Original Research Article

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

ABSTRACT

Baramati Tehsil is the rural part of the Pune district which have an 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 purposes. The present study studied, fluoride from Dug well water of the Baramati Tehsil area. A total of 15 groundwater samples of dug well were collected in post-monsoon (POM) winter 2015 to pre-monsoon (PRM) summer 2017 for four seasons by using standard methods of American Public Health Association (APHA). The various physico-chemical parameters such as pH, Electrical conductivity (EC), Total Dissolved Solids (TDS), Total Hardness (TH), Calcium (Ca^{2+}), Magnesium (Mg^{2+}), Sodium (Na^{+}), Potassium (K^{+}), Bicarbonate (HCO_3^{-}), Chloride (Cl^{-}), Sulphate (SO_4^{2-}) and Fluoride (F^{-}) were determined using standard procedures of APHA suggested for determination of the water quality for drinking purpose. The results obtained from the analysis was used for interpretation of fluoride and other ions concentration and the effect on human health. Results obtained indicate that the fluoride concentrations in POM and PRM were within the maximum permissible limit of WHO and BIS recommended for drinking. World Health Organization (WHO 2006) has fixed a safe limit of fluoride from 0.5 to 1.5 mg/l in drinking water. The maximum fluoride concentration in the study area was 0.68 mg/l while the minimum was 0.12 mg/l and the average fluoride concentration was 0.41 mg/l in all four seasons. The drinking water intake with fluoride content less than 0.5 mg/l can cause tooth decay. The groundwater of all wells was suitable for drinking purposes without treatment for fluoride removal at the time of analysis.

Keywords: Dug Well, Fluoride Concentration, Groundwater, Pre-monsoon, WHO

1. INTRODUCTION

The irrigated and non-irrigated regions of the Baramati Tehsil have slightly differing climates. The winter season runs from December to the middle of February, and the summer season runs from March to May. The southwest monsoon season lasts from June to September, with the post-monsoon season lasting from October to November. The average minimum temperature is 12°C, while the average maximum temperature is 39°C. The average annual rainfall for the period 2003 to 2012 of Baramati Tehsil was 505.76 mm [1].

Fluorides are substances that include fluorine and another material, usually a metal. Fluoride monofluorophosphate (MFP fluoride), sodium fluoride, and stannous fluoride are examples. Fluorides can be found naturally in soil, air, and water, though fluoride levels can vary greatly. Fluoride is also found in plant and animal food sources. When food material enters the body, fluorides are absorbed into the blood through the digestive tract. They travel through the blood and collect in areas high in calcium, such as the bones and teeth. Water and other liquids, food, and fluoride-containing dental products are the most common sources of fluoride for most people (toothpastes, mouth rinses, etc.). Because dental products are rarely consumed (unless by very young children), they are less likely to create health problems. 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], South Africa [16], South 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 the formation of dental enamel [23-24]. However, prolonged high fluoride intake in drinking water can cause fluorosis [25-27].

It is further estimated that the 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) to 17 per 1000 population in India [28-29].

Fluoride in drinking water has a healthy level of 0.5 to 1.5 mg/l, according to the World Health Organization [30]. Furthermore, tooth decay can be caused by drinking water with a fluoride level of less than 0.5 mg/l. When fluoride levels in drinking water exceed 1.5 mg/l, it is dangerous for human ingestion, causing dental and skeletal fluorosis [31-33]. By considering the importance of fluoride in human health the study of fluoride concentration in dug wells of the Baramati Tehsil area is undertaken.

2. MATERIAL AND METHODS

Baramati Tehsil belongs to the western part of Maharashtra. It belongs to the Pune division, located 100 km east of district headquarters Pune, 240 km east of state capital Mumbai (Figure 1). Baramati Tehsil has its headquarter at Baramati city. Baramati Tehsil lies between 18º04´ to 18°32´ north latitudes and 74°26´to 74°69´ east longitudes. It is located at an altitude of 550 meters above sea level [34]. Dug well water samples from fifteen different wells of the Baramati Tehsil 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 the non-canal-irrigated area) were selected to collect water samples for fluoride analysis and other parameters necessary for drinking purposes.

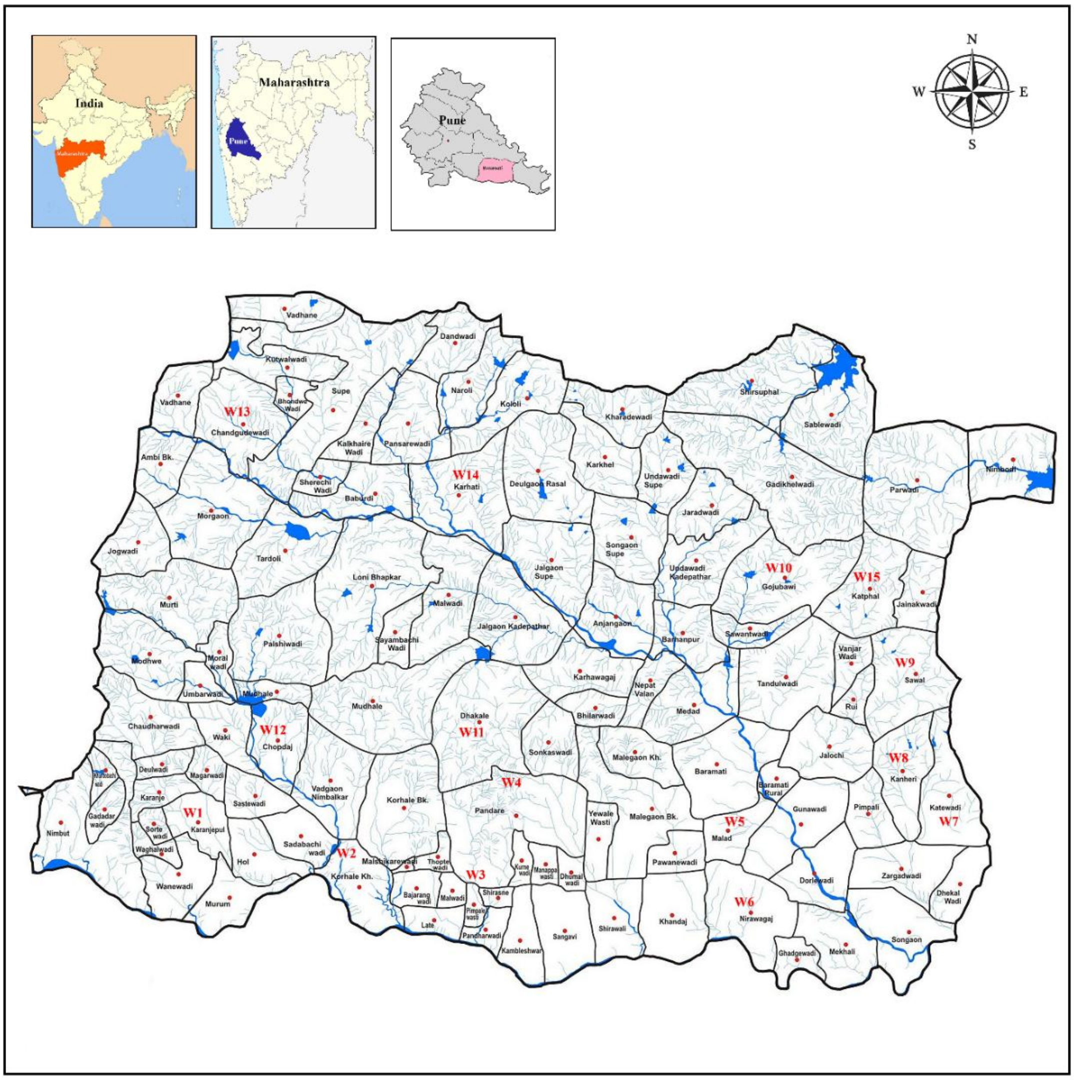


Figure 1: Study area map showing the location of the sampling sites

Physico-chemical parameters like pH, Electrical conductivity (EC), Total dissolved solids (TDS), Total Hardness (TH), cations: Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, anions: 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 a 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 (F^{-}) by SPANDS method (Method for fluoride determination involves the reaction of fluoride with a red zirconium-dye solution) 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 World Health Organisation (WHO) (1.5 mg/l) and Bureau of Indian Standards (BIS) [35] (1.0 mg/l). The chemical composition of water in diverse bodies of water changes with the seasons. As a result, it is vital to test the water on a regular basis in order to detect any minor changes in water quality. Seasonal changes in environmental circumstances were also known to be a key source of variation in water chemistry [36]. Given this, it was decided to collect the dug well water samples from fixed sampling stations twice a year covering pre-monsoon (summer) and post-monsoon (winter) seasons for two years.

Because of the creation of zirconium fluoride when fluoride (HF) combines with zirconium (SPANDS) solution (under acidic circumstances), the colour of SPANDS is bleached ($ZrF2-$). Because bleaching is caused by fluoride ions and is proportional to fluoride concentration, it follows Beer's rule in reverse [37-38]. A series of standard fluoride solutions were prepared by diluting the stock solution with distilled water. 100 ml volumetric flasks, numbering from 1 to 10 is taken and 1 to 10 ml standard sodium fluoride solution is added in the flask 1 to 10 respectively. Added 4 ml con. HCl and 1 ml Zirconium alizarin reagent in every flask, and diluted to 100 ml with distilled water. Mixed and allowed standing overnight. Absorbance was noted at 520 nm on a Bio Era's UV-Visible Spectrophotometer. The same procedure was used for the analysis of fluoride from groundwater. Compared the absorbance of groundwater samples with a standard curve and determined the fluoride concentration [39].

3. RESULTS AND DISCUSSION

The physicochemical composition of the groundwater samples for four seasons was analysed and the average results of various parameters are given in table 1 with Maximum, Minimum and Average values. The pH of the groundwater sample ranges from 6.63 to 8.32 with an average of 7.78, indicating 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 of 1777, which shows higher TDS in the study area and which is not suitable for drinking purpose. The maximum permissible limit recommended by BIS and WHO for TDS in drinking water is 500 mg/l. The TDS can be reduced by using RO system in purification of the water for drinking purposes.

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

Table 1: Average Physico-chemical data of the groundwater of Baramati Tehsil area, Pune, India (Winter 2015 to Summer 2017 Average Values)

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

Fluoride causes mottling of teeth, skeletal fluorosis, forward bending of the spinal column, distortion of knee joints and other portions of the body, and even paralysis when the concentration is high (paraplegia, quadriplegia). Mottled enamel is appears on the teeth of children who drink water containing too much fluoride during the period when permanent teeth are formed. A lower concentration of fluoride in a 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 preventing dental care. The degree of fluorosis generally increases as fluoride concentration increases above 2.4 mg/l [40-42]. The fluoride concentrations in all water samples in study area were within the permissible limit of WHO and BIS.

Table 2: Fluoride concentration of dug well water of Baramati Tehsil 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		

In the study area variations in fluoride in four seasons are observed, shown in figure 2. Natural water qualities fluctuate during the monsoon and summer seasons. Rainwater, by replenishing the aquifer and generating a dilution effect, can alter the geochemical characteristics. In the summer, however, the evaporation process causes a salt concentration in the natural water. During the summer, drying clay minerals above the water table causes oxidation, which improves mineral solubility by infiltrating water during the monsoon season [43-44].

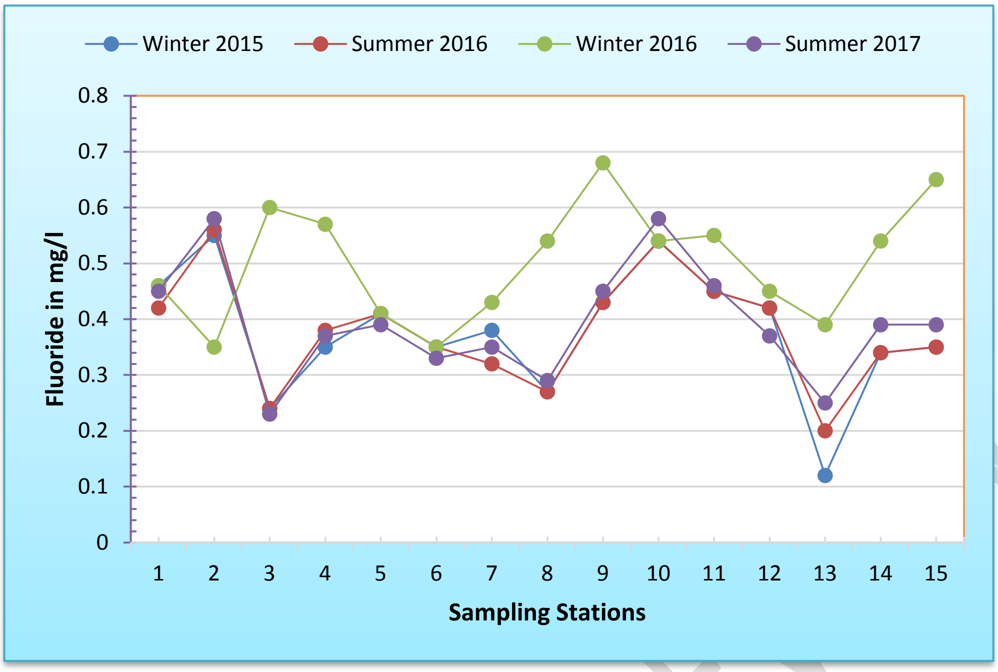


Figure 2: Spatio-temporal variation in Fluoride of groundwater from Baramati Tehsil area, Pune, India

Table 3: Correlation coefficient (r) among various groundwater parameters of samples from Baramati Tehsil 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

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 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 water parameters 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].

In the study area TDS showed moderate correlation with Ca^{2+} (0.66) and strong correlation with Mg^{2+} (0.79), Na^{+} (0.87), Cl^{-} (0.82), SO_4^{2-} (0.77) and TH (0.79). Ca^{2+} showed strong correlation with Mg^{2+} (0.87), Cl^{-} (0.81), SO_4^{2-} (0.77), TH (0.91) and moderate correlation with Na^{+} (0.47). Mg^{2+} showed strong positive correlation with Na^{+} (0.69), Cl^{-} (0.93),) and SO_4^{2-} (0.92).

The relation between F^{-} and other chemical parameters provide significant geochemical information and also help to know the controlling factors and its mechanism of F^{-} enrichment in the groundwater [47-48]. The correlation of fluoride with other parameters is shown in figure 3 to figure 9. F^{-} concentration shows a positive correlation with pH (0.17) (Table 3 and figure 3).

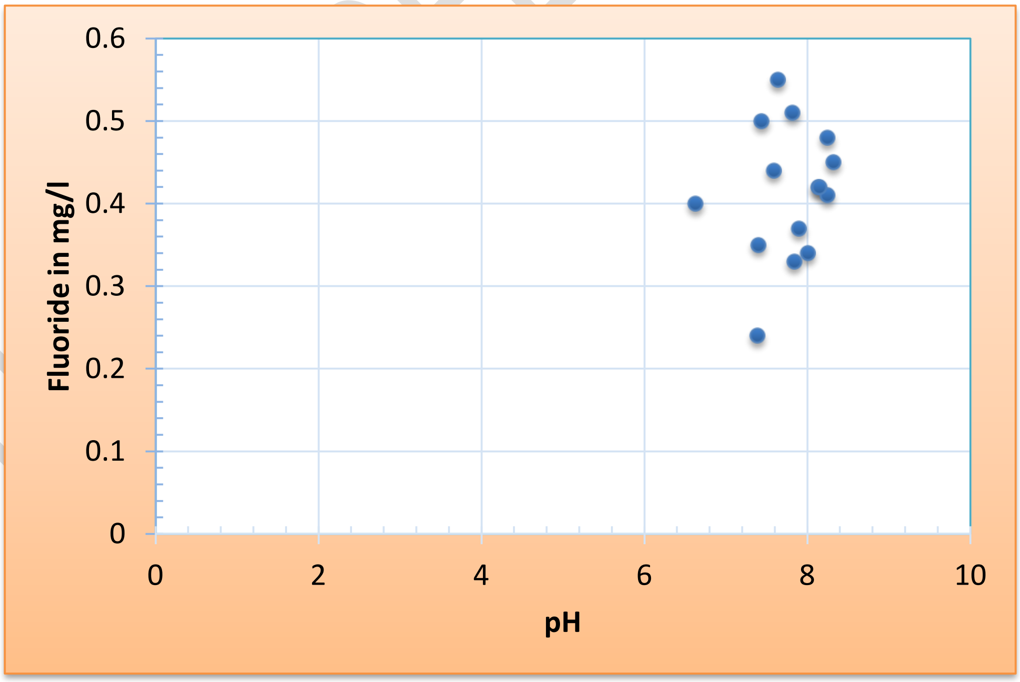


Figure 3: Correlation between Fluoride and pH of Groundwater samples from Baramati Tehsil area

Figure 3 indicates that the higher alkaline nature of water accelerates the enrichment of F^- concentration and thus typically affects the concentration of F^- in the groundwater [49-53]. A significant positive correlation is noticed between F^- with HCO_3^- (0.07) (Table 3 and figure 4), which declares that the alkaline environment is the dominant controlling chemical mechanism for leaching of F^- from the fluoride bearing minerals in the groundwater of the study area [54-55].

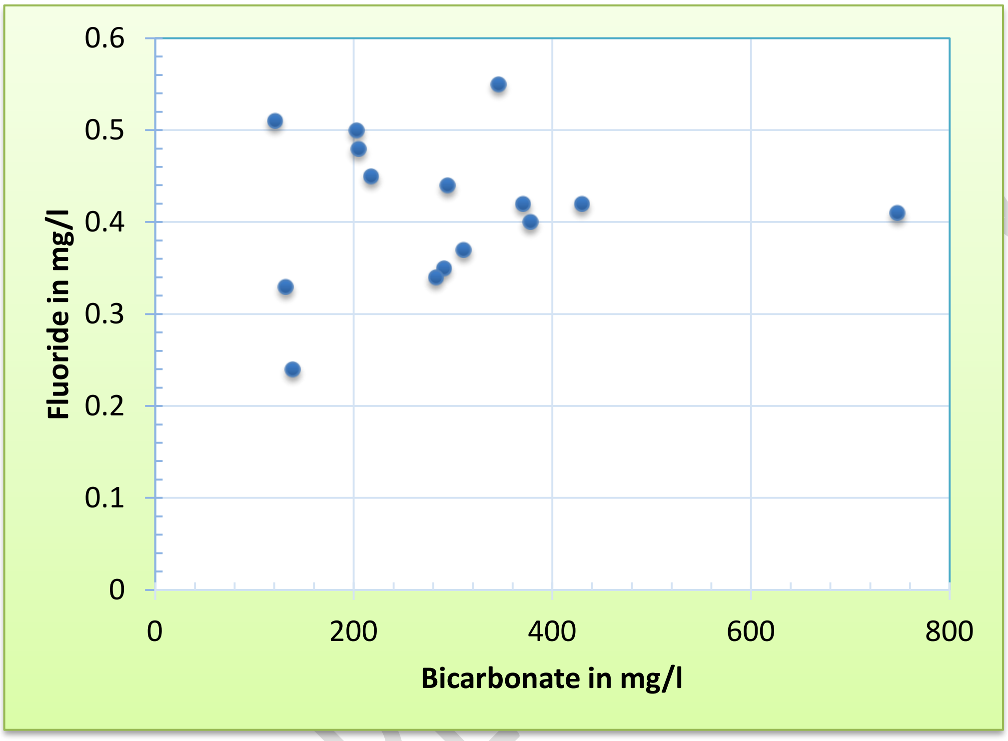


Figure 4: Correlation between Fluoride and Bicarbonate of Groundwater samples from Baramati Tehsil area

Moreover, as shown in Table 3 and Figure 5, the correlation of F^- and Ca^{2+} (0.02), clearly indicates that high Ca^{2+} content favored low F^-. The positive correlation of F^- is also observed with K^+ (0.060) (Table 3 and figure 6). It is observed that the major role of the precipitation process is a vital mechanism for the enhancement of F^- occurrence in groundwater [56]. The negative correlation of F^- observed with Na^+ (-0.15), Cl^- (-0.12) (Table 3 and figure 7) and Mg^{2+} (-0.10), SO_4^{2-} (-0.10) (Table 3 figure 8) is an agreement as established by Reddy et al. [57].

A number of research have shown that favourable connections between F- and pH, HCO3- and Na+, as well as an inverse link between F- and Ca2+, which indicates the concentration in fluorite saturated groundwater, often accelerate the F- concentration in groundwater [58]. EC and TDS correlation with F^- are not positive like pH, HCO_3^-, and Ca^{2+} in the study area (table 3 figure 9), which divulges that a higher affinity of F^- with pH and HCO_3^- rather than EC and TDS. Other studies, on the other hand, show that high EC and TDS concentrations are invariably associated with higher F- concentrations, with identical findings in different locales [59-60].

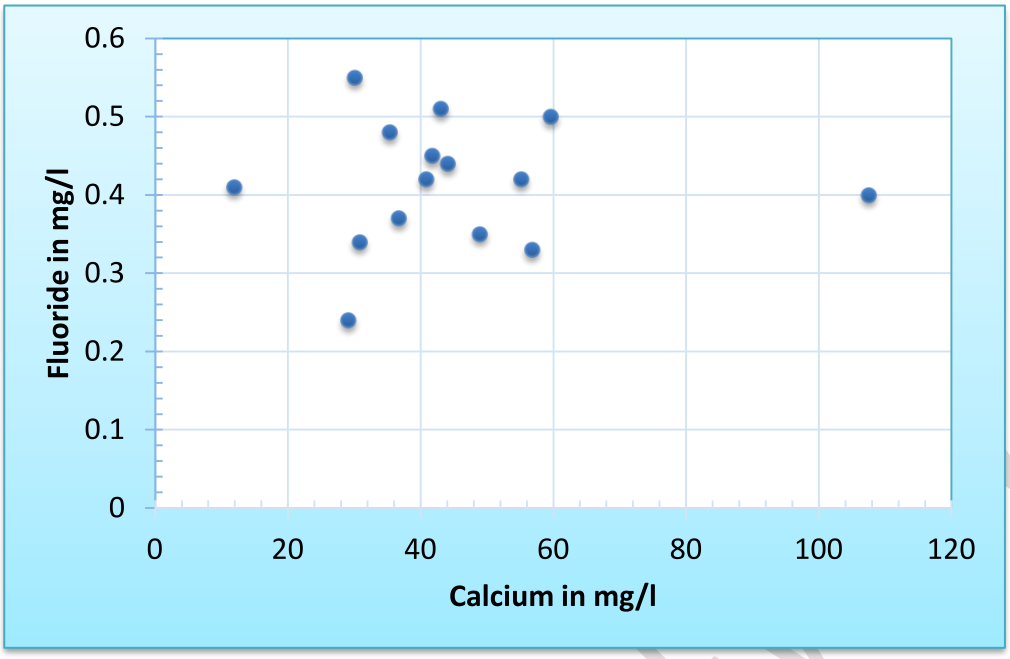


Figure 5: Correlation between Fluoride and Calcium of Groundwater samples from Baramati Tehsil area

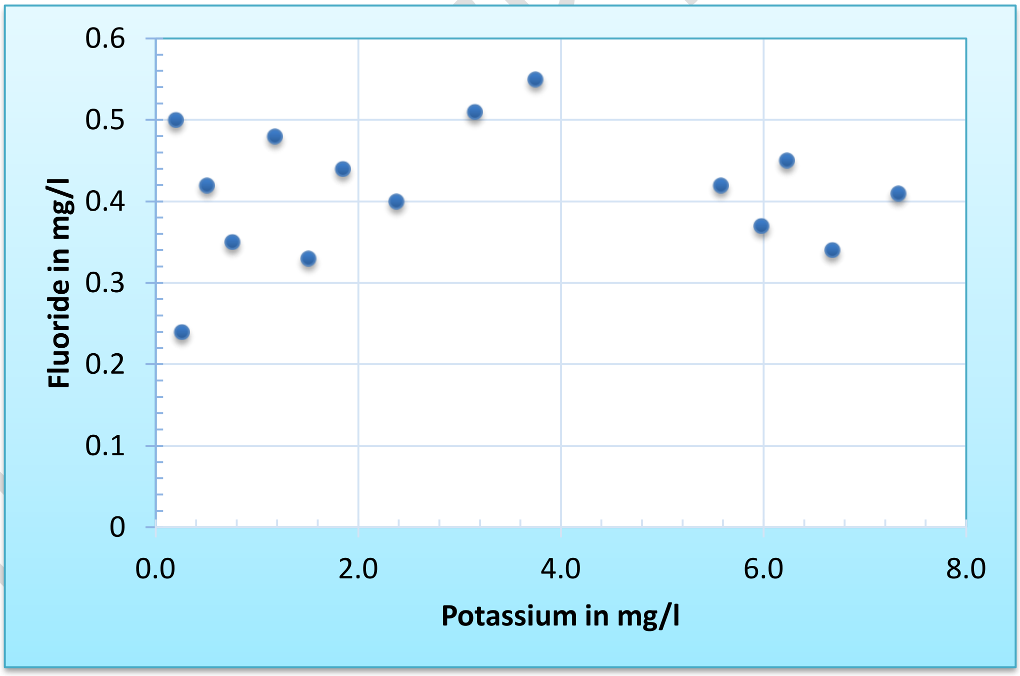
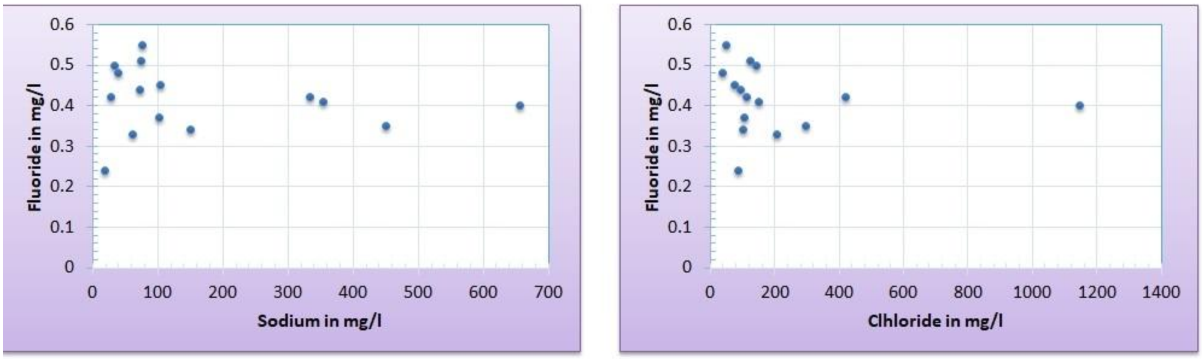
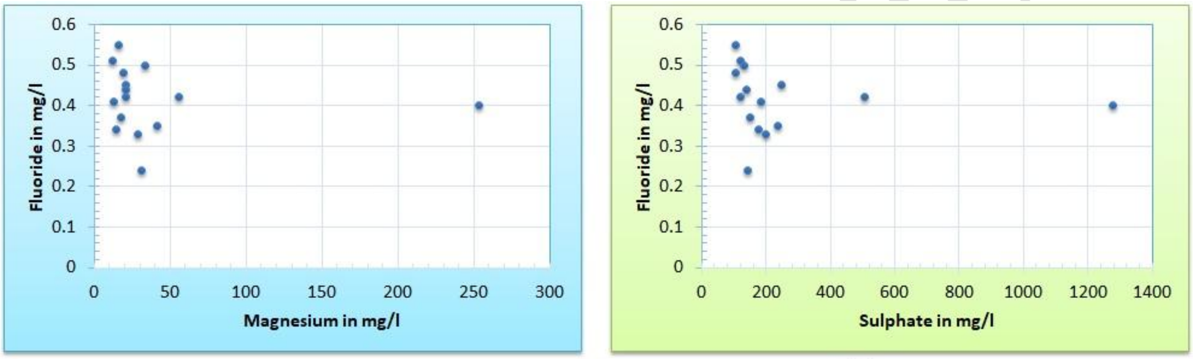


Figure 6: Correlation between Fluoride and Potassium of Groundwater samples from Baramati Tehsil area



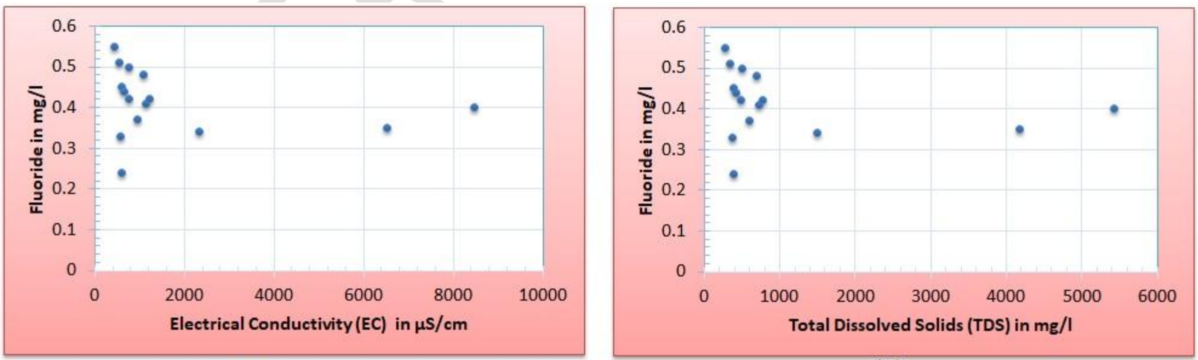
(A) (B)

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



(A) (B)

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



(A) (B)

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

4. CONCLUSION

Fluoride concentrations in the study area range from 0.12 to 0.68 mg/l, showing that fluoride levels in the Baramati Tehsil area were below WHO's permitted limits (1.5 mg/l) and BIS (1.0 mg/l). It indicates that the fluoride concentration does not have any health hazards due to the consumption of groundwater. But the lower concentration of fluoride in certain areas causes tooth decay. The intake of drinking water with fluoride content less than 0.5 mg/l can cause tooth decay. The TDS in the study area varies from 282 to 5422 mg/l. The majority of the research area's TDS is unfit for human consumption. The TDS in drinking water can be lowered by utilising a RO system to purify the water.

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