



Science

GEOCHEMICAL PROVENANCE AND SPATIAL VARIATION OF FLUORIDE IN GROUNDWATER OF SINDHUDURG DISTRICT, WESTERN MAHARASHTRA

N. Suneetha ¹, G. Gupta ^{*1}, G. Shailaja ¹

^{*1} Indian Institute of Geomagnetism, New Panvel (W), Navi Mumbai 410218, India

Abstract

The present study focuses on the occurrence and intensity of fluoride in drinking water from the Malvan-Vijaydurg coastal tract in Sindhudurg district, Maharashtra. Concentration of fluoride (F⁻) from 36 dug wells/bore wells sampled during this study ranged from 0.04 mg/L to 1.6 mg/L. The results indicate that about 47% of the water samples from the study area have fluoride content greater than 1 mg/L, which exceeds the permissible limit of BIS (1 mg/L), while 4 samples exceeded the World Health Organization standards of 1.5 mg/L. Spatial variation in F⁻ concentration indicates that most of the sampling sites having comparatively high fluoride also have high values of pH, EC and TDS concentration measured in groundwater. High fluoride levels are associated with Ca-Mg-Cl-SO₄, Na-Cl and Na-HCO₃ type water produced by calcite precipitation and/or base ion exchange. The groundwater with higher fluoride concentrations is accompanied by lower Ca⁺² levels. This study augments our understanding on geochemical behaviour of F⁻ concentration and highlights the importance of spatial variability of F⁻ in coastal aquifers. It is also pertinent to monitor the water quality periodically in this region and consequently discontinue using the wells which contain high fluoride concentration.

Keywords: Fluoride; Groundwater; Saturation Index; Konkan; Sindhudurg; Maharashtra.

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1. Introduction

Groundwater quality is a major concern today as it is mostly the only source used for drinking purpose worldwide. Tremendous strides in industrialization, urbanization and agricultural practices are primarily responsible for groundwater contamination. Apart from these, the incidence of fluoride in groundwater is also due to natural geogenic reasons. As per¹, the maximum permissible limit for F⁻ in drinking water is 1.0 mg/L whereas² has assigned a maximum permissible limit of 1.5 mg/L.

Amongst all the chemical elements in the halogen group, Fluorine is the lightest and is one of the most reactive. It is also the most electronegative of all the elements³, which suggests that it has a strong propensity to gain a negative charge, and in solution forms F^- ions. Fluoride ions have the same charge and nearly the same radius as hydroxide ions and may replace each other in mineral structures³. Fluoride therefore forms mineral complexes with a number of cations, and some common minerals of low solubility contain fluoride.

It is reported that fluoride is found at significant levels in a wide variety of minerals, like fluorite, fluorapatite, hydroxylapatite, rock phosphate, cryolite, mica, hornblende etc. These minerals may occur in a variety of rocks including basalts, granites, gneisses, schists, charnockites, khondalites. Anthropogenic sources of fluoride include mining, chemical fertilizers, sewage, sludge etc⁴. Fluorite (CaF_2) is a common fluoride mineral of low solubility occurring in both igneous and sedimentary rocks. Fluoride is also usually linked with volcanic activity and fumarolic gases. Water having high pH is also rich in fluoride⁵.

In India F^- above permissible limit in groundwater has been reported from parts of 19 states⁶, with a combination of higher rate of evaporation and longer interaction of water with aquifer materials under alkaline environment⁷⁻¹¹. Some anthropogenic activities such as use of fertilizers, pesticides and sewage and sludge have also been indicated for an increase in fluoride concentration in groundwater. Fluoride is known to cause significant effects in human beings from mild dental fluorosis to crippling skeletal fluorosis as the level and period of exposure increases¹².

A detailed study of fluoride concentration in groundwater was carried out by different researchers¹²⁻¹⁴ amongst others in different geological formations in India. In the state of Maharashtra, high fluoride contaminated groundwater has been reported from parts of trap-covered Sindhudurg district¹⁴. These studies were mainly focused in the gneisses terrain in the coastal belt, but the occurrence of fluoride in the immediate vicinity of the coastline along the Sindhudurg district is unreported so far. The main objective of this study is to investigate the occurrence of fluoride in groundwater, both in the coastal tract and the inland parts of northern Sindhudurg district so as to assess its suitability for drinking water purposes.

2. The Study Area

The study area lies between $73.3^\circ E$ and $73.8^\circ E$ latitude and $16.0^\circ N$ and $16.5^\circ N$ longitude in the coastal district of Sindhudurg, Maharashtra (Fig. 1). This part of Konkan experiences heavy monsoon rains (average 2500-3500 mm/yr) and the weather is mostly humid throughout the year. Occurrence and movement of groundwater is influenced by geomorphological features such as mountains, hills, rivers, slopes, escarpments, plateaus, valleys and alluvial terrace etc, controlling the hydrogeological conditions¹⁵. This region essentially lies between the Sahyadris to the east and the Arabian Sea to the west. The area is covered by flat topography, dissected by several east-west trending ridges, some of which reach right up to the coast. The region is also traversed by several rivers, with steep gradient, which have their origin in the Sahyadris. The major rivers in this region are the Kasul, Gad, Achara, Piyali, and Kharada, flowing west-ward and meeting the Arabian Sea. These rivers also form major creeks in the study area¹⁶ (Fig. 1).

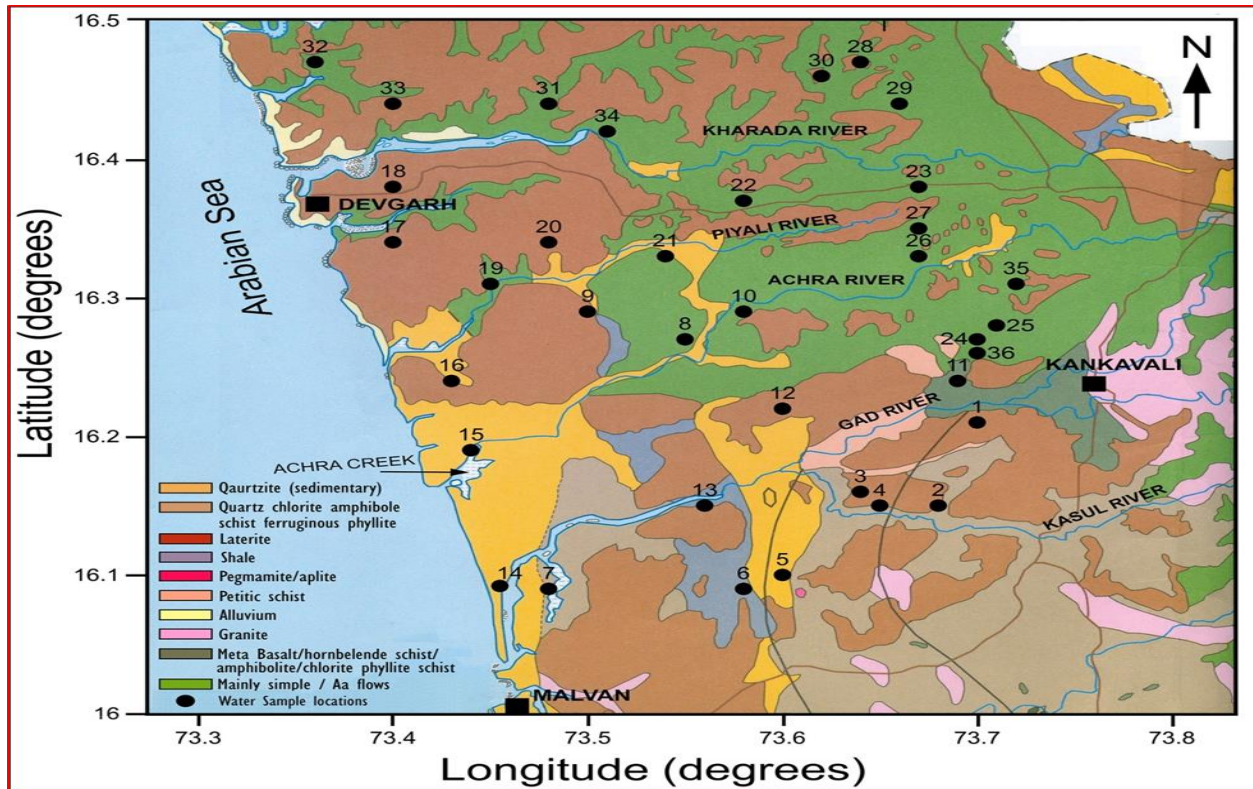


Figure 1: Location map of the study area. Also shown are the water sample sites (Redrawn after¹⁶)

The major rock types in the area are basalt, quartzite, sandstone, shale and associated limestone, laterites and alluvium (Table 1). In Deccan Traps regions the lava flows belong to Ambenali Formation, Mahabaleshwar Formation and litho-stratigraphically into the Purandargarh Formation¹⁵. Laterites and its soils of varying thickness are found extensively in the study area. The Deccan Traps are exposed in the eastern part, near Kankavli. Patches of quartzites are present along the coastal stretch and parts of inland. The alluvial deposits are mainly along the river course and near the sea shore. Along the Gad River basin rocks from Precambrian to Recent age are exposed¹⁴.

Table 1: General geological succession in Sindhudurg district, Western Maharashtra, India (Source CGWB report¹⁵)

Geological time	Formation
Recent to sub recent	Alluvium beach sand
Pleistocene	Laterite and lateritic spread
Miocene	Shale with peat and pyrite nodules
Cretaceous to Eocene	Deccan trap basalt lava flows
Upper Pre-Cambrian	Kaladgi series, quartzite, sandstone shale, and associated limestone
Dharwar Super Group	Phyllite, conglomerate, quartzite

The aquifers in the region occur in weathered/fractured/jointed/vesicular/massive basalts, under phreatic and semi-confined to confined conditions. The depth to water level varies from 2 to 10 m below ground level (bgl) in shallow aquifers and from 20 to 50 m bgl in deeper aquifers¹⁵. Rainfall is the main source of groundwater recharge in this region.

3. Materials and Methods

Total 36 water samples were collected during pre-monsoon period of May 2016 in the study area in order to understand the water quality. The groundwater sample locations are shown in Fig. 1. Samples are analyzed for the physico-chemical attributes like pH, electrical conductivity (EC), total hardness (TH), total dissolved solids (TDS), major ions (Ca^{2+} , Mg^{2+} , Na^+ , K^+ , Cl^- , HCO_3^- , CO_3^{3-} and SO_4^{2-}). The methods used for the analysis were standardized as per procedures¹⁷. F^- was estimated by *SPADNS* spectro-photometric *method*.

Chemistry of the groundwater samples mainly depends on the geology as well as on the geochemical processes which take place within the groundwater system. Spatial mapping of groundwater facies based on classification of Piper¹⁸ diagram can help in visualizing the progressive fluctuations in water chemistry. In the present study, the groundwater samples were classified hydrochemically using major cations and anions with conventional Piper trilinear diagram using AquaChem 2014.2, to determine the similarities between groundwater.

Gibbs diagram¹⁹ is widely used to establish the relationship between lithological characteristics and water composition of aquifers. In this the ratio of dominant anions [$\text{Cl}/(\text{Cl}+\text{HCO}_3)$] and cations [$(\text{Na}+\text{K})/(\text{Na}+\text{K}+\text{Ca})$] are separately plotted against TDS in order to get the possible sources of dissolved chemical constituents in groundwater aquifers from essentially three different fields such as, atmospheric precipitation dominance, evaporation dominance and rock–water interaction dominance.

Saturation indices (SI) are used to estimate the chemical stability of a mineral in water. It is a useful parameter to evaluate the equilibrium state for various solubility reactions, which describe the tendency of the solution to dissolve or precipitate in a set of minerals²⁰. Changes in the saturation state are useful to distinguish different stages of hydrochemical evolution and help identify which geochemical reactions are important in controlling water chemistry²¹⁻²². In the present study $\text{SI}_{\text{fluorite}}$ and $\text{SI}_{\text{calcite}}$ were calculated using PHREEQC Interactive Software (ver. 3.3.8-11728)²³. It is reported that if the water is exactly saturated with the dissolved mineral, SI equals to zero, while positive values of SI signify super-saturation and the mineral would tend to precipitate, and negative one implies under-saturation and the mineral would tend to dissolve²⁴.

4. Results and Discussions

The statistical analysis of the chemical composition of groundwater samples are given in Table 2. The pH values range between 6.2 to 8.7 with an average value of 7.2, indicating alkaline nature of water. EC and TDS ranges from 174-94920 $\mu\text{S}/\text{cm}$ and 77-4667.0 mg/L respectively with an average value of 686.5 $\mu\text{S}/\text{cm}$ and 335.9 mg/L. The minimum and maximum values along with average values and standard deviation (SD) of Na, Cl, Ca, SO_4 , Mg, CO_3 , HCO_3 , K and F^- are given in Table 2.

Table 2: Geochemical data of groundwater samples from Sindhudurg district

Well	pH	EC	TDS	Ca	Mg	Na	CO ₃	HCO ₃	SO ₄	Cl	F
1	7.48	317	144	28	9	15	40	67	82.61	32	0.84
2	6.27	284	182	62	78	35	32	138	90.77	52	1.28
3	7.4	300	134	26	7	20	34	62	83.09	48	0.9
4	7	537	239	36	9	40	50	95	109.5	53	0.96
5	7.2	543	241	37	25	34	32	65	76.84	32	0.72
6	6.95	333	149	21	15	18	4	73	107.1	56	0.96
7	7.1	2151	955	91	32	30	33	201	119	125	0.96
8	7.44	186	84	53	15	10	32	48	86.45	18	1.5
9	7.4	178	80	51	15	8	17	45	146.4	36	1
10	6.43	174	77	35	18	48	16	38	183.4	42	0.8
11	7	400	178	97	20	19	30	29	205.6	28	0.84
12	7.2	514	231	28	2.1	67	14	56	210	32	0.88
13	7.62	187	83	39	7	41	26	87	79	15	1.2
14	7.42	517	231	103	23	46	18	165	248	119	0.8
15	7.44	875	391	160	34	38	62	99	211.6	29	1.6
16	6.74	246	110	22	16	39	34	144	204.5	28	0.96
17	7.43	517	231	34	4	42	54	165	149.8	56	1.12
18	7.05	338	151	65	13	44	34	68	209.8	49	1.08
19	6.19	401.7	179.2	60	11	16	38	78.68	130.1	29.36	0.4
20	6.38	255.3	98.13	76	14	9	30	71.05	129	21.43	1.24
21	6.58	246.7	104.4	49	5	34	12	122.9	86.45	31	1
22	7.1	325	143.8	91	12	38	28	65.12	126	21	0.71

23	7.05	547.4	244	144	36	42	64	86	115	67	1.04
24	7.15	620	277	25	9	50	36	130	131	39	1.08
25	7.5	342	156	24	16	10	3	102	122	45	1.24
26	7.33	319.4	139.2	46	9	5	28	203.61	184	15	1.48
27	8	243	107.9	43	6	9	22	88.14	210	51	1.28
28	8.66	440.1	201.3	52	5	21	14	41.21	245	67	0.4
29	7.1	238.7	109.3	75	18	32	36	156.87	177	48	1.56
30	6.7	246	90.6	28	7	23	18	62.89	152	61	0.84
31	6.95	741.9	330.3	142	67	92	64	59.87	117	82	1.44
32	7.3	9420	4667	640	107	108	12	82.41	150	249	0.67
33	7.24	479	254	275	102	89	12	84	87	96	0.92
34	6.8	346	139	90	57	34	34	35	110	35	0.84
35	7.6	484	810	189	13	25	26	110	163.2	28	0.9
36	7.2	389	145	29	18	12	8	64	131	39	1.4
Min.	6.2	174	77	21	2.1	5	3	29	76.8	15	0.4
Max.	8.7	9420	4667	640	107	108	64	203.6	248	249	1.6
Mean	7.2	686.5	335.9	85.2	23.7	34.5	29.1	91.4	143.6	52.1	1
SD	0.5	1534.2	764.3	109.7	26.2	23.9	15.8	45.5	49.9	42.5	0.3

From Table 2, it can be noticed that the concentration of F⁻ ranges from 0.4 to 1.6 mg/L with an average value of 1 mg/L. It is further observed that 47% of the water samples exceed the limit of the BIS standards (1 mg/L). However according to², the permissible limits of F⁻ in drinking water is 1.5 mg/L. The recommended limit of F⁻ in the water depends on the climatic conditions of an area²⁵, because the amount of water consumed and the amount of F⁻ ingested is influenced primarily by ambient temperature. Intake of F⁻ below 0.5 mg/L causes dental caries², it is reported that if F⁻ is greater than 1 mg/L, it may cause dental and skeletal fluorosis²⁵. In the present case only two samples (well nos. 19 and 28) reveal low F⁻ concentration of 0.4 mg/L, while all other samples have values high enough to cause human damage (Fig. 2).

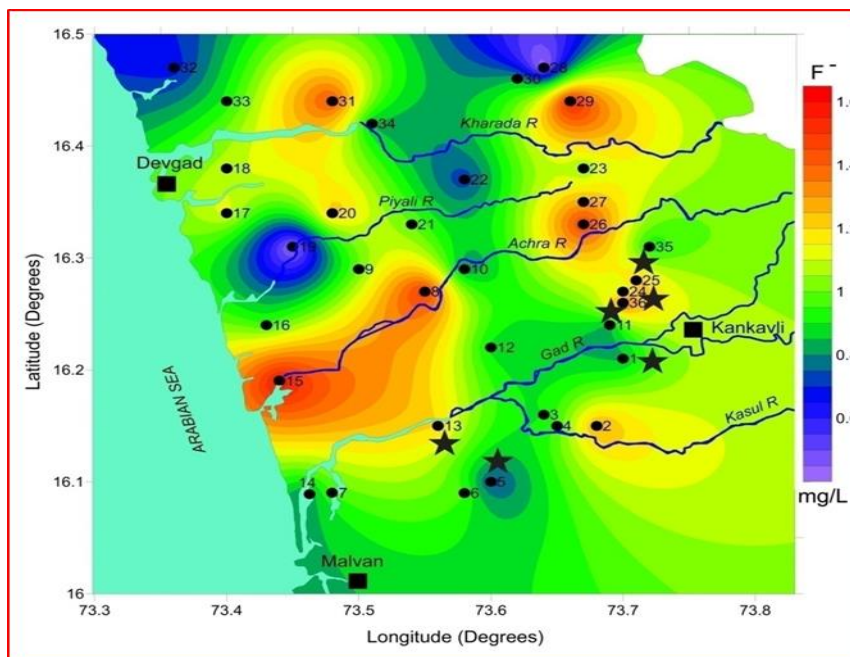


Figure 2: Spatial variation map of fluoride concentration. The closed circles are the water sample locations. Stars are the hotspots of fluoride concentration as reported by¹⁴

Spatial Distribution of Fluoride

The spatial distribution map of fluoride in the study area is shown in the Fig. 2. In order to analyze the factors controlling the fluoride content in groundwater, one should be aware of soil cover, lithology, climate, geomorphology, anthropogenic activity and aquatic behavior (marine source). From Fig. 2, it is observed that high F^- is wide-spread along the creeks and the inland part of the study area. For the sake of discussion, the study area is classified into two parts based on the sample locations: one is inland and the other is coastal. In this area, the inland part was earlier studied from parts of Gad River basin around Kankavli¹⁴. This study reported very high fluoride concentration (5 mg/L) in the groundwater over laterite, basalt and the Precambrian gneissic aquifers. High fluoride concentration is attributed to the presence of tourmaline bearing pegmatites in the Precambrian basement. Also deep circulation of fluoride-rich groundwater between the lateritised basalts and the underlying basement could be responsible for the occurrence of fluoride in both the shallow and deeper aquifers of the region. The wells sampled in that study have subsequently been abandoned. In the present study, water samples were collected from the earlier wells sampled¹⁴. Water samples were also collected from the coastal tract. From Fig. 2 it is evident that the samples 2, 24, 25 and 36, located in the basaltic and gneissic aquifers, show relatively high F^- concentration (1.28, 1.08, 1.24 and 1.4 mg/L respectively) and is greater than the BIS permissible limit. High fluoride values ranging from 1.6 to 2.5 mg/L was reported earlier in wells near to the present sampling sites (2, 24, 25 and 36) thereby reconfirming the fluoride hotspots¹⁴. Besides these, high F^- values (>1 mg/L) are observed at the sampling sites 23, 26, 27 and 29, which is located near the rivers Achara, Piyali and Khadra within the study.

The coastal tract samples 8, 9, 13, 15, 17, 18, 20, 21 and 31 reveals higher F^- values (1.08 to 1.6 mg/L) along the creeks and near to sea coast. It can be noted from the above nine coastal sites that except sampling points 20, 21 and 31, others have pH value greater than 7, indicating alkaline nature of water (Table 2). Also sampling points 15 and 18 reveals high SO_4 values (211 and 210

mg/L respectively), exceeding the permissible limit (200 mg/L), while station 15 also reveals high EC value (875 $\mu\text{S}/\text{cm}$) exceeding the permissible limit of 750 $\mu\text{S}/\text{cm}$. Perhaps the high fluoride content at sampling point 15 is due to the sea water intrusion.

Hydrochemical facies

Hydrochemical analysis of the water samples from the study area were classified graphically using trilinear diagram (Fig. 3) in order to assess the hydrochemistry of groundwater and to establish the source of fluoride concentration¹⁸. All the major cations and anions are expressed in meq/L. The total cations are plotted on the left triangle while anions are plotted on the right triangle (Fig. 3). Each point is then projected into the upper field along a line parallel to the upper margin of the field, and the point where the extension intersects indicates the character of the water as represented by the association among Na^+ , K^+ , Ca^{2+} , Mg^{2+} , CO_3^{2-} , HCO_3^- , Cl^- and SO_4^{2-} ions²⁶. From Fig. 3 it is observed that majority of the samples fall in the Ca-Mg-Cl- SO_4 and Na- HCO_3 type. A few samples also fall in the Ca-Mg- HCO_3 and Na-Cl types of water.

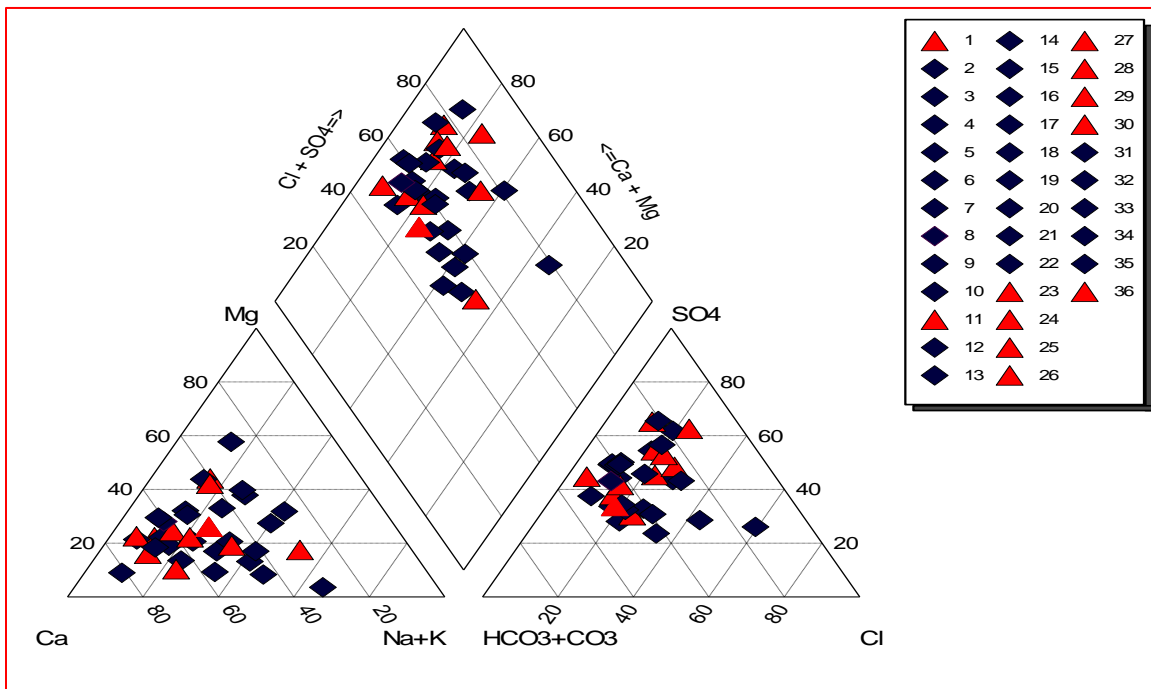


Figure 3: Hydrochemical facies of water types

Rock-water interaction

Water sample data was plotted explicitly for the purpose of understanding the prevailing controlling factors in groundwater aquifers¹⁹. From Fig. 4a,b it is evident that the grouping of data points reveal that 90% of the samples fall under the chemical weathering of rock forming minerals, which is the dominant factor controlling the groundwater chemistry in the study area, while 10% are in the evaporation domain.

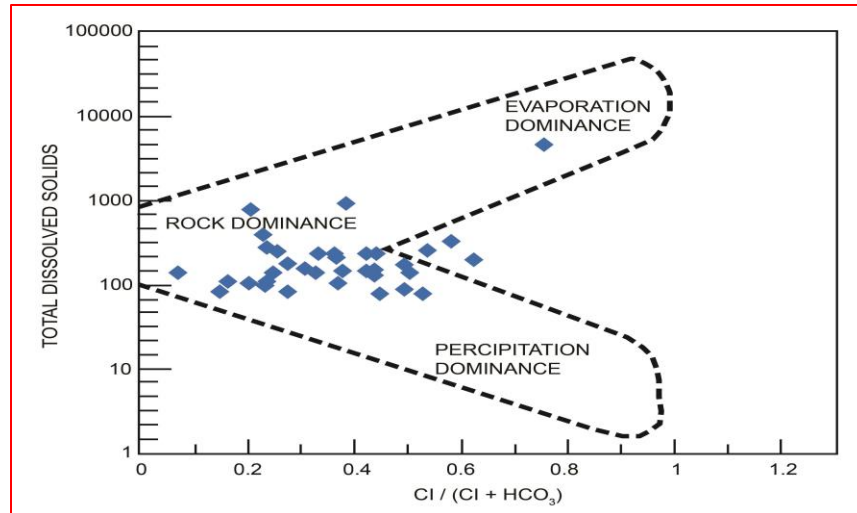


Figure 4a: Gibb's diagram of cation

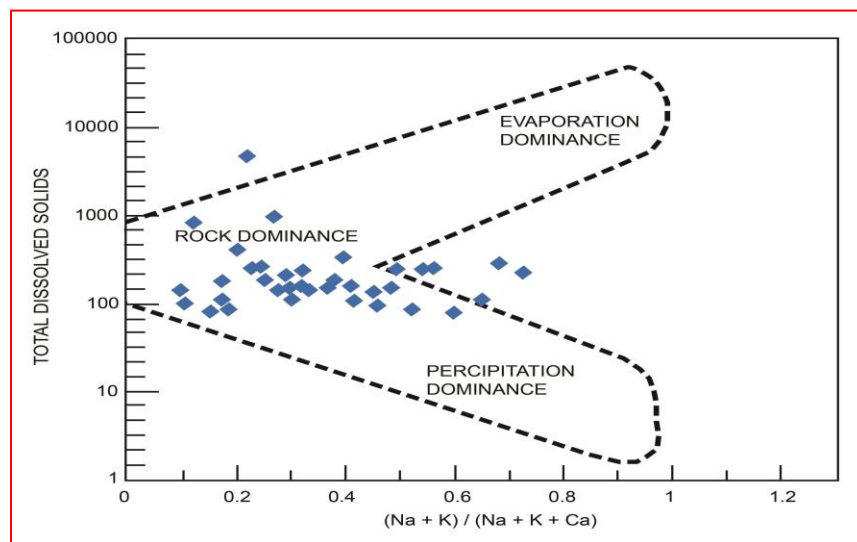


Figure 4b: Gibb's diagram of anion

The rock-water interaction phenomena are probably caused by longer residence time of groundwater. The weak evaporation dominance is a result of intensive rainfall and long-term leaching of fluorine from rocks due to weathering effects. Evaporation during summer is high and is supported by Na-Cl type of groundwater. In the present study, fluoride concentration in the groundwater is not very high, probably as a result of heavy rainfall, leaching and/or precipitation of calcium fluoride from rocks as compared to other areas.

Saturation indices of fluoride minerals

The groundwater chemistry depends on the interaction between rain and the rock near the Earth's surface. The concept of speciation modelling has been used to study the chemical equilibrium existing in groundwater and to recognize the source of high fluoride concentration in groundwater²⁷. The most important results of speciation calculations are saturation indices (SI) for minerals, which indicate whether a mineral dissolves or precipitates. The solubility limits for fluorite and calcite provide natural controls on water composition with a view that calcium,

fluoride and carbonate activities are interdependent¹⁰. The data for the saturation indices (SI) of fluorite (CaF₂) and calcite (CaCO₃) in the groundwater samples were calculated using PHREEQC Interactive software and shown in Table 3.

Table 3: Saturation indices (SI) of fluorite and calcite in the groundwater samples

Well	Ca (mg/L)	F (mg/L)	SI_Fluorite	SI_Calcite
1	28	0.84	-1.4551	-0.6599
2	62	1.28	-0.9589	-1.5958
3	26	0.9	-1.4238	-0.8106
4	36	0.96	-1.2582	-0.9538
5	37	0.72	-1.5338	-0.8897
6	21	0.96	-1.4835	-1.3469
7	91	0.96	-0.9809	-0.187
8	53	1.5	-0.7116	-0.5929
9	51	1	-1.081	-0.6807
10	35	0.8	-1.4451	-2.0995
11	97	0.84	-1.0057	-1.0736
12	28	0.88	-1.4122	-1.048
13	39	1.2	-1.0187	-0.2723
14	103	0.8	-1.0732	0.1326
15	160	1.6	-0.3155	0.106
16	22	0.96	-1.4783	-1.3009
17	34	1.12	-1.1489	-0.2676
18	65	1.08	-0.9338	-0.8064
19	60	0.4	-1.8035	-1.9097
20	76	1.24	-0.7377	-1.5745
21	49	1	-1.0815	-1.2332
22	91	0.71	-1.1627	-0.6313
23	144	1.04	-0.7338	-0.4247
24	25	1.08	-1.3111	-0.802
25	24	1.24	-1.2119	-0.538
26	46	1.48	-0.7845	-0.1545
27	43	1.28	-0.9197	0.1634
28	52	0.4	-1.8558	0.5545
29	75	1.56	-0.5824	-0.3438
30	28	0.84	-1.4555	-1.5906
31	142	1.44	-0.5264	-0.7302
32	640	0.67	-0.7658	0.2921
33	275	0.92	-0.7402	-0.0339
34	90	0.84	-1.1269	-1.2967
35	189	0.9	-0.7125	0.3947
36	29	1.4	-1.0268	-0.982

Saturation indices (SI) of fluorite (CaF_2) and calcite (CaCO_3) are plotted in Fig. 5, which shows that except six samples (nos. 13, 15, 27, 28, 32 and 35) which accounts for 16% of water samples, all the other samples are under-saturated with respect to calcite and fluorite. The highest F^- concentrations observed are at sample nos. 13, 15 and 27 (F^- values 1.2, 1.6, 1.28 mg/L respectively), which are very near to the creeks.

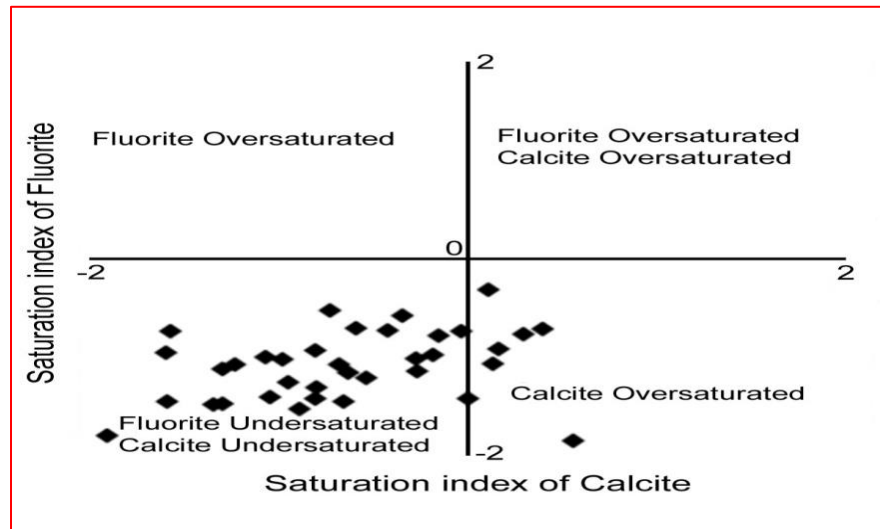


Figure 5: Plot of calcite saturation index and fluorite saturation index

Thus solubility control on the higher concentration of fluoride can be explained due to the fact that fluoride ions in groundwater can be increased as a consequence of precipitation of CaCO_3 at high pH, which removes Ca^{2+} from solution allowing more fluorite to dissolve²⁸. The average pH in the study area is 7.2 (Table 2). The released Ca^{2+} ions combine with CO_3^{2-} ions to further increase the precipitation of CaCO_3 .

It is also revealed that the samples which shows high calcite is located near to coast and is super saturated with the Ca^{2+} . At the same time, the inland samples having high fluoride also indicate the super saturation of Ca^{2+} , where the water type is Cl^- dominant. This is replaced with Ca/Mg by Base Exchange due to rock water interaction or evaporation which increases the ionic strength with respect to the least soluble minerals, and finally the oversaturated solution may precipitate out. This shows that Base Exchange process is an important factor for controlling the F^- in the groundwater²⁹. As mentioned earlier, six samples in the vicinity of creeks and coast reveals a positive relation between pH and F^- . However, poor correlation between pH and F^- observed at other sampling sites may be due to high dilution by rainwater, variable fresh-saltwater interaction in the coastal aquifers or lesser time of water residence or water-rock interaction in the aquifers³⁰.

5. Conclusions and Recommendations

The groundwater of the coastal Sindhudurg district, Maharashtra was assessed for fluoride concentration, as its deficiency or excess has a direct vulnerability on human health and related societal repercussions. Therefore information regarding the status of fluoride in the coastal part of the State is crucial. The spatial variation map of F^- reveals that about 47% of the groundwater samples show elevated fluoride concentration (1.08-1.6 mg/L), exceeding the BIS permissible

limit. This study further indicated that fluoride originates from the weathering of minerals in the aquifer matrix and possibly the exchange of OH⁻ for F⁻ weathered rocks under high pH condition. Rock-water interaction is the dominant process affecting about 90% groundwater quality, which could be due to longer residence time of groundwater. About 10% samples fall in evaporation domain, owing to intensive rainfall and long-term leaching of fluorine from rocks due to weathering effects. The saturation indices of the samples reveal that they are supersaturated with respect to calcite containing high fluoride. It is possible that the removal of calcium from solution permits fluoride concentrations to increase. Some calcium is removed from solution through base ion exchange, thus increasing the fluoride concentrations associated with NaCl- and Na-HCO₃ type water.

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*Corresponding author.

E-mail address: gupta_gautam1966@ yahoo.co.in