Spatiotemporal distribution of Fluoride in groundwater of five South-West districts of Haryana, India.

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ABSTRACT

The fluoride concentration and other water quality parameters were analyzed in ground water of five south-west districts of Haryana namely Rohtak, Jhajjar, Rewari, Mahendergarh and Bhiwani, where it was used as a source of drinking water. Two sets of total 362 samples were collected in pre-monsoon and post-monsoon seasons. The range of fluoride in the area during pre-monsoon and post-monsoon were 0.03-7.7 mg/L^{-1} and 0.02-9.97 mg/L^{-1}, respectively. The World Health Organization(WHO) regards a concentration of more than 1.5 milligrams per liter as the threshold level. The number of samples which had values higher than the threshold in pre-monsoon and post-monsoon was 126 and 119, respectively. The main source of such level of fluoride is geogenic as the study area is having granite as bedrock, which is a fluoride bearing mineral. The spatial distribution of fluoride showed high concentration in major parts of Bhiwani and minor parts of Jhajjar and Rohtak districts of Haryana. The temporal analysis of the data revealed that there was decrease in the concentration of fluoride due to monsoon rainfall infiltration in all districts except in central part of Jhajjar, which showed an increase. This can be attributed to high soil pH of the area favoring dissolution of fluoride from the parent soil material. To avoid the health impact of fluoride contamination, suitable intervention is required in these districts.

Keywords- Ground water, fluoride, spatial distribution, temporal distribution, Haryana.

1. INTRODUCTION

The one common problem in many areas throughout the globe is groundwater contamination consequential from existence of high fluoride in groundwater [1]. As per the report of World Bank Report 2012, nearly 85% population of India relies on the groundwater for drinking, therefore groundwater quality becomes an important issue. In India, the excessive fluoride in groundwater was noticed in 177 districts covering 21 states, affecting 62 million people, including 6 million children [2]. In Haryana Rewari, Mahendergarh, Hisar, Fatehabad, parts of Gurgaon and Faridabad districts were put on fluoride red alert (Geological Survey of India, 2003).

The presence of limited amount of fluoride is necessary (0.6- 1.0 mg/l) to prevent dental caries, however excess consumption of fluoride causes fluorosis (>1.20 mg/l), so it is crucial to have ideal fluoride concentration in drinking water from 0.6 – 1.20 mg/l. The study was undertaken to assess the water quality parameters in five south-west districts of Haryana, where ground water is still used as a source of potable water, hence had been selected for the study.

2. Materials and Methods

2.1 Study area

The present research work was carried out in five districts of Haryana viz. Rohtak, Jhajjar, Rewari, Mahendergarh, and Bhiwani. The geographical location of the area is in between Latitude 27^o 799' N- 29^o 087' N and longitude 75^o 474' E – 76^o958' E covering an area of 7304.93 km^2(Figure-1) and the climate is arid to semi-arid with hottest months May and June and coldest months are December and January. The mean annual rainfall of area is 524 mm and southwestern monsoon is the primary source of rainfall [3].

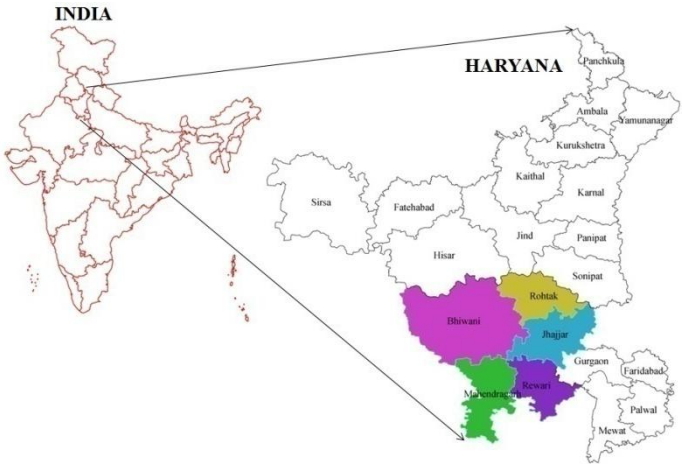

Fig-1: Study area map

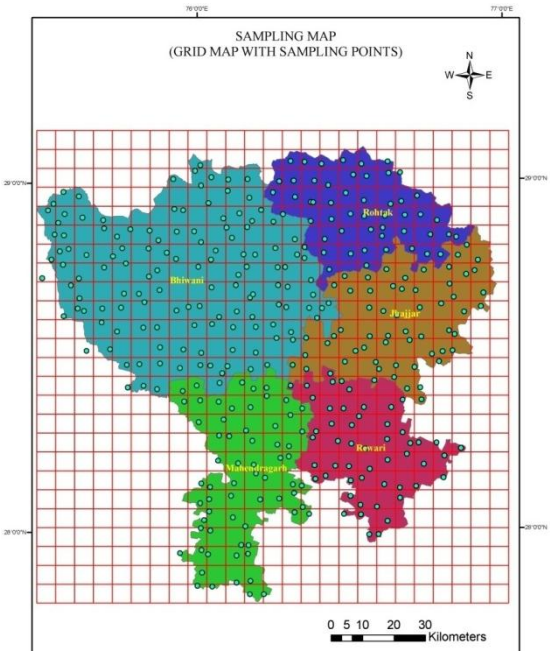

Fig- 2: 6x6 Km Grid map with sampling site.

2.2 Sampling of Groundwater

For sampling, whole area was divided into 6 x 6 Km grids and one sample of ground water was taken from a water source nearest to the centroid of each grid (Figure 2). The water samples were collected from drinking water sources (bore well/hand pump) at 362sites in consecutive pre-monsoon and post monsoon season of year 2017. The co-ordinates of the sampling sites were recorded using GPS (Garmin e Trex 20x). The samples were collected in high-density polyethylene (HDP) bottles of 1 L capacity. Before sampling, hand pump/bore well was pumped/switched on for approximately10 minutes to tap ground water and then HDP bottles were rinsed minimum three times with ground water before cramming with the sample. Some of the parameters were recorded at site only and for rest the bottles were carried to the laboratory and stored at room temperature till the completion of analysis.

2.3 Analysis of water parameters

The various physio-chemical parameters were analyzed as pH, electrical conductivity (EC), total dissolved solids (TDS), nitrate and salinity (*in- situ*) and total hardness, calcium hardness, magnesium hardness, alkalinity, fluoride, chloride, sulphate, phosphate, carbonate, bicarbonate (*ex-situ*). The parameters were analyzed as per standard methods of APHA,1995[4]. Fluoride was estimated by SPANDS method; nitrate by portable Hanna (HI 96728) nitrate meter, sulphate by gravimetric method, phosphate by stannous chloride colorimetric method, chloride by Volhard method, alkalinity by HCl titrimetric method, total and calcium hardness by EDTA titrimetric method and magnesium was calculated from total and chemical hardness.

Suitable statistical techniques and graphical representations were used for analysis. All the experiments were carried out in triplicate and the results were reproducible within ±5%error limit. Spatial analyst modeling tool (ArcGIS 10.3) was used to make thematic map of fluoride concentration.

3. RESULT AND DISCUSSION

The collected ground water samples had no color, odor, and turbidity. The data of various physio-chemical parameters of the collected samples compared to WHO/BIS standards was given in Table 1.

Table.1. Physio-chemical parameters of groundwater of Rohtak, Jhajjar, Rewari, Mahendergarh and Bhiwani districts of Haryana.

Water quality parameters	WHO/ BIS*		Pre-monsoon			Post-monsoon			% exceeding above allowable limit	
	Most Desirable	Max. allowable limit	Range	Median	Mean	Range	Median	Mean	Pre-monsoon	Post-monsoon
Rohtak (N=53)										
pH	6.5-8.5	9.2	5.9-7.9	7.2	7.2	7.0-7.9	7.3	7.4	-	-
TDS (mg L^{-1})	600	1000	162-7208	1352	1839	132-5585	937	1289	50	45
EC (µS cm^{-1})	-	1500	323-14416	2704	3679	254-11147	1968	2616	74	57
TH (mg L^{-1})	300	600	18.2-2765	57.9	240	15.5-1426	57.4	162.6	12	8

Ca^{2+}(mg L^{-1})	75	200	12.9-83.9	40.6	41.3	7.6-747.7	44.3	83.2	-	8
Mg^{2+} (mg L^{-1})	50	150	0-2721	8.1	198.9	0.13-919.7	14.2	79.4	16	9
HCO_3^- (mg L^{-1})	-	300	12.5-96	46.5	46.4	9.1-289.6	39.3	57.3	-	-
Cl^- (mg L^{-1})	250	1000	51.1-11542	384.3	829.1	38.1-4346	206.3	395.6	16	6
SO_4^{2-} (mg L^{-1})	150	400	1-3748	147.7	433.6	2.8-1470	96.2	223.8	32	21
PO_4^{3-} (mg L^{-1})	-	-	<0.1-0.19	0.08	0.08	<0.1-41	<0.1	1.5	-	-
NO_3^- (mg L^{-1})	45	-	<0.5-57.8	3.6	8.8	<0.5-43.2	2.1	6.45	2	-
F^- (mg L^{-1})	1.2	1.5	0.14-2.5	1.31	1.26	0.12-9.97	0.85	0.96	34	4
Jhajjar (N=60)										
pH	6.5-8.5	9.2	6.5-8.3	7.3	7.3	6.9-9.1	7.5	7.5	-	-
TDS (mg L^{-1})	600	1000	170.9-4158	1542	1740	204-3800	1135	1365	67	55
EC (µS cm^{-1})	-	1500	341.9-8316	3086	3484	391-7600	2230	2732	70	63
TH (mg L^{-1})	300	600	22.1-2203	131.3	231.6	20.5-2436	108.0	307.4	8	15
Ca^{2+}(mg L^{-1})	75	200	22.1-552.1	89.4	110.4	20.5-1237	86.6	188.7	12	19
Mg^{2+} (mg L^{-1})	50	150	0-1862	22.5	113.8	0-1656	24.3	118.7	17	16
HCO_3^- (mg L^{-1})	-	300	24.5-555.8	95.6	112.8	28.7-894.6	94.7	171.9	3	15
Cl^- (mg L^{-1})	250	1000	142.9-5126	925.9	1483	47.9-2166	372.5	564.0	48	18
SO_4^{2-} (mg L^{-1})	150	400	55.5-1059	244.2	332.4	24.7-784.5	123.9	185.9	32	8
PO_4^{3-} (mg L^{-1})	-	-	<0.1-0.92	0.15	0.17	<0.1-0.59	0.05	0.07	-	-
NO_3^- (mg L^{-1})	45	-	1.3-290.7	9.8	26.3	0-97.7	5.3	14.8	15	13
F^- (mg L^{-1})	1.2	1.5	0.5-2.1	1.2	1.2	<0.05-7.3	0.8	0.9	27	8
Rewari (N=49)										
pH	6.5-8.5	9.2	6.1-8.1	7.5	7.5	7.03-7.9	7.6	7.6	-	-
TDS (mg L^{-1})	600	1000	201-7336	1147	1578	191-5729	981	1309	57	49
EC (µS cm^{-1})	-	1500	402-14672	2295	3155	376-11450	1953	2608	65	63
TH (mg L^{-1})	300	600	46-3462	229	501	44-2194	218	431	29	29
Ca^{2+}(mg L^{-1})	75	200	35-483	116	123	44-987	149	250	4	39
Mg^{2+} (mg L^{-1})	50	150	0-3352	112	378	0-2097	76	182	43	35
HCO_3^- (mg L^{-1})	-	300	33-478	119	130	22-666	129	197	2	16
Cl^- (mg L^{-1})	250	1000	1-3430	390	584	41-2178	243	406	20	12
SO_4^{2-} (mg L^{-1})	150	400	1-1260	104	209	19-703	97	135	16	8
PO_4^{3-} (mg L^{-1})	-	-	<0.1-0.32	0.26	14.395	<0.1-0.55	0.12	0.01	-	-
NO_3^- (mg L^{-1})	45	-	<0.5-27	7.2	8.5	<0.5-25	4.0	5.6	-	-
F^- (mg L^{-1})	1.2	1.5	<0.05-2.7	0.45	0.6	0-1.1	0.2	0.3	6	-
Bhiwani (N=137)										
pH	6.5-8.5	9.2	6.8-8.5	7.3	7.4	6.9-8.4	7.2	7.3	-	-
TDS (mg L^{-1})	600	1000	135-7336	1060	1366	133-6458	981	1225	59	50
EC (µS cm^{-1})	-	1500	270-34296	2146	2868	266-12916	2000	2560	73	66
TH (mg L^{-1})	300	600	43-2348	272	405	22-2116	256	355	25	18
Ca^{2+}(mg L^{-1})	75	200	22-1555	187	283	19-1145	184	256	55	42
Mg^{2+} (mg L^{-1})	50	150	0-1014	187	125	0.9-970	47	99	34	22
HCO_3^- (mg L^{-1})	-	300	3.6-1238	140	196	15-1150	110	176	19	15
Cl^- (mg L^{-1})	250	1000	39-2174	336	438	12-2015	205	322	9	2
SO_4^{2-} (mg L^{-1})	150	400	20-1033	160	233	12-947	138	170	21	9
PO_4^{3-} (mg L^{-1})	-	-	<0.1-0.24	0.1	0.07	0-0.21	0.1	0.07	-	-
NO_3^- (mg L^{-1})	45	-	0-130	5.9	10.4	0-59.6	5.1	7.8	4	3
F^- (mg L^{-1})	1.2	1.5	0.03-7.7	1.3	1.9	<0.02-7.1	1.03	1.3	49	23
Mahendergarh (N=63)										
pH	6.5-8.5	9.2	6.9-8.3	7.6	7.6	6.5-8.34	7.4	7.4	-	-
TDS (mg L^{-1})	600	1000	319-2456	1076	1136	316-2294	950	1117	54	46
EC (µS cm^{-1})	-	1500	638-4912	2152	2272	632-4588	1900	2216	25	21
TH (mg L^{-1})	300	600	48-4045	227	376	57-882	242	313	14	11
Ca^{2+}(mg L^{-1})	75	200	12-373	107	119	0.249-613	115	155	2	16
Mg^{2+} (mg L^{-1})	50	150	0-3672	111	264	0.2-538	92	158	40	48
HCO_3^- (mg L^{-1})	-	300	3.7-228	104	112	4-397	101	129	-	5
Cl^- (mg L^{-1})	250	1000	45-1085	255	354	43-1046	271	344	3	3
SO_4^{2-} (mg L^{-1})	150	400	6.9-292.1	80.6	101	5-443	82	107	-	2
PO_4^{3-} (mg L^{-1})	-	-	<0.04-0.445	0.1	0.12	<0-0.4	0.1	0.11	-	-
NO_3^- (mg L^{-1})	45	-	0.28-116.6	4.6	9.56	0.2-101	4.6	8.6	3	3
F^- (mg L^{-1})	1.2	1.5	0.05-3.26	0.84	0.976	<0.04-3.1	0.7	0.8	13	14

3.1 Fluoride

The level of F^-concentration in ground water samples of Rohtak, Jhajjar, Rewari, Mahendergarh and Bhiwani districts in pre-monsoon varied from 0.14-2.5mg/L,0.5-2.1 mg/L, <0.5-2.7 mg/L, 0.05-3.26 mg/L and 0.03-7.7 mg/L with mean values of 1.26 mg/L, 1.20 mg/L, 0.60 mg/L, 0.98 mg/L and 1.90 mg/L (Table. 1), respectively. In post monsoon season, the F^-concentration in these districts ranged from 0.12-9.97 mg/L, <0.05-7.3 mg/L, 0.1-1 mg/L, <0.04-3.1 mg/L and <0.02-7.1 mg/L with mean values of 0.96 mg/L, 0.90 mg/L, 0.30 mg/L, 0.80 mg/L and 1.30 mg/L respectively. It had been observed that in Rohtak, Jhajjar, Rewari, Mahendergarh and Bhiwani districts 34%, 27%,6%, 13%, and 49% samples had higher concentration of F^-respectively than WHO/BIS standard limit of 1.5 mg/L during pre-monsoon, whereas during post monsoon 4%, 8%, 14% and 23%of samples from Rohtak, Jhajjar,

Mahendergarh, and Bhiwani respectively were exceeding the WHO/BIS standard limit. However, in Rewari district post monsoon all samples were within WHO/BIS standard limit.

The cumulative percentage of samples lying between 0.51-1.5 mg/L was 70.68% for the pre-monsoon and 86.70% for post monsoon season in the study area (Figure-3). The mean value of F^- concentration was found to be higher in pre-monsoon as compared to post monsoon season (Figure-4).The trend of decrease in F^-concentration in ground water during the post-monsoon was due to rain fall infiltration and similar results were also reported in many studies [5,6,7].

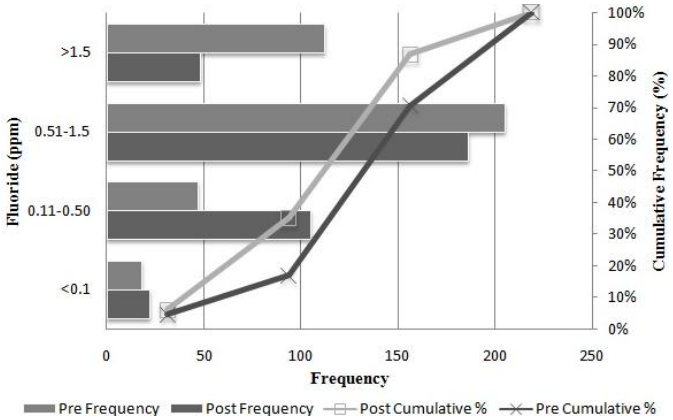

Fig-3: The frequency and cumulative frequency distribution of fluoride in pre-monsoon and post-monsoon season of Rohtak, Jhajjar, Rewari, Mahendergarh and Bhiwani district.

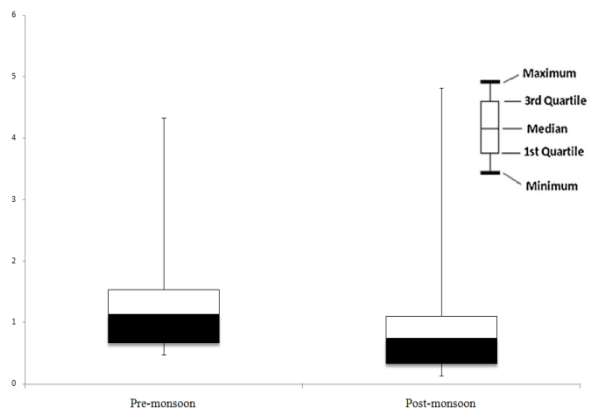

Fig-4:Box and whisker Plot showing fluoride concentration in Rohtak, Jhajjar, Rewari, Mahendergarh and Bhiwani districts.

3.2 Other water quality parameters

The mean value of pH for the ground water samples of all five districts ranged between 7.2-7.6. A small increase of mean pH was observed in the post monsoon season. According to WHO the standard range for pH is 6.5 to 8.5, so it was found that the water was slightly alkaline that favored the dissolution of fluoride bearing minerals. The EC is important indicator of TDS and salinity on which taste of water depends [8]. The EC in the ground water samples of the study area ranged between 270-34296µS/cm during the pre-monsoon and 254-12916µS/cm in post-monsoon season. The mean values of pre-monsoon samples were 3679 µS/cm, 3484 µS/cm, 3155 µS/cm, 2275 µS/cm, and 2868 µS/cm in Rohtak, Jhajjar, Rewari, Mahendergarh, and Bhiwani district respectively and the corresponding values for post-monsoon sampling were 2616 µS/cm, 2732 µS/cm, and 1953 µS/cm, 2216 µS/cm and 2560 µS/cm respectively. The higher mean values observed in pre-monsoon samples could be due to the concentration of salts due to surface evaporation effect in the hot pre-monsoon season [9]. The decrease in the EC during post-monsoon sampling suggested a dilution effect of rainwater. Same trend was followed by TDS and salinity as it is directly proportional to EC. The percentage of TDS of samples exceeding the allowable limit were very high being 50% in pre-monsoon and 45% in post-monsoon in case of Rohtak, the corresponding values for pre and post monsoon were 69% and 55% for Jhajjar, 57% and 49% for Rewari, 54% and 46% for Mahendergarh and 59% and 50% for Bhiwani district respectively. Approximately half of the samples analyzed were having higher concentration of TDS throughout the year, so consumption of this water may cause cardiovascular problems and kidney stones in local population [10].This also leads to metallic taste of water and made pipes more prone to corrosion. Among the other anions analyzed in the sample, chloride was found to be dominating followed by sulphate. The mean values of chloride during pre-monsoon were 829 mgL^{-1}, 1483 mgL^{-1}, 584 mgL^{-1}, 354 mgL^{-1}, and 438 mgL^{-1} for Rohtak, Jhajjar, Rewari, Mahendergarh, and Bhiwani districts, respectively. The same trend of decreasing concentration was found in the post-monsoon and the mean values being reduced to 396 mgL^{-1}, 564 mgL^{-1}, 406 mgL^{-1}, 322 mgL^{-1} and 322 mgL^{-1} for Rohtak, Jhajjar, Rewari, Mahendergarh, and Bhiwani. In sulphate the mean values for Rohtak, Jhajjar, Rewari, Mahendergarh and Bhiwani during pre-monsoon were 434, 332, 209, 101, and 233 mgL^{-1} and in post-monsoon decreased to 224, 186, 135, 107, and 170 mgL^{-1}. In case of nitrate the number of samples exceeding WHO limit of 45 mgL^{-1}, were 2% (Rohtak), 15% (Jhajjar), 3% (Mahendergarh), and 4% (Bhiwani) in pre-monsoon whereas, in post-monsoon 13% (Jhajjar), 3% (Mahendergarh), and 3% (Bhiwani) samples exceeded WHO limit. All the samples of the study area analyzed for phosphate were found to be under standard limits set by the WHO.

The samples having higher values of EC were also found to be having higher concentrations of calcium, magnesium, and bicarbonate ions. In general, it was concluded that these samples belong to the category of hard water.

3.3 Spatial distribution of Fluoride

For the generation of spatial distribution map, the spatial analyst modeling tool (ArcGIS 10.3) was used. This modelling tool predicted unknown value of F^- concentration from known point on the geographical area based on spatial autocorrelation/Inverse distant weightage (IDW) interpolation method after generating raster surface. The temporal and spatial distribution of F^- in groundwater has been shown in Figure- 5.

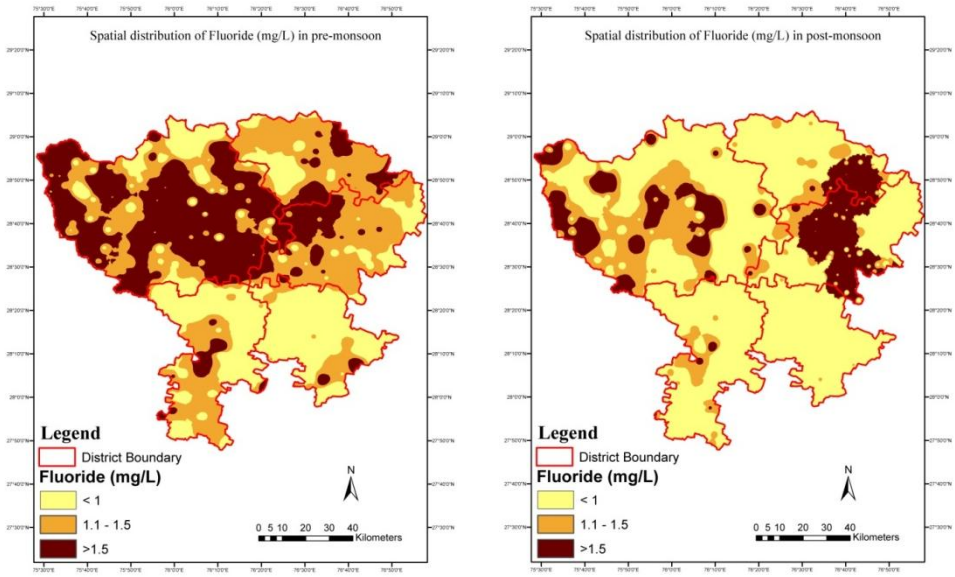

Fig.-5:Spatial distribution of F^- in pre-monsoon and post-monsoon season.

The generated map showed an elevated level of F^- towards the north-west portion (Rohtak, Bhiwani, and Jhajjar) of the study area and highest being in Bhiwani district. The higher levels of F^- ions in groundwater was majorly attributed to the lithology and geology of the area. As per Geological Survey of India 2012 report [11] the bedrock of the study area consists of granite. Granitic bedrock is the most common source of the F^- [12,13] and the range of F^- concentration is 500-1400 mg/kg [14,15] Major fluoride minerals in these rock type are fluoroapatite [$Ca_5(PO_4)_3F$], fluorite [CaF_2], biotite [$K(Mg,Fe)_2(AlSi_2O_{10})(F,OH)_2$] [16,17,18] etc. These minerals are easily soluble in water, hence higher concentration of fluoride was observed in groundwater that interacts with them during percolation [19].

The maps also clearly depicted that concentration of F^-decreased in post-monsoon as compared to pre-monsoon which was due to dilution with rainwater infiltration during monsoon season [20, 21, 22]. However, in central part of Jhajjar the F^- concentration increased in post-monsoon season. This increase can be attributed to high pH of ground water in central part of Jhajjar (> 8.0) as alkaline pH is more favorable for fluoride dissolution from soil profile [23].

4. CONCLUSION

Fluoride analysis in ground water had been done in 362 samples among which 126 (pre-monsoon) and 119 (post-monsoon) samples had F^- level above standards limits. In spatial analysis, where the bed rock is granite type the concentration of F^- had been found higher. In temporal analysis it was found that due to monsoon rainfall infiltration F^- concentration decreased. The F^-concentration had been positively correlated with pH, total alkalinity, chloride, nitrate, sulphate, TDS and negatively with total hardness.

The range of fluoride in the study area varied between 0.03-7.7 mg/L in pre-monsoon and 0.04-9.97 mg/L in post-monsoon season. Some of the areas of Mahendergarh, Bhiwani, Jhajjar and Rohtak had very high levels of fluoride in the ground water throughout the year, so may lead to a substantial increase in prevalence of dental fluorosis among children and prolonged exposure leads to osteo/skeletal fluorosis among people after the age of 30 years.

Water de-fluoridation should be a priority in those areas and suitable treatment of groundwater may be targeted to improve water quality.

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