

Fluoride Contamination in Groundwater: A Pilot Study on Dug Well Recharge System for In situ Mitigation

L. Elango and G. Jagadeshan

Abstract Dissolved ions concentration in groundwater beyond the recommended limits is a major problem as they make the water unsuitable for drinking purpose. Fluorine commonly found in certain rocks is released into groundwater due to the processes of rock–water interaction. This leads to increase in the concentration of fluoride in groundwater which is a major problem in several parts of the world including India. Presence of fluoride beyond the prescribed limits causes health problems to humans due to prolonged consumption of water, which is common in many parts of India. Dental and skeletal fluorosis is observed due to prolonged drinking of water with fluoride concentration above 1.5 mg/l. The objective of the study is to know how fluoride get released from the host rock and spot out suitable location for installing a dug well recharge system to decrease the fluoride concentration in groundwater. Several methodologies exist for in situ or exsitu removal of fluoride from groundwater. Exsitu methods can be enforced at community level or even at household level for the reduction of fluoride before its consumption, through ion exchange, reverse osmosis, adsorption, electrodialysis, coagulation, Nalgonda technique, electrodialysis, coagulation, precipitation, etc. Even artificial recharging structures can also be built in suitable location for diluting fluorite concentration in groundwater. Rainwater harvesting is also found effective to reduce the fluoride concentration of groundwater in existing wells. A pilot study was carried out by construction of a dug well recharge system in Dharmapuri district, Tamil Nadu, India. The study successfully demonstrated the applicability of dug well recharge system at a carefully selected site based on the systematic long-term hydrogeochemical studies to solve the problem of fluoride contamination affecting millions of rural people.

Keywords Groundwater • Fluoride contamination • Rainwater harvesting
Hard rock aquifers • Geogenic contamination

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1 Introduction

Geogenic and anthropogenic contamination may make groundwater unsuitable for various uses including its use as drinking water. Consumption of such water may cause many health problems. Presence of fluoride, arsenic, and iron above recommended limits is found in groundwater due to geogenic contamination in many parts of India and world (Mukherjee et al. 2015). Release of arsenic from arseno-pyrite minerals is responsible for higher concentration of arsenic in groundwater of Bengal basin (Farooq et al. 2011; Saha 2009). As about 80% of rural population of India depends on the use of groundwater for domestic use, the water needs to be treated before it is used for drinking purpose. The initial concentration of fluoride, occurrence and removal of co-contaminants, and disposal of sludge, etc., decide treatment methods of water to be treated (Brindha et al. 2016). Bhagavan and Raghu (2005) studied the effect of increase in recharge by check dams, and Pettanati et al. (2014) highlighted the use of percolation ponds in regions with fluoride problem in groundwater. Some studies have also indicated an increase in fluoride concentration in groundwater during the process of recharge when check dams are constructed (Bhagavan and Raghu 2005). Thus, contradicting findings are also observed from recharge for in situ mitigation of fluoride concentration (Brindha et al. 2016). Hence, it is essential to reveal the release mechanism of fluoride during the process of recharge based on a long-term temporal variation in fluoride concentration and in the level of groundwater in the area. The objective of this study is to understand the release processes of fluoride and identify a suitable location for construction of dug well recharge system in the area for reducing the concentration of fluoride in groundwater to an acceptable level. This pilot study was carried out in Dharmapuri district, Tamil Nadu (Fig. 1), where about 72,000 out of about 190,000 school students were found having dental fluorosis (UNICEF 2009).

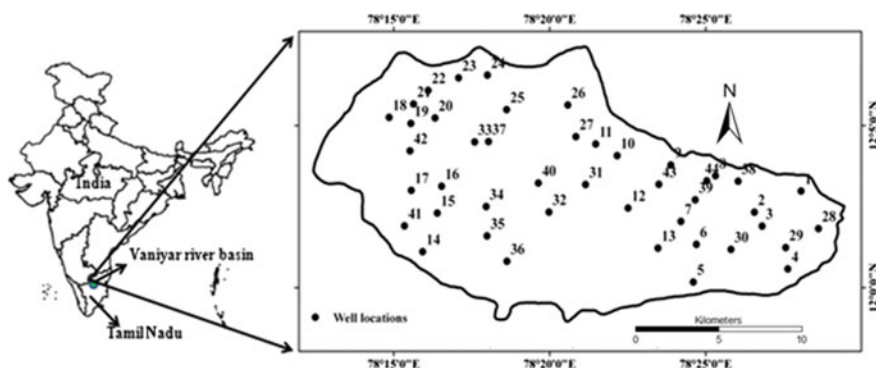


Fig. 1 Location map of study area and sampling wells

2 Methodology

Systematic investigation during June 2011 helped in identifying 44 key monitoring well to represent the area (Fig. 1). These key wells were used to collect bimonthly groundwater level and water sample between June 2011 and August 2014. Largely, the key wells are >30 m in depth and are utilized for obtaining drinking and agricultural water. Water level indicators (Solinist) were used to collect water level data. Pre-washed and treated 1000-ml capacity clean polyethylene bottles were used to collect groundwater samples after rinsed with sample. Merck 0.22- μm filter paper (Merck 755004722) were used to filter the samples before collection. To know the in situ temperature, EC and pH of the groundwater in field portable digital meter (YSI 556MPS) were used. Total dissolved solids (TDS) were obtained using $\text{EC} = \text{TDS}/0.64$ (Carroll 1962). Aquamerck (1.11109.0001) test kit was used to measure CO_3^{2-} and HCO_3^{-} in the field itself. Ion chromatograph (Metrohm 861) was used for measuring the concentrations of Na^+ , K^+ , Ca^{2+} , Mg^{2+} , Cl^- , F^- , NO_3^- , and SO_4^{2-} of samples in the laboratory. The concentration of silica was determined by using spectrophotometer (Systronics 201) for the groundwater samples collects in August 2014. Batch test experiment's results were obtained to know the fluoride release processes from the rocks/soil, at laboratory.

3 Sources of Fluoride in Groundwater

Fluoride-bearing minerals are common in nature; therefore, contamination of fluorite is vast. Sometime it is intensive and became alarming in many places. The permissible range for fluoride in drinking water as per the Bureau of Indian Standards (BIS 2012) is from 0.6 to 1.5 mg/l. A desired concentration of fluoride in drinking water is favorable to determine the physiological activities in human bodies, whereas the consumption of fluoride above or below the permissible limits may cause health problems such as fluorosis of dental and skeletal structure. In India, northwestern and southern states are severely affected with fluorosis (Agarwal et al. 1997; Ali et al. 2016). Weathering of rocks rich in fluoride is the common natural cause for the fluoride in groundwater. The marine sediments and sediments of foothill areas (WHO 2002; Fawell et al. 2006), as well as marginal alluvial terrain of the Gangetic Plains (Saha et al. 2008; Saha and Alam 2014), are more vulnerable for water with high concentration of fluoride. Fluorine occurs naturally in igneous and sedimentary rocks. Common fluoride minerals are sellaite, fluorite or fluorspar, cryolite, fluorapatite, apatite, topaz, phlogopite, biotite, epidote, tremolite and hornblende, mica, clays, villuanite, and phosphorite (Brindha and Elango 2011; Haidouti 1991; Gaumat et al. 1992; Gaciri and Davies 1993; Kundu et al. 2001). Nalgonda district, Andhra Pradesh, known for its fluorite

contamination is due to the inherent fluoride-rich granitic rocks. The fluoride content in granitic rocks in Nalgonda district varies from 325 to 3200 mg/kg with a mean of 1440 mg/kg. Thus, the Nalgonda granites having higher fluoride content than world granite, where it occurs average 810 mg/kg (Brindha and Elango 2011), whereas in Hyderabad granites, it is estimated 910 mg/kg (Brindha and Elango 2011; Ramamohana Rao et al. 1993). However, groundwater in northeastern districts of Tamil Nadu has high fluoride because of their fluoride-rich mineral such as epidote, hornblende, biotite, apatite, carbonatite, mica, and fluorapatite (Jagadeshan 2015; Jagadeshan et al. 2015a, b; Brindha et al. 2016). Jagadeshan et al. (2015b) carried out a study of rocks of Vaniyar river basin in Dharmapuri district, Tamil Nadu, to understand the mineralogical composition. He made thin sections of the charnockite and epidote hornblende biotite gneiss rocks (Fig. 2), where plagioclase, hypersthene, biotite, quartz, hornblende, orthoclase, and muscovite opaque (probably iron oxide) were the major minerals. Biotite which consists of fluorine was also identified in charnockite rock.

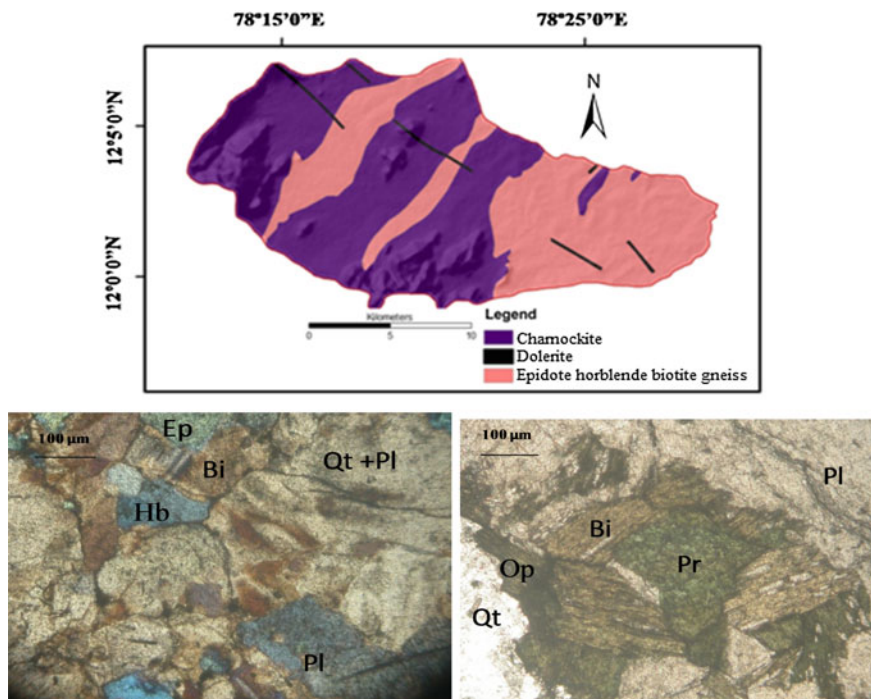


Fig. 2 Geological map of the study area and thin section photographs (X5 magnification, field of view 2500 μ m wide, plan polarized illumination) showing quartz (*Qt*), plagioclase (*Pl*), epidote (*Ep*), biotite (*Bi*), hypersthene (*Op*), hornblende (*Hb*), pyroxene, and other opaque minerals (*Op*) in epidote hornblende gneiss (*left*) and charnockite (*right*)

4 Geochemical Analysis of Groundwater

Largely, the elevated amount of fluorite in groundwater is due to its release from host rock. The climatic conditions, pH, host rock mineralogy, and hydrogeological condition of an area are generally governing the release of fluoride into groundwater (Shan et al. 2013; Raju et al. 2009). The phosphatic fertilizers use may also lead to rise in fluoride concentration in groundwater anthropogenically.

Utilization of fertilizers and industrial activities such as brick kilning are some of the other causes for elevated fluoride concentration in groundwater (Selvam 2015). The weathering of these rocks results in increased fluoride content in groundwater. Longer residence time in aquifers with fractured fluoride-rich rocks enhances fluoride levels in the groundwater. Granite and granitic gneisses in Nalgonda, India, contain fluoride-rich minerals such as fluorite (0–3.3%), biotite (0.1–1.7%), and

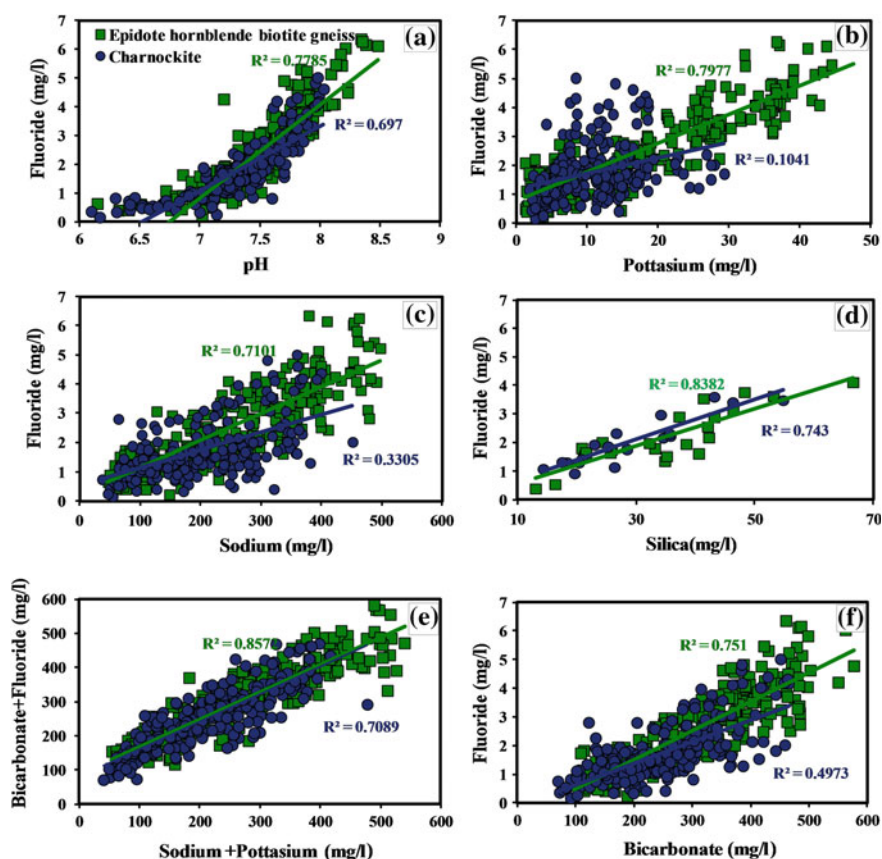
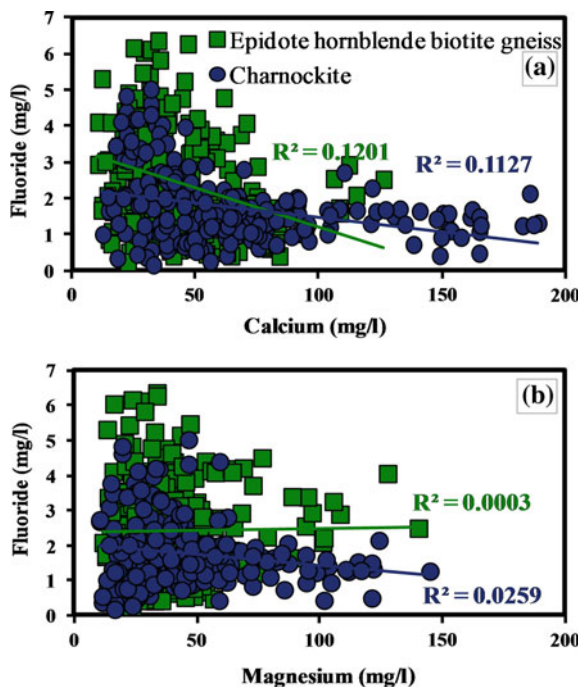


Fig. 3 Bivariate diagram of fluoride versus pH (a), potassium (b), sodium (c), silica (d), bicarbonate + fluoride versus sodium + potassium (e), and bicarbonate versus fluoride (f)

Fig. 4 Bivariate diagram of fluoride versus **a** calcium and **b** magnesium



hornblende (0.1–1.1%) (Brindha and Elango 2011; Brindha et al. 2016; Ramamohana Rao et al. 1993). A strong positive correlation is found among sodium, potassium, silica, bicarbonate + fluoride versus sodium + potassium and bicarbonate versus fluoride (Fig. 3).

However, negative correlation was obtained for fluoride versus calcium and magnesium (Fig. 4) (Jagadeshan et al. 2015b). These diagrams support the leaching of fluoride from minerals.

During laboratory experiments Jagadeshan et al. (2015b) found leaching of fluoride from charnockite and epidote hornblende biotite gneiss rocks resulted in enhanced fluoride in water from 1.41 to 3.54 mg/l, respectively, after 2000 hours (Fig. 5).

For reducing or removal of fluoride concentration from drinking water, there exist various methods at domestic or community level. The various techniques of fluoride removal are ion exchange method, precipitation method, adsorption method, Nalgonda method, and reverse osmosis. Thus, the existing treatment methods of water to remove fluoride for domestic use have several limitations. Further the people's mind-set is also not to use treatment methods as it is time consuming, expensive, and also requires trained personal. To overcome the problems and limitation of fluoride removal methods from the water, induced recharge may be adopted. David et al. (2014) reported in tropical countries such as India, rainwater harvesting could be useful for dilution of high-fluoride groundwater.

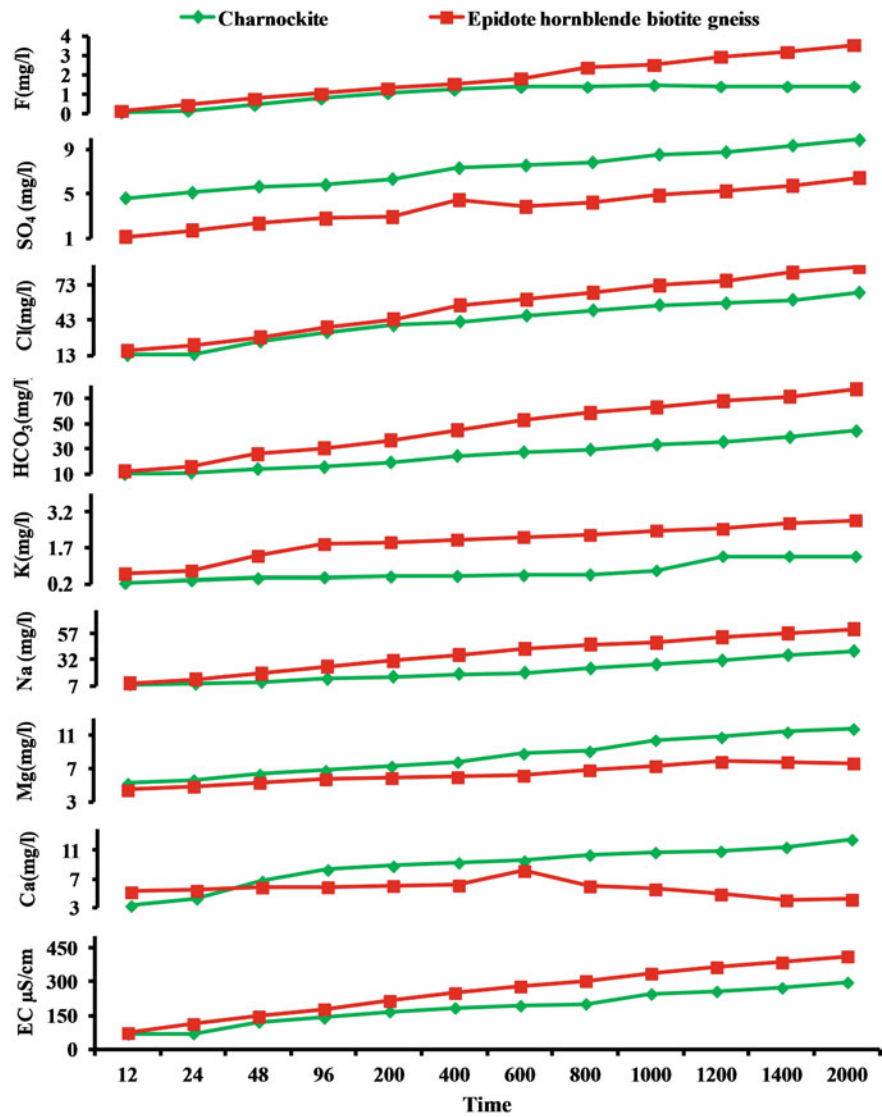


Fig. 5 Concentrations of ions released during laboratory experiment with respect to time from powder of (45 μ size) charnockite and epidote hornblende biotite gneiss

The recharge from the storage tank (Gupta and Deshpande 1998), subsurface barrier (Rolland et al. 2011), and check dams (Bhagavan and Raghu 2005) has reduced the concentration of fluoride in groundwater by dilution.

5 Pilot Study

A pilot study was carried out by the construction of a dug well recharge system in Dharmapuri district, Tamil Nadu. A few photographs taken during the construction of this structure are shown in Fig. 6. The schematic diagram of the constructed recharge structure is shown in Fig. 7. The site was chosen on the basis of three-year temporal variation in the fluoride concentration and fluctuation in groundwater level (Jagadeshan et al. 2015b). The groundwater level in induced recharge well (Well no. 44) raised to 9.1 m from 14.5 m (bgl), and the electrical conductivity has decreased from 1342 to 945 $\mu\text{S}/\text{cm}$ (Jagadeshan et al. 2015b). Geochemical results indicate that fluoride concentration has decreased from 3.1 to 1.41 mg/l in well no. 44 due to dilution by the rainwater passing through induced recharge structure into dug well. This clearly indicates that the raise in groundwater level reduces the electrical conductivity and fluoride concentration of groundwater. The induced recharge from the structure constructed benefited an area of about 1 km² (Fig. 8). This study promises that the concentration of fluoride in groundwater can be decreased in other fluoride-affected areas if such low-cost induced recharge structures are constructed.



Fig. 6 Photographs taken during the construction

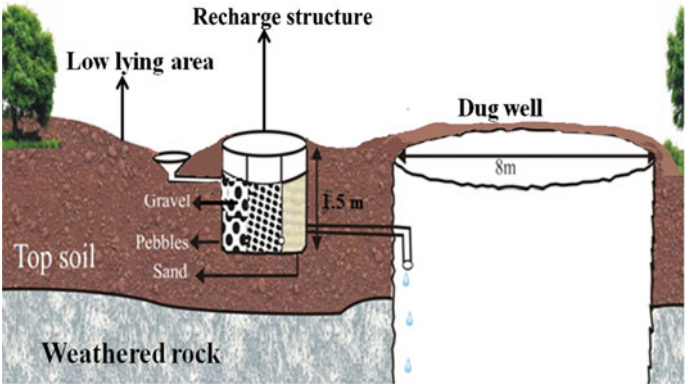


Fig. 7 Conceptual diagram of induced recharge structure

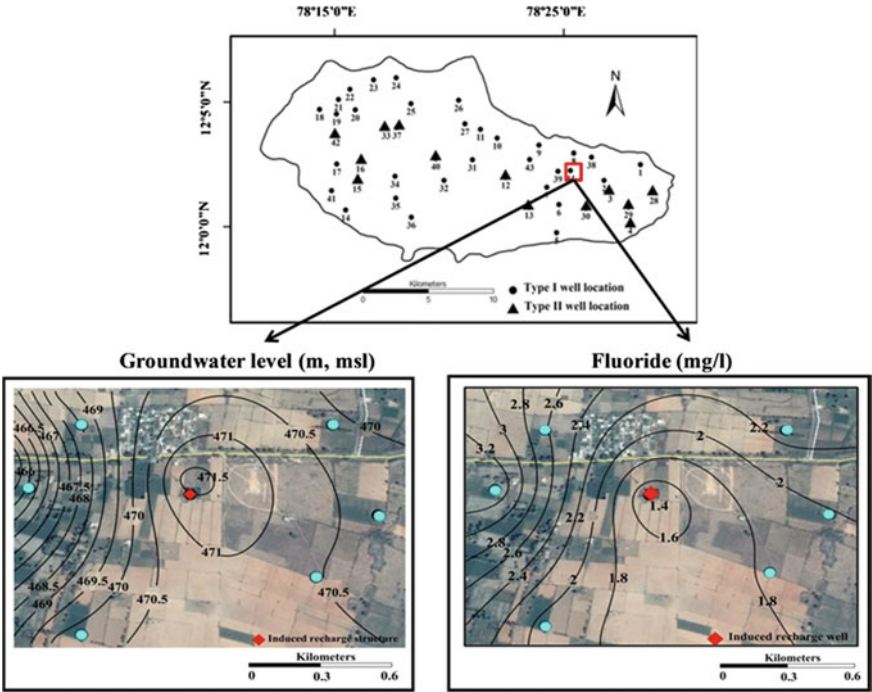


Fig. 8 Spatial variation diagram for groundwater level (m msl) and fluoride concentration (mg/l) around the induced recharge structure

6 Conclusion

Groundwater with high fluoride is a major problem as evident from studies carried out by numerous researchers in different parts of the world. The host rock rich in fluorine is the major source of fluoride in groundwater. Concentration of fluoride in groundwater increases due to host rocks weathering and prolonged residence time of water. Treatment of the high-fluoride groundwater is possible by several methods, but they have limitations and cost intensive. Technique such as rainwater harvesting, constructing check dams and percolation ponds, and facilitating artificial recharge of rainwater through existing wells are suitable on-site treatment available. A pilot study carried out by a dug well recharge system in Dharmapuri district, Tamil Nadu, demonstrated the potential of this scheme in reducing the concentration of fluoride in groundwater over an area of about 1 km². The recharge from the structure has reduced concentration of fluoride in groundwater from 3.1 to 1.44 mg/l due to dilution. The study successfully demonstrated the applicability of dug well recharge system at a carefully selected site based on the systematic long-term hydrogeochemical studies to solve the problem of fluoride contamination affecting millions of rural people.

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