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Research Article

EVALUATING FLUORIDE CONTAMINATION IN GROUND WATER SAMPLES OF DHARMAPURI DISTRICT, TAMIL NADU, INDIA

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ABSTRACT

Water is the one of the most precious resource in the earth. In that groundwater is the major resource of fresh water to all living organism. Most of the rural and urban population uses groundwater for domestic purposes. The people living in rural areas are more exposed since there is no centrally supplied treated water in these areas. Instead, groundwater accessed through dug wells, is their only water supply. Many people are not at all aware of the water borne diseases affecting their health due to high concentration of fluoride in drinking water which causes dental and skeletal fluorosis to humans, Based on (WHO, 2008) Standard Fluoride beyond desirable amounts (0.6 to 1.5 mg/l) in groundwater causes serious health hazards to humans and irreversible damage to plants. Ground water samples of open wells and bore wells collected from different locations in Dharmapuri Taluk in Dharmapuri District were analyzed for their physico-chemical characteristics. This study provide an overview of the fluoride content in drinking water and the extent of human exposure to different level of fluoride contamination in the blocks of Dharmapuri district, Tamilnadu. Most of the people in these area suffer from dental and skeletal flurosis such as mottling of teeth, osteosclerosis of pelvis and vertebral column, chronic joint pain. The ground water samples were studied during pre-monsoon and post-monsoon seasons from twelve different villages. The present study was undertaken to characterize the physico-chemical parameters such as temperature, pH, Total Alkalinity (TA), Electrical conductivity (EC), salinity, Calcium Hardness (CH), Magnesium Hardness (MH), Total Hardness (TH), Total Dissolved Solids (TDS), Total Suspended Solids (TSS), Total Solids (TS) and fluoride. Each parameter was compared with the standard permissible limit of the parameter as prescribed by World Health Organization (WHO). The Langelier Saturation Index (LSI) shows that majority of samples have the border line corrosive nature with negative LSI values. The study reveals that in few villages, water has high hardness and fluoride content. Hence, ground water must be used for drinking after proper treatments viz., softening and defluoridation.

INTRODUCTION

Water is an essential natural resource for sustaining life and environment but over the last few decades the water quality is deteriorating due to it's over exploitation. Water quality is essential parameter to be studied when the overall focus is sustainable development keeping mankind at focal point. Water is the precious gift of nature to all the living beings for sustenance. The suitability of water for domestic, agricultural and industrial purposes mainly depends on the chemical composition of surface and subsurface. Ground water is the major and preferred source for drinking all over the world even though its contribution is very less (only 0.6%) to the total water resources on earth. Water quality is essential parameter to be studied when the overall focus is sustainable development keeping mankind at focal point, since it is directly linked with human welfare. (Saxena and Saxena, 2013)

The degradation of water quality is mainly due to the increasing population, urbanization, industrialization and over-utilization of water resources. The ground water is getting polluted because of disposal of industrial effluents, hazardous wastes, sewage disposal and deep percolation of pesticides and fertilizers from activated fields. (Meenakshi *et al.,* 2006). Fluoride is an essential trace element for human metabolism. Its concentration in drinking water is the prime factor to decide whether fluoride is beneficial or harmful. In India, research on the assessment of water quality especially with reference to fluoride has been carried out by various workers (Suthar *et*

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*al.,*2008; Yadav *et al.,* 2009; Jha *et al.,* 2010; Patil *et al.,* 2010; Meta, 2010). Ingestion of excess of fluoride, most commonly in drinking water, can cause fluorosis which affects the teeth and bone.

Among the water quality parameters, fluoride ion exhibits unique properties in drinking water that its concentration in optimum dose is advantageous to health whereas its overdose affects the health (Saravanan *et al.,* 2008). High fluoride concentration in the water sources is a major concern for the mankind. The properties of Calcium, Sodium and Fluoride are interrelated that the higher the fluoride level, cause the lower is that of Calcium. This may be as a result of the substitution of Na by Ca as well as due to high affinity of Fluoride ion towards Calcium (Diabl *et al.,* 2005).

Presence of various hazardous elements like arsenic, nitrate, sulphate, fluoride, other heavy metals etc., in underground water have been reported from different parts of India and world.(Deepti Mishra *et al.,*2009; Khaiwal Ravindra *et al.,* 2007; Mufeed and Batarseh, 2006). A systematic study of correlation of the water quality parameters not only helps to assess the overall water quality but also to quantify relative concentration of various pollutants in water and provide necessary cue for implementation of rapid water quality management programmes (Dash *et al,* 2006).

WHO (2008) and BIS (1991) has set a allowable concentration for fluoride in drinking water. The permissible limits of fluoride in drinking water by various organizations are given in table 1. When administered in more than 1 ppm level of concentration, fluoride has been reported to cause depressions in DNA and RNA synthesis in cultured cells (Shivasankara, 2009).

Table 1 Range of allowable Fluoride concentration in drinking water

S. No	Name of the Organizations	Permissible limit of Fluoride (mg/l)
1	World Health Organization (WHO)	1.5
2	Bureau of Indian Standards (BIS)	1.0
3	The committee on Public Health Engineering (PHE), Govt. of India	1.0
4	Indian Council of Medical Research (ICMR), Govt. of India	1.0

Fluoride content some of the District in Tamil Nadu

In Tamil Nadu, the high concentration of fluoride in groundwater is found to be in Dharmapuri and Salem district closely followed by Coimbatore, Madurai, Trichy, Dindigul and thuthugudi district (Lilly Florence, 2012). The districts having low fluoride are Tirunelveli, Pudukottai, Vellore, and Ramnad. The district wise fluoride status in Tamil Nadu is presented in (**Table 2).**

Table 2 Distribution of Fluoride content some of the district in Tamilnadu state

S. No	Status	Districts
1	Severe	Dharmapuri, Salem
2	Moderate	Coimbatore, Madurai, Trichy, Dindigul, Thuthugudi
3	Less	Pudukottai, Tirunelveli, Vellore , Ramnad

Since there were no major studies in the recent past, the present study was carried out to understand the present status of the qualitative analysis of some physicochemical parameters of ground water quality in Dharmapuri district in Tamil Nadu.

MATERIALS AND METHODS

Study Area

The study area Dharmapuri District, Tamilnadu is situated 12.12° N 78.26° E, which is bounded by Tiruvannamalai and Villupuram Districts on the east, Salem District on the South, Krishnagiri District on the north and Cauvery River on the west. The famous Hogenakkal waterfalls is in Dharmapuri district. The sub basin area is bounded by Harur, Palakodu, Pennagaram, Dharmapuri, Pappireddipatti Taluks. Dharmapuri district goes through a hot climate during the summer (March-May) with a temperature upto 36°C and in winter it varies between 16°C - 12°C. The district has an average annual rainfall of about 895.56 mm.

Water samples collection

Ground water samples were collected from 24 different villages of Dharmapuri district as given in the Table 4. Ground water samples were collected in polyethylene bottles of two liter capacity with necessary precautions. (Brown *et al.,* 1974). The samples were collected, during July 2014 to June 2015 from manually operated hand pumps, open wells and bore wells. Standard procedures of APHA to avoid unpredictable changes in their characteristics (Sudhakar 2013, APHA, 1998).

Ground water samples of a total of 24 villages in dharmapuri district were collected in pre-cleaned and rinsed polythene bottles of two litre capacity with necessary precautions.

Sample preservation and handling

The collected samples were preserved in an icebox to avoid retarding biological action, deterioration, hydrolysis of chemical compounds and complexes, volatility of constituents and then brought to the laboratory for the determination of both physico-chemical parameters. The preservation techniques of various parameters of water were given by U.S. Environmental Protection Agency (USEPA 1983).

Physico-chemical parameters analysis

All the samples were analyzed for the following Physico-chemical parameters; pH, Electrical Conductivity (EC),

Table 3 Parameters and methods employed in the physicochemical examination of water samples

S. No	Parameters	Unit	Method Employed
1	pH	-	Digital pH-meter
2	Electrical Conductivity	µmhos/cm	Digital Conductivity-meter
3	Total Alkalinity	Mg/L	Titrimetric method (With HCl)
4	Total Hardness (as CaCO3	Mg/L	Titrimetric method (with EDTA)
5	Calcium Hardness (as CaCO3)	Mg/L	Titrimetric method
6	Magnesium Hardness (as CaCO3)	Mg/L	Titrimetric method
7	Chloride (as Cl-)	Mg/L	Titrimetric method (With AgNO3)
8	Nitrate (as NO3-)	Mg/L	Spectrophotometric method
9	Fluoride (as F-)	Mg/L	Ion Selective Electrode
10	Total Dissolved Solids	Mg/L	Digital Conductivity-meter

Total Alkalinity (TA), Total Hardness (TH), Calcium hardness (Ca H), Magnesium hardness (Mg H), Chloride, Nitrate, Fluoride and Total Dissolved Solid (TDS). The analysis of water samples were carried out in accordance to standard analytical methods (APHA, 2005). All the chemicals were used AR grade and double distilled water for preparation of solutions. Details of the analysis methods are summarized in Table-3.

Table 4 Standards for drinking water quality

S. No	Parameters	BIS:1999	ICMR:1975	WHO:2006
1	pH	6.5-8.5	7.0-8.5	6.5-8.5
2	EC (µmhos/cm)	-	-	1400
3	Total Alkalinity	600	600	120
4	Total Hardness	600	600	500
5	CL-	1000	200	200
6	NO_3-	10	50	45
7	F	1.54	1.5	1.5
8	TDS	2000	1500	500

Statistical Analysis

In the present study Minimum, Maximum, Average, Standard Deviation and Correlation coefficient (r) has been calculated for each pair of water quality parameters by using Excel spreadsheet for the experimental data. The standard formulae were used in the calculation for statistical parameters are as follows (Gupta, 1999).

$$Mean\ (\mu) = \frac{\sum x}{N}$$

x = Value of Observation

N = Number of Observation

$$\text{Standard Deviation } (\sigma) = \sqrt{\frac{n\sum x^2 - \left(\sum x\right)^2}{n(n-1)}}$$

x = Values of Parameter

n = Number of Observations

$$\text{Karl Pearson's Coefficient of Correlation } r = \frac{n\sum xy - \sum x \sum y}{\sqrt{n\sum x^2 - \left(\sum x\right)^2}\sqrt{n\sum y^2 - \left(\sum y\right)^2}}$$

x,y = Values of array 1 and array 2 respectively.
n = Number of Observations

RESULT AND DISCUSSION

The values of all water quality parameters are illustrated as mentioned in Table-2. All the results are compared with standard permissible limit recommended by the Bureau of Indian Standards (BIS), Indian Council of Medical Research (ICMR) and World Health Organization (WHO), depicted in Table-3. The ground water samples of study area were estimated at every month for the period of one year at twenty four different stations of groundwater in and around dharmapuri district. The period of study is from July 2014 to June 2015.the Physico - chemical parameters of the selected stations are given in the Table.4

pH

The pH is measure of intensity of acidity or alkalinity of water. All chemical and biological reactions are directly dependent upon the pH of water system (Rao, 2006). In our findings pH varied between 6.5-7.8. Maximum pH was recorded at station 15 in village Balajangamanahalli and minimum pH was recorded at station 7 in village Adilam , which are not within the permissible limit prescribed by BIS, ICMR and WHO. The variation of pH in ground water samples of study area is depicted in Figure-1, which shows that most of the samples are alkaline in nature. The hydrogen ion concentration affects the taste of the water (Garg *et al.,* 2007). The pH of water is very important indication of its quality and provides information in many types of geochemical equilibrium or solubility calculations (Mitharwal *et al.,* 2009).

Electrical Conductivity

The electrical conductivity of water depends upon the concentration of ions and its nutrient status. Based on electrical conductivity values the water quality can be classified as poor, medium or good (Gulta, Sunita, & Saharan, 2009). Generally, Groundwater tends to have high electrical conductivity due to the presence of high amount of dissolved salts (Prakash and Somashekar, 2006). In the present investigation maximum conductivity 6278 µmhos/cm was observed at S12 in village Chinthalpadi and minimum 605 µmhos/cm at S21 in village Adilam and Athimuthu. The permissible limit exceeds at chithalpatti, Belamaranahalli, A.pallipatti and Manchavadi.. The maximum limit of EC in drinking water is prescribed as 1400 µmhos/cm (WHO: 2006), Sathish Kumar (2007) has also revealed 75% of groundwater of the area exceeds the permissible limits. The variability of electrical conductivity may be due to the natural concentration of the ionized substances present in the water samples are exceeding the permissible limit are shown in Figure- 2.

Total Alkalinity

In the present study total alkalinity ranges from 385 mg/L to 419 mg/L was recorded the maximum value was recorded in Chikkamarandahalli (S19) and minimum in village Akkamanahalli (S3). Variation in total alkalinity of ground water samples were represented in Figure- 3 which clearly depicts that these values are more than the permissible limits of BIS, ICMR and WHO. The alkalinity was recorded in low amounts at non-industrial area and recorded marginally high at industrial area. Similar observations were observed by Deka *et al.* (2007) in Barpeta District, Assam and Swarnalatha *et al.,* (2007) in AP. In ground water, most of the alkalinity is caused due to carbonates and bicarbonates. The high alkalinity of the water may not only lead to caustic embattlement but also to the precipitation of sludge and deposition of scales (Khan *et al.,* 2005). The alkalinity ranged between 20-710 mg/L as CaCO3 indicated high alkaline nature of water. The excess of alkalinity could be due to the minerals, which dissolved in water from mineral rich soil (Prakash and Somashekar, 2006).

Total Hardness

Hardness is the property of water which prevents lather formation with soap and increases the boiling point of water. Hardness of water mainly depends upon the amount of calcium or magnesium salt or both (Singh *et al.,* 2012).

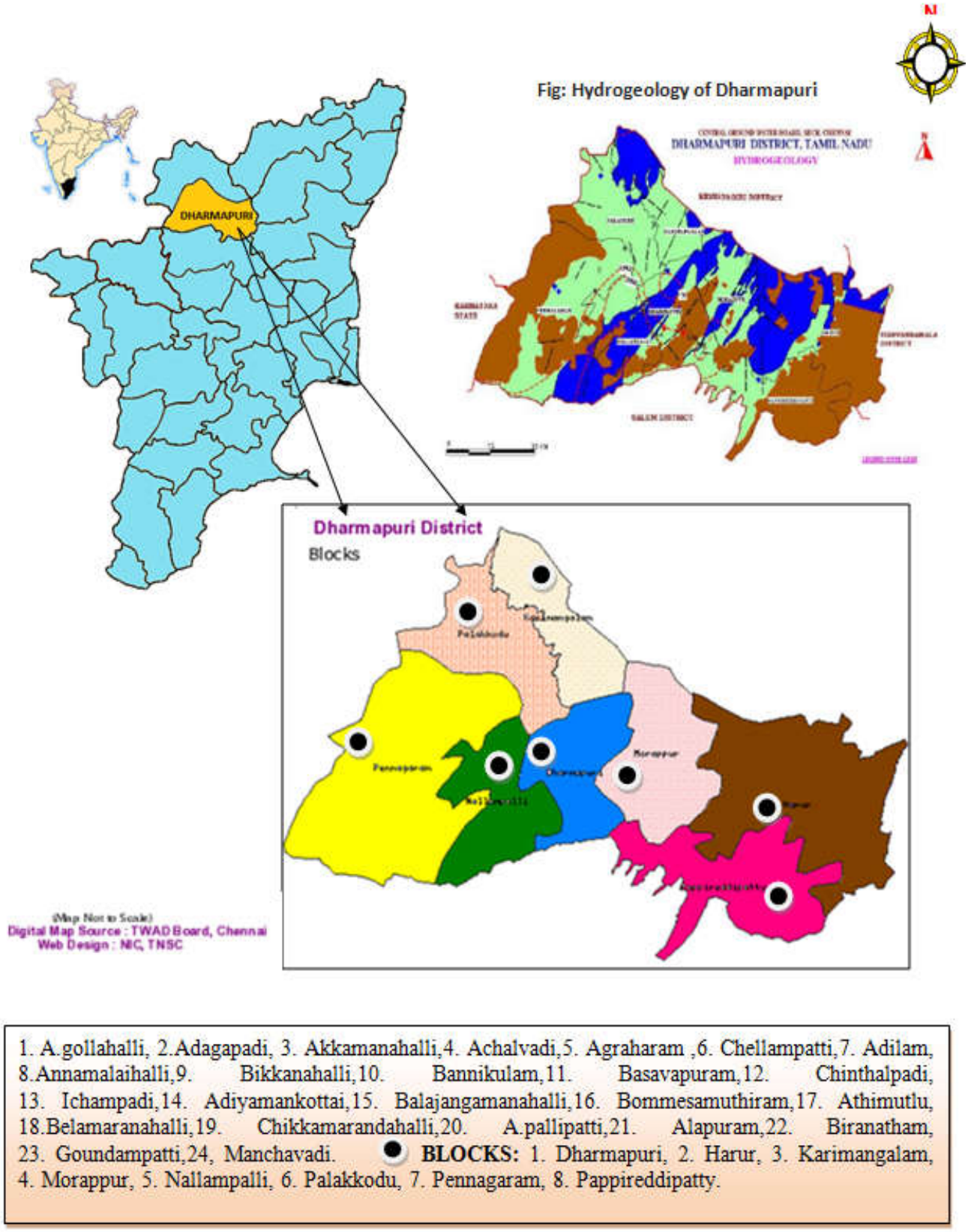

Fig 1 Map Showing Selected Sampling Stations In Dharamapuri District

fluctuates from 269 mg/L to 989 mg/L (Figure-4), which are beyond the permissible limit as prescribed by BIS, ICMR and

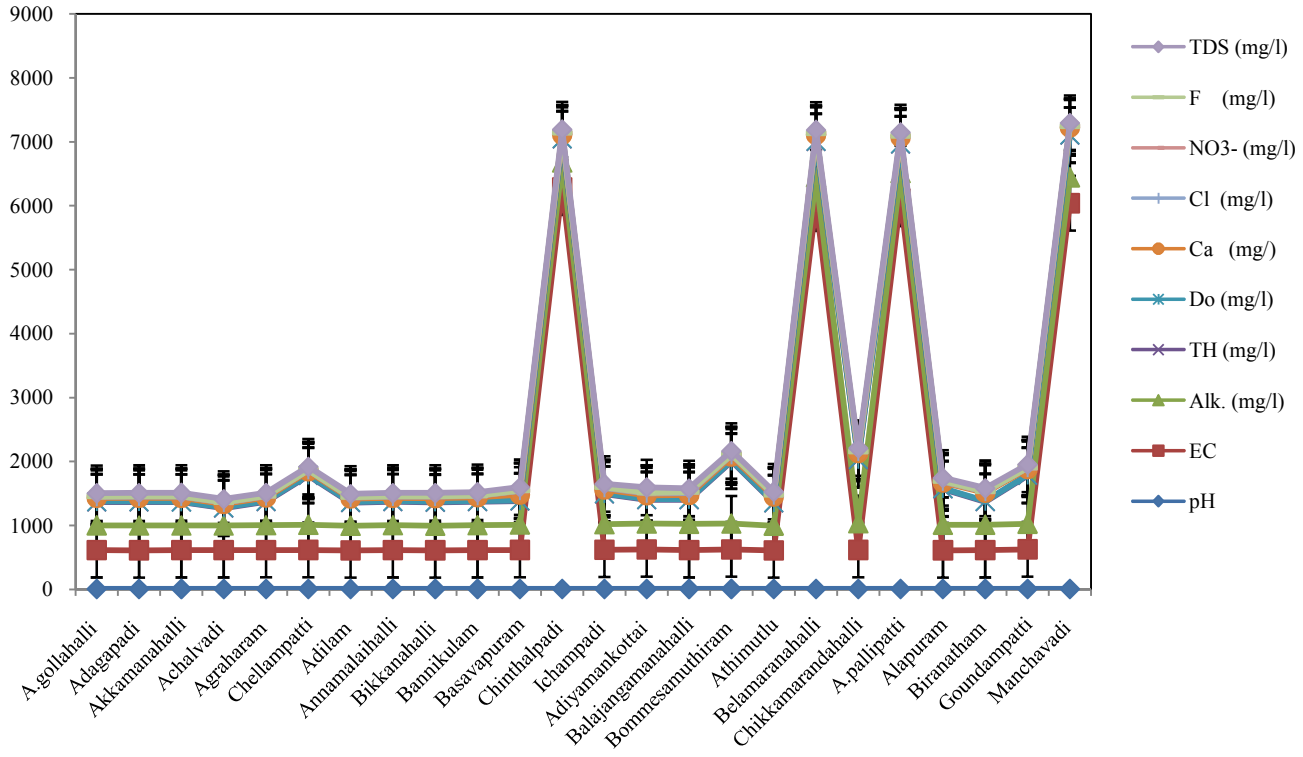

Figure 2 Variation in of physical parameters in and around villages of sampling sites in dharmapuri district from July 2014 to June 2015

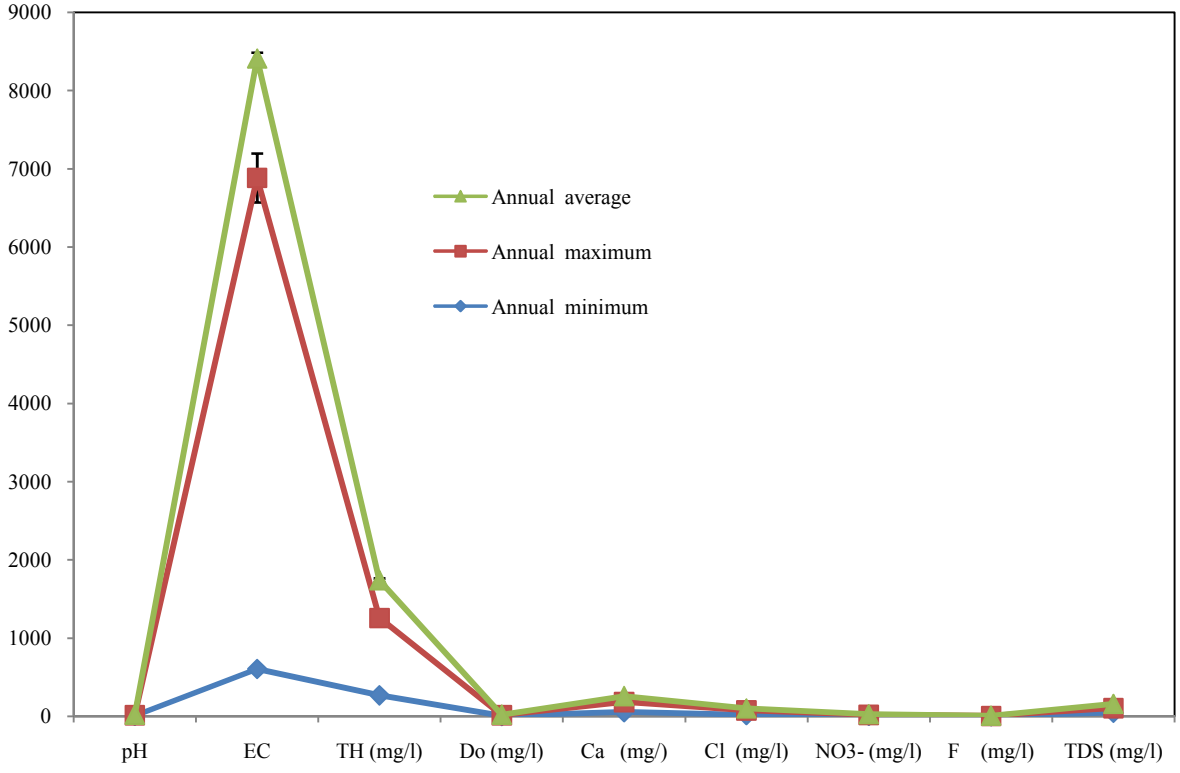

Figure 3 Variation in of physical parameters in and around villages sampling sites in dharmapuri district from July 2014 to June 2015

It is an important criterion for determining the usability of water for domestic, drinking and many industrial supplies (Mitharwal *et al.,* 2009). In our findings the value of hardness WHO. The minimum value was found in S4 (village - Achalvadi) and maximum value was found in samples S19

(village - Chikkamarandahalli).The variation between stations are shown in Fig.4.

Calcium Hardness

Hardness is caused by compounds of calcium and magnesium and by a variety of other minerals The high quantity of calcium may be due to mixing of sewage and industrial wastes.

Fig 4 Dental Fluorosis in Dharmapuri district

Dissolved oxygen

The amount of oxygen dissolved in water is referred as dissolved oxygen. It is one of the most important parameter in analyzing water quality and it is an index of physical and biological processes occurred in water. Total dissolved oxygen at selected stations ranges from 4.6 mg/L to 9.7 mg/L the minimum value was recorded in Adiyamankottai (S14) and maximum in village Chellampatti (S6). The dissolved oxygen value varies depending upon water temperature and the partial pressure of oxygen in its gas phase. Dissolved oxygen value is reduced by higher temperature, biological impurities, ferrous iron, ammonia, nitrites and substances such as hydrogen sulfide. Oxygen saturated water has pleasant taste (Pandey and Nand, 2003).

Calcium concentration nearly 30% of groundwater exceeds the permissible limit. Calcium Hardness varies from 57 mg/L to 129.3 mg/L as illustrated in Figure-5. It may be due to the presence of high amounts of calcium salts in ground water samples. Water system using ground water as a source are consened with water. Harddness, since as water source through soil and rock it disso raise. Some agent as naturally occurring minerals and carries the in to ground water

Chloride

EPA secondary drinking water regulations recommend a maximum concentration of 250mg/L for chloride ions.it is an essential electrolyte, trafficking in and out of cells through chloride channels and playing a key role in maintaining cell homest as isan transmitting action potentials in seasonal. The

chloride is in the form of chloride ions, which is one of the major inorganic anion in water. Naturally chloride occurs in all types of waters. The contribution of chloride in the groundwater is due to minerals like apatite, mica and hornblende and also from the liquid inclusions of igneous rocks (Prakash and Somashekar, 2006). Chloride contents in fresh water are largely influenced by evaporation and precipitation. Chloride ions are generally more toxic than sulphate to most of the plants and are best indicator of pollution (Rao, 2006). Chloride found high during the study ranged from 17.0 mg/l to 59.7 mg/l (Figure -6). Minimum value was observed at samples S23 (Goundampatti) and maximum value was observed at S11 in village Basavapuram. These unusual concentrations may indicate pollution by organic waste. Chloride salts in excess of 100 mg/1 give salty taste to water and when combined with calcium and magnesium, may increase the corrosive activity of water (Tatawat and Singh Chandel, 2007).

Nitrate

During the study, Nitrate fluctuated between 4.3 to 15.1 mg/l (Figure -7). The minimum (4.3) was recorded at (station 22).Biranatham and maximum (15.1) was recorded at (station 15). Balagangamanahalli. Nitrate (No_3) are an essential source of nitrogen (N) for plants when nitrogen fertilizers are used to enrich soils.Nitrates may be carried by rain irrigation and other surface waters through the soil in to ground water. Human and animal water can also contribute to nitrate contamination of ground water. Potential serious health effects associated with consuming water containing nitrate above the permissible level. Which are beyond the permissible limit of BIS, ICMR and WHO. In presence of high concentration of nitrate in drinking water is toxic. The long-term consumption of drinking water with nitrate concentrations even below the maximum contaminant level may stimulate the endogenous formation of nitrosamines (Townsend *et al.,* 2003) and it has been linked to higher risks for non-Hodgkin's lymphoma in bladder and ovarian cancer (Weyer *et al.,* 2001). Due to higher concentration (over 100 mg/L) of nitrate in water, infants, less than six month old, are suffering from methamoglobinemia or blue baby disease. Due to higher concentration (Greer and Shanon (2005) have also been reported that breastfeeding infants are the at risk of methemoglobinemia when mothers ingest water with very high nitrate concentrations (100 mg NO3-N/L).

Fluoride

Fluorides may be properly defined as binary compounds or salts of fluorine with other elements (like Na, K, etc). Fluorine is the lightest member of the halogen family which is the most electronegative element. Fluoride is 13th in the order of abundance of elements in the earths crust.Fluoride is important in human nutrition for the normal development of bones. The required level of fluoride is 1.0 to 1.5 mg/L. Higher concentration of fluoride in ground water appears to create dental, skeletal and non-skeletal fluorosis. Fluoride concentration in sampling sites ranges from 1.0 to 4.3 mg/L in ground water samples, with lowest value1.0 mg/L (, S6, S10, S11, S16, and S19) in village banikulam , Chellampatti, Basavapuram, Bommesamuthiram, and Chikkamarandahalli and the highest value 4.3 mg/L (S20) in village A.pallipatti. As shown in Figure-8 most of the samples are having fluoride concentration more than the permissible limit and suffering from the acute fluoride problems. The occurrence of fluoride in the drinking water in different concentration has been reported from almost all parts of the world (Jain *et al.,* 2005). The level of fluoride in water exceeds 1.5 mg/L, yellowing of teeth followed by pitting and chipping of tooth enamel are common symptoms of dental fluorosis. Fluoride, in excess is the most exclusive bone-seeking element existing, owing to its great affinity for calcium phosphate leading to deformation of bones termed as skeletal fluorosis (Srivastava *et al.,* 2007).the present study also revealed that the elevated level of fluoride also evidenced by the dental fluorosis

Total Dissolved Solids

Total dissolved solids are composed mainly of carbonates, bicarbonates, chlorides, phosphates and nitrates of Calcium, Magnesium, Sodium, Potassium, Manganese, organic matter salt and other particles (Siebert *et al.,* 2010). Beyond certain limit it imparts a peculiar taste to water and reduce its portability. TDS in water less than 1000 mg/L has been classified as non-saline. In the present finding TDS value varied from 47.6 to 85.5 mg/L (Figure-9), which is also not within the prescribed permissible limits. TDS minimum recorded at S6 in village Chellampatti and Maximum at S14 in village Adiyamankottai. TDS concentration in groundwater, except at few stations, were showing below the permissible limits as prescribed by WHO (2003).Total dissolved solid is an important parameter for drinking water and water to be used for other purposes beyond the prescribed limit, it imparts a peculiar taste to water and reduce its potability (Sandeep Mitharwal *et al.,* 2009).

CONCLUSION

The analysis of ground water samples collected from different villages of dharmapuri District revealed that, the water quality parameters like (pH, electrical conductivity, total alkalinity, total hardness, calcium hardness, magnesium hardness, chloride, nitrate, fluoride and TDS are beyond the permissible limit as per BIS, ICMR and WHO standards. In comparison to all other parameters there is an acute problem of extremely high levels of Fluoride, Nitrate, Total Dissolved Solids and Chloride. As only 55% of ground water samples have fluoride content within the permissible limit (> 1.5 mg/L, WHO) and remaining 46% of villages are having very high fluoride concentrations. The favourable factor which contributes to rise of fluoride in ground water is presence of fluoride rich rock salt system. Excess fluoride may lead to tooth decay and kidney disease. The need for new institutional economics approach to deal with current and emerging problems has become very crucial. The results of current study indicate that the drinking water, used by the people residing in villages of dharmapuri district, is not potable. So, the proper environment management plan must be adopted to control drinking water pollution immediately. Based on these results and analysis of water samples, it is also recommended to use water only after boiling and filtering or by Reverse Osmosis treatment for drinking purpose by the individuals to prevent adverse health effects.

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