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L E T T E R S

613.8 : 661.482 (540.27)

FLUORIDE CONTAMINATION IN SOUTHERN BLOCK OF SONBHADRA DISTRICT, UTTAR PRADESH, INDIA

1. Fluorine being the most electronegative and reactive elements of the periodic table, it does not occur in its elemental form in nature. In the solution state, fluorine tends to ionize in the anionic form fluoride (F^-). Since fluoride is similar in size and charge as that of hydroxide ion (OH^-), it tends to replace OH^- ion in the mineral lattice (Hem, 1985). This process of exchange of ion leads to the presence of fluoride in various minerals (Farwell *et al*., 2006; WHO, 2011) *viz*., apatite, fluorspar, hornblende, cryolite and mica (Murray, 1986). The igneous and sedimentary rocks commonly have fluoride-containing minerals. These minerals dissolve in due course of time and result in an increase of fluoride content in the sub-surface water resources (Apambire *et al*., 1997; Apambire, 2000; WHO, 2004, 2011). According to, world health organization (WHO), the permissible limit of fluoride ranges from 0.6 to 1.5 mg/l in potable water. The potable water having low concentrations of fluoride reduces the occurrence of dental caries (Fawell *et al*., 2006; WHO, 2011) in the human population. On the other hand, even little increment in the content may cause dental fluorosis in children (0.9-1.2 mg/l; Dean, 1942; WHO, 2011 or even lower; Galagan and Lamson, 1953; Apambire, 2000) and in adolescents (1.5 mg/l) leading to permanent discoloration of the teeth, erosion of enamel and pitting (Whitford, 1997) which is a worldwide phenomenon (Fawell *et al*., 2006). Exposure for a longer time with a high concentration of fluoride can lead to adverse health hazards *viz*., skeletal fluorosis, increased risk of bone fractures, increased bone density, calcification of the ligaments and tendons and bone deformity like crippling skeletal fluorosis (NRC, 2006). Similar, reports have appeared from different parts of the world *viz*., India, China (Wei and Wei, 2015; Wang *et al.*, 2007), Africa (Fawell *et al*., 2006; USEPA, 2010a; Craig, 2015), East Africa (Joan and Nielsen, 1999), Central Rift Valley, Ethiopia (Datturi *et al*., 2017) and United States (USEPA, 2010a). In India High concentration of fluoride in groundwater has been reported from different regions of Assam, Bihar, Chhattisgarh, Gujarat, Madhya Pradesh, Odisha, Rajasthan, West Bengal and Uttar Pradesh (Bhattacharya and Chakrabarti, 2011; Suthar, 2008; PHED, 2007; Sankararamakrishnan, 2008; Salve, 2008; Mishra, 2009; Kundu and Mandal, 2009). The impact of fluoride intake causing skeletal fluorosis is less common than that of dental fluorosis (NRC, 2006). The concentration of fluoride ion between 3 to 6 mg/l in drinking water is responsible for skeletal fluorosis while concentrations higher than 10 mg/l cause skeletal crippling in the human population (WHO, 1984, 2011).

In formulating the standards and targets for the developing nations, it is important to keep in view some factors *viz*., climate, fluoride sources, lifestyle and food habits (WHO, 2011). Since, water is the chief source of fluoride (Dean, 1942; NRC, 2006), average daily intake of water has been taken into account in defining standards for fluoride in drinking water (WHO, 1993; USEPA, 2010b). Although, water consumption data are available mainly for countries in temperate climate zone *viz*., Canada, United Kingdom and USA yet in many countries, there is a challenge to set national standards by collecting representative water consumption data at a massive scale. In countries like India, there is an urgent need to pursue a detailed study and monitor the standards already set to modify if needed. Since India does not have a proper water distribution system, the major population depends on different sources *viz*., wells, rainwater catchment bodies, lakes and streams. The estimation of exact water consumption by individuals is difficult, as the users are not accustomed to using the exactly measured amount of water. So, it is difficult to know the exact amount of water consumed on a daily basis.

The Sonbhadra district UP is bounded by three states *viz*., MP, Chhattisgarh and Jharkhand. The block Duddhi is well connected by road with Varanasi and can be reached in 3-4 hours (Fig. 1).

2. The district of Sonbhadra geologically comprised of Duddhi granitoid complex, Vindhyan Supergroup, Mahakoshal group and the recent alluvium which occur along Son River and other streams in narrow strips. The Semri and Kaimur Group of rocks occurring in this area is comprised of limestone and sandstone. The lithology of the Semri group includes basal conglomerates and Limestone, underlain unconformably by the schist, phyllite of the Mahakoshal group and over the granites of Duddhi granitoid complex (Srivastava *et al*., 2000).

The area under consideration situated in Duddhi block experiences a semi-arid and arid climate. The average annual temperature range between 9 °C (minimum) to 48 °C (maximum) respectively. In this

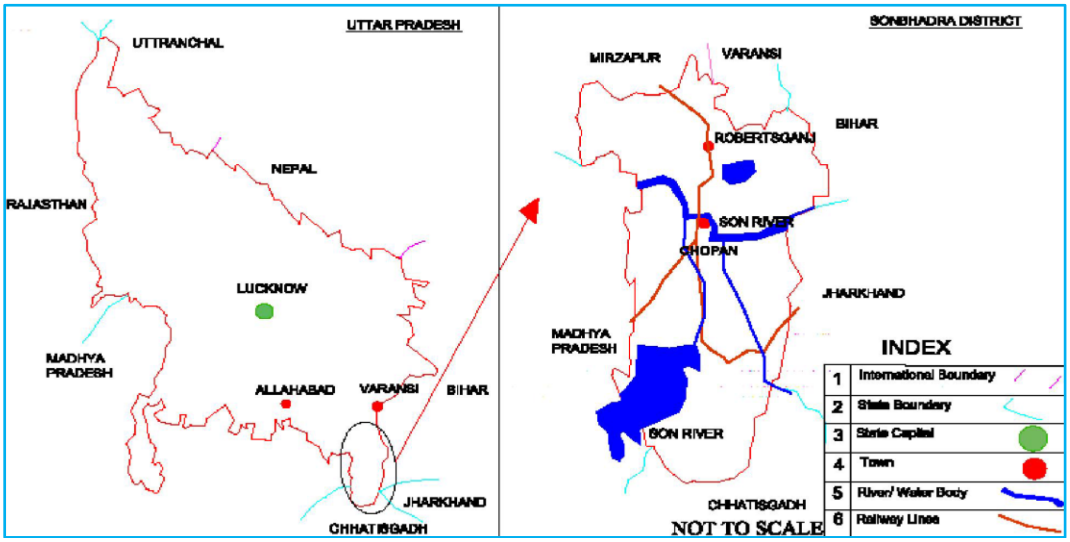

Fig. 1. Physical map of the study area

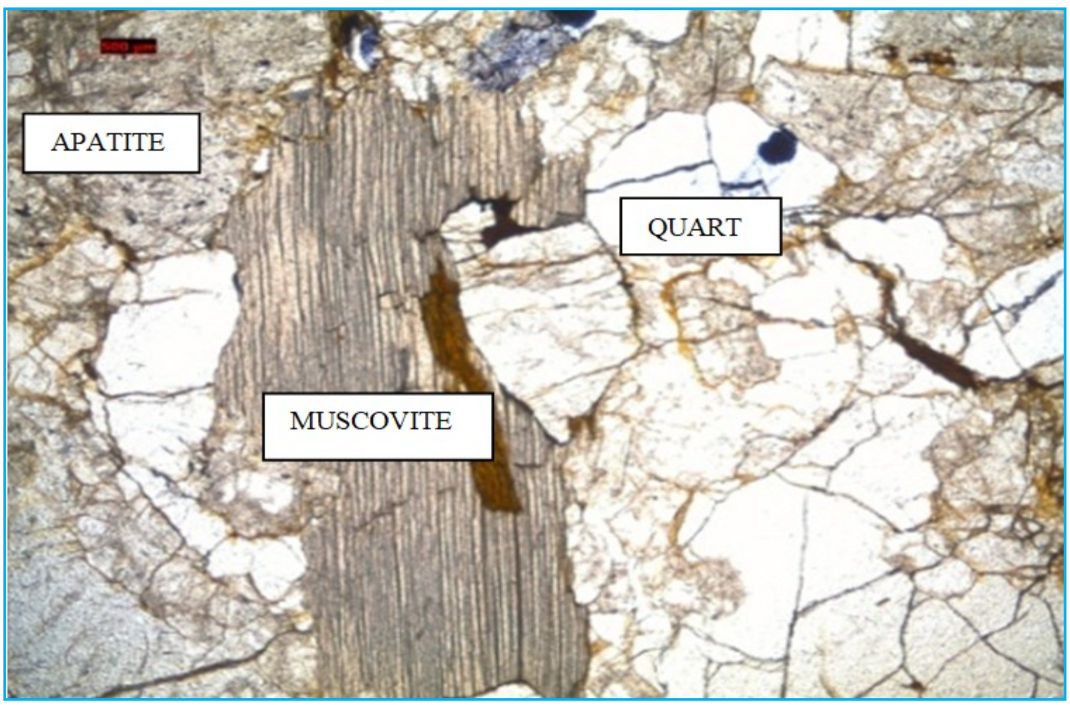

Fig. 2. Photomicrograph of granite

region, the main source of groundwater recharge is precipitation (average annual rainfall being 750 mm). There is a scarcity of groundwater in the study area. Therefore, agricultural activities mainly depend on rainfall. The major litho-unit of the area comprised of granitic and phyllitic rocks, overlain by red sandy soil cover. The drainage pattern varies from dendritic to sub-dendritic. Secondary intrusives such as dyke, pegmatite and quartz veins also occur to a limited extent. Here, groundwater occurs under the phreatic condition in the weathered and fractured zones. The water table lies at a depth ranging between 5.7 to 12.2 m below the ground surface. Further, the depth and the diameter of the dug wells range between 8 to 13 m and 2 to 8 m respectively. The maximum depth of hand pumps is generally 40 m below the groundwater level. The microscopic study of granite of the study area reveals that it consists of more than one-third of feldspar as alkali feldspar, Mica (muscovite and biotite) and Quartz as a major mineral with apatite, monazite and aplite as a minor mineral (Fig. 2).

TABLE 1

Location and Geochemical analysis of Groundwater samples in the study area

Duddhi Block	PH	Turbiditty (NTU)	TDS (PPM)	TH (PPM)	Alkalinity (PPM)	Fluoride (PPM)	Chloride (PPM)	Nitrate (PPM)	Sulphate (PPM)	Iron (PPM)	Water level (MBGL)
Dumardiha	7.52	Absent	500	380	340	5.1	370	15	260	0.6	9.5
Khursi Tola	7.6	Absent	540	340	340	4.97	330	17	210	0.5	9.7
Rajpahaad	7	Absent	570	370	260	0.85	340	20	130	0.7	10.5
Jharokhurd	7	Absent	530	310	230	0.81	305	22	290	0.8	10.5
Jharokala	7.2	Absent	520	305	370	1.5	290	20	220	0.1	10.1
Majhauli	7.6	Absent	570	320	130	0.3	310	19	150	0.5	10.7
Kaadal	7.7	Absent	590	360	300	5.63	280	18	205	0.2	9.4
Javar	7	Absent	595	390	150	0.43	250	16	265	3.3	10.6
Vasin Tola	7	Absent	560	310	260	0.81	370	17	180	0.3	10.5
Mudisemar	7	Absent	570	305	170	0.58	390	20	210	0.9	10.5
Chhatarpur	7.1	Absent	565	320	280	1.9	360	21	280	0.1	10.1
Harpura	7.6	Absent	520	360	330	4.55	350	22	210	0.5	9.7
Kudari	7.5	Absent	510	390	270	3.52	360	17	290	0.7	9.2
Khairkhar	7	Absent	565	310	150	0.42	370	23	180	0.1	10.1
Ghorpa	7	Absent	560	310	210	0.71	390	21	210	0.1	10.2
Bome	7.1	Absent	570	320	350	1.5	290	16	220	0.7	9.9
Jatajua	7.6	Absent	540	350	350	4.17	280	20	290	0.2	9.6
Badhmandva	7.3	Absent	510	310	260	1.87	280	17	290	0.5	10.1
Jorukhar	7.5	Absent	570	370	230	3.63	270	19	270	2.7	9.1
Fulwaar	7.4	Absent	530	310	250	2.16	310	20	180	0.1	9.2
Kevtaan Tola	7.2	Absent	550	320	240	1.95	320	16	210	0.9	9.2
Kanhaiya Ghutra	7	Absent	560	350	130	0.08	310	19	250	0.3	10.5
Duddhi	7.3	Absent	390	490	310	1.5	290	19	310	1.5	10.1
Chak Garadarwa	7.2	Present (5)	210	390	340	1	300	25	220	0.1	10.1
Birar	7.1	Present (5)	420	300	240	0.5	375	10	210	0.3	10.5
Kalkali Tola	7.4	Present (5)	420	225	260	1.5	375	25	205	0	9.7
Dharti Dolawa	7	Present (5)	375	300	230	1	375	10	265	0.3	10.1
Chatawa	7.3	Present (5)	375	225	370	1.5	275	25	270	0.1	10.2
Jampani	7	Present (5)	300	330	160	0.5	275	30	210	0.3	10.7
Koragi	7	Present (10)	900	330	130	0.32	275	10	180	0	10.7
Jharokhurd	7	Present(5)	405	315	130	0.5	500	10	210	0.5	10.6
Ghiwahi	7.2	Present(5)	600	300	260	1.5	250	10	290	0.1	10.1
Bairkhar	7.5	Present(5)	510	375	270	3.1	250	10	270	0	9.5
Kharwari Tola	7.1	Present(5)	480	225	280	0.5	225	25	210	0.5	10.4
Jharo Kala	7.2	Present(5)	540	450	310	1.5	350	25	250	0	10.1
Karoudhi	7	Present(5)	330	375	330	0.9	500	10	250	0.1	10.2

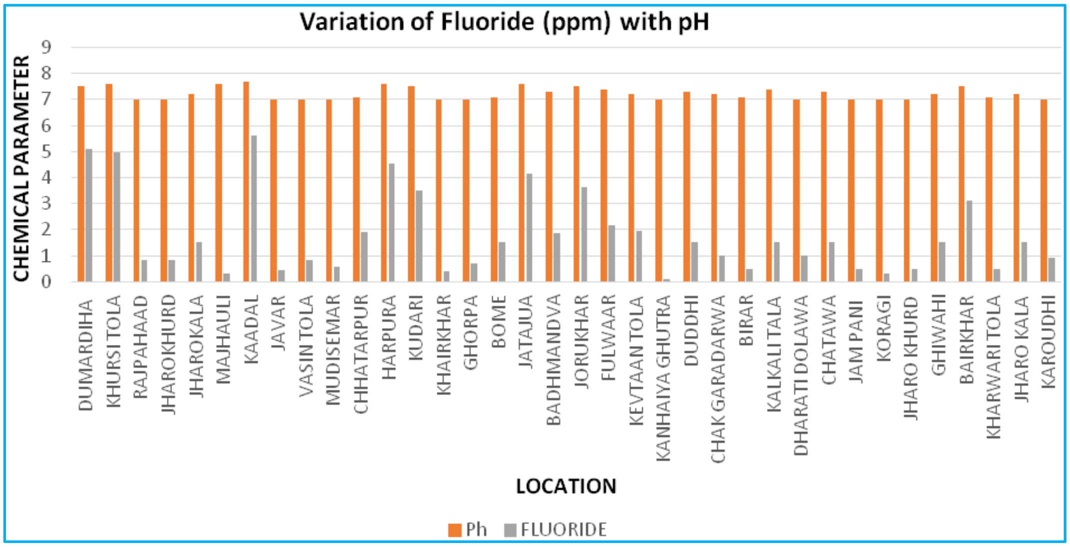

Fig. 3. Variation of Fluoride (ppm) with pH in groundwater

3. The important fluoride-bearing minerals include fluorapatite, fluorite (fluorspar), cryolite, mica *viz.*, biotite, muscovite and lepidolite, tourmaline, hornblende series minerals, glaucophane-riebeckite, asbestos (chrysotile, actinolite and anthophyllite), sphene, apophyllite, zinnwaldite, etc. These minerals have sufficient fluorine in their crystal lattice. The soil derived from such fluoride-rich minerals is responsible for an increase in the fluoride content of the groundwater.

The groundwater samples were collected from 36 hand pumps and bore wells taking all necessary precautions. The study includes the analysis of water samples for pH, TDS, EC, Hardness, Turbidity, Alkalinity, Chloride, Nitrate, Sulphate, Iron and Fluoride to know their correlation and impact on human health. The samples were then analyzed as per the prescribed guidelines and methods for major ion chemistry (APHA, 1995). The pH has been measured using pH meters while the total dissolved solids (TDS) have been determined by an ionic calculation method. Further, alkalinity has been determined by titrating the groundwater samples with a standard HCL solution. Total hardness (TH) were analyzed titrimetrically using standard EDTA solution. Turbidity is measured by Nephelometric Turbidity Units. Iron has been measured by Atomic absorption spectrometer (AAS), Chloride (Cl) has been estimated using standard Silver Nitrate ($AgNO_3$) titration and Sulphate (SO_4) has been measured using the turbidimetric method at Department of Civil Engineering, IIT-BHU Varanasi. The fluoride content was measured using fluoride ion-selective electrode (model 96-09 with 720 pH/ISE meter; Orion, USA) following APHA, 1998.

4. The result of the chemical analysis of the groundwater samples is presented in Table 1. The pH value of groundwater ranges between 7.0 and 7.7. The comparative study (Fig. 3) of fluoride concentration with pH of groundwater of different villages reveals that alkaline pH increases the solubility of fluoride from fluoride bearing minerals resulting in the high concentration of fluoride in groundwater (Saxena and Ahmed, 2003). In acidic medium, fluoride is adsorbed on clay while in an alkaline medium, it is desorbed. Thus, alkaline pH is more favorable for fluoride dissolution as shown in Fig. 3. Fluoride has a unique chemical behavior with water where most of the anions are easily replaced even under normal temperature and pressure. In general, apatite and fluorite, besides the replacement of hydroxyl ion by fluoride ions in mica, hornblende and soil mostly consisting of clay minerals, are major sources of fluoride in circulating water (Hem, 1985).

Fig. 4 represents a higher concentration of fluoride at shallow depths and as the water level goes down, there is a decrease in fluoride concentration. This is due to the higher rate of weathering of fluoride bearing rocks in the shallow well. Correlation matrix (Table 2) has value ranges between +1 and -1. The value near to +1 shows a strong and positive relationship between the element and value near to zero indicates no relationship between the elements while a value near -1 shows a strong negative relation between the elements. The correlation matrix of the different chemical parameters of groundwater indicates that fluorine concentration is positively correlated with pH, alkalinity and sulphate in the study area. The correlation value of iron clearly indicates that it does not

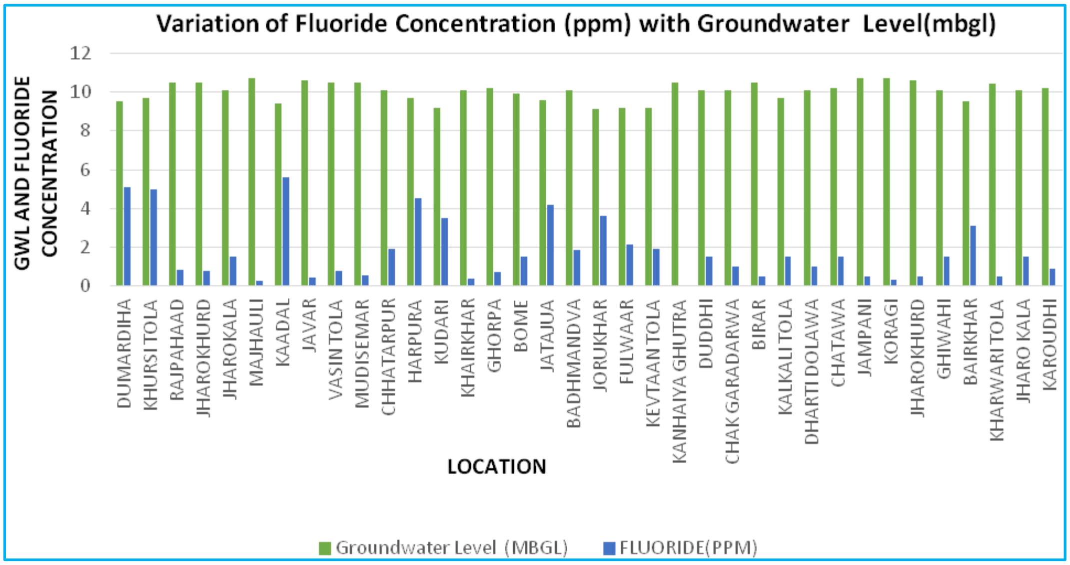

Fig. 4. Variation of Fluoride concentration with groundwater Level (GWL)

TABLE 2

Correlation matrix between Chemical parameter of Groundwater Sample

	pH	Turbidity	TDS	TH	Alkalinity	Fluoride	Chloride	Nitrate	Sulphate	Iron
PH	1	-	-	-	-	-	-	-	-	-
Turbidity	-0.01432	1	-	-	-	-	-	-	-	-
TDS	0.032281	-0.0983	1	-	-	-	-	-	-	-
TH	0.183223	-0.02804	-0.01412	1	-	-	-	-	-	-
Alkalinity	0.432156	-0.03685	-0.31043	0.1197203	1	-	-	-	-	-
Fluoride	0.836733	-0.09044	0.084072	0.2675251	0.55562375	1	-	-	-	-
Chloride	-0.26672	-0.13147	-0.25743	0.0152123	-0.1084217	-0.13165	1	-	-	-
Nitrate	0.056876	-0.31631	-0.25126	-0.110363	0.12763705	-0.04509	-0.25075	1	-	-
Sulphate	0.134073	0.086308	-0.19681	0.2732234	0.30032989	0.243814	-0.19258	-0.13372	1	-
Iron	-0.00088	-0.30466	0.115577	0.327842	-0.2113723	0.032185	-0.23201	-0.04607	0.2346	1

have any significant correlation with fluoride contamination, while chloride shows a negative correlation with fluoride.

According to 'Dean's classification index the degrees of dental fluorosis, zero (0) has been correlated as *no dental fluorosis,* One (*i*) for *questionable fluorosis*, two (*ii*) for *mild dental fluorosis,* three (*iii*) for *moderate dental fluorosis* while four (*iv*) as *severe dental fluorosis'* (Dean, 1942). As per the 'Dean's classification index 2 and index 4 of dental fluorosis has been observed in the study area. The majority of the people have mild dental fluorosis while some of the people are severely affected. The spectrograph of the residents residing in the Sonbhadra area clearly shows the shreds of evidence of the degree of dental fluorosis (Fig. 5). A lesser intake or even lack of fluoride intake in children (up to nearly 5 years) does not affect tooth development but may result in increased sensitivity of enamel to acid attacks. Excess intake of fluoride content initiate gastric and kidney disorder and can be fatal (http://www.fluoridealert.org/health). It can intervene with calcium metabolism and enzymic activities, activating both proteolytic and glycolytic functions and cell respiration by inhibiting

Fig. 5. Dental Fluorosis

Fig. 6. Skeletal fluorosis among the children of the study area

Na+/K+ -ATPase and can be lethal on the intake of 5 to 10 g in adults and 500 mg in children (Lech, 2011).

5. It is evinced by the study that fluoride is distributed non-uniformly in groundwater of study area. The study reports reveal that 35% of villagers of this area are at high risk due to excessive intake of fluoride while 65% of villagers are safe from fluoride-related health hazards. The Socioeconomic condition of the populace of the study area also has some impact on the percentage of fluoride debility or increase. Higher-income groups of society could escape the ill effects of fluoride contaminated water.

In the Duddhi Block, the fluoride concentration range in groundwater is 0.08 to 5.63 mg/l and its source appears to be geogenic. This is due to weathering of fluoride bearing rocks, rate of water flow, amount of rainfall, pH of the water and climatic conditions. Dental fluorosis (Fig. 5) is characterized by discolored, blackened, mottled or chalky white teeth which are clear indications of overexposure to fluoride during childhood. Chronic intake of undue fluoride leads to the severe and permanent bone and joint deformations termed as skeletal fluorosis (Fig. 6). Although initially no symptom of fluoride deficiency is identified, the excess concentration causes the number of physiological and morphological changes in the human body. To cope up with the growing problem of fluorosis, it is of utmost importance to understand the distribution and occurrence of fluoride in water resources and accordingly to work out the strategies for its mitigation and management with the techniques available for removal of fluoride from drinking water. There are four kinds of deflouridation techniques known *viz*., Nalgonda Technique (NEERI, 1987), Activated Alumina Technique, Reverse Osmosis and Ion Exchange. It is easy to adopt all these techniques. There are seventeen villages having fluoride concentration is within the permissible limit *viz*., Kanhaiya ghutra, Majhauli, Koragi, Khairkhar, Javar, Birar, Jampani, Jharokhurd, Kharwari tola, Mudisemar, Ghorpa, Jharokhurd, Vasin tola, Rajpahaad, Karoudhi, Chak garadarwa, Dharti dolawa (Fig. 4). Borewells and Dugwell which have a high content of fluoride (>1.5 mg/l) are to be identified and confined to the local habitant. Borewell/ Dugwell water with a low concentration (< 0.5 mg/l) of fluoride can be blended with high fluoride water (up to 2 mg/l) before being supplied for the domestic purpose where there is no alternate source of domestic water supply. The fluoride level in groundwater can also be brought down by rainwater harvesting and artificial recharge of groundwater.

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