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Role of Rock–Water Interaction in Groundwater Fluoride Enrichment

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Abstract: *Groundwater is an important source of drinking water and irrigation in many arid and semi-arid regions, but its chemical quality can be strongly influenced by interactions between groundwater and the rocks through which it flows. Fluoride enrichment is a widespread geochemical problem because naturally occurring fluoride-bearing minerals can release fluoride into groundwater through mineral dissolution, ion exchange, desorption, and other water–rock reactions.*
The concentration of fluoride is controlled not only by the abundance of fluoride-bearing minerals but also by groundwater residence time, pH, alkalinity, temperature, ionic composition, evaporation, and the availability of competing ions such as hydroxyl, bicarbonate, calcium, and phosphate. Minerals such as fluorite, apatite, micas, amphiboles and certain clay minerals may act as sources or sinks of fluoride depending on the prevailing hydrogeochemical conditions.

Keywords: Groundwater, Fluoride enrichment, Rock–water interaction, Fluorite dissolution, Mineral weathering, Ion exchange.

I. INTRODUCTION

Groundwater chemistry develops through a complex combination of atmospheric inputs, recharge processes, mineral dissolution, precipitation, adsorption–desorption, ion exchange, oxidation–reduction reactions and mixing. Among naturally occurring groundwater contaminants, fluoride is particularly important because elevated fluoride concentrations may occur without any direct anthropogenic source. Fluoride is present in many geological formations and can be released into groundwater when water interacts with minerals containing fluorine. Consequently, understanding the relationship between geology and groundwater chemistry is fundamental to explaining spatial and temporal variations in fluoride concentration.

Fluoride occurs naturally in a variety of igneous, metamorphic and sedimentary rocks. Fluoride-bearing minerals include fluorite (CaF_2), fluorapatite, biotite, muscovite, amphiboles and several clay minerals. The weathering of these minerals can introduce fluoride into groundwater. However, the presence of fluoride-bearing minerals alone does not necessarily result in high fluoride concentrations. Fluoride enrichment depends on the mineral's susceptibility to weathering, groundwater chemistry, residence time, temperature, pH, calcium activity and other geochemical conditions.

The role of rock–water interaction becomes especially important in crystalline-rock aquifers, where groundwater may circulate through fractures and weathered zones for relatively long periods. During this circulation, groundwater progressively acquires dissolved constituents from the surrounding rocks. Fluoride can therefore increase along groundwater flow paths as water becomes chemically evolved. In many fluoride-affected regions, groundwater is characterized by alkaline pH, relatively high bicarbonate concentrations, low calcium concentrations and elevated sodium concentrations. Such conditions can favor fluoride mobility.

The relationship between fluoride and calcium is particularly significant. Fluorite dissolution can be represented by:

$$CaF_2 \rightleftharpoons Ca^{2+} + 2F^-$$

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When groundwater contains sufficient calcium, fluoride activity can be limited by fluorite saturation. However, if calcium is removed from solution through ion exchange or precipitation of calcium-bearing minerals, fluorite dissolution may continue, increasing dissolved fluoride. This mechanism helps explain why high fluoride concentrations are often associated with relatively low calcium concentrations.

II. CONCEPT OF ROCK–WATER INTERACTION

Rock–water interaction refers to the physical and chemical processes occurring when groundwater comes into contact with geological materials. As groundwater moves through pores, fractures and weathered rock zones, it can dissolve minerals, exchange ions with mineral surfaces and either precipitate new minerals or adsorb dissolved constituents onto mineral surfaces.

The intensity of rock–water interaction depends on several factors:

1. mineralogical composition of the aquifer;
2. groundwater residence time;
3. water–rock ratio;
4. temperature;
5. pH;
6. redox conditions;
7. groundwater flow rate;
8. fracture density;
9. specific surface area of minerals; and
10. initial chemical composition of recharge water.

Fresh recharge water generally contains relatively low concentrations of dissolved ions. As it moves through the subsurface, interaction with rocks increases the concentration of major and trace elements. Therefore, groundwater located farther along a flow path may have a markedly different chemical composition from recently recharged groundwater.

Fluoride enrichment is particularly sensitive to these interactions because fluoride can be simultaneously released from minerals and removed through adsorption, precipitation and mineral incorporation.

III. GEOLOGICAL SOURCES OF FLUORIDE

1. Fluorite

Fluorite is one of the most important fluoride-bearing minerals. Its dissolution provides a direct source of fluoride:

$$CaF_2 + H_2O \rightarrow Ca^{2+} + 2F^-$$

Fluorite is relatively important in hydrogeochemical studies because its dissolution and precipitation can control fluoride concentrations under suitable calcium and fluoride activities.

2. Fluorapatite

Fluorapatite is common in igneous and metamorphic rocks and can release fluoride during weathering. Its dissolution is more complex than fluorite dissolution because phosphate and calcium are also involved.

3. Micas

Biotite and muscovite may contain significant amounts of fluorine. Weathering of these minerals can release fluoride, particularly under conditions favoring mineral alteration. The transformation of primary micas into secondary clay minerals can therefore contribute to fluoride mobilization.

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4. Amphiboles and Other Silicate Minerals

Certain amphiboles and other silicate minerals contain structural fluorine. Weathering can release fluoride gradually over prolonged periods. In crystalline-rock aquifers, these minerals may therefore represent long-term sources of dissolved fluoride.

5. Clay Minerals

Clay minerals can both release and retain fluoride. Their large specific surface area provides abundant sites for adsorption and ion exchange. Depending on pH and ionic composition, fluoride may either be adsorbed onto or desorbed from clay surfaces.

IV. MECHANISMS OF FLUORIDE ENRICHMENT

1. Mineral Dissolution

Mineral dissolution is one of the fundamental mechanisms responsible for fluoride release. Groundwater becomes enriched in fluoride when it dissolves fluorine-bearing minerals. The degree of dissolution depends on mineral stability, groundwater acidity or alkalinity, temperature and contact time.

In areas where groundwater residence time is long, continuous dissolution may progressively increase fluoride concentrations.

2. Alkalinity and pH

pH is an important control on fluoride mobility. Under alkaline conditions, fluoride adsorption onto mineral surfaces may decrease, increasing the concentration of fluoride in groundwater. Hydroxyl ions may compete with fluoride for adsorption sites, promoting fluoride desorption.

Thus, alkaline groundwater commonly provides favorable conditions for fluoride mobilization.

3. Calcium Control

Calcium is a major control on fluoride concentrations. When calcium is abundant, fluoride can be limited through fluorite saturation and precipitation. Conversely, low calcium concentrations can allow fluoride to remain dissolved.

The removal of calcium through cation exchange can therefore indirectly promote fluoride enrichment.

4. Cation Exchange

Cation exchange occurs when dissolved ions exchange with ions attached to mineral surfaces. In many groundwater systems, calcium and magnesium may be exchanged for sodium attached to clay minerals.

A simplified representation is:

$$2Na–X + Ca^{2+} \rightleftharpoons Ca–X_2 + 2Na^{+}$$

When calcium is removed through such reactions, the capacity of groundwater to dissolve fluoride-bearing minerals may increase. This process is one reason why sodium-rich and calcium-poor groundwater frequently exhibits elevated fluoride.

5. Desorption

Fluoride may be adsorbed on the surfaces of minerals such as iron and aluminum oxides and clay minerals. Changes in pH, ionic strength and competing anions can cause fluoride to desorb.

Bicarbonate and hydroxyl ions can influence surface reactions and thereby modify fluoride mobility.

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6. Evaporation

Evaporation does not directly generate fluoride, but it increases the concentration of dissolved constituents. In arid and semi-arid environments, intense evaporation can therefore enhance fluoride concentrations when groundwater recharge is limited.

7. Residence Time

Long groundwater residence time provides greater opportunity for water–rock interaction. Groundwater that remains underground for extended periods can acquire higher concentrations of dissolved minerals than recently recharged water.

Consequently, fluoride concentrations may increase along groundwater flow paths.

V. HYDROGEOCHEMICAL EVOLUTION AND FLUORIDE ENRICHMENT

Groundwater generally evolves chemically from dilute recharge water toward more mineralized groundwater. During this evolution, several processes can interact simultaneously.

Fresh groundwater may initially contain relatively high calcium and bicarbonate derived from carbonate dissolution. With increasing residence time, ion exchange can transform calcium-rich water into sodium-rich water. At the same time, dissolution of silicate and fluoride-bearing minerals can increase fluoride concentrations.

A typical fluoride-enriched groundwater may therefore exhibit:

1. high pH;
2. high bicarbonate;
3. high sodium;
4. relatively low calcium;
5. low magnesium in some settings;
6. elevated total dissolved solids; and
7. high fluoride concentration.

The exact composition varies according to geology and climate.

Role of Different Rock Types

Table 1. Major Geological Sources and Their Potential Role in Fluoride Enrichment

Rock/mineral type	Important fluoride-bearing minerals	Main process	Potential contribution to fluoride
Granitic rocks	Biotite, muscovite, apatite, fluorite	Weathering and dissolution	High
Gneiss	Biotite, amphiboles, apatite	Mineral alteration	Moderate–high
Schist	Micas, amphiboles	Weathering and ion exchange	Moderate–high
Volcanic rocks	Fluorapatite, volcanic glass, micas	Weathering/dissolution	Moderate
Sedimentary rocks	Fluorite, apatite, clay minerals	Dissolution/desorption	Variable
Carbonate rocks	Fluorite, apatite and accessory minerals	Dissolution and geochemical interaction	Variable
Alluvial sediments	Clay minerals, transported minerals	Desorption/ion exchange	Variable
Basaltic rocks	Apatite and fluorine-bearing silicates	Weathering	Low–moderate
Pegmatites	Fluorite, micas and accessory minerals	Mineral dissolution	Potentially high

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The table demonstrates that fluoride enrichment cannot be attributed to one mineral or rock type. Instead, it results from the interaction between mineralogical composition and groundwater chemistry.

VI. MAJOR FACTORS CONTROLLING ROCK–WATER FLUORIDE INTERACTION

Table 2. Hydrogeochemical Factors Affecting Fluoride Mobility

Factor	Effect on fluoride	General influence
pH	Controls adsorption/desorption	Alkaline conditions generally favor mobility
Calcium	Controls fluorite saturation	Low Ca may favor higher dissolved F^-
Bicarbonate	Influences mineral reactions and surface competition	Often associated with fluoride enrichment
Sodium	Indicator of ion exchange and evolved water	Frequently associated with high F^-
Residence time	Increases water–rock contact	Longer time may increase F^-
Temperature	Influences reaction kinetics	Higher temperature may enhance weathering
Evaporation	Concentrates dissolved ions	Can increase F^- concentration
Mineralogy	Determines fluoride sources and sinks	Strong geological control
Groundwater flow	Determines contact duration	Slow flow can promote enrichment
Clay content	Controls adsorption and ion exchange	Can either remove or release F^-
Phosphate	Competes for adsorption sites	May influence fluoride desorption
Alkalinity	Modifies mineral and surface reactions	Often favors fluoride mobility

VII. RELATIONSHIP BETWEEN FLUORIDE AND OTHER MAJOR IONS

Hydrogeochemical interpretation often uses relationships between fluoride and major ions to identify the processes responsible for enrichment.

1. Fluoride–Calcium Relationship

An inverse relationship between fluoride and calcium can indicate fluorite-controlled processes. Low calcium may permit increased fluoride concentrations because the groundwater is undersaturated with respect to fluorite.

2. Fluoride–Sodium Relationship

A positive relationship between fluoride and sodium can indicate cation exchange and groundwater evolution. Sodium-rich groundwater may develop as calcium is exchanged onto mineral surfaces.

3. Fluoride–Bicarbonate Relationship

High bicarbonate concentrations are frequently associated with fluoride-rich groundwater. Bicarbonate can influence mineral dissolution and surface adsorption processes and is also an indicator of chemically evolved groundwater.

4. Fluoride–pH Relationship

Increasing fluoride concentrations under alkaline conditions may indicate enhanced desorption and reduced fluoride retention on mineral surfaces.

VIII. CONCEPTUAL MODEL OF FLUORIDE ENRICHMENT

A conceptual model of fluoride enrichment can be represented as:

Recharge → Infiltration → Rock–Water Interaction → Mineral Weathering → Ion Exchange → Calcium Reduction → Fluoride Mobilization → Groundwater Evolution → Fluoride Enrichment

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During recharge, relatively dilute water enters the aquifer. As it moves through the geological formation, it interacts with fluoride-bearing minerals. Weathering releases fluoride, while ion exchange may remove calcium from solution. Under alkaline conditions, fluoride adsorption decreases and desorption increases. Long residence times and evaporation can further enhance fluoride concentrations.

This conceptual model demonstrates that fluoride contamination is often a natural geochemical process rather than a single-source pollution event.

IX. GEOCHEMICAL INDICATORS OF FLUORIDE ENRICHMENT

Several hydrogeochemical indicators can be used to identify rock–water processes:

1. **Na^+/Ca^{2+} ratio:** Higher values can indicate cation exchange and evolved groundwater.
2. **Ca^{2+} versus F^- relationship:** Useful for evaluating fluorite equilibrium.
3. **F^- versus pH:** Helps evaluate the influence of alkaline conditions.
4. **F^- versus HCO_3^-:** Helps identify the association between alkalinity and fluoride.
5. **Sodium adsorption relationships:** Useful for assessing ion-exchange processes.
6. **Saturation indices:** Thermodynamic calculations can determine whether groundwater is saturated or undersaturated with respect to fluorite and other minerals.
7. **Gibbs diagrams:** Can help distinguish precipitation, rock dominance and evaporation as broad controls on groundwater chemistry.
8. **Ion ratios:** Ratios such as Na^+/Cl^- and Ca^{2+}/Mg^{2+} can provide evidence of geochemical evolution.

X. COMPARATIVE SUMMARY OF FLUORIDE MOBILIZATION MECHANISMS

Table 3. Principal Rock–Water Processes Responsible for Fluoride Enrichment

Mechanism	Process description	Favorable conditions	Expected effect
Fluorite dissolution	CaF_2 releases Ca^{2+} and F^-	Undersaturation, suitable pH	Direct F^- increase
Silicate weathering	Fluoride-bearing silicates weather	Long residence time	Gradual F^- release
Fluorapatite dissolution	Releases F^- with Ca and phosphate	Weathering conditions	F^- enrichment
Cation exchange	Ca^{2+} exchanged for Na^+	Clay-rich aquifers	Indirect F^- increase
Fluoride desorption	F^- released from mineral surfaces	High pH, competing ions	F^- increase
Evaporation	Concentrates dissolved constituents	Arid climate	Apparent F^- enrichment
Calcite precipitation	Removes Ca^{2+} from groundwater	Suitable carbonate chemistry	May enhance F^- mobility
Long residence time	Prolonged mineral contact	Slow groundwater flow	Increased mineralization
Mixing	Combination of different waters	Recharge/groundwater interaction	Variable
Anthropogenic contribution	Fertilizers/industrial inputs	Developed areas	Local F^- enrichment

XI. ENVIRONMENTAL AND PUBLIC HEALTH SIGNIFICANCE

Fluoride is an essential trace element at low concentrations but excessive long-term exposure can result in adverse health effects, particularly dental and skeletal fluorosis. Therefore, understanding the natural mechanisms responsible for fluoride enrichment is essential for groundwater management.

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The identification of geological and hydrogeochemical controls allows vulnerable aquifers to be mapped before extensive groundwater development occurs. Areas containing fluoride-bearing crystalline rocks, alkaline groundwater and long residence times may require particular attention.

Fluoride risk assessment should therefore combine geological mapping, groundwater sampling, hydrochemical analysis and geochemical modeling rather than relying solely on fluoride concentration measurements.

XII. IMPLICATIONS FOR GROUNDWATER MANAGEMENT

Rock–water interaction should be incorporated into groundwater-quality management strategies. Important measures include:

1. systematic mapping of fluoride-bearing geological formations;
2. regular monitoring of groundwater fluoride;
3. identification of high-fluoride zones;
4. monitoring of pH, calcium, sodium and bicarbonate alongside fluoride;
5. evaluation of groundwater flow paths and residence times;
6. controlled groundwater abstraction;
7. use of alternative low-fluoride water sources where necessary; and
8. application of appropriate fluoride-removal technologies for affected communities.

Because fluoride enrichment is frequently controlled by natural hydrogeochemical processes, simply reducing anthropogenic pollution may not eliminate the problem.

XIII. RESEARCH GAPS AND FUTURE DIRECTIONS

Although considerable progress has been made in understanding fluoride enrichment, several research gaps remain. The relative contribution of individual fluoride-bearing minerals under different hydrogeochemical conditions requires further investigation. Long-term monitoring is also needed to understand how groundwater extraction changes residence time, flow paths and mineral–water equilibrium.

Geochemical modeling using tools such as PHREEQC can provide valuable information on mineral saturation and reaction pathways. Combining such modeling with geophysical investigations, mineralogical analysis and isotopic techniques could improve understanding of groundwater evolution. Machine-learning approaches may also help predict fluoride-risk zones by integrating geological, hydrochemical, climatic and groundwater-level data. However, predictive models should be supported by sound geochemical interpretation rather than treated as substitutes for field-based hydrogeological investigation.

XIV. CONCLUSION

Rock–water interaction is a fundamental control on natural fluoride enrichment in groundwater. Fluoride is released through the weathering and dissolution of fluoride-bearing minerals such as fluorite, fluorapatite, micas and amphiboles. However, mineral dissolution alone cannot explain the distribution of fluoride. Groundwater pH, calcium concentration, alkalinity, sodium enrichment, ion exchange, adsorption–desorption, evaporation and residence time collectively determine whether fluoride is retained in the aquifer or mobilized into groundwater.

Low calcium and alkaline conditions are particularly important because they can favor continued fluoride mobilization and reduce fluoride removal through mineral precipitation. Long groundwater residence times increase the opportunity for mineral–water reactions, while evaporation can further concentrate dissolved constituents. Consequently, fluoride enrichment should be interpreted as the outcome of a coupled geological and hydrogeochemical system.

A comprehensive assessment should therefore integrate mineralogical characterization, groundwater chemistry, saturation-index calculations, ion-exchange analysis and groundwater-flow information. Such an integrated approach can improve identification of fluoride-prone aquifers and support sustainable groundwater management and public-health protection.

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