



Hydrogeochemistry of fluoride concentration in groundwater and human health risk assessment: A study from some parts of Rarh Bengal, Eastern India

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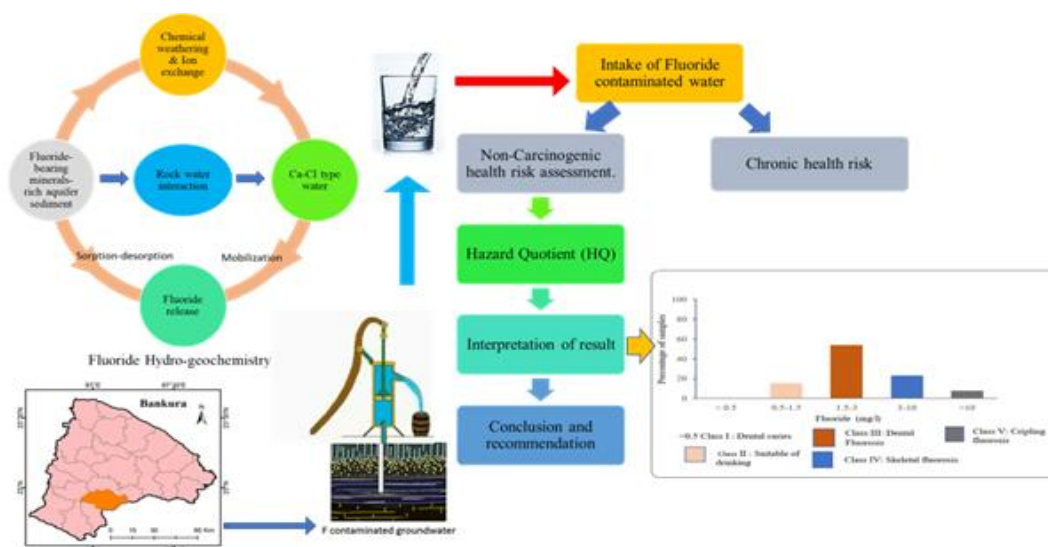
1. Abstract:

Excess fluoride (F⁻) in drinking water results in serious health risks in many parts of the world as well as western parts of Rarh Bengal, India. The current study was carried out to investigate the relationship between fluoride and other physiochemical parameters, as well as to assess the health risk of residents of the Simlapal Block, Bankura District. In this study, 26 water samples collected from the five highly fluoride-contaminated villages were tested for fluoride and other chemical characteristics. The result shows that fluoride correlates considerably negatively with Na (-0.07), Ca (-0.12), Cl (-0.19), Mg (-0.10), and SO₄ (-0.09) but significantly positively with pH (0.88), TDS (0.82), EC (0.98), TH (0.89), K (0.19), and HCO₃ (0.91). Based on the geochemical classification of groundwater, the three main hydro-chemical facies are Ca-Cl, Na-HCO₃, and NaHCO₃-Ca. The Gibbs diagram demonstrates that rock water interaction and evaporation are the primary controlling mechanisms for adjusting the water quality in this hard rock aquifer. Notably, 85 percent of water samples revealed fluoride concentrations over the permitted limit of 1.5 mg/L. The study found that the groundwater fluoride content in the area ranged from 2.93 mg/L to 10.51 mg/L, resulting in dental, skeletal, and other fluorosis. Children and teenagers are more vulnerable to fluoride poisoning than adults, according to the hazard quotient of fluoride (HQ_{Fluoride}) index. To provide clean drinking water and lower the risk of fluorosis, meticulous planning is necessary. Findings of this research and recommendation may be useful for planners and policy makers for providing public health facilities in the areas.

Highlights:

- Fluoride (F^-) and other geochemical concentrations of groundwater samples have been measured.
- F^- concentrations range from 0.89 to 10.51 mg/L, with 85% of samples exceeding the 1.5 mg/L threshold.
- F^- correlates positively with pH, TDS, EC, TH, K and HCO_3 , but negatively correlated with Na, Ca, Cl, Mg and SO_4 .
- The children and teenagers are more susceptible to fluoride poisoning than adults.
- Appropriate safety precautions must be implemented to control health risks in this region.

Graphical Abstract:



1. Introduction

The degradation of groundwater quality and the availability of safe drinking water are serious issues that affect the entire planet. Many pollutants have recently been released into the environment from both surface and subsurface sources, contaminating groundwater. (Podgorski et al., 2018; Nizame et al., 2021; Rashid et al., 2021; Qiu et al., 2023). Fluoride is a naturally occurring element found in various geological formations, and its presence in groundwater has been a topic of growing concern worldwide due to its potential impact on human health. The presence of fluoride in groundwater primarily originates from the weathering of minerals such as fluorite, apatite, and mica in the geological formations (Rudra

2012). Understanding the intricate interplay between geological factors, hydrogeological processes, and climatic conditions is essential in comprehending the distribution of fluoride in groundwater. Moreover, anthropogenic activities such as agricultural practices, industrial discharges, and unregulated groundwater extraction can exacerbate fluoride contamination, exacerbating the risk to human health. According to WHO (2011), more than 260 million people are suffering from different kinds of fluorosis diseases as a result of long-term ingestion of high fluoride contaminated water. Fluoride contamination of groundwater is extremely common, particularly in arid and semi-arid regions of the world, including China, India, Africa (Rift Valley Zone), parts of South America, Nigeria, parts of Iran, Kenya, Pakistan, Sri Lanka, and parts of Nigeria (Adimalla and Wu, 2019; Ahmed et al., 2022). About 19 states in India have high fluoride contamination (CGWB, 2010) and endemic fluorosis has been prevalent in India's diverse regions since 1937 (Short et al. 1937). Due to the abundance of fluoride-bearing minerals, volcanic rock prone zones are highly fluoride-contaminated where fluoride concentration in groundwater is anticipated to be quite high (Samal et al. 2020; Subramaniyan et al., 2022).

Fluoride is an important micronutrient for human health, particularly for the tissues, skeleton, and teeth (Yousefi et al., 2019; Chandrajith & Diyabalanage, 2022). However, when its concentration exceeds permissible limits in drinking water, it can pose significant health risks, leading to a range of debilitating conditions. Excessive fluoride use causes a variety of health problems, including skeletal and dental fluorosis, excessive thirst, low haemoglobin levels, skin rashes, melancholy, anxiety, and muscle fibre degradation (Xiao et al., 2022; Jiang et al., 2019).

In recent years, the hydrogeochemistry of fluoride in groundwater has emerged as a critical concern, particularly in regions where groundwater is the primary source of potable water. Eastern India, specifically the Rarh Bengal region, is no exception to this global challenge. The Rarh Bengal region, situated in the eastern part of India, is marked by its unique geological and hydrogeological characteristics. The region's aquifers are predominantly composed of sedimentary rocks, including sandstone, shale, and clay, which vary widely in terms of their fluoride-bearing capacity. Additionally, the semi-arid to sub-humid climate of Rarh Bengal further complicates the hydrogeochemical dynamics, as climatic factors such as temperature and precipitation can influence the dissolution of fluoride-bearing minerals.

In West Bengal, groundwater supplies are mostly used for household and agricultural purposes (Chakrabarti et al. 2012; De et al., 2022). Eight districts of the state are having fluoride contaminated groundwater, whereas western parts of Rarh Bengal like Purulia, Bankura, and Birbhum districts are most prevalent (Rudra, 2021; Mandal & Sanyal, 2022; Das et al., 2022). Fluoride poisoning has been a serious problem in the Bankura area of West Bengal's groundwater for the past

twenty years. The district's Simlapal block is severely fluoride affected. Although the characteristics of ground water in the Simlapal Block have been studied and analysed in a few previous studies (Rudra and Khan, 2018), there has not been a thorough investigation into the nature of groundwater and the geochemical parameters that make them suitable for drinking. People in the study area's various villages have recently been diagnosed with various fluorosis diseases. As a result, a thorough examination of the health risk assessment is also required. Thus, the current study focuses on the geochemical properties of local groundwater to assess drinking water quality in terms of fluoride pollution.

2. Materials and methods

2.1 Study area

Simlapal block of Bankura district is, bordered on the west by the Khatra block and the West Medinipur district, and on the north and south by the Taldangra and Sarenga blocks. It stretches from 22°59'38.84"N to 22°50'34.42"N latitude and from 86°55'20.25"E to 87°13'06.10"E longitude. Simlapal, Parsola, Machatora, Laxmisagar, Bikrampur, Dubrajpur, and Mondalgram are the seven-gram panchayats in this block with 204 villages (Fig 1). With an average elevation of 57 meters, this block's entire geographic area is 309.28 square kilometers (119 square miles, or 1144.04 hectares) (187 feet).

The Bankura district, which accounts for 7.75 percent of West Bengal's land area, is divided into three topographical areas: an alluvial plain on the district's eastern side where Simlapal Block is located; a mountainous region on the western side; and an undulating area in the middle. Shilabati and Joypanda River proceed beyond the obstruction.

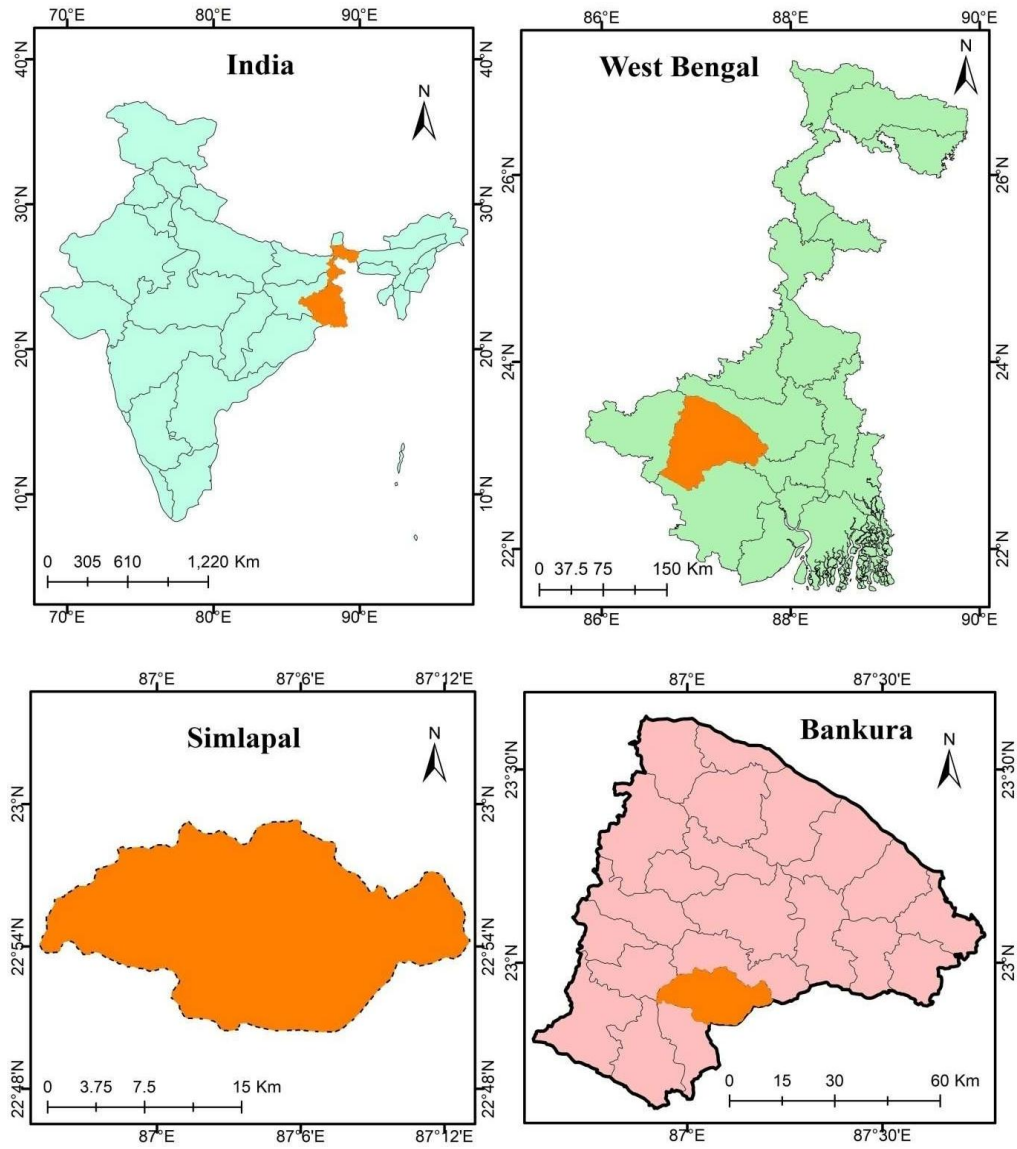


Fig. 1: Location map of the study area

Geologically Bankura district is composed of hard rock structure with ranging from metamorphic gneisses and schists of the Archaean age of the western part to recent alluvium in the eastern part. Basement rocks consisting of Precambrian granites and granite-gneisses emerge at relatively shallow depths. The basement crystalline rocks possess only secondary porosity formed by joints, fractures/fissures, and intergranular porosity (Ojo et al. 2015; Amadi and Olasehinde 2010). The majority of rock types found in Bankura include granite gneisses of various grades and kinds, mica schists, hornblende schists, pyroxenites, anorthosites, quartzites, and younger

dolerite dykes (Fig. 2). Satellite data may be used to show different structural elements and landforms.

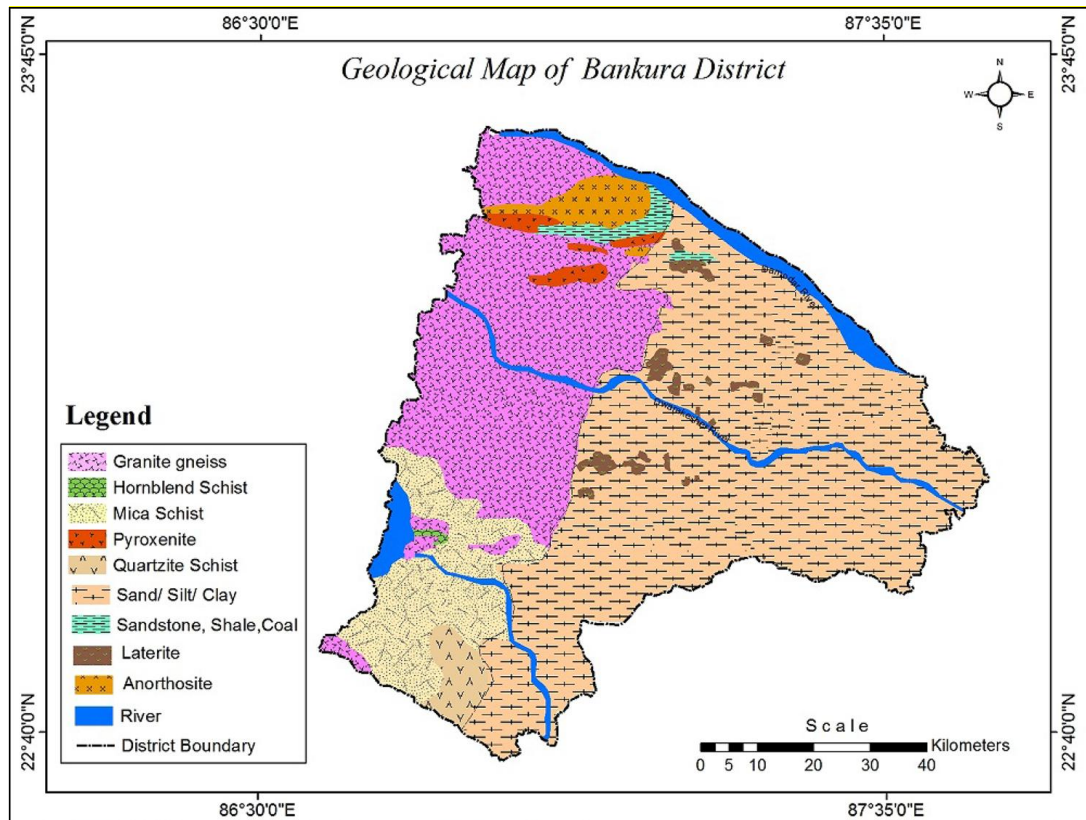


Fig. 2 Geological map of Bankura district. Source: GSI District Resource Map 2001

The Simlapal block is densely forested and is designated as a tribal dwelling (Mondal et al. 2016). According to the 2011 census, 1,43,038 people are living in the Simlapal block, with 73,008 men and 70,030 women. The Scheduled Caste (SC) community makes up 26.38 percent of the population in this area, while the Scheduled Tribe (ST) community accounts for 14.86 percent. Even though the overall literacy rate is 68.44, the gender disparity in literacy is higher than 20%. Due to a lack of alternatives, the area's rural residents use groundwater-based tube wells to obtain their water, which is highly fluoridated. A huge number of people, primarily children and the elderly, suffer from dental and skeletal fluorosis disorders.

2.2 Sampling design:

Total of 26 water samples was taken from the water sources in the spring season at around 9 am by random sampling method from the five villages named Jamda, Sainidanga, Laxmisagar, Brindabanpur and Machatora of the Simlapal blocks of Bankura district. The source, collection date, and other pertinent details were written

on the clean polyethene bottles in which the water samples were taken. In this study, fluoride is given major emphasis but at the same time other chemicals like TDS, P^H , Total Hardness, Electric conductivity, Sodium, Potassium, Chloride, Calcium, Magnesium, Sulphate and Bicarbonate were also analysed for discussion of the real picture of fluoride distribution throughout the block. Based on well-designed and previously prepared questionnaires, door-to-door surveys of roughly 50 randomly selected households in each village were also conducted to ascertain the socioeconomic position and fluorosis prevalence of the rural population in villages affected by fluoride.

2.3 Analysis of chemical parameters:

The concentration of fluoride in the samples of groundwater was analysed by using the instrument fluoride ion-selective electrode (Model No. - Orion 9609 BNWP). TDS and pH of all the samples were measured with the help of Digital TDS and pH meter (Model No. HFTDS-M7). EDTA (ethylene diamine tetra acetic acid) is a very simple and recommended method for the determination of TH, Ca, and Mg of water. EC of all the samples was restrained with the help of a Digital Electric Conductivity meter (Model No. HFTDS-M7). By using the flame photometer 284, the sodium concentration in the sample water has been measured. The concentration of potassium was also experimented with the same as per sodium by using a flame photometer. The concentration of sulphate in sample water was measured by the turbidimetric method using a spectrophotometer (Saxena, 1990).

2.4 Preparation of maps and diagrams:

To get preliminary data on topography, drainage patterns, the presence of surface water bodies, and other topics, the Survey of India (SoI) toposheets with the numbers 73 I/12, 73 I/16, and 73 I/13 with a 1:50,000 scale were employed. The current 1:250,000 scale Geological Survey of India (GSI) map was used to create a map displaying geological characteristics. Map showing the spatial distribution of fluoride were prepared using ArcGIS tools. Piper diagram, Durov plot and Gibbs diagram were prepared by the grapher 13 tools and other diagrams and correlation matrix table were also prepared with the help of Jamovi software.

2.3 Human health risk assessment

Fluoride can enter the body through the skin, ingestion (through food and drink), and inhalation (through breathing). However, drinking accounts for roughly 75–90% of the overall consumption and is the main mechanism of groundwater fluoride exposure (WHO, 2010). Furthermore, non-carcinogenic fluoride exposure through the skin and inhalation has little consequences on health (Mukherjee et al., 2019; Noor et al., 2021; Sahu & Debsarma, 2023). As a result, the standard technique was used to evaluate the health risk associated with drinking water that contains too

much fluoride (USEPA, 1993). In the first step of this procedure, the chronic daily intake (CDI) of fluoride in a person is determined using the following equation:

$$CDI \text{ (mg/kg/d)} = \frac{[Fc \times DID \times TEF \times ED]}{[ABW \times AET]} \dots\dots\dots (i)$$

Where Fc: Fluoride content in groundwater samples (mg/L); DID: Daily Ingestion Dose of drinking water (L/day); TEF: Total Exposure Frequency of drinking water; ED: Exposure Duration; ABW: Average Body weight; and AET: Average Exposure Time (calculated as the product of the number of years and days) and the values of the parameters are provided in Table 1.

Finally, the equation (ii) is used to determine the Hazard Quotient of fluoride due to consumption of fluoride-contaminated water, which is a non-carcinogenic health risk.

$$HQ_{\text{Fluoride}} = \frac{CDI}{RfD} \dots\dots\dots (ii)$$

Where RfD is the USEPA (1993) recommended reference dose for chronic oral fluoride exposure which is taken as 0.06mg/kg/d.

Table 1: Numerical values of the parameters used in the calculation of the fluoride health risks

Parameter	Physical significance	Values	Units	References
Fc	Fluoride concentration	0.89-10.51	Mg/L	This study
DID	Daily ingestion dose	Males:4 Females:3 Children:1	L/day	Naz et al. (2016)
TEF	Total exposure frequencies	365	Day/year	Ahada and Suthar (2019), USEPA (1999)
ED	Exposure duration	Males: 64 Females: 67 Children: 12	Year	WHO (2013)
ABW	Average body weight	Males: 65 Females: 55 Children: 15	Kg	ICMR (2009)
AET	Average Exposure time	Males:23360 Females:24455 Children:4380	Day	WHO (2013)

3. Result and discussion

3.1 Fluoride concentration and spatial distribution

Simlapal Block is the most fluoride-affected area in the Bankura district. Within this block, Bikrampur G.P. is the hardest hit, with approximately 82.35% of its villages having unsafe levels of fluoride (over 1.5 mg/L), and only 17.65% are considered safe. Conversely, in Mondalgram G.P., about 74.29% of the villages have safe fluoride levels, while 25.71% are unsafe. According to PHED data from 2015-16 (Lokepore), 125 villages in Simlapal Block are deemed unsafe, whereas 79 villages are safe (Table 2). The distribution of fluoride concentration in groundwater is uneven across the Simlapal block, with Laxmisagar G.P. recording the highest levels. Figure 3 illustrates the nature and spatial distribution of fluoride concentrations in the groundwater of Simlapal Block.

Table 2: Safe and unsafe villages in different GP of Simlapal block (2015-16)

GP	No. of Village			Percent of Village	
	Safe	Unsafe	Total	Safe	Unsafe
Laxmisagar	7	22	29	24.14	75.86
Bikrampur	6	28	34	17.65	82.35
Simlapal	6	21	27	22.22	77.78
Dubrajpur	10	11	21	47.62	52.38
Parsola	13	15	28	46.43	53.57
Mandalgram	26	9	35	74.29	25.71
Machatora	11	19	30	36.67	63.33
Total	79	125	204	38.73	61.27

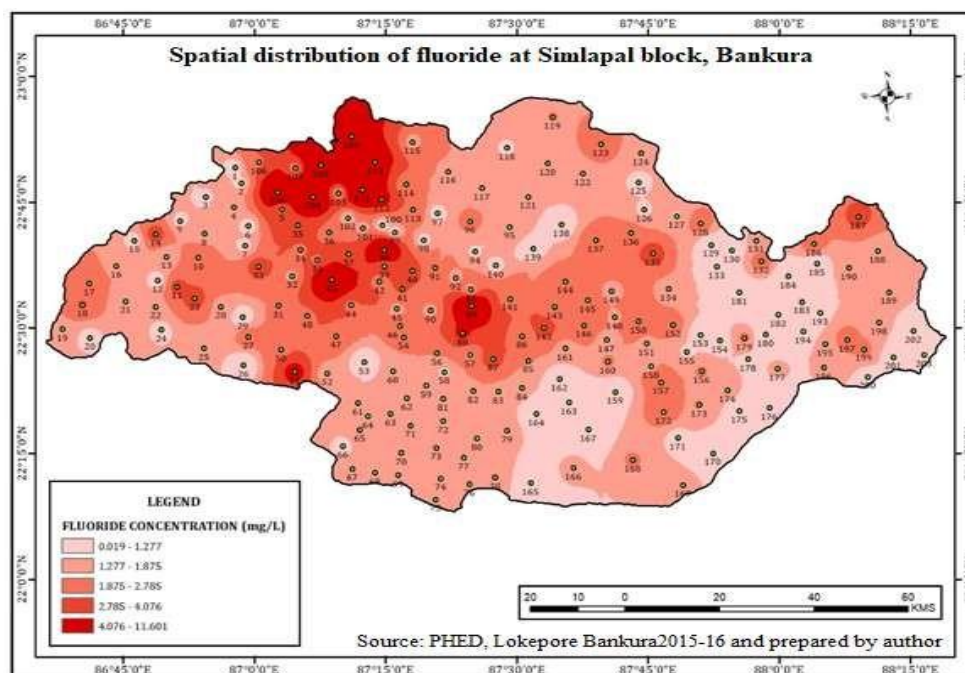


Fig. 3: Spatial distribution of fluoride at Simlatal block.

The main sources of waters are well, tube well and submersible. The table 3 shows the pattern of fluoride distribution of some selected villages (Jamda, Laxmisagar, Sainidanga, Brindabanpur and Machatora) with respect to depth of ground water.

Table 3: Nature of the water sources and fluoride level

Name of the village	Sample name	Sample No	Source of water	Depth (mts)	Fluoride (mg/L)
Jamda	J1	1	Tube Well	210	3.27
	J2	2	Well	42	1.69
	J3	3	Pond	4	0.89
	J4	4	Well	39	1.51
	J5	5	Submersible	190	2.87
	J6	6	Tube Well	180	10.51
Laxmisagar	L1	7	Well	38	2.22
	L2	8	Tube Well	175	2.78
	L3	9	Submersible	215	1.93

	L4	10	Tube Well	185	3.07
	L5	11	Well	37	1.53
Sainidanga	S1	12	Tube Well	200	4.64
	S2	13	Tube Well	180	3.96
	S3	14	Well	44	1.68
	S4	15	Well	39	1.43
	S5	16	Submersible (school)	190	2.14
Brindabanpur	B1	17	Tube Well	215	3.27
	B2	18	Well	41	1.59
	B3	19	Well	43	1.39
	B4	20	Tube Well (school)	220	2.98
	B5	21	Well	39	1.69
Machatora	M1	22	Well	43	1.41
	M2	23	Tube Well	190	10.39
	M3	24	Submersible	210	2.11
	M4	25	Tube Well (school)	185	3.17
	M5	26	Tube Well	195	2.14

The density concentration and the correlation between depth and fluoride concentration (mg/L) are positive (Fig. 4). Generally, water from greater depths is associated with higher levels of fluoride, while shallower water tends to have lower fluoride concentrations. This variation is likely due to rock-water interactions. Depth-wise fluoride variations in different water sources among the sampled villages are shown in Fig. 5a and Fig. 5b. Fluoride levels vary across different water sources, with the highest concentrations found in tube wells and the lowest in shallow sources like ponds. Fig. 5b also indicates that all water sources, except ponds and wells, generally have high depths.

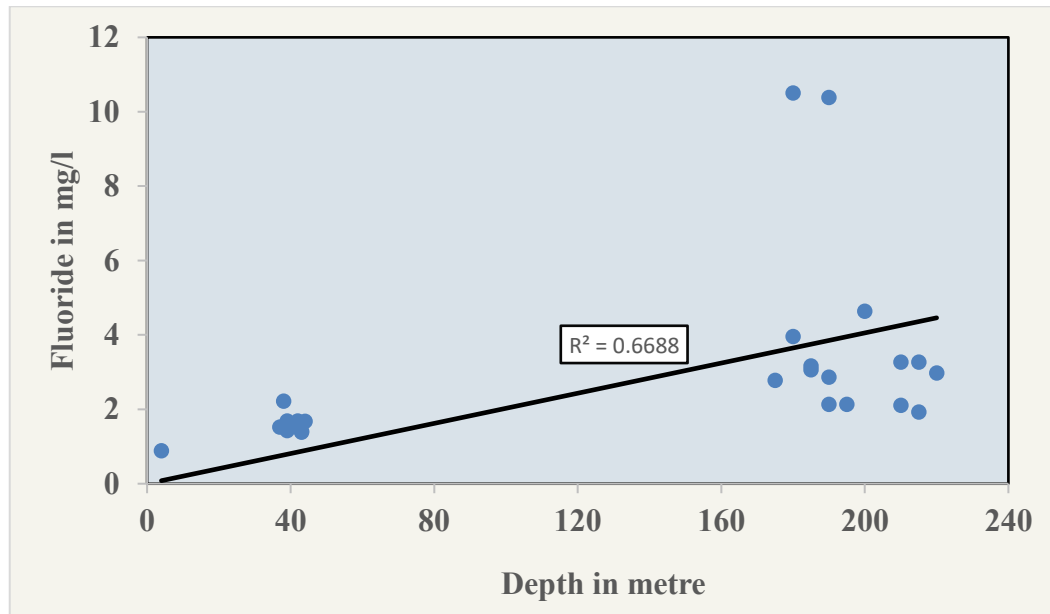


Fig. 4: Correlation between depth (meter) and fluoride concentration (mg/L) of sample water

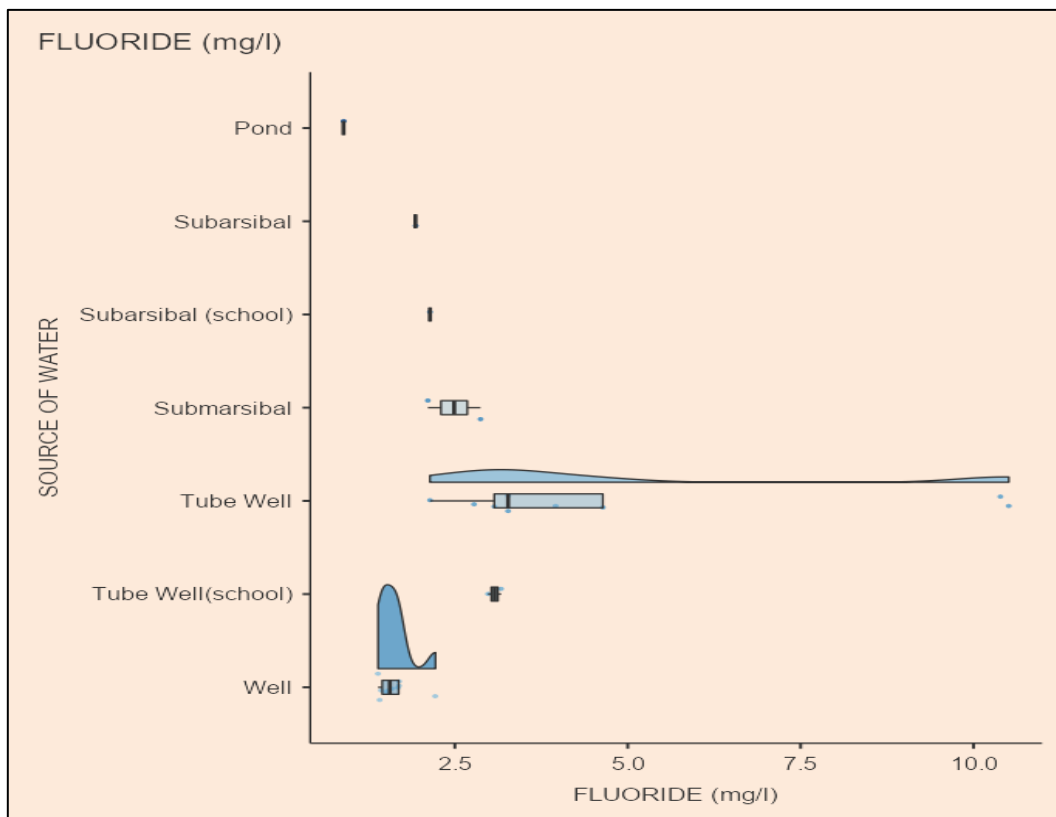


Fig. 5a: Source-wise fluoride variation among the sample water

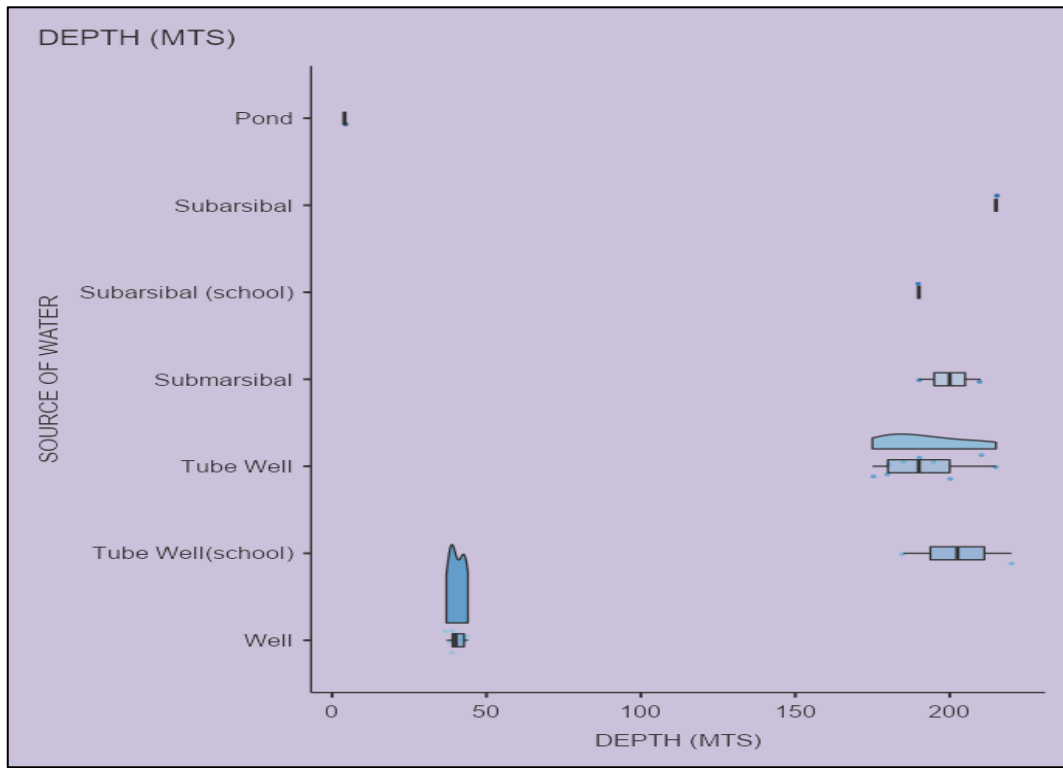


Fig. 5b: Different source-wise depth variations of sample water

3.2 Groundwater chemistry

3.2.1 Hydrochemical parameters

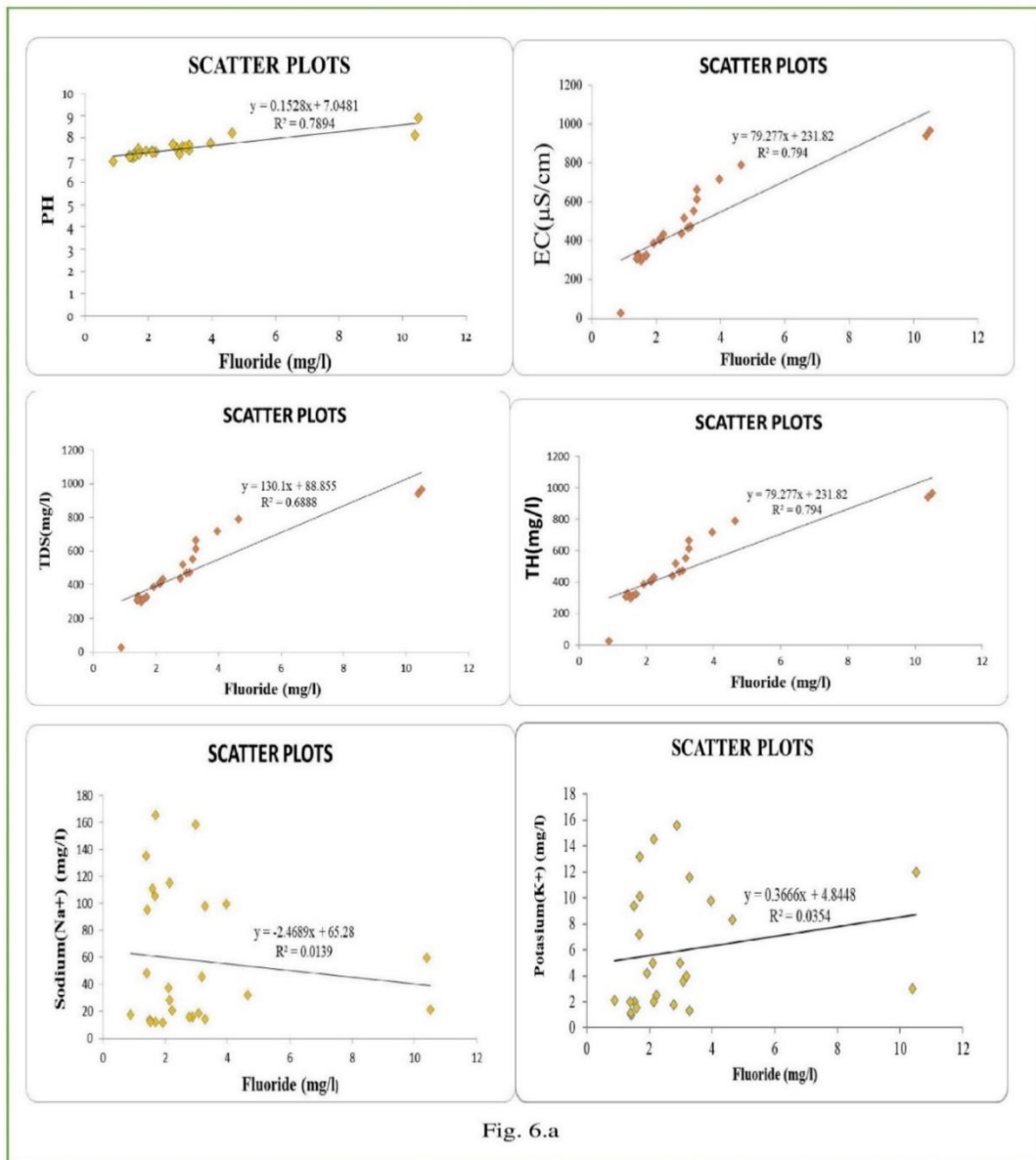
The experimental results of the hydrochemical parameters revealed that the geological and geochemical conditions of the research area caused significant fluctuations in all parameters, including fluoride (F-) concentration in the groundwater. Table 4 shows the distribution of different fluoride levels (mg/L) in the various samples collected from the selected villages in the study area. About 84.62% of the samples exceed the permissible limit for drinking water, while only 15.38% are considered safe. Most samples, 53.85%, fall within the range of 1.5 to 3.0 mg/L. Additionally, 23.08% of samples have fluoride levels between 3.0 and 5.0 mg/L, and 7.69% exceed 10.0 mg/L.

Table 4: Distribution of samples according to fluoride concentration

Fluoride level (mg/L)	Sample No	Total	Percent of Samples
<1.5	3, 15, 19, 22	4	15.38
1.5-3.0	2, 4, 5, 7, 8, 9, 11, 14, 14, 16, 18, 20, 21, 24, 26	14	53.85

3.0-5.0	1, 10, 12, 13, 17, 25	6	23.08
5.0-10.0	0	0	
>10.0	6, 23	2	7.69

Descriptive statistics for fluoride and other chemical parameters of the experimental samples are given in Table 5. Table 6 represents the correlation matrix (CM) in the experimented groundwater samples of Simlapal block, Bankura district. The degree of connection between the several physicochemical parameters that affect groundwater quality in the study area is ascertained using the correlation matrix (CM). A measure of how well one parameter predicts the value of another is provided by correlation analysis, which involves statistical computations (Kurumbein and Graybill, 1965). According to Chapman (1996), CM is a well-accepted and practical statistical method in the field of water chemistry for determining the positive and negative associations between ions. Strong positive correlations can indicate that certain ions originate from the same sources, whether they are man-made or natural (Islam et al., 2017). Conversely, weak correlations suggest that the sources of ions are unrelated to one another. Strong correlations are those where the correlation coefficient (r) is greater than 0.7; moderate correlations are those where the correlation coefficient (r) is between 0.3 and 0.7, and weak correlations are displayed by r values less than 0.3. Table 6 shows a positive association between EC and TDS ($r = 0.88$), F ($r = 0.89$), HCO_3 ($r = 0.94$), and TH ($r = 0.98$). This suggests that the dissolving of salts accelerates the electrical process of weathering. Rainfall and human-induced activities have an impact on the Mg, Na, TDS, Cl, and SO_4 inputs in the studied region. Furthermore, there is a substantial correlation shown by the following variables, such as TDS with TH ($r = 0.94$), HCO_3 ($r = 0.80$), and F ($r = 0.83$). In addition, TH shows a very significant positive correlation with both F ($r = 0.89$) and HCO_3 ($r = 0.86$). Moreover, there is a good correlation between Na and Ca ($r = 0.77$) and between HCO_3 and F ($r = 0.92$). A moderate link has been found between Mg and Na ($r = 0.63$) and Ca ($r = 0.50$), SO_4 and Na ($r = 0.35$), K ($r = 0.38$), Cl ($r = 0.37$) and Ca ($r = 0.38$). Furthermore, the substantial correlation with irrigation has been validated by Ca, Mg, Na, and Cl, EC, TH, and TDS. Fluoride is positively correlated with pH, EC, TDS, TH, K^+ , and HCO_3^- , while Na^+ , Cl^- , Ca^{2+} , Mg^{2+} , and SO_4^- are negatively correlated (Fig 6.a & Fig. 6.b).



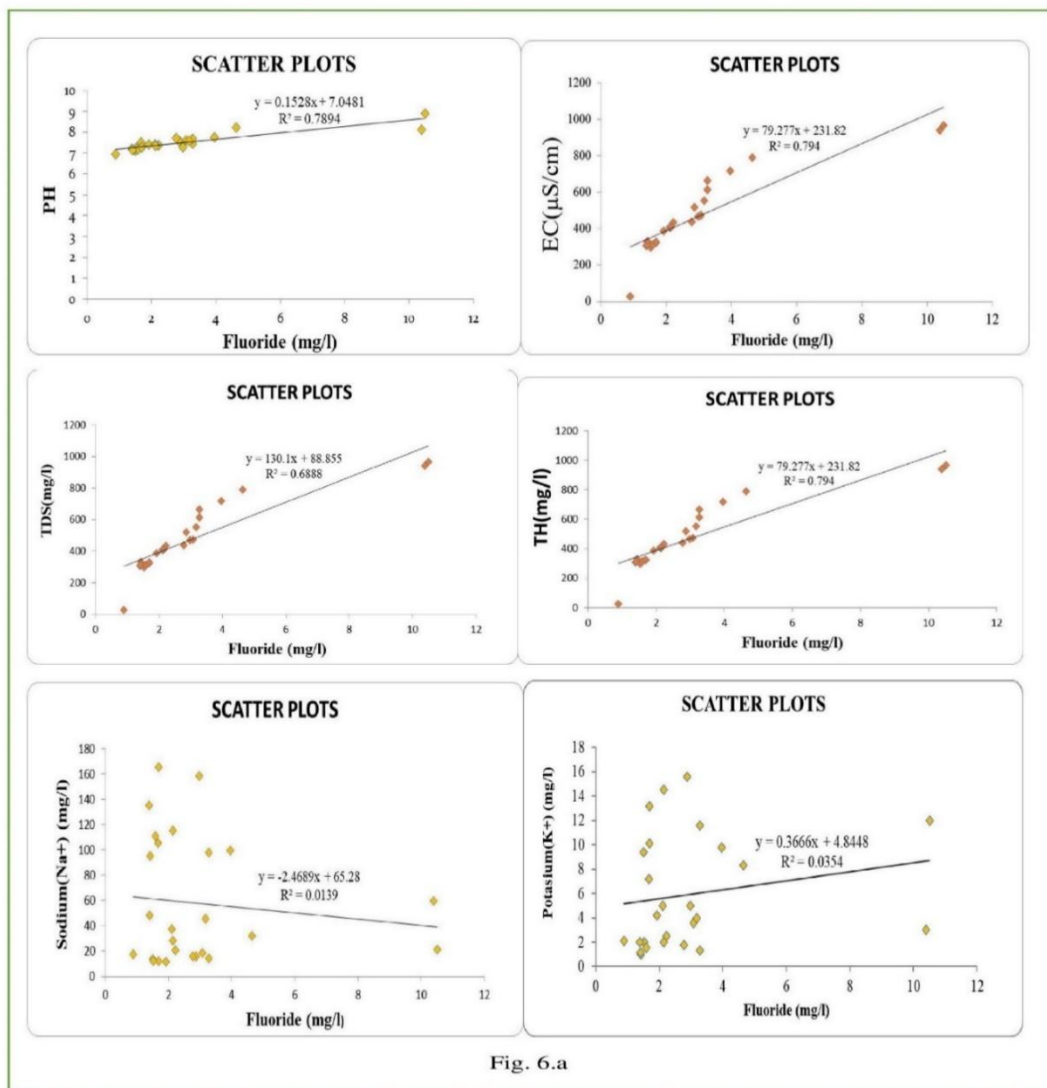


Fig. 6: Scatterplots showing relation between fluoride and other chemical parameters

Table 5: Summarized chemical values of collected water samples

	<i>P H</i>	<i>EC</i>	<i>TDS</i>	<i>TH</i>	<i>Na</i> +	<i>K</i> ⁺	<i>Cl</i> ⁻	<i>Ca</i> ²⁺ +	<i>Mg</i> ²⁺ +	<i>SO</i> ₄ ²⁻	<i>HCO</i> ₃ ⁻	<i>F</i> ⁻
<i>Max.</i>	8.89	3189.00	1209.60	967.00	130.00	15.60	232.20	165.20	88.63	40.30	298.00	10.51
<i>Min.</i>	6.93	337.90	33.00	27.00	4.00	1.00	13.60	11.80	9.88	2.12	59.00	0.89
<i>Mean</i>	7.50	974.73	470.46	464.35	48.50	5.92	133.11	58.04	29.55	16.27	125.58	2.93
<i>SD</i>	0.40	740.55	366.81	208.18	37.34	4.56	68.76	49.06	18.67	11.46	57.77	2.34
<i>CV</i> (%)	5.37	75.98	77.97	44.83	76.99	77.03	51.66	84.54	63.20	70.47	46.00	79.78

Note: EC is expressed as $\mu\text{S}/\text{cm}$, and all other parameters are expressed as mg/L

Table 6: Correlation matrix of the analysed chemical parameters

	pH	EC	TDS	TH	NA ⁺	K ⁺	CL ⁻	CA ²⁺	MG ²⁺	SO ₄ ²⁻	HCO ₃ ⁻	F ⁻
pH	1.00											
EC	0.90	1.00										
TDS	0.87	0.88	1.00									
TH	0.90	0.92	0.94	1.00								
NA ⁺	-0.14	-0.07	-0.06	-0.01	1.00							
K ⁺	0.29	0.19	0.24	0.26	-0.12	1.00						
CL ⁻	0.01	-0.13	-0.07	-0.05	-0.31	0.28	1.00					
CA ²⁺	-0.18	-0.09	-0.11	-0.07	0.77	-0.23	-0.23	1.00				
MG ²⁺	-0.09	-0.13	-0.06	-0.03	0.63	-0.44	-0.20	0.50	1.00			
SO ₄ ²⁻	0.02	0.00	-0.04	0.04	0.35	0.38	0.37	0.38	0.09	1.00		
HCO ₃ ⁻	0.86	0.94	0.80	0.86	0.06	0.09	-0.08	0.05	-0.01	0.09	1.00	
F ⁻	0.89	0.98	0.83	0.89	-0.07	0.19	-0.19	-0.12	-0.10	-0.09	0.92	1.00

Bold values indicate significant correlations among physicochemical parameters.

3.2.2 Types of groundwater based on hydrochemical characteristics

The Piper diagram was designed to depict the nature and quality of water from 26 samples collected throughout the study area, and it represents several chemical properties, primarily anions and cations. Two triangular sectors and one diamond-shaped sector are prominent in this diagram. The sector of cation parameters is

depicted as a single point in the left-hand triangle, with a total cation percentage in mg/L and anions in the right-hand triangle (Piper 1953).

Each valuable point is projected into the diamond-shaped field along a line parallel to the top sector, and the intersections represent the nature of water quality in the relationship between K^+ , Na^+ , Mg^{2+} , Ca^{2+} , HCO_3^- , CO_3^{2-} , and Cl^- , SO_4^{2-} ions. This diagram also emphasizes the homogeneity and heterogeneity of the different water samples because homogeneous water samples tend to plot as a single group. It also emphasizes that 75% of the groundwater samples were placed in the south-central part of the diamond sector which indicated the non-domination of only one cation or anion point (Fig. 7). The piper diagram depicts the geochemical facies of the various water samples collected from the study area. Samples are concentrated in the calcium-magnesium and sulphate-chloride zones. It is possible to conclude that the water in the study area is of the calcium chloride type.

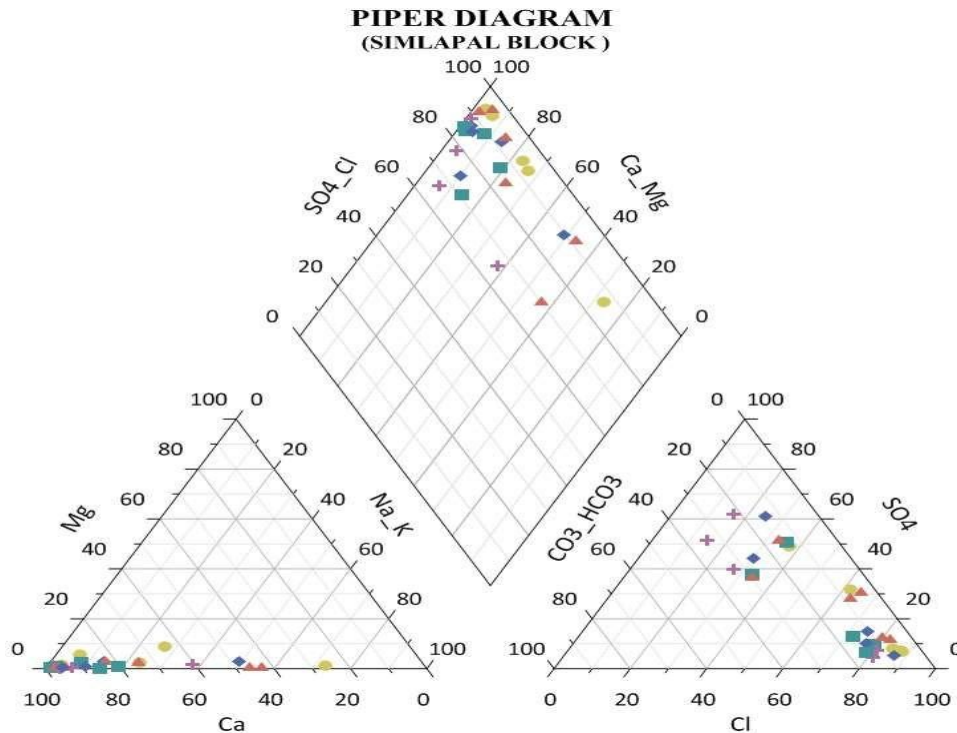


Fig. 7: Piper diagram showing the nature of the water

The samples plotted towards the dissolving or mixing line on the Durov diagram supported the assertion that mixed water types predominate in the research area (Fig. 8). According to Lloyd and Heathcoat's classification (1985), this trend can be attributed to fresh, recently recharged water that exhibits simple dissolving or mixing with no dominant major anion or cation. Most of the samples, however, are plotted in

fields 2, 4, and 5, indicating that the samples were either simply dissolved by the reverse cation exchange process or were directly recharged with surface water.

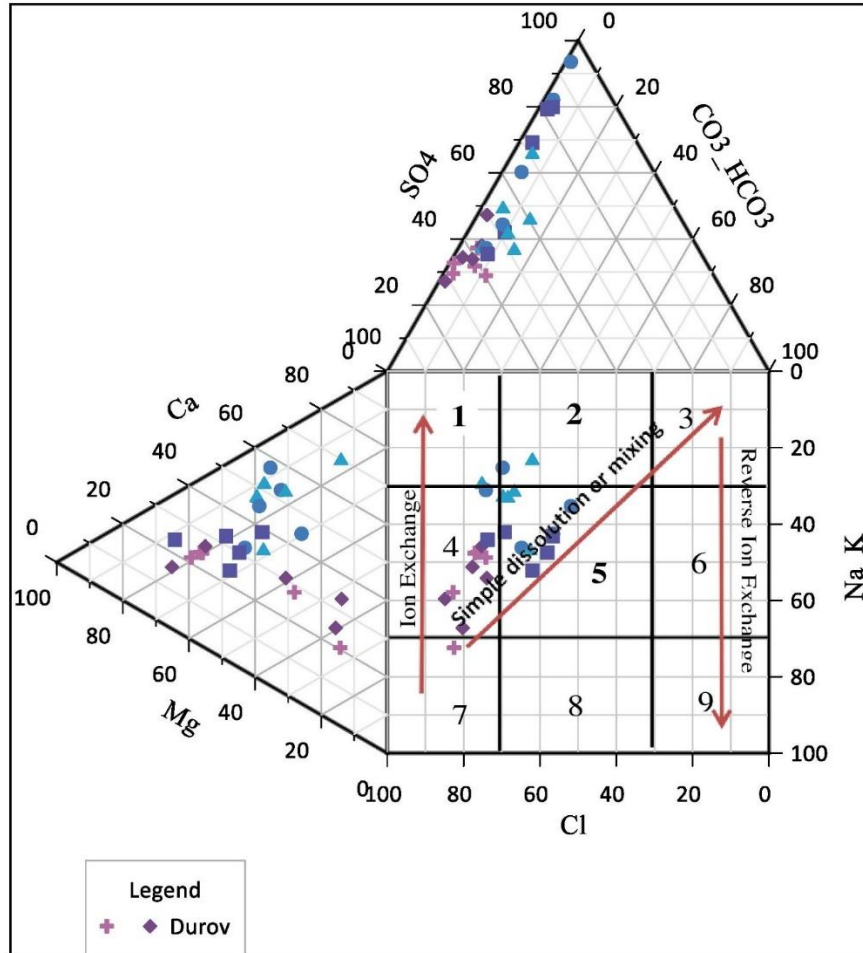


Fig. 8: Durov plot showing the involved hydrochemical processes

3.2.3 Rock water interaction of the groundwater

The Gibbs diagram depicts the interplay of rock and water in any given location. In fact, the type of lithological diversity is intimately related to water composition, as depicted by a Gibbs diagram. This diagram highlights three key fields: precipitation dominance, evaporation dominance, and rock water interaction dominance (Gibbs, 1970). Most of the water samples assembled from the study area fall into the rock dominance of the diagram (Fig. 9). The rock water sector denotes the collaboration between the infiltrated water chemistry and the different rock chemistry presence in the subsurface area also. In this figure, as the samples are intense rock water contact dominance sector the water chemistry is determined by the changes of facies at the time of groundwater interface with the nature of aquifer

materials though the interaction varies with the depth.

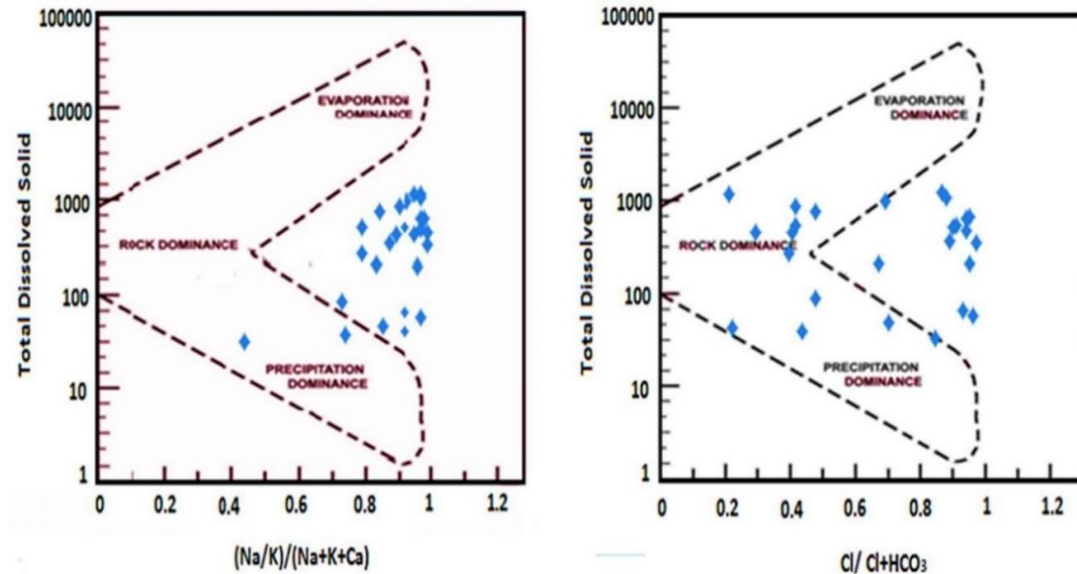


Fig. 9: Gibbs diagram of groundwater samples

3.3 Groundwater quality assessment

3.3.1 Groundwater quality for drinking

All natural water sources are essentially saturated with calcium carbonate, apart from rain and the ocean. The reason for this is that these natural supplies often contain a 10 mg/L concentration of fluoride, which is the concentration above which it becomes insoluble. We are all very familiar with the whitish-gray deposit that forms around faucet aerators, shower heads, tea kettles, and other appliances. This deposit is primarily calcium carbonate scale. To forecast the propensity of a specific water supply to generate a calcium carbonate scale, Langelier-Saturation Index (LSI) is used.

If the value of LSI is positive, a calcium carbonate scale may form but if the value is LSI is negative, the water will dissolve scale. These numbers, like pH, are logarithmic and every unit is a factor of ten. The groundwater quality of Simlapal block was evaluated using the following Langelier Saturation Index method. The LSI is calculated based on the carbonate equilibrium in groundwater (Taghavi et. al, 2019) and it is tabulated in Table 7.

$$LSI = pH + \log \left(\frac{K_a \cdot \gamma_{Ca^{2+}} \cdot [Ca^{2+}] \cdot \gamma_{HCO_3^-} \cdot [HCO_3^-]}{\gamma_{H^+} \cdot K_{sp}} \right) \dots\dots\dots(iii)$$

As per the Saturation Index the nature of variation and indication of water has also been shown in Table 8. About the 69.23% of samples are under < 0 and 30.77% of samples are under > 0 in respect of saturation index. It indicates that 34.62% of samples are serious corrosion and slightly corrosion but not scale forming respectively. On the other hand, slightly scale forming and corrosion and scale forming but non-corrosive of samples are under 30.77%.

Table 7: Saturation index of the water samples

Saturation Index	Samples fallen	Percentage	Remarks
< 0	1, 2, 3, 4, 5, 7, 8, 9, 10, 11, 15, 16, 18, 19, 21, 22, 24, 25,	69.23	In terms of calcium carbonate, water is undersaturated. Existing calcium carbonate protective coatings in pipes and equipment tend to be removed by undersaturated water.
0	Nil	0.0	Water is seen as neutral. Neither a scale maker nor a scale remover.
> 0	6, 12, 13, 14, 17, 20, 23, 26,	30.77	Scale formation is possible because water is supersaturated in calcium carbonate (CaCO_3).

Table 8: Nature of the water based on Saturation Index

Saturation Index	Samples fallen	Percentage	Indication
-2.0 to -0.50	1, 2, 3, 4, 5, 7, 9, 11, 22	34.62	Serious corrosion
-0.50 to 0	8, 10, 15, 16, 18, 19, 21, 24, 25	34.62	Slightly corrosion but non-scale forming
0	0	0	Balanced but pitting corrosion is possible
0 to 0.50	14, 17, 20, 26,	15.38	Slightly scale forming and corrosive
0.50 to 2.0	6, 12, 13, 23	15.38	Scale forming but non-corrosive

3.3.2 Groundwater quality for agriculture

In the Bankura district, groundwater is also the main supply of water for agricultural irrigation. When using groundwater for irrigation, salt and alkali dangers must be considered (Asadi et al, 2020, Zhao et al, 2020). Therefore, it is crucial to assess the

quality of groundwater before using it for irrigation in order to ensure the well-being of the soil and plants. A typical indicator to measure potential salt dangers in agricultural soil owing to irrigation is the sodium percentage (Na%) (Zhang et al, 2020). The high concentration of Na⁺ in irrigation water may impede air and water circulation in plant roots, increase the osmotic pressure of the soil, decrease soil permeability, and impact plant growth.

In this regard, sodium is one of the most striking components of water, as it affects soil penetrability and fertility. Irrigation is also dependent on the presence of sodium. The importance of irrigation can be estimated based on the sodium percentage.

The formula of sodium percentage is-

$$\text{Na}\% = \frac{(\text{Na} + \text{K}) \times 100}{(\text{Ca} + \text{Mg} + \text{Na} + \text{K})} \text{ mg/L} \dots\dots\dots (\text{iv})$$

Table 9: Water suitability classes according to sodium percent.

Na%	Description	Samples fallen	% of samples
< 20	Excellent	7, 9, 10	15.38
20 – 40	Good	8, 11, 12, 13, 14, 15, 18	38.46
40 – 60	Permissible	1, 2, 3, 4, 6, 17, 19	38.46
60 – 80	Doubtful	5,16	7.69
> 80	Unsuitable	0	0

In this calculation, the concentration of ions is expressed in mg/L. The summarized result is shown in Table 9 and highlights that only 7.69% of samples are doubtful in position in respect of irrigation, 38.46% of the samples are permissible and about 53.84% (15.38+38.46) of samples are good for irrigation purposes.

3.4 Human health risk assessment

3.4.1 Non carcinogenic health risk

Different samples from the study area have significant amounts of fluoride contamination. Due to ingesting the contaminated water for an extended length of time, the inhabitants in the research area frequently experience the detrimental health effects of fluoride content in drinking water (Ahada and Sathur, 2019; Kadam et al., 2022).

The health impacts on research participants' residents according to different amounts of fluoride exposure from drinking groundwater are shown in Fig. 10.

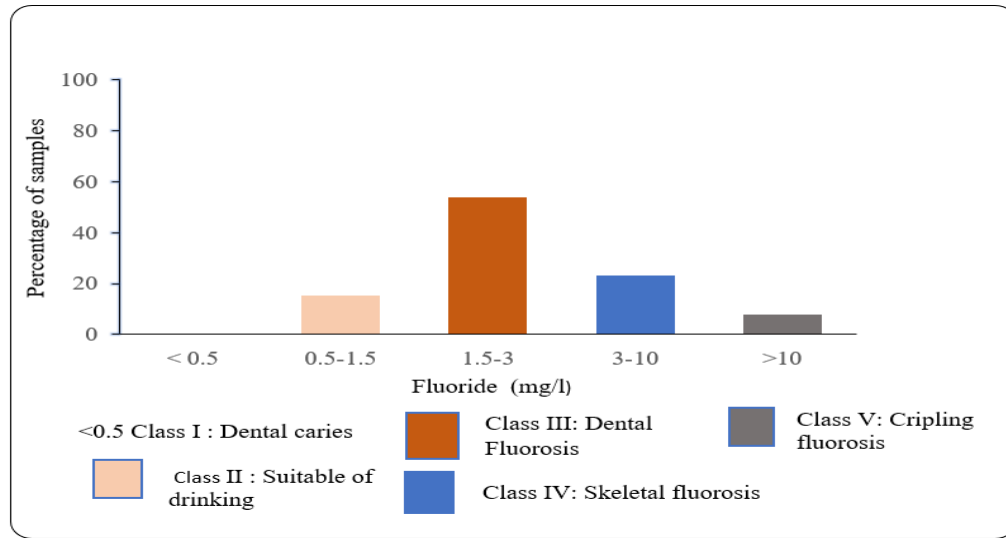


Fig. 10 Effects of fluoride toxicity due to different levels of fluoride intake

There is no class I sample, indicating that there is no risk of dental issues. There are 6 samples in class II, or 15.38% of all samples, with fluoride concentrations between the recommended range (0.5 and 1.5 mg/L) for excellent human health. 53.85% of samples fall under class III, which could lead to various forms of dental fluorosis. Contrarily, class IV and class V together make up roughly 30.77% of the total samples, suggesting that extended consumption of groundwater may represent harm to one's health by causing calcification of ligaments, non-skeletal fluorosis disorders, and crippling of the bones (Genu valgum and Genu verum).

The highest fluoride concentration (10.51 mg/L and 10.39 mg/L) found in the groundwater samples of this area is compatible with the maximum CDI value seen in both adults and children at Jamda and Machatora localities, while it is medium in the villages of Sainidanga and Brindabanpur.

For a better assessment of fluoride health risk to the human individual residing in the Simlapal block of Bankura district, the magnitude hazard index i.e., HQ_{Fluoride} was calculated for the different age and sex groups separately (Table 9). The magnitude of hazard index, HQ_{Fluoride} of more than one indicates a high chance of development of fluoride induced health hazards to the individual living. HQ_{Fluoride} of less than one indicates less health risk.

However, compared to adult people, children have the highest levels of HQ_{Fluoride} because children's have smaller bodies, which tend to store more pollutants, due to fluoride exposure (He and Wu, 2019).

Table 9: Hazard Quotient of Fluoride for various people

	CDI (Chronic Daily Intake) mg/kg/day	HQ _{Fluoride}
Male	0.05-0.65	0.9-10.8
Female	0.05-0.57	0.8-9.6
Children	0.09-0.70	0.99-11.7

The majority of groundwater samples taken from the Simlapal block have HQ_{Fluoride} values above the unitary value, resulting in fluorosis-related health concerns for both children and adults living there. However, fluoride toxicity is affected by several factors, including the amount of fluoride in groundwater, the rate of consumption, the duration of fluoride exposure, and the regional climate (particularly temperature).

Similarly, the groundwater fluoride health risk to individual in the current study is higher or similar to most of the global fluoride endemic hotspots viz China (Chen et al. 2017), Iran (Yousefi et al., 2018), Kenya (Mwiathi et al., 2022), Pakistan (Noor et al., 2021), Mexico (Fernandez-Macias et al., 2020), Tunisia (Guissouma et al., 2017), and Vietnam (Nguyen et al., 2021) including Northern and Southern India (Ahada and Sathur, 2019 and Kumar et al., 2019; Nizam et al., 2022; Shukla and Saxena, 2022).

4.3.2 Implication for chronic health risk

The various chronic health conditions, such as liver damage, high blood pressure, respiratory failure, blood in the stool, paralysis, and chronic fluoride poisoning, which causes anemia, tingling, and weight loss, are frequently brought on by drinking high fluoride water for an extended period of time. High fluoride intake also has an impact on men's reproductive and fertility systems (Ortiz-Perez et al., 2003). Children's visuospatial skills are severely impacted by fluoride intake of 2-4 mg/L, which results in poorer IQ scores on the real-time exam (Arvinda et al., 2016). The widespread presence of high fluoride levels in the groundwater systems of Simlapal block in the Bankura district indicates that the situation is likely to worsen soon. To deal with the current fluoride contamination scenario, an appropriate water management plan must be implemented immediately.

4. Conclusions

This paper provides a general review of groundwater pollution by fluoride and related health risks. The groundwater samples' hydrogeochemical properties and spatial distribution underwent rigorous examination. About 84.62% of the groundwater samples examined had fluoride concentrations more than 1.5 mg/L. Higher concentrations are found in deeper aquifers than in shallower aquifers, which is likely due to contamination moving downward through the surface. The

most common type of water contains calcium chloride. The main factors influencing groundwater hydrochemical properties are rock-water interaction and rock weathering. According to the Saturation Index, 34.62% of samples exhibit severe corrosion and minor corrosion but not scale formation. On the other side, samples that are only mildly corrosive, scale-forming, and/or scale-forming but not corrosive are under 30.77%. According to Na%, just 7.69% of samples are in doubt regarding their suitability for irrigation, 38.46% of samples are acceptable, and approximately 53.84 percent of samples are suitable. The residents of the study region were exposed to a persistent fluoride health risk, as shown by groundwater fluoride concentration and probabilistic health risk. According to the HQ_{Fluoride} index, children and teenagers are more susceptible to fluoride poisoning than adults. The government and management organizations can use the information provided by this study to safeguard the local people who are exposed to groundwater in the study area.

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