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Toxicity of Chemical Substances in Groundwater of Jhunjhunu District of Rajasthan, India: A Physicochemical Analysis

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Abstract

Freshwater habitats are a lifeline of all living organisms. According to the ICMR standard, a 0.6 mg/l fluoride concentration is required for human teeth and bones while more than 1.5 mg/l fluoride concentration is responsible for various human diseases. 42 groundwater and deep wells samples have been collected from Chirawa, Buhana, and Surajgarh of the Jhunjhunu district, Rajasthan for the meta-assessment of fluoride concentration. All water samples are analyzed through physiochemical methods. Fluoride concentration was determined with the help of the APHA-23nd 2017 method. The highest concentration of fluoride was found at 1.4 mg/l of Kakoda (Sample number 16) while the lower concentration of fluoride was found at 0.7 1.4 mg/l of three places as Pilani water box, Morva, and Chirawa CHC (Sample numbers 1, 10, 30). The fluoride concentration of the remaining places was found suitable for human beings. The lower total hardness has been found at 120 mg/l in S-1 while a maximum of 296 mg/l in S-14.

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1. Introduction

Globally, maintaining water quality is a major challenge due to increasing industrialization, over-exploitation, deforestation, urbanization, ecosystem degradation, and depletion of the water bodies due to the increasingly sophisticated living standards of a growing population.[1,2] These freshwater habitats are less than 1% of the world's total surface area and are mostly stored in glaciers, rivers, lakes, reservoirs, ponds, streams, and the underground of the earth. Yet these 10% houses of all known animals and up to 40% of all known fish species.[3,4] Natural portable water of excellent quality is found in some places the world over while few places have impurities included in the water, therefore, people cannot drink it directly. The impurities include arsenic, fluoride,[5,6] and some heavy metals and organic and inorganic chemicals dissolved directly or indirectly.[7,8] Few countries are not providing purified water for their citizens due to a lack of facilities,[9,10] but pretreatment of water should be required for drinking.[11-13] 70% of the total groundwater is used for irrigation, and the quantity of groundwater taken out is estimated to be 1100 km^3 per year by 2050 as compared to 800 km^3 per year as of 2010.[14] Fluorine is the more electronegative element in the periodic table so it has high reactivity. Therefore, it is not found in elemental form rather it is found in ionic form. If water has a pH value less than 5, it tends to form complexes with metal ions, while at higher pH values it tends to exist as a F^- ion. Natural sources are connected to various types of rocks and to volcanic activity. Agricultural (use of phosphatic fertilizers) and industrial activities (clays used in ceramic industries or burning of coals) also contribute to high fluoride concentrations in the groundwater.[15-17] Low or high fluoride ion concentrations are a major issue as they make groundwater unsuitable for various purposes. Fluoride ion causes health problems in people in about 25 countries around the world. At least 0.6 mg/l fluoride concentration is required for human consumption. It strengthens teeth and bones. More than 1.5 mg/l fluoride concentration in groundwater, on the other hand, results in issues such as acute to chronic dental fluorosis where the tooth

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becomes colored from yellow to brown. It also causes skeletal fluorosis which manifests as weakness and bending of the backbone also called "Hunch back disease". The World Health Organization (WHO) has recommended a 0.5 to 1 mg/L fluoride concentration limit for drinkable water. The pooled fluoride concentration in India is approximately 2.37 mg/L with a 95% confidence interval (1.46–3.28 mg/L), which is higher than the WHO and national standard limit of 1.5 mg/L. According to the meta-analysis of data from a rural area in India, fluoride concentration is 1.85 times higher than in urban areas. The fluoride concentration in groundwater decreased pointedly ($p < 0.05$) with the increase in rainfall and longitude. The risk assessment via oral and dermal route exposure reveals that the consumers in the majority of the regions are at considerable non-carcinogenic risk and children are at higher risk than adults (total hazard quotient > 1).[18-20] According to a 2012 World Bank report, 85% population of India is dependent on groundwater for drinking; therefore, the quality of groundwater becomes an important issue. Groundwater in India was assumed to be generally safe and widely used for drinking without proper risk assessment. Globally, 3.6 billion people do not get adequate drinkable water for at least 1 month a year.[14] Further, 260 million people are drinking water with high fluoride concentrations, above the WHO standard, and fluoride concentrations were found greater than 1.5 mg/l in drinking water.[21] Fluoride concentration in groundwater undergoes seasonal fluctuation; it tends to increase in the pre-monsoon season. The medium brown sandy soil and deep dark brown sandy soil consist of high fluoride concentration. The kharif and rabi crops are grown in the Jhunjhunu district, but the three types of crops are also grown in some regions with a high level of fluoride. Fertilizers are also expected to contribute toward high fluoride concentration.[22] The present research finds that Pearson's correlation coefficient s(r) was calculated among fluoride and other parameters. It was observed that fluoride content is exhibiting a statistically significant positive correlation with sodium, iron, and copper contents. The results are helpful in identifying the affected areas of India.

2. Experimental

2.1 Study area

Jhunjhunu district is situated in the northern portion of Rajasthan. It is bounded in the northwest of the Churu district, in the eastern state of Haryana, and in the southwest by Sikar. It stretches between 27º38'13.88" to 28º31'11.09" North latitude and 75º01'30.74" to 76º06'01.47" East longitude covering an approximate 5,911.1 sq km area. A systematic drainage system is not developed in the major part of the district, except for a strip in the center of the district running northwest to southeast, which is part of the Shekhawati River Basin, the remaining part of the district both in the east and west the region forms part of a so-called Outside Basin.

2.2 Climate condition

The climate of the Jhunjhunu district is largely arid. Extremely hot summers and cold winters with generally poor but occasionally good rainfall are observed during the southwest monsoon period. In May and June, the maximum temperature may sometimes go up to 48 °C. Winter temperatures drop down to 3 °C to 4 °C. The potential evapotranspiration rates are quite high, especially during May and June. The average annual rainfall of this district is about 440.0 mm.

2.3 Materials and methods

The area of Chirawa, Buhana, and Surajgarh tehsils of Jhunjhunu district comprises different types of Archaean crystalline formation. 2-liter plastic bottles have been washed with dilute HNO_3 and then ringed to distilled water before the sample collection. 42 samples of ground-water were randomly collected in precleaned polyethylene (Thermo-plastics) bottles from villages and towns of three tehsils including Chirawa, Buhana, and Surajgarh of Jhunjhunu district of Rajasthan. All water samples were collected in 2-liter plastic bottles because plastic reactivity was zero to groundwater samples and their dissolved minerals. All sample analyses have been completed within 8 to 10 hours. All samples were determined for physicochemical parameters using a standard technique: a water testing kit (APHA-23ND, 2017) and APHA, 2005, as shown in Table 2. All values of samples are shown in Tables 1 and 2. pH values and electrical conductivities of water are determined through a pH meter and digital conductivity meter. Fluoride concentration was determined with the help of APHA-23nd 2017, and the statistical analysis was carried out by Karl Pearson's coefficient method, shown in Tables 3 and 4.

3. Results and discussion

3.1 Physico-chemical properties of the groundwater samples

The groundwater qualities should be determined for domestic, industrial, and agricultural use before the supply. The physicochemical properties of all water samples are shown in Supporting Materials 1. The lower pH value has been found at 7.46 for sample S-26 while the maximum is 7.75 for sample S-8. The IS-3025(P-11) method has been used for pH calculation. The lower conductance of water has been found at 568 μS/cm in sample S-1 while the maximum is 1216 μS/cm in sample S-42. IS-3025(P-14) method has been used for the conductance of water. The IS-3025(P-10) method was used for the calculation of turbidity values of water. The lower turbidities of water have been found at 1. 5 mg/l in S-8, S-9, and S-10 while the maximum is found at 6 mg/l in S-41 and S-42. The total dissolved solid in water has been measured with the help of the IS-3025(P-16) method. TDS is a very important property of water. Lots of water purifier companies have focused on TDS. In the current research, 926 mg/l maximum TDS has been found in S-14 while lower 426 mg/l TDS has been found in S-1. The total hardness of water is an important property. The APHA-23nd Ed-2017 method has

Table 1. Physico-chemical parameters and their refereed methods.

S. No.	Biochemical Attribute	Refereed Method	Desirable Limit	Permissible Limit	Minimum values in the present research	Maximum values in the present research
1	pH	Digital pH Meter	6.5 – 8.5	No Relaxation	7.4	7.90
2	Electrical Conductance	Digital Conductivity Meter	400 µS/cm	No Relaxation	568 µS/cm	1216 µS/cm
3	Turbidity, NTU.	2130 B. Nephelometric Method (APHA, 2005)	1 NTU	5 Max	2 NTU	6 NTU
4	Total Dissolved Solids	2540 B. Total Solids Dried at 103–105 °C (APHA, 2005)	500 mg/litre	2000 Max	426 mg/litre	926 mg/litre
5	Total Hardness	2340 C. EDTA Titrimetric Method (APHA, 2005)	200 mg/litre	600 Max	120 mg/litre	296 mg/lite
6	Total Alkalinity	2320 B. Titration Method (APHA, 2005)	200 mg/litre	600 Max	190 mg/litre	390 mg/litre
7	Sulphate Content	4500-SO_4^{2-} E. Turbidimetric Method (APHA, 2005)	200 mg/litre	400 Max	28.5 mg/litre	58.8 mg/litre
8	Nitrate Content	4500-NO^{3-} B. Ultraviolet Spectrophotometric Screening Method (APHA, 2005)	45 mg/litre	No Relaxation	4.5 mg/litre	7.4 mg/litre
9	Total Phosphate Content	4500-P D. Stannous Chloride Method (APHA, 2005)	10 – 40 µg/litre	No Relaxation	1.0 mg/litre	1.6 mg/litre
10	Chloride Content	4500-Cl^{-} B. Argentometric Method (APHA, 2005)	250	1000 Max.	68 mg/litre	490 mg/litre
11	Fluoride Content	4500-F^{-} D. SPADNS Method (APHA, 2005)	1.0 mg/litre	1.5 Max	0.7 mg/litre	1.4 mg/litre
12	Sodium Content	3500-Na B. Flame Emission Photometric Method (APHA, 2005)	30 µg/litre	No Relaxation	107 mg/litre	186 mg/litre
13	Potassium Content	3500-K B. Flame Photometric Method (APHA, 2005)	12 µg/litre	No Relaxation	91 mg/litre	170 mg/litre
14	Ferrous Content	3111 B. Direct Air-Acetylene Flame Method (APHA, 2005)	0.3 mg/litre	No Relaxation	0.44 mg/litre	1.22 mg/litre
15	Copper Content	3111 B. Direct Air-Acetylene Flame Method (APHA, 2005)	0.1 mg/litre	0.3 Max	1.75 mg/litre	4.01 mg/litre

been used for measuring total hardness. The total hardness is found below 75 mg/l, which is generally considered soft, 76 to 150 mg/l moderately hard, and 151 to 300 mg/l hard, if more than 300 mg/ is very hard. The lower total hardness has been found at 120 mg/l in S-1 while a maximum of 296 mg/l in S-14. The lower total hardness has been found approximately equal to the moderate hard standard limit. The total alkalinity of water has been measured by IS-3025(P-23) method. The maximum 390 mg/l alkalinities of water have been found in S-15 while the lower 190 mg/l in S-28 and S-29. All sulfate contents are calculated through APHA-23nd Ed-2017 methods. The 58.8 mg/l higher sulfate contents are obtained from S-14, and 28.5 mg/l from S-1 and S-10. All nitrate contents are measured by IS-3025(P-34) methods. The lower 4.8 mg/l nitrate contents are found from S-1, while higher 6. 9 mg/l nitrate contents are obtained from S-14 and S-16. the rich contents of the nitrate and total phosphate in the sampling sites of the Surajgarh and Buhana tehsils are being influenced by the agriculture practices of the nearby Haryana state. The total phosphate content such as PO_4 HPO_4, and H_2PO_4 is measured by the APHA-23nd Ed-2017 method. The maximum 1.6 mg/l phosphate content is obtained from S-14 while the lower values at 1 mg/l from S-8, S-9, S-10, S-11, S-12, S-31, S-32, S-33, and S-35. A possible reason is predicted that unlawfully, sewage in urban areas is being disposed into the groundwater. It is a popular practice, where proper disposal facilities for sewage are not maintained. This action may be causing a hike in total phosphate content in the study area.

The chloride contents are determined through the APHA-23nd Ed-2017 method. The higher 486 mg/l chloride contents are obtained from S-14 while the lower 68 mg/l contents from S-31. The APHA-23nd Ed-2017 method has been used for fluoride contents. The lower 0.7 mg/l fluoride contents are obtained from S-30 while 1.4 mg/l from S-16. The fluoride content is lower or equal to the standard limits of all samples. The sodium contents are determined with the help of the IS-3025(P-45) method. The higher 186 mg/l values of sodium contents are obtained from S-14, S-15, and S-42 while the lower value is 112 mg/l from S-34. Further, all minimum and maximum values of current research have been shown in Table 1. The F^- concentration in groundwater from various regions in India have higher than the maximum permissible limits prescribed by BIS and WHO (*i.e.* 1.5 mg/L) such as 5.98 mg/L in Madhya Pradesh, 4.67 mg/L in Punjab, 3.80 mg/L in Chattisgarh, 3.30 mg/L in Rajasthan, 2.56 mg/L I Karnataka, 2.03 mg/L in Andhra Pradesh, 1.70 mg/L in Maharashtra, 1.65 mg/L in Haryana, 1.64 mg/L in Uttar Pradesh, and 1.54 mg/L in Bihar.[23]

3.2 Statistical analysis

Correlation refers to the statistical relationship between two entities. In other words, it is how two variables move in relation to one another, or correlation denotes the association between two quantitative variables. Correlation is one of the most common and useful statistics. A correlation is a single number that describes the degree of relationship between two variables. The degree of association is measured by a correlation coefficient, denoted by "r" and measures the linear association. The correlation coefficient is measured on a scale that varies from "+1" through "0" to "-1". A complete correlation between two variables is expressed by either "+1" or "-1". In the present research work, the correlation among the various water quality parameters was analyzed to know the relationship and status of an individual water quality parameter. Generally, water quality parameters exhibit a relationship with other water quality parameters due to chemical or physical affinities. For such purposes, statistical software calculated Pearson's correlation coefficients (r). The Pearson's correlation coefficient (r) values varied from "+1" to "-1". The "+1" value reveals a strong positive correlation and the "-1" value reveals a strong negative correlation. Statistical analyses were carried out with the help of Pearson's coefficient. And their results were examined in graphic images in Figs. 1-4. The correlation was calculated among different physico-chemical parameters to approach precise and conclusive results. Based on statistical significance, the Pearson's correlation coefficient (r) values are expressed in Table 2.

Table 2. The Pearson's correlation coefficient (r) values and their statistical significance.

S. No.	r -value	Statistical Conclusion
1	0.9 - 1.0	Very high positive (negative) correlation
2	0.7 - 0.9	High positive (negative) correlation
3	0.5 - 0.7	Moderate positive (negative) correlation
4	0.3 - 0.5	Low positive (negative) correlation
5	0.0 - 0.3	Negligible correlation

The bedrock of granite/gneisses consists of 0–3.3% fluorite, 0.1–1.7% biotite, and 0.1–1.1% hornblende fluoride-containing minerals. These bedrocks are covered by the Delhi supergroup having quartzite and schists, having a high concentration of biotite and muscovite, and having high solubility. Hence, the major reason for elevated fluoride concentration in the study area can be attributed to weathering of rocks or rock-water interactions. The hydrogeological description by CGWB (2017) shows that quartzite rocks are exposed to the surface in the eastern and northern parts and these are regions where an elevated fluoride concentration is noted. Generally, a higher pH value favors the enrichment of fluoride in groundwater as OH^- replaces F^- of clay minerals. The fluoride content of groundwater has increased with an increase in pH values. When groundwater with an elevated pH interacts with underlying rocks the OH− replaces the fluoride causing an enrichment in fluoride content. The interactions of rocks such as biotite, muscovite, and fluorite with groundwater are elucidated in equations 1-4.[20-22]

$$KMg_3AlSi_3O_{10})F_2 \rightarrow 2\ OH^- \ KMg_3AlSi_3O_{10})\ (OH^-)_2 + 2F^- + CO_2 \quad (1)$$

$$\text{Biotite } KAl_2AlSi_3O_{10})F_2 + 2\ OH^- \rightarrow KAl_2AlSi_3O_{10})\ (OH^-)_2 + 2F^- \quad (2)$$

$$\text{Muscovite } CaF_2 + 2{\cdot}HCO_3 \rightarrow CaCO_3 + 2F^- + H_2O + CO_2 \quad (3)$$

$$\text{Fluorite } CaF_2 + 2NaHCO_3 \rightarrow CaCO_3 + 2Na^+ + 2F^- + H_2O + CO_2 \quad (4)$$

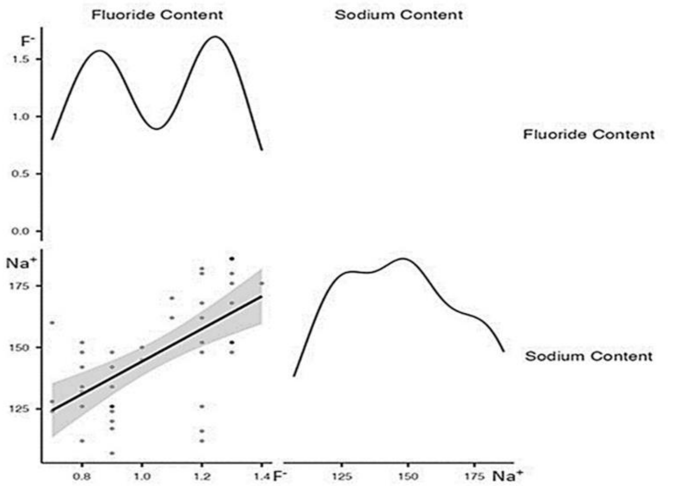

Fig. 1 Correlation curve between fluoride content and sodium content.

In the present research work, correlation coefficient values are calculated in a sequential manner by keeping stable a physico-chemical property or water quality parameter against the other physico-chemical properties as shown in Figs. 1-4. The detailed analysis of various physico-chemical properties is covered in successive headings. Pearson's correlation coefficients (r) were calculated among fluoride content and other water quality parameters as shown in Table 3. It was observed that fluoride content exhibits a statistically significant positive correlation with sodium, iron, and copper content. Pearson's correlation coefficients (r) were calculated among pH and other water quality parameters. It was observed that in the first place, pH does not express any statistically significant correlation with other physico-chemical parameters, since pH does not directly affect the several physico-chemical parameters. The correlation study for the

Table 3. Correlation coefficient (r) value among different Physico-chemical parameters.

r Value	pH	EC	Turbidity	TDS	TH	TA	SO_4^{2-}	NO_3^-	T.P.	Cl^-	F^-	Na^+	K^+
pH	1												
EC	0.122	1											
Turbidity	0.173	0.911	1										
TDS	0.141	0.989	0.906	1									
TH	0.402	0.765	0.738	0.782	1								
TA	-0.038	0.677	0.419	0.695	0.485	1							
$SO4^{2-}$	0.049	0.908	0.844	0.905	0.578	0.638	1						
NO_3^-	-0.087	0.816	0.799	0.796	0.473	0.471	0.823	1					
T.P.	0.195	0.776	0.700	0.791	0.594	0.590	0.746	0.531	1				
Cl^-	0.206	0.928	0.833	0.941	0.812	0.727	0.833	0.714	0.780	1			
F^-	0.007	0.839	0.797	0.824	0.561	0.459	0.790	0.755	0.503	0.709	1		
Na^+	0.099	0.807	0.598	0.816	0.556	0.816	0.746	0.567	0.713	0.831	0.612	1	
K^+	-0.100	0.560	0.363	0.564	0.179	0.763	0.574	0.463	0.490	0.573	0.420	0.852	1

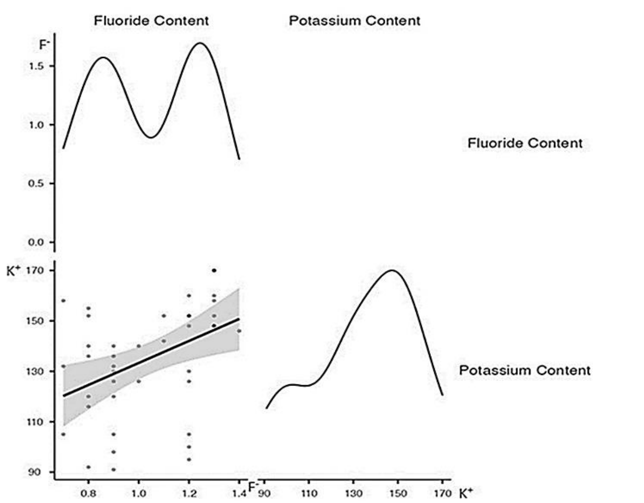

Fig. 2 Correlation curve between fluoride content and potassium content.

physico-chemical analysis is indicating that conductance keeps a positive relationship with other water quality parameters. Therefore, the measurement of EC can reveal the water quality in the first place.

Pearson's correlation coefficients (r) were calculated among EC and other water quality parameters. It was observed that EC is exhibiting a statistically significant positive correlation with all the physico-chemical parameters including turbidity, total dissolved solids, total hardness, total alkalinity, sulfate content, nitrate content, total phosphate content, chloride content, fluoride content, sodium content, potassium content, iron content, and copper content. Pearson's correlation coefficients (r) were calculated among turbidity and other water quality parameters. It was observed that turbidity is exhibiting an artistically significant correlation with total dissolved solids, total hardness, sulfate content, nitrate content, total phosphate content, chloride content, fluoride content, sodium content, iron content, and copper content. Pearson's correlation coefficients (r) were calculated among total dissolved solids and other water-quality parameters. It was observed that total dissolved solids are exhibiting a statistically significant positive correlation with total hardness, total alkalinity, sulfate content, nitrate content, total phosphate content, chloride content, fluoride content, sodium content, potassium content, iron content, and copper content. Pearson's correlation coefficients (r) were calculated among total hardness and other water quality parameters. It was observed that total hardness is exhibiting a statistically significant positive correlation with sulfate content, total phosphate content, chloride content, fluoride content, sodium content, iron content, and copper content. Pearson's correlation coefficients (r) were calculated among total alkalinity and other water quality parameters. It was observed that total alkalinity is exhibiting a statistically significant positive correlation with sulfate content, total phosphate content, chloride content, sodium content, potassium content, iron content, and copper content.

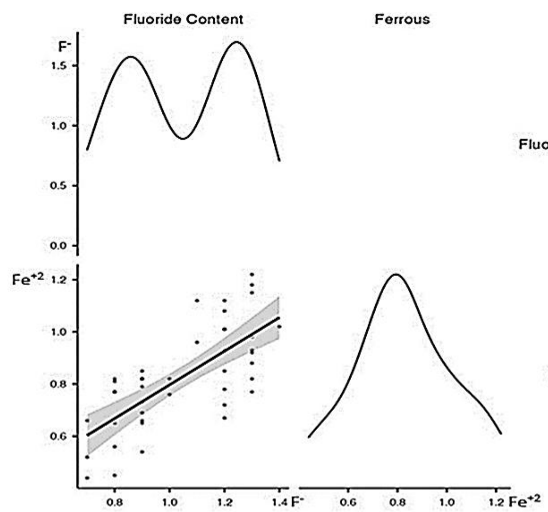

Fig. 3 Correlation curve between fluoride content and ferrous content.

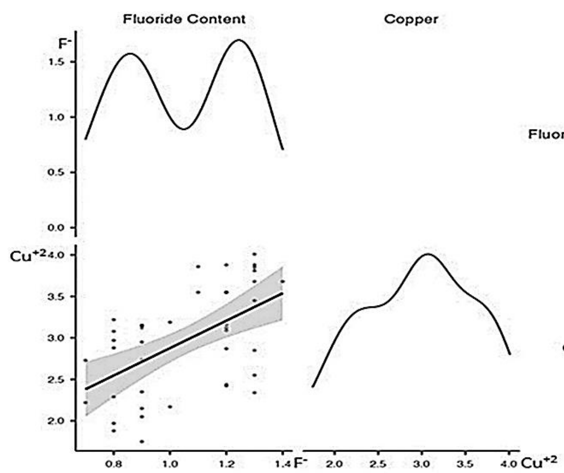

Fig. 4 Correlation curve between fluoride content and copper content.

Pearson's correlation coefficients (r) were calculated among sulfate content and other water quality parameters. It was observed that sulfate content is exhibiting a statistically significant positive correlation with nitrate content, total phosphate content, chloride content, fluoride content, sodium content, potassium content, iron content, and copper content. Pearson's correlation coefficients (r) were calculated among nitrate content and other water quality parameters. It was observed that nitrate content is exhibiting a statistically significant positive correlation with total phosphate content, chloride content, fluoride content, sodium content, potassium content, iron content, and copper content. Pearson's correlation coefficients (r) were calculated among total phosphate content and other water quality parameters. It was observed that total phosphate content is exhibiting a statistically significant positive correlation with chloride content, fluoride content, sodium content, iron content, and copper content.

Pearson's correlation coefficients (r) were calculated among chloride content and other water quality parameters. It was observed that chloride content is exhibiting a statistically significant positive correlation with fluoride content, sodium content, potassium content, iron content, and copper content. Pearson's correlation coefficients (r) were calculated among sodium content and other metallic ions. It was observed that sodium content exhibits a statistically significant positive correlation between potassium, iron, and copper content. Pearson's correlation coefficients (r) were calculated among potassium content and other metallic ions. It was observed that potassium content is exhibiting a statistically significant positive correlation between iron content and copper content. Pearson's correlation coefficients (r) were calculated among iron content and other metallic ions. It was observed that iron content is exhibiting a statistically significant positive correlation between iron content and copper content. The correlation study for the physicochemical analysis reveals that all the studied metallic ions (Na, K, Fe, Cu) keeps a strong association with chloride instead of fluoride. The albite mineral might be the possible additional source of Na^{+} over Cl^{-}. Further both Na^{+} and Cl^{-} ions were obtained from clay. The rich source of Na^{+} in groundwater had a significant contribution in controlling groundwater F^{-} geochemistry. The higher F^{-} concentration could have been attributed to the high Na^{+} and low Ca^{2+} contents in groundwater. A positive correlation between Na^{+} and F^{-} ($r = 0.14$) had been observed as evidence of this statement. The leaching precipitated secondary salts due to the increase of HCO_3^{-} decreased the F^{-} ions in the groundwater. The negative correlation between F and both Ca and Mg ions (−0.13 and −0.28) showed the precipitation of CaF_2 and MgF_2 due to an abundance of calcium and magnesium. According to 2005, sepiolite and palygorskite as Mg sink in Rajasthan are responsible for the release of F^{-} under alkaline conditions. The negative correlation between F^{-} and NO^{3-} ($r = -0.24$) indicated geogenic sources responsible for fluoride levels in groundwater rather than anthropogenic ones. The high HCO^{3-} and $Na^{+,}$ concentrations, and high pH favor the release of fluoride from the aquifer matrix into groundwater. F^{-} anion is sorbed onto positively charged surfaces and shows greater desorption at higher pH. Since the groundwater is alkaline, therefore the solubility of fluoride-bearing minerals had been increased in groundwater.[22]

4. Conclusion

The present research work was carried out to know the water quality parameters of the groundwater of the Surajgarh, Chirawa, and Buhana tehsils of the Jhunjhunu district of Rajasthan. The groundwater of the Buhana tehsil is less suitable for drinking purposes because (Sampling sites S-35 to S-42) exhibit higher values for the various physicochemical. The total hardness of all samples was found equal to the hard water limit. The groundwater of the study area exhibits equal and higher values than the permissible limit and their values specified by the BIS for various physicochemical parameters such as electrical conductivity, turbidity, TDS, total hardness, total alkalinity, sulfate, nitrate, total phosphate, chloride, fluoride, sodium, potassium, iron, and copper. The high fluoride, iron, and copper concentration in all the samples reveal that the study area suffers from the toxicity of such chemical substances. The tehsil-wise analysis of the physicochemical parameters reveals that Surajgarh tehsil (Sampling sites S-1 to S-26) exhibits more diverse values for the various physicochemical parameters. Besides, Chirawa tehsil (Sampling sites S-27 to S-34) exhibits modest values. parameters.

Conflict of Interest

There is no conflict of interest.

Supporting Information

Applicable.

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