Original Research Article

Fluoride Contamination in Groundwater and its Effect on Human Health: Study of Baramati Tahsil Area, India

ABSTRACT

Baramati Tahsil is the rural part of Pune district which have arid to semi-arid region. Groundwater is the main source of drinking water for the people living in this area. The groundwater is being removed from a dug well and borewells in the study area for drinking purpose. In the present study, fluoride from Dug well water of Baramati Tahsil area was studied. A total 15 groundwater samples of dug well were collected in the period of post-monsoon (POM) winter 2015 to pre-monsoon (PRM) summer 2017 for four seasons by using standard methods of APHA. The various physico-chemical parameters such as pH, Electrical conductivity, Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, Total Dissolved Solids (TDS), Total Hardness (TH), HCO_3^-, Cl^-, SO_4^{2-} and F^- were determined using standard procedures of APHA suggested for determination of quality of water for drinking purpose. The results obtained from analysis was used for interpretation of fluoride and other ions concentration and its effect on human health. Results obtained indicates that concentration of fluoride in POM and PRM was within the maximum permissible limit of WHO and BIS recommended for drinking purpose. World Health Organization (WHO 1984) has fixed a safe limit of fluoride from 0.5 to 1.5 mg/l in drinking water. The maximum fluoride concentration in study area was 0.68 mg/l while minimum was 0.12 mg/l and average fluoride concentration was 0.41 mg/l in all the four seasons. The intake of drinking water with fluoride content less than 0.5 mg/l can cause tooth decay. The groundwater of all wells was suitable for drinking purpose without treatment for removal of fluoride at the time of analysis.

Key words: Dug Well, Fluoride Concentration, Groundwater, Pre-monsoon, WHO

1. INTRODUCTION

The climate of the Baramati tahsil area is slightly different in irrigated and non-irrigated area. The winter season is from December to about the middle of February followed by summer season which last up to May. June to September is the south-west monsoon season, whereas October and November constitute the post-monsoon season. The mean minimum temperature is about 12°C and mean maximum temperature is about 39°C. The average annual rainfall for the period 2003 to 2012 of Baramati tahsil was 505.76 mm [1].

Fluorides are compounds that combine the element fluorine with another substance, usually a metal. Examples include fluoride monofluorophosphate (MFP fluoride), sodium fluoride and stannous fluoride. Some fluorides occur naturally in soil, air or water, although the levels of fluoride can vary widely. Fluoride is also found in plant and animal food sources. When food material enters inside the body, fluorides are absorbed into the blood through the digestive tract. They travel through blood and tend to collect in areas high in calcium, such as the bones and teeth. The major sources of fluoride for most people are water and other beverages, food, and fluoride-containing dental products (toothpastes, mouth rinses, etc.). Because dental products are generally not swallowed (except, perhaps, by younger children), they cause less concern for possible health issues. Water fluoridation began in some parts of the United States in 1945, after scientists noted that people living in areas with higher water fluoride levels had fewer cavities. Starting in 1962, the United States Public Health Service (PHS) recommended that public water supplies contain fluoride to help prevent tooth decay [2].

Nearly 200 million people from 25 nations are affected by the deadly disease of fluorosis [3-4]. Fluorosis-affected regions are reported from China [5-7], India [8-15], Africa [16], Korea [17], Mexico [18], Kenya [19] and Nigeria [20]. In India, a number of people suffer from fluorosis due to intake of high fluoride content through drinking water, Approximately the excessive fluoride in groundwater is noticed in 177 districts covering 21 states, affecting 62 million people, including 6 million children [21-22]. A small amount of fluoride is essential to maintain bones and formation of dental enamel [23-24]. However, prolonged intake of high fluoride in drinking water can surely cause fluorosis [25-27].

It is further estimated that Indian population of 18,197,000 are affected with dental fluorosis and 7,889,000 with skeletal fluorosis. The study further indicates skeletal fluorosis has attributed Disability-Adjusted Life-Years (DALY) of 17 per 1000 population in India [28-29].

World Health Organization (WHO) [30] has fixed a safe limit for fluoride from 0.5 to 1.5 mg/l in drinking water. Moreover, the intake of drinking water with fluoride content less than 0.5 mg/l can cause tooth decay. Larger than 1.5 mg/l fluoride content in drinking water is risky for human consumption which leads to dental fluorosis and skeletal fluorosis when exceeds 3.0 mg/l [31-33]. By considering the importance of fluoride in human health the study of fluoride concentration in dug wells of Baramati Tahsil area is undertaken.

2. MATERIAL AND METHODS

Baramati Tahsil belongs to western part of Maharashtra. It belongs to Pune division. It is located 100 km towards east from district headquarters Pune. 240 km from state capital Mumbai towards east (Figure 1).

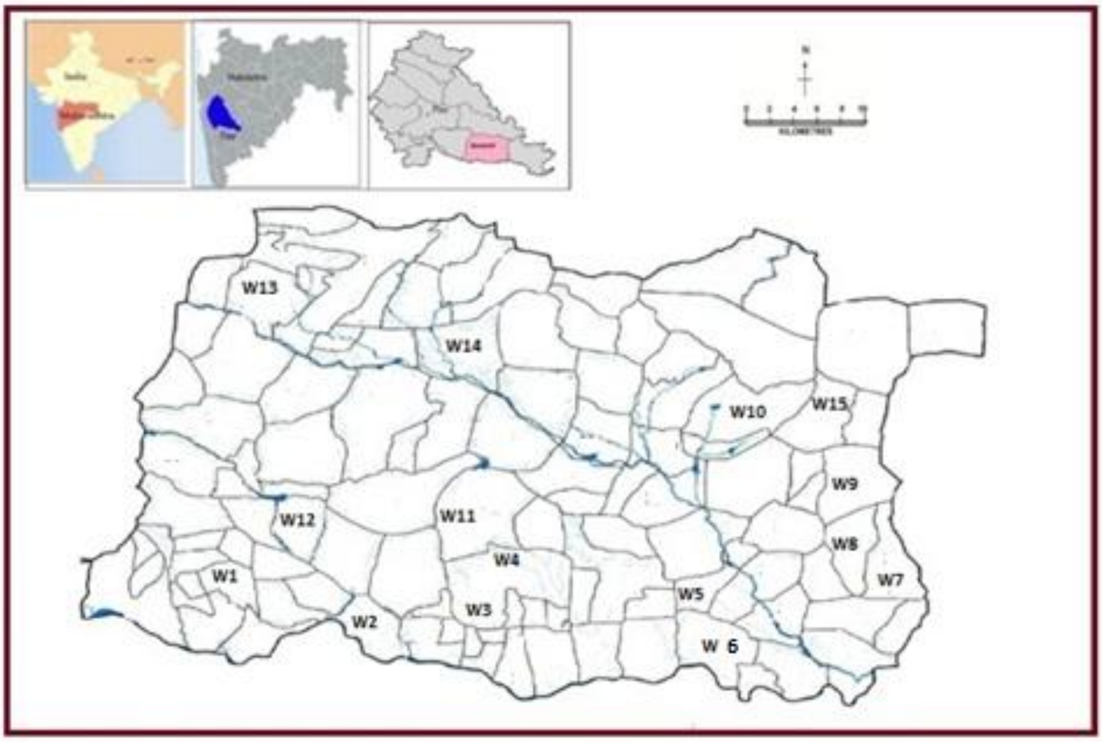


Figure 1: Study area map showing location of the sampling sites

Baramati Tahsil has its head quarter at Baramati city. Baramati Tahsil lies between 18º04´ to 18°32´ north latitudes and 74° 26´to 74° 69´ east longitudes. It is located at altitude of 550 meters above means sea level [34]. Dug well water samples from fifteen different wells of Baramati Tahsil area were selected randomly by considering the topography and anthropological activities of the study area (Figure 1).

Dug well Water samples from the selected sites were collected in a good quality polyethylene bottle of one-litre capacity during the period POM (winter 2015) to PRM (summer 2017) for four seasons in triplicate. Fifteen dug well water samples (W1- W8 from canal irrigated area and W9-W15 from non-canal-irrigated area) were selected for collection of water samples for analysis of fluoride and other parameters necessary for drinking purpose.

Physico-chemical parameters like pH, EC, TDS, Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, $Cl^{–}$, $SO_4^{2–}$, $CO_3^{2–}$, $HCO_3^{–}$, NO_3^{-}, F^{-} etc. were analysed in the laboratory by using standard methods recommended by APHA. Various physical parameters like pH, EC, and TDS were determined immediately within two hours of collection of sampling with the help of digital portable pH meter and Conductivity meter in the laboratory. Calcium (Ca^{2+}), Magnesium (Mg^{2+}), Chloride ($Cl^{–}$), Carbonate ($CO_3^{2–}$), Bicarbonate ($HCO_3^{–}$) and Sulphate ($SO_4^{2–}$) were determined by volumetric titration methods; while Sodium (Na^{+}) and Potassium (K^{+}) by Flame Photometry and fluoride by SPANDS method as recommended by APHA. The respective average values of all four seasons are reported in Table 1.

The fluoride concentration found in four seasons (Table 2) were compared with standard parameters recommended by the WHO (1.5 mg/l) and BIS [35] (1.0 mg/l). The chemical composition of water in various water bodies varies in different seasons. Therefore, it is necessary to test water frequently so that any small change in water quality is noted. It is also known that a seasonal change in environmental conditions were the major source of variation in the water chemistry [36]. In view of this, it was decided to collect the dug well water samples from fixed sampling stations twice in a year covering pre-monsoon (summer) and post-monsoon (winter) seasons for two years.

Fluoride (HF) reacts with zirconium (SPANDS) solution (under acidic condition) and the colour of SPANDS gets bleached due to formation of zirconium fluoride (ZrF_2^-). Since bleaching is a function of fluoride ions and directly proportional to the fluoride concentration. It obeys beer's law in reverse manner [37-38]. A series of standard fluoride solutions were prepared by diluting stock solution with distilled water. 100 ml volumetric flask, numbering from 1 to 10 is taken and added 1 to 10 ml standard sodium fluoride solution in the flask 1 to 10 respectively. Add 4 ml con. HCl and 1 ml Zirconium alizarin reagent in every flask, and dilute to 100 ml with distilled water. Mix and allow standing overnight. Absorbance was noted at 520 nm on Spectrophotometer. Same procedure was used for analysis of fluoride from groundwater. Compare the absorbance of groundwater samples with standard curve and find out concentration of fluoride [39].

3. RESULTS AND DISCUSSION

The physicochemical composition of the groundwater samples for four seasons were analysed and the average results of various parameters are given in table 1 with Maximum, Minimum and Average values. The pH of groundwater sample ranges from 6.63 to 8.32 with an average 7.78, indicated the alkaline nature of groundwater. Electrical Conductivity (EC) is the most important parameter of water to indicate TDS and its suitability for drinking purpose. The EC varies from 440 to 8473 µS/cm with an average 1777 which shows higher TDS in study area and which is not suitable for drinking purpose. Maximum permissible limit recommended by BIS and WHO for TDS in drinking water is 500 mg/l.

Table 1: Average Physico-chemical data of the ground water of Baramati Tahsil area, Pune, India (Winter 2015 to Summer 2017)

Sr No	PH	EC	TDS	Ca^{2+}	Mg^{2+}	Na^+	K^+	HCO_3^-	Cl^-	SO_4^{2-}	NO_3^-	TH	F^-
		µS/cm	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l
W1	8.32	600	384	42	21	105	6.2	218	75	248	37	191	0.45
W2	7.82	533	341	43	12	74	3.2	121	123	121	38	157	0.51
W3	7.84	580	371	57	29	61	1.5	132	208	200	44	262	0.33
W4	8.14	765	490	55	56	28	0.5	430	113	119	43	368	0.42
W5	8.25	1128	722	12	13	355	7.3	748	149	185	48	83	0.41

W6	7.4	6523	4174	49	41	450	0.8	291	298	236	41	292	0.35
W7	7.9	943	603	37	18	102	6.0	311	104	152	47	166	0.37
W8	8.01	2338	1496	31	14	151	6.7	283	102	178	38	136	0.34
W9	7.44	773	494	60	34	34	0.2	203	142	131	27	287	0.50
W10	7.64	440	282	30	16	76	3.8	346	50	104	28	142	0.55
W11	8.25	1085	694	35	19	40	1.2	205	39	104	22	169	0.48
W12	8.15	1215	778	41	21	334	5.6	371	422	507	25	188	0.42
W13	7.39	613	392	29	31	19	0.3	139	88	142	25	200	0.24
W14	6.63	8473	5422	108	254	656	2.4	378	1147	1278	25	1313	0.40
W15	7.59	650	416	44	20	73	1.9	295	94	138	15	194	0.44
Avg	7.78	1777	1137	45	40	170	3.2	298	210	256	34	277	0.41
Max	8.32	8473	5422	108	254	656	7.3	748	1147	1278	48	1313	0.55
Min	6.63	440	282	12	12	19	0.2	121	39	104	15	83	0.24
Med	7.84	773	494	42	21	76	2.4	291	113	152	37	191	0.42
SD	± 0.45	± 2396	± 1533	± 21	± 60	± 189	± 2.6	± 156	± 278	± 300	± 10	± 296	± 0.08

Table 2: Fluoride concentration of dug well water of Baramati Tahsil area, Pune, India (Winter 2015 to Summer 2017)

Sr No.	**Winter 2015 (POM)**	**Summer 2016 (PRM)**	**Winter 2016 (POM)**	**Summer 2017 (PRM)**	**Average**	**SD**
	mg/l	mg/l	mg/l	mg/l		
W1	0.46	0.42	0.46	0.45	0.45	±0.02
W2	0.55	0.56	0.35	0.58	0.51	±0.11
W3	0.24	0.24	0.6	0.23	0.33	±0.18
W4	0.35	0.38	0.57	0.37	0.42	±0.10
W5	0.41	0.41	0.41	0.39	0.41	±0.01
W6	0.35	0.35	0.35	0.33	0.35	±0.01
W7	0.38	0.32	0.43	0.35	0.37	±0.05
W8	0.27	0.27	0.54	0.29	0.34	±0.13
W9	0.43	0.43	0.68	0.45	0.50	±0.12
W10	0.54	0.54	0.54	0.58	0.55	±0.02
W11	0.45	0.45	0.55	0.46	0.48	±0.05
W12	0.42	0.42	0.45	0.37	0.42	±0.03

W13	0.12	0.2	0.39	0.25	0.24	±0.11
W14	0.34	0.34	0.54	0.39	0.40	±0.09
W15	0.35	0.35	0.65	0.39	0.44	±0.14
Average	0.38	0.38	0.50	0.39		
Maximum	0.55	0.56	0.68	0.58		
Minimum	0.12	0.20	0.35	0.23		
Median	0.38	0.38	0.54	0.39		
Standard Deviation	±0.11	±0.10	±0.10	±0.10		

Fluoride concentration of groundwater of study area in winter 2015 ranges from 0.12 to 0.55 mg/l having average 0.38 mg/l. Standard deviation was ± 0.11 while median of the data in this season is 0.38. In summer 2016 fluoride value ranges from 0.20 to 0.56 mg/l having average 0.38 mg/l and deviation of fluoride concentration was found to be ± 0.10. Standard deviation of fluoride in winter 20106 season was ± 0.10 and values ranges from 0.35 to 0.68 mg/l having average of 0.50 mg/l. In summer 2017 fluoride concentration value ranges from 0.23 to 0.58 mg/l having average 0.39 mg/l and deviation in the readings found to be ± 0.10. (Table 2).

When fluoride concentration is high, it causes mottling of teeth or dental fluorosis, skeletal fluorosis, forward bending of vertebral column, deformation of knee joints and other parts of body and even paralysis (paraplegia, quadriplegia). Mottled enamel is appeared on the teeth of children who drink water containing too much fluoride during the period when permanent teeth are formed. Lower concentration of fluoride in certain area may causes tooth decay. The intake of drinking water with fluoride content less than 0.5 mg/l can cause tooth decay. Fluoride concentration between 0.7 to 1.5 mg/l is effective in the prevention of dental cares. The degree of fluorosis generally increases as fluoride concentration increases above 2.4 mg/l. [40-42]. Fluoride concentration of all water samples in study area were within the permissible limit of WHO and BIS.

In the study area variations in fluoride in four seasons is observed which is shown in figure 2. The monsoon and summer seasons show changes in properties of natural water. Rainwater can change the geochemical properties by recharging the aquifer and causing dilution effect. On the other hand, in summer, the process of evaporation leads to concentration of salts in the natural water. Drying up of clay minerals above the water table during summer season leads to oxidation, which increases the solubility of minerals by the infiltrating water during monsoon season [43-44].

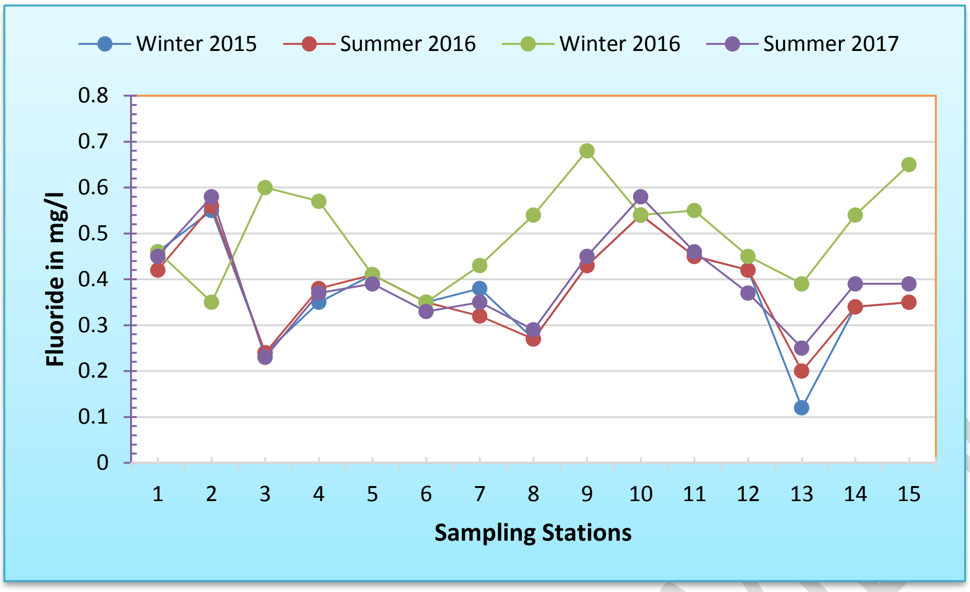


Figure 2: Spatio-temporal variation in Fluoride of groundwater from Baramati Tahsil area, Pune, India

The Correlation coefficient (r) among various groundwater parameters was calculated. Correlated data of four seasons are given in table 3. The statistical analysis has been performed using standard methods. The correlation coefficient among the different parameters will be true when the value of correlation coefficient (r) is high and approaching to one. Correlation, the relationship between two variables, is closely related to prediction. The greater the association between variables, the more accurately we can predict the outcome of events. The correlation coefficients (r) for some parameters of water were evaluated and presented in Table 3. The degree of correlation was distributed in four types i.e. Perfect positive correlation (r = + 1), Perfect negative correlation (r = - 1), moderately positive correlation (0 < r < 1) and moderately negative correlation (- 1 < r < 0) [45]. More precisely it can be said that parameters showing r > 0.7 are considered to be strongly correlated whereas 'r' between 0.5 and 0.7 shows moderate correlation [46].

Table 3: Correlation coefficient (r) among various groundwater parameters of samples from Baramati Tahsil area (Winter 2015 to Summer 2017)

Para meter	**PH**	**EC**	**TDS**	**Ca^{2+}**	**Mg^{2+}**	**Na^{+}**	**K^{+}**	**HCO_3^-**	**Cl^-**	**SO_4^{2-}**	**NO_3^-**	**TH**	**F^-**
pH	1.00	-0.67	-0.67	-0.69	-0.73	-0.46	0.48	0.18	-0.67	-0.60	0.37	-0.73	0.17
EC		1.00	1.00	0.66	0.79	0.87	-0.13	0.16	0.82	0.77	-0.04	0.79	-0.23
TDS			1.00	0.66	0.79	0.87	-0.13	0.16	0.82	0.77	-0.04	0.79	-0.23
Ca^{2+}				1.00	0.87	0.47	-0.42	-0.19	0.81	0.77	-0.21	0.91	0.02
Mg^{2+}					1.00	0.69	-0.21	0.13	0.93	0.92	-0.20	1.00	-0.10

Na^+						1.00	0.20	0.48	0.84	0.81	0.03	0.66	-0.15
K^+							1.00	0.47	-0.06	0.05	0.38	-0.26	0.06
HCO_3^-								1.00	0.18	0.18	0.30	0.07	0.07
Cl^-									1.00	0.98	-0.18	0.92	-0.12
SO_4^{2-}										1.00	-0.22	0.91	-0.10
NO_3^-											1.00	-0.20	-0.23
TH												1.00	-0.08
F^-													1.00

In the study area TDS showed moderate correlation with calcium (0.66) and strong correlation with magnesium (0.79), sodium (0.87), chloride (0.82), sulphate (0.77) and total hardness (0.79). Calcium showed strong correlation with magnesium (0.87), chlorine (0.81), sulphate (0.77), total hardness (0.91) and moderate correlation with sodium (0.47). Magnesium showed strong positive correlation with sodium (0.69), chlorine (0.93),) and sulphate (0.92).

The relation between fluoride and other chemical parameters provide significant geochemical information and also help to know the controlling factors and its mechanism of fluoride enrichment in the groundwater [47-48]. The correlation of fluoride with other parameters is shown in figure 3 to figure 9. Fluoride concentration shows positive correlation with pH (0.17) (Table 3 and figure 3), which indicates that the higher alkaline nature of water accelerates the enrichment of fluoride concentration and thus typically affects the concentration of fluoride in the groundwater [49-53]. A significant positive correlation is noticed between fluoride with bicarbonate (0.07) (Table 3 and figure 4), which declares that the alkaline environment is the dominant controlling chemical mechanism for leaching of fluoride from the fluoride bearing minerals in the groundwater of the study area [54-55].

Moreover, as shown in Table 3 and Figure 5, the correlation of fluoride and calcium (0.02), clearly indicates that the presence of high calcium content favored low fluoride. The positive correlation of fluoride is also observed with potassium (0.060) (Table 3 and figure 6). It is observed that the major role of precipitation process that is a vital mechanism for enhancement of fluoride occurrence in groundwater [56]. The negative correlation of fluoride observed with sodium (-0.15), chlorine (-0.12) (Table 3 and figure 7) and Magnesium (-0.10), sulphate (-0.10) (Table 3 figure 8) is an agreement as established by Reddy et al [57].

A number of studies have demonstrated that the positive correlations between fluoride and pH, bicarbonate and sodium typically accelerate the fluoride concentration in groundwater as well as an inverse relationship between fluoride and calcium, which reveals the concentration in fluorite saturated groundwater [58]. In study area EC and TDS correlation with fluoride is not positive like pH, bicarbonate, and calcium in the study area (table 3 figure 9), which divulges that a higher affinity of fluoride with pH and bicarbonate rather than EC and TDS. However, other workers study reveals that the high concentration of EC and TDS is always linked with greater fluoride concentration and the similar observations noticed in different regions [59-60].

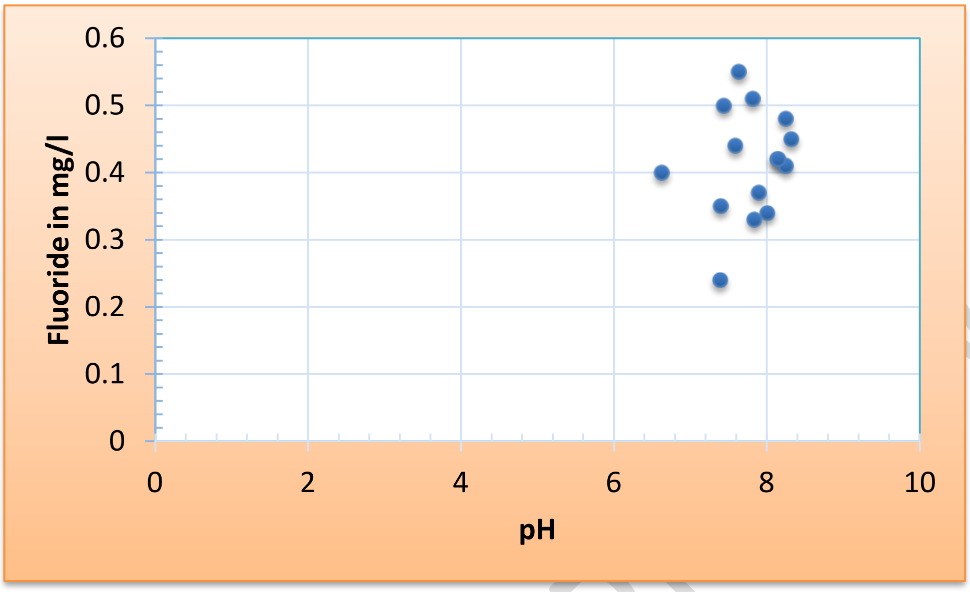


Figure 3: Correlation between Fluoride and pH of Groundwater samples from Baramati Tahsil area

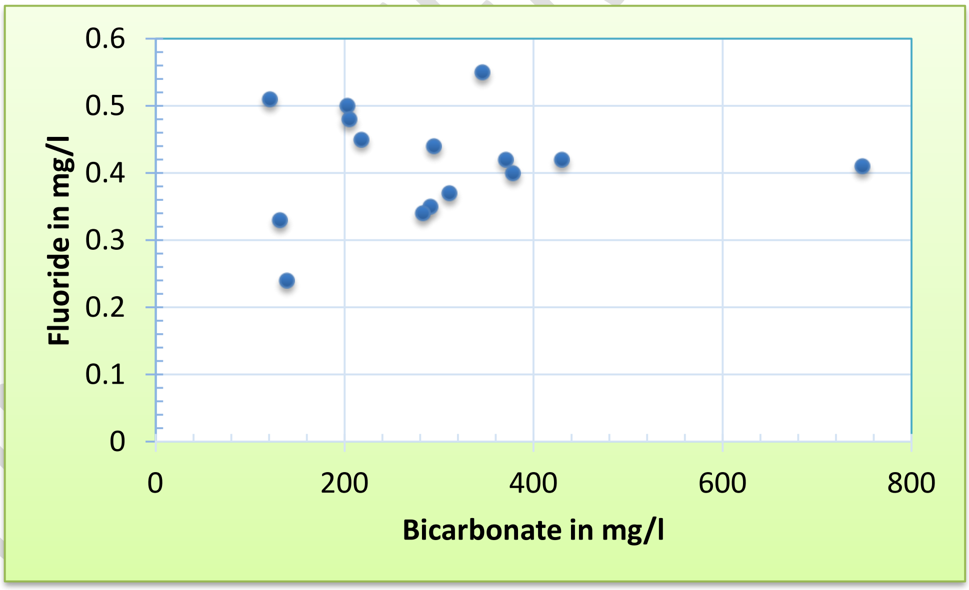


Figure 4: Correlation between Fluoride and Bicarbonate of Groundwater samples from Baramati Tahsil area

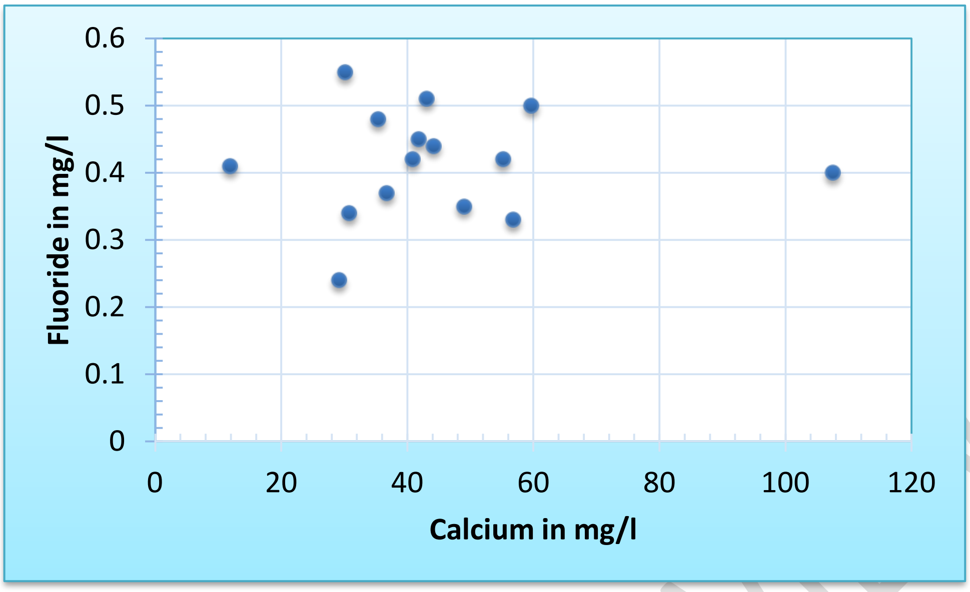


Figure 5: Correlation between Fluoride and Calcium of Groundwater samples from Baramati Tahsil area

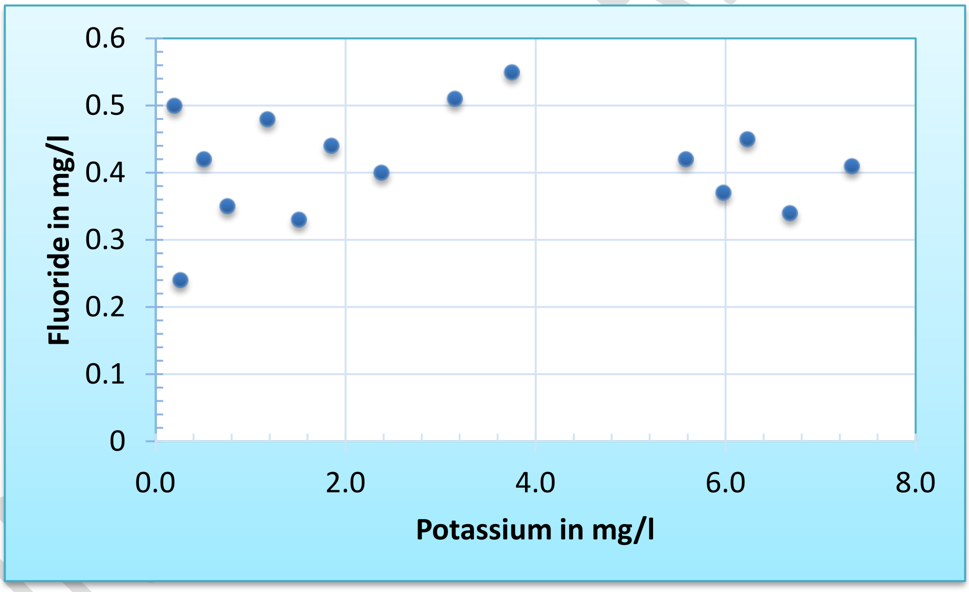
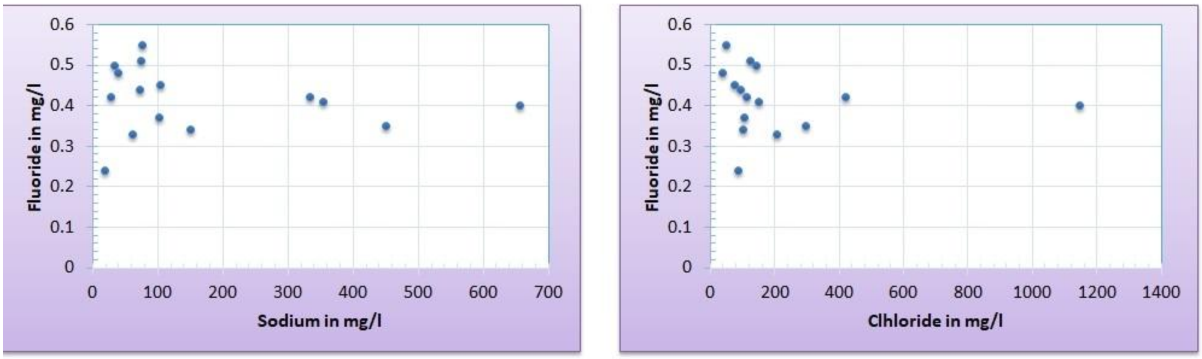
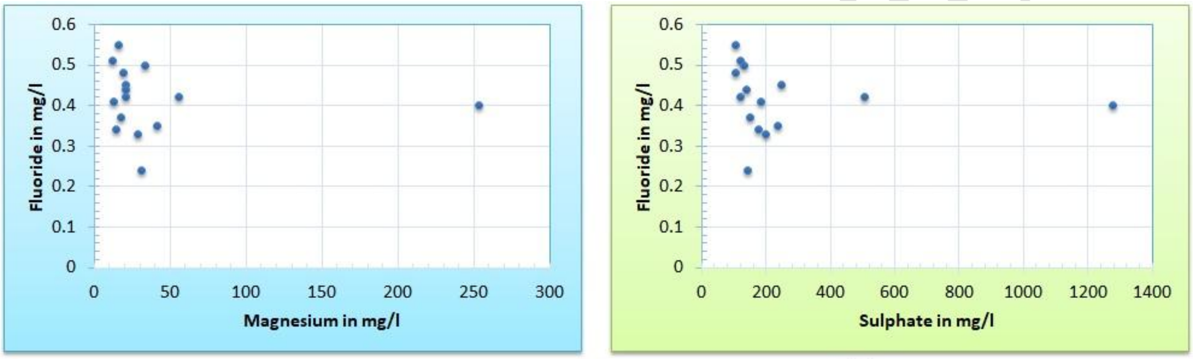


Figure 6: Correlation between Fluoride and Potassium of Groundwater samples from Baramati Tahsil area



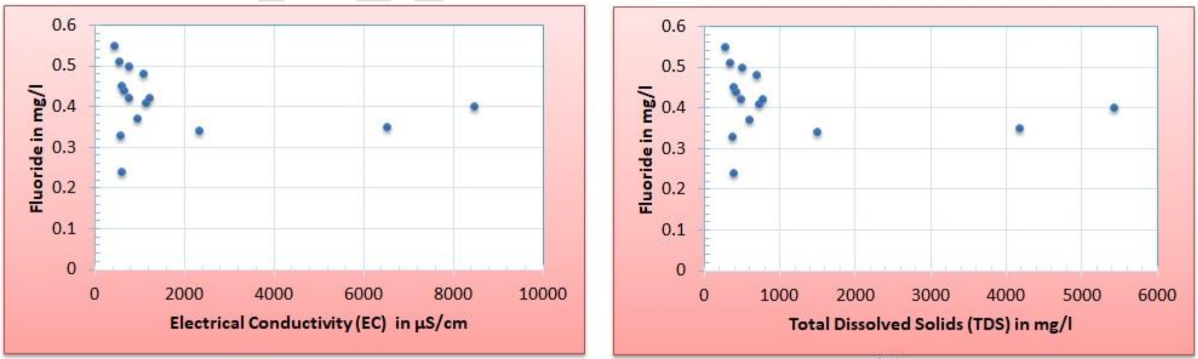
(A) (B)

Figure 7: Correlation between (A) Fluoride and Sodium, (B) Fluoride and Chloride of Groundwater samples from Baramati Tahsil area



(A) (B)

Figure 8: Correlation between (A) Fluoride and Magnesium, (B) Fluoride and Sulphate of Groundwater samples from Baramati Tahsil area



(A) (B)

Figure 9: Correlation between (A) Fluoride and EC, (B) Fluoride and TDS of Groundwater samples from Baramati Tahsil area

4. CONCLUSION

In study area fluoride concentration varies from 0.12 to 0.68 mg/l/. This indicates the fluoride concentration in Baramati tahsil area was in permissible limit of WHO (1.5 mg/l) and BIS (1.0 mg/l). This not causes adverse effect of fluoride due to consumption of groundwater, indicating the fluoride concentration is not any health hazard. But lower concentration of fluoride in certain area may causes tooth decay. The intake of drinking water with fluoride content less than 0.5 mg/l can cause tooth decay.

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