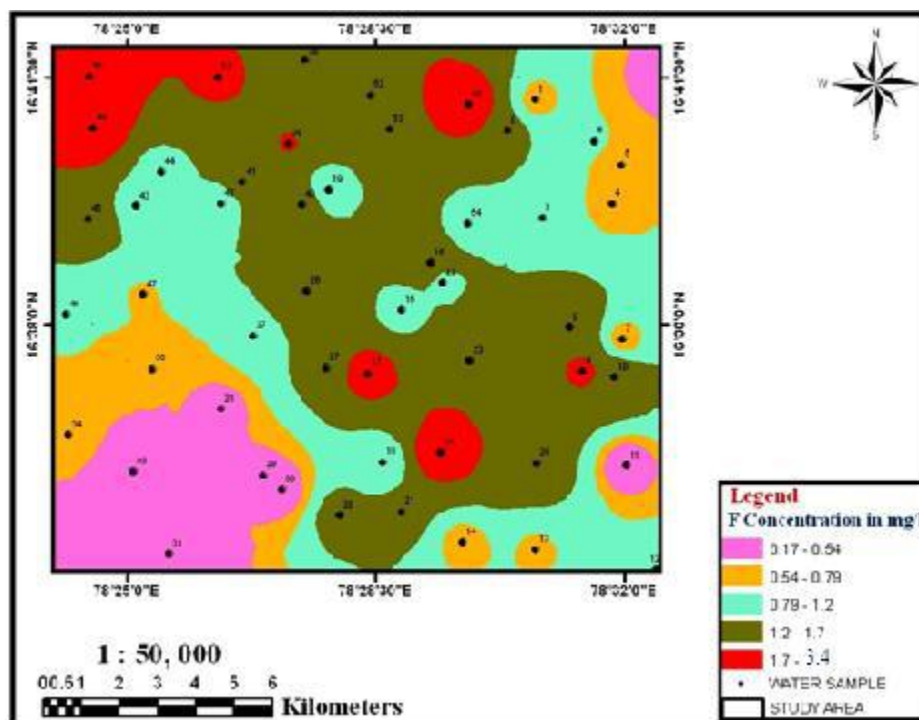


Fig. 5 Fluoride distribution map of Kalwakurthy area, Mahabubnagar district, Telangana, India

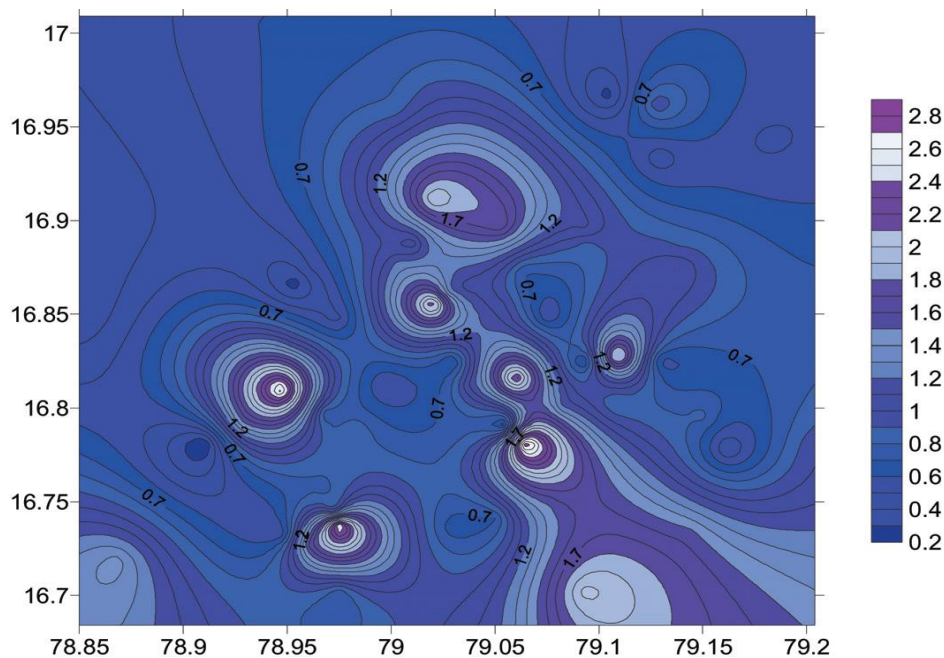


Fig. 6 Fluoride distribution map of Devarakonda area, Nalgonda district, Telangana, India

Fluoride: it's health implications: Fluoride ingested with drinking water is almost completely absorbed by the human body. It is rapidly distributed and mainly retained in skeleton. A small portion may be retained in teeth. It can interfere with carbohydrate, lipid, protein, vitamin, enzyme and mineral metabolism when the dosage is high. Due to the binding effect between fluoride and calcium present in the skeleton, acute fluoride intoxication symptoms are manifested in the form of bends in vertebral column and bone joints. Fluoride is primarily excreted in urine. However, skeletal deformation and weakening of joints is typical form of fluoride excretion at high levels of fluoride intake [26]. Dental fluorosis is a defect in the formation of tooth enamel in children, which results in mottled teeth. This is not reversible and remains for life. Dental fluorosis can occur in communities consuming water with fluoride content less than 0.4 mg/L. Early symptoms of skeletal fluorosis are back stiffness, pains in the bones and joints, sensations of burning, pricking, and tingling in the limbs, muscle weakness, chronic fatigue, gastrointestinal disorders, and reduced appetite. Osteoporosis develops in long bones and bony outgrowths may occur. Eventually the victim may be crippled and the vertebrae fuse together. Other ailments due to consumption of excess fluoride include destruction of about 60 enzymes including cytochrome C cholinesterase that handle oxygen, genetic change in sperm

and other cells, 250% increase in Downs Syndrome of with 70% developing cataracts, increase in infant mortality, spontaneous abortions and miscarriages, infant birth defects and Goiter [8].

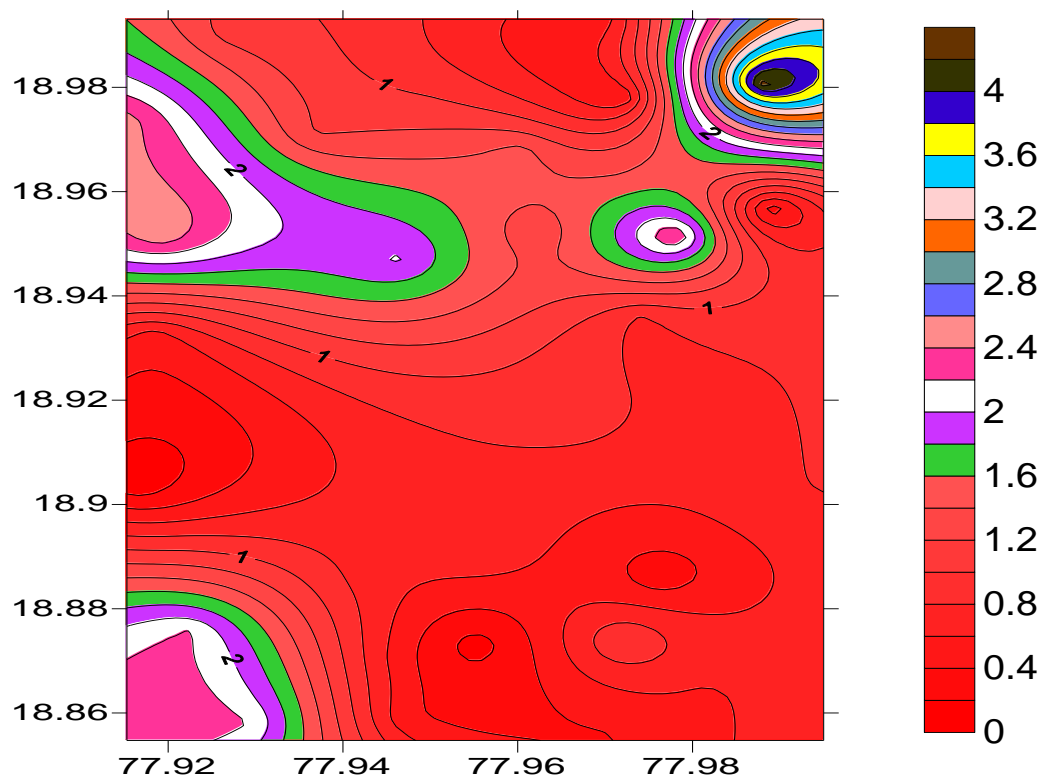


Fig. 7 Fluoride distribution map of Basara area, Adilabad district, Telangana, India

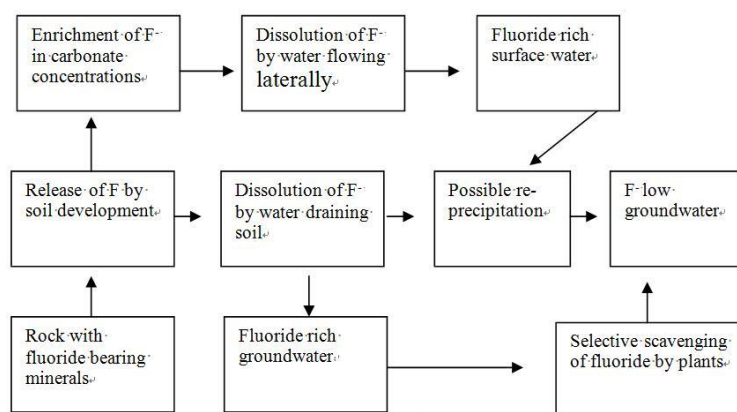


Fig. 8 Mechanism of fluoride ingestion into natural waters [34]

Fluoride was found to be an equivocal carcinogen [27]. Further studies have confirmed a 6.9 fold increase in bone cancer in young males [28]. Earlier studies had found a 5% increase in all types of cancers in fluoridated communities [29]. Cancer researchers have found a 17% rise in 16 years of cancers in towns in the USA that is fluoridated in comparison with those that are not. A rise in a rare bone cancer, osteosarcoma, was recorded in men under 20 in fluoridated areas. The US National Toxicology Program confirmed this in studies on rats in 1989 [30]. Studies carried out at National Institute of Nutrition, Hyderabad, India brought to light that in several villages of Telangana, Andhra Pradesh and Tamil Nadu, crippling bone deformity in the form of Genu-Valgum can occur in adults and even children under 10 years of age among communities exposed to high (4-13 mg/L) levels of fluoride [31]. In addition to drinking water source, fluoride also enters into the human body through other routes (food and air) and climatic conditions, daily water intake, diet and malnutrition also play vital role on the health. Similar research in Sri Lanka revealed that in its dry zone parts children were affected by fluorosis at fluoride concentration levels of 0.4 – 1.0 mg/L [22].

Mechanism of fluoride ingestion: Solubility studies have indicated that transport of fluorine in aqueous solution is dependent on the solubility of fluorite (CaF_2). Quantity of fluoride is dependent on the presence of other electrolytes in aqueous solutions that are partially ionized [23]. The ratio of the concentration of calcium to that of fluoride ions in solution

appears to be important. This is supported by the presence of low fluorine content in high calcium rocks. The concentration of fluoride depends on degree of weathering and presence of leachable fluoride in a particular geological terrain. The leachability is controlled by pH of the draining solutions, alkalinity, CO_2 and PCO_2 of the soil/rock it is draining through [22, 32]. Weathering of fluoride minerals in recharge area of the watershed/basin creates a situation wherein there can be initiation of ingestion into waters of the basin. It is observed that fluoride concentrations could be higher in discharge areas or valley portions of a basin than in the recharge portions [32]. Wells located in valley portions of one of the basins of Nalgonda district of Telangana showed very high concentrations of fluoride whereas those located in recharge area (where the village is located) showed allowable limits of fluoride [33]. The mechanism of fluoride ingestion into waters of hard rock terrain in South India has been studied and the presented in Fig. 8 and suggested a mechanism, as shown in Fig. 8 [34]. The relationship between fluoride concentration and depth of the groundwater source has been studied in Punjab and reported that shallow groundwater contained permissible concentrations of fluoride while deeper ones had objectionable levels of fluoride. Presence of clay layers above the shallow water aquifer was found responsible for prevention of fluoride leaching while the deeper layers probably got its fluoride enrichment from the source, some distance away in the recharge area [35].

IV. CONCLUSIONS

Hydrogeochemical investigations carried out in parts of granitic tracts of Telangana regions revealed that the fluoride concentrations in the groundwater are much higher than the prescribed limit. It is observed that fluorine bearing minerals like fluorite and apatite present in granitic rocks are the major source for the excessive fluoride in groundwater. It is observed that geology of the area and climatic conditions play vital role for the high concentrations of fluoride in groundwater. In addition, depletion of groundwater levels due to climatic changes is another factor for high incidence of fluorosis. It is recommended to avoid high fluoride bearing groundwater zones in the endemic areas and tap alternate source of water for drinking and domestic purposes.

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Prof. Sudarshan is associated with various scientific bodies in India. He is the Editor of the Journal “Environmental Geochemistry” brought out from the Association of Environmental Geochemists of India since 1998. He also served as Editor of the Journal of the Indian Academy of Geoscience for ten years. Prof. Sudarshan is awarded prestigious Basic Scientific Research Faculty Fellowship by the University Grants Commission, Government of India for a period of three years from 2015 for outstanding contribution in the field of Earth Sciences. He is also a member in the State Pollution Control Board, Telangan, India in the Consent for Establishment & Consent for Operations committees. He has been on Boards of selection committees in various Universities & research institutions.