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Groundwater quality assessment with focus on fluoride in different parts of Haryana State, India.

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Abstract: A systematic physico-chemical assessment of the groundwater at eight different locations in Haryana, India has been taken up to evaluate its suitability for drinking purpose. The data revealed considerable variations in the water samples with respect to chemical composition. The samples at eight sites were collected in triplicate at 3 different depth levels. Overall water quality was found unsatisfactory for drinking purposes. Some parameters viz. Hardness, Chloride and Fluoride were above the permissible limits of drinking water standards. The fluoride concentration varies from 0.27 mg/l to 2.87 mg/l. All the samples at the depth > 40 m bgl exceeded the BIS standard prescribed limit of 1.0 mg/l fluoride in drinking water. Fluoride contamination increases as we move from shallow to deep aquifers. Among the study area, 46 percent samples have more than maximum permissible limit (1.5 mg/l) of fluoride concentration. High concentration of fluoride in underground water may be attributed to geological (fluoride-bearing rocks) and anthropogenic (brick-kiln industries, phosphatic fertilizers) environment of the area. Sustainable and economic treatment techniques and good diet (calcium & vitamin c enriched) can control the adverse effects of fluorosis epidemic in this region.
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1. Introduction

For drinking and irrigation, people of Haryana are dependent upon ground water resources to a great extent due to less availability and non- acceptance of surface water. In many areas of the state, ground water is the only source for drinking water. Gupta and Mishra, 2014 stated more than 75 % of Haryana's population depends on ground water for domestic needs. In such places the major health problems are the result of inorganic chemicals contamination, poor sanitary conditions and illness brought about by disease causing organisms. Underground aquifers are more vulnerable to degradation as it remains as unusable or even hazardous condition for decades or centuries. The typically low velocity of groundwater prevents it from dilution; henceforth, a contaminant zone may maintain a high concentration as it slowly moves from points of recharge to zone of discharge (Pattyjohns, 1979). One of the major contaminant is fluorine due to which, dental fluorosis has become a common disease. Nearly 200 million people from 25 nations are affected by the epidemic of fluorosis at global level. In India, the excessive fluoride in groundwater is noticed in 177 districts covering 21 states, affecting 62 million people, including 6 million children (Adimalla and Venkatayogi, 2018). Dental fluorosis in adults and children is very common in Haryana state and symptoms of skeletal fluorosis are prominent among adults in some districts having high fluoride concentration (Amanjeet et al, 2017).

In past decades, due to intensive farming practices, deforestation, low & erratic rainfall, declining surface water resources; the people are more dependent upon underground water resources in this agrarian state. Eventually, groundwater is becoming more vital water resource primarily for drinking, domestic, and other usages. Thus, dental and skeletal health problems become more endemic in this region (CGWB, 2017). Fluoride in groundwater mostly occurs because of the geomorphology of the aquifer below the groundwater. Most of the districts of Haryana have rock bed of quartzites, mica and clay, which is the preliminary case of fluoride existence in groundwater. Besides, it is also observed that at some sites, fluoride concentration varies from one to other. This may be attributed to the fluctuations in water table at those sites. (Gupta and Misra, 2014).

Permissible limit for F- concentration is 1-1.5mg/l according to health organizations (WHO, 2006). As per BIS, 2012 permissible limit of fluoride in potable water is 1.0 mg/l. A bibliographic survey has shown in Table 1, indicated a high concentration of fluoride in Haryana. Further, some researchers conducted work on prevalence of fluorosis among children (Haritash et al, 2018; Kumar et al, 2017; Yadav et al, 2009). They concluded that due to consumption of underground water school going children are affected by dental fluorosis. Fluoride (F-) concentration varied from place to place and mostly dependent on the geological and environmental factors. Till now, no efforts have been explored for variation in fluoride content at different depths. Henceforth, this study was conducted to assess the variation of water parameters with special focus on fluoride ion at different depths in central parts of Haryana. It will attract further research in investigating the fluorosis endemic problems in this region.

Table 1: Fluoride concentration in groundwater in different parts of Haryana, India.

Sr. No.	Author and Year	Study Area	Findings
1	Meenakshi et al. (2004)	Jind	0.3 – 6.9 mg/l
2	Rout and Sharma, 2011	Ambala	0.14 – 0.90 mg/l.
3	Singh, 2011	Dabwali	0.90 – 34.50 mg/l
4	Manjeet et al. 2014	Gurgaon	0.02 – 6.4 mg/l.
5	Ravish et al, 2019	Ambala / Yamunanagar	0.2 – 1.7 mg/l
6	Haritash et al, 2018	Hisar rural	0.05 – 2.4 mg/l
7	Kumar & Sharma, 2017	Hisar city	0.5 – 2.98 mg/l
8	Kumar, S. et al, 2017	Jhajjar	1.63 – 3.33 mg/l
9	Kumar, R. et al, 2017	Mahendergarh	0.6 – 5.1 mg/l
10	Bishnoi and Malik, 2008	Panipat	0.24 – 9.27 mg/l
11	Singh.S et al, 2012	Jind	0.11 – 2.93 mg/l
12	CGWB, 2017	Whole Haryana	0.1 – 30 mg/l

2. Material and Methods

Fig.1: Study area map (Haryana State, India).

2.1. Study Area:

Haryana is a major agricultural state of India with an area of 44,212 Km^2, and it has regional areas of high concentration of fluoride in groundwater throughout its stretch. The main sources of groundwater (hand-pump, tube-well, and submersible pump) in central parts of Haryana fulfill the domestic water needs of inhabitants. The whole Jind district and parts of Hisar, Karnal, Kaithal, Panipat, Sonepat and Rohtak represents the central parts of the state.

2.2. Collection and analysis of water samples:

Water samples from various drinking groundwater sources (hand pumps, open wells and tube wells) were collected from various sites of central parts of Haryana to determine groundwater quality. The sampling bottles were well rinsed before sampling and tightly sealed after collection and labeled in the field. Sampling was carried out without adding any preservative. The samples at each site were collected in triplicate at 3 depth levels viz. **a) < 10m bgl b) 10-40m bgl c) > 40m bgl.**

The physio-chemical analysis of water samples was carried out for various quality parameters such as pH, electrical conductivity (EC), total dissolved solids (TDS), total alkalinity (TA), total hardness (TH), Chloride, Nitrate and Fluoride as per the standard procedure described by the "Standard methods for the examination of water and wastewater - American Public Health Association (APHA, 2005)."

3. Results and discussion:

The underground water samples were colorless, odorless and without turbidity. Although most of the water samples collected were tasteless but it was slightly brackish at some of the locations due to hardness. The characterization of different parameters in sample sites at diverse depths is provided in Table-2.a and comparative analysis can be observed in Table-2.b. The results indicate that the quality of water varies considerably at different depths. It has been also concluded that the underground water quality varied geo-spatially also.

Table 2 a: Physico-chemical characteristics of ground water at different depths in sampling locations.

Parameters	**pH**	**TDS**	**EC**	**TA**	**TH**	**Cl^-**	**NO_3^-**	**F^-**
Sampling Sites		mg/l	µs/cm	mg/l	mg/l	mg/l	mg/l	mg/l
Salwan (Karnal)								
Depth < 10m bgl	7.95	1594	2390	420	580	180	12	0.27
Depth- 10-40m bgl	7.86	1149	1723	360	540	140	2.9	0.79
Depth > 40m bgl	7.8	834	1251	360	560	120	3.5	1.39
Mean	7.87	1192.33	1788	380	560	146.66	6.13	0.81
Didwara (Panipat)								
Depth < 10m bgl	7.97	1219	1828	550	240	164	3.1	0.86
Depth- 10-40m bgl	8.02	1013	1520	620	240	52	2.3	1.56
Depth > 40m bgl	7.8	1219	1828	530	380	280	2.4	1.66
Mean	7.93	1150.33	1725.33	566.66	286.66	165.33	2.6	1.36
Kaithal City								
Depth < 10m bgl	8.1	2493	3739	780	440	560	2.7	0.8
Depth- 10-40m bgl	7.9	1127	1690	400	200	380	1.9	0.94
Depth > 40m bgl	8.06	2541	3810	820	480	550	15.2	1.02
Mean	8.02	2053.66	3079.66	666.66	373.33	496.66	6.6	0.92
Jhanj Kalan (Jind)								
Depth < 10m bgl	8.35	419.5	629	88	200	80	1.2	0.71
Depth- 10-40m bgl	7.99	1671	2505	320	420	360	2.6	0.99
Depth > 40m bgl	8.32	2281	3420	440	440	560	2.2	2.62
Mean	8.22	1457.16	2184.66	282.66	353.33	333.33	2	1.44
Ashraf Garh (Jind)								
Depth < 10m bgl	7.82	3255	4880	472	680	800	7.1	0.78
Depth- 10-40m bgl	7.87	3097	4640	440	880	1200	6.4	0.9
Depth > 40m bgl	7.81	4389	6580	460	1720	2000	10.3	1.71
Mean	7.83	3580.33	5366.66	457.33	1093.33	1333.33	7.93	1.13
Dhani Garn (Hisar)								
Depth < 10m bgl	8.02	668	1001	320	200	48	1.8	2.02
Depth- 10-40m bgl	7.9	1447	2169	680	240	160	3.7	2.7
Depth > 40m bgl	7.94	2428	3640	450	720	680	3.8	2.74
Mean	7.95	1514.33	2270	483.33	386.66	296	3.1	2.48
Pauli (Rohtak)								
Depth < 10m bgl	7.54	1012	1517	240	560	156	3.4	1.61
Depth- 10-40m bgl	7.32	1842	2761	224	470	180	3.2	2.54
Depth > 40m bgl	7.58	1440	1646	256	584	378	3.5	2.87
Mean	7.48	1431.33	1974.66	240	538	238	3.36	2.34
Bali (Sonepat)								
Depth < 10m bgl	8.06	207	310	80	180	16	2.9	0.63
Depth- 10-40m bgl	7.94	1125	1686	440	300	280	2.7	0.81
Depth > 40m bgl	7.5	679	1018	260	220	140	2.1	1.92
Mean	7.83	670.33	1004.66	260	233.33	145.33	2.56	1.12

Table 2 b: Comparative chart of groundwater quality of sampling sites with water standards (BIS and WHO).

	Range of samples				BIS Standards		WHO Limits
Parameters	**Min.**	**Max.**	**Mean**	**Std. Dev.**	**Acceptable**	**Permissible**	**Min.-Max**
pH	7.32	8.35	7.89	0.22	6.5-8.5	6.5-8.5	6.5-8.5
TDS	207	4389	1631.22	956.46	500	2000	500
EC	310	6580	2424.20	1441.33	250	500	NA
T. Alkalinity	80	820	417.08	177.37	200	600	200
T. Hardness	180	1720	478.08	309.39	200	600	100
Chloride	16	2000	394.33	426.02	250	1000	200
Nitrate	1.2	15.2	4.28	3.2	25	45	25
Fluoride	0.27	2.87	1.45	0.73	1	1.5	1
Standard drinking water specification: Ref. BIS (IS: 10500:2012) & WHO, 2006.							
Values are expressed in mg/l except pH while EC is expressed in µs/cm.							

Table 2 c. Correlation matrix for different groundwater quality parameters.

	pH	**TDS**	**EC**	**T.Alkalinity**	**T.Hardness**	**Chloride**	**Nitrate**	**Fluoride**
pH	1	-0.018	0.0049	0.2542	-0.222	-0.002	-0.014	-0.348
TDS		1	0.9978	0.438	0.8291	0.9248	0.6085	0.0416
EC			1	0.4503	0.82	0.9218	0.6093	0.0147
T.Alkalinity				1	0.0865	0.2557	0.411	-0.0623
T.Hardness					1	0.8877	0.5637	0.0508
Chloride						1	0.5142	0.127
Nitrate							1	-0.2775
Fluoride								1

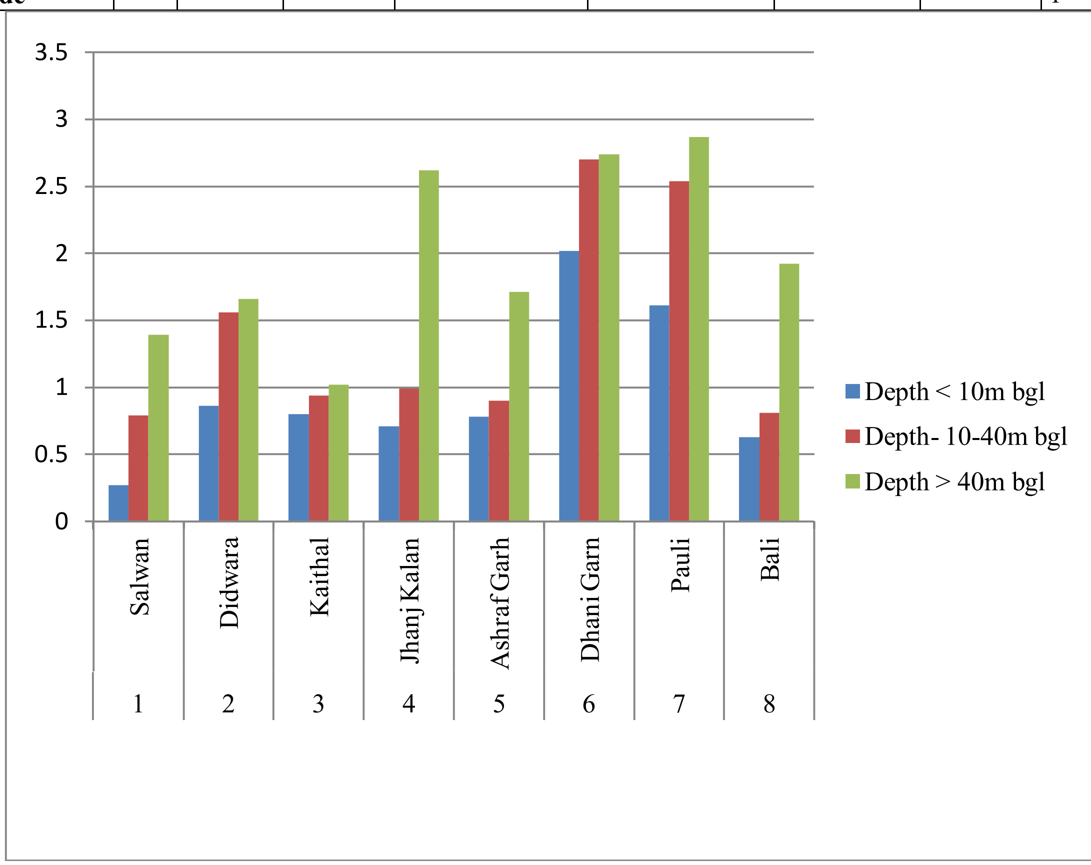

Fig. 2: Comparative graphical portray of fluoride concentration at different depths in all sites.

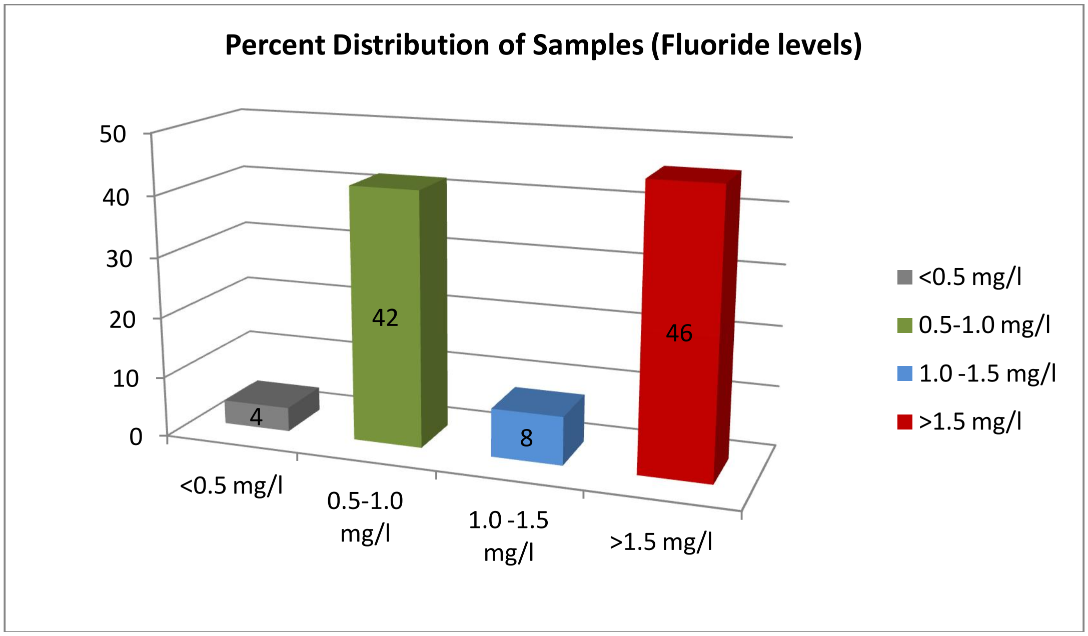

Fig. 3: Percent distribution of sampling locations according to F^- levels.

Table 3: Effects of Fluoride on Human Health (IPCS, 2002).

Sr. No.	Fluoride Concentration (mg/l)	Effects on Health
1	Nil	Limited growth and fertility
2	< 0.5	Dental caries
3	0.5 – 1.5	Promote dental health and prevent tooth decay
4	1.5 – 4.0	Dental Fluorosis (mottling and pitting of teeth)
5	4.0 – 10.0	Skeletal Fluorosis
6	> 10.0	Crippling Fluorosis

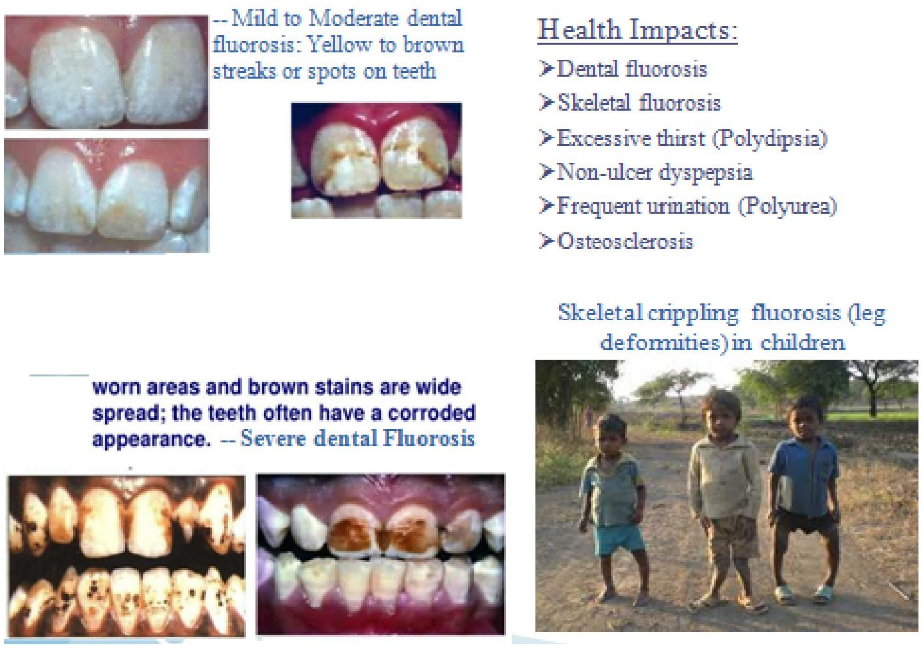

Fig.4: Human health implications of excess fluoride intake.

Chemical analysis shows that ground water is slightly to moderate alkaline with pH ranging between 7.32 to 8.35 pH units with a mean value of 7.8; hence found within permissible limits. TDS values are varying from 207mg/l to 4389mg/l with a mean at 1631.22 mg/l which is much higher than the permissible limits. EC, a primary indicator of dissolved mineral content, is found to vary widely with a minimum value of 310µS/cm at Bali in district Sonepat and a maximum value of 6580 µS/cm at 25°C at Ashraf Garh in district Jind. The major concern parameter total hardness varies from 180mg/l to 1720 mg/l with an average 478.08mg/l. The desirable limit is 200 mg/l. Hardness may causes urolithiasis, kidney stone and other problems in urinary and excretory system. Similar results were observed by Singh, S et al, 2011; Bishnoi and Malik, 2008. Chloride varies from 16.0 mg/l to 2000 mg/l and Nitrate varies from 1.2 to 15.2 mg/l. The fluoride concentration fluctuates from 0.27 mg/l to 2.87 mg/l. All the samples at the depth > 40 m bgl exceeded the standard prescribed limit of 1.0 mg/l fluoride in drinking water (BIS, 2012).

Further it can be observed from the Fig. 2.a that fluoride contamination increases as we move from shallow to deep aquifers. Similarly, high fluoride content was observed in deep aquifers as compared to shallow aquifers in Orissa state by Das et al, 2000. Among the study area, lowest concentration was found in Karnal samples and highest was in Rohtak region samples (Table 2.a). Among the study area, 46 percent samples have more than maximum permissible limit (1.5 mg/l) fluoride concentration (Fig. 3).

High fluoride concentration in drinking water led to dental and skeletal fluorosis. In Haryana state, dental and skeletal fluorosis epidemic is observed among human population due to consumption of underground water with high content of fluoride. In Table 3, effect of fluoride consumption on human health is given. In Fig. 4 some images of fluorosis impact on human can be observed. Correlation matrix indicates that hardness, electrical conductivity and chloride content are positively correlated with fluoride content (Table 2.c). Various other studies also indicates a positive correlation of fluoride concentration with Na^+, HCO_3^-, Ca, TDS, TA, EC (Singh.S. et al, 2012; Bishnoi and malik, 2008; Haritash et al, 2018). It signifies the possible presence of fluorspar, fluoraptite, cryolite in under soil strata/rocks. Thus, high concentration of fluoride in underground water may be attributed to geological and anthropogenic environment of the area. Different studies support these facts. Agarwal et al. 1997, stated that the major sources of fluoride in groundwater are fluoride-bearing rocks such as fluorspar, cryolite, fluorapatite and hydroxylapatite. Fluoride ions from these minerals leach into the groundwater and contribute to high fluoride concentrations (Latha et al., 1999; Ramesam and Rajagopalan 1985). Further overexploitation of underground water for irrigation needs in central Haryana enhances the problem by lowering water table. Every year level of ground water continuously goes down as per CGWB, 2017 reports.

The likely causes for high fluoride in ground water may be due to leaching of phosphatic fertilizers and depletion of calcium either due to precipitation or exchange phenomenon. Among the various sources of fluoride in the environment, those of anthropogenic origin have occasionally been considered to be major ones such as brick-kiln industries, phosphatic fertilizer plant and agri-run-off (Jha et al. 2013). Various cost-effective and simple procedures (Nalgonda Technique; F- Adsorbents; Recharging of groundwater with rain water) for water defluoridation are already known, but the benefits of such techniques have not reached the rural areas due to various limitations. An effective alternate to mitigate the problem of fluorosis is nutrient enriched diet to affected population. As per Yadav et al, 2018; foodstuff having rich content of calcium and vitamin C can prevent fluorosis to a certain extent.

4. Conclusion:

On the basis of present physico-chemical analysis of groundwater of Haryana state central parts, it has been concluded that groundwater quality in the studied zone of the area varies geo-spatially and depth wise also. Higher values of some parameters (Hardness, Chloride, Fluoride) above permissible limits at certain locations indicate that the groundwater on those specific locations is not suitable for domestic use. These high values of certain groundwater constituents may be due to geological and anthropogenic reasons. This study demonstrates that central parts of Haryana comprise higher levels of fluoride in underground water samples. The concentration increases with depth and as the water table continuously depletes hence implications of high fluoride content to population also increases. Sustainable defluoridation techniques must be adopted to ensure safe drinking water for the inhabitants. Apart from removal techniques, affect of excess intake of fluoride can be reduced by good diet and nutrition (Calcium & vitamin C enriched) which can control the damaging effect of fluorosis.

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