Distribution and Co-enrichment Genesis of Arsenic and Fluoride in Groundwater of the Manas River Basin, Xinjiang: Postprint

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Abstract

Arsenic (As) and fluoride (F) contamination in water bodies and their co-occurrence represent a prominent issue in the environmental field, posing significant threats to public health upon long-term exposure. Taking the Manas River Basin in Xinjiang as the study area, this research analyzes the hydrochemical and spatial distribution characteristics of high-As and high-F groundwater, and further elucidates the hydrogeochemical processes governing the source, transport, and enrichment of As and F in groundwater by integrating geological conditions, occurrence environments, and anthropogenic influences. The results indicate that the groundwater in the study area is generally fresh water under weakly alkaline and reducing conditions, with As and F mass concentrations ranging from 1.13~41.35 g · L^{-1} and 0.06~8.02 mg · L^{-1}, respectively. High-As and high-F groundwater samples account for 62.9% and 45.7% of the total water samples, respectively, while the co-enrichment of arsenic and fluoride represents 37.1% of the total samples. Groundwater with co-enriched arsenic and fluoride is primarily distributed in the northern part of the plain area near the desert margin, with hydrochemical types dominated by $HCO_3 \cdot Cl$ and $HCO_3 \cdot SO_4 \cdot Cl$. High-As and high-F rock strata in the southern mountainous region of the study area constitute the primary source of As and F in groundwater, with regional geological structure and hydrogeological conditions representing important factors influencing arsenic and fluoride enrichment in groundwater. High-As groundwater in the Manas River Basin is characterized by high pH values, low $\gamma SO_4{}^{2-}/\gamma Cl^-$ ratios, and low Eh values, indicating that weakly alkaline reducing environments favor As enrichment in groundwater; high-F groundwater exhibits high pH values, high $HCO_3{}^-$, high Na^+, and low Ca^{2+} concentrations, suggesting that alkaline environments with high Na^+ and low Ca^{2+} facilitate F enrichment in groundwater.

Full Text

Abstract

Arsenic (As) and fluoride (F) pollution in water bodies, along with their coexistence, represent critical environmental concerns, with long-term exposure posing significant threats to public health. This study focuses on the Manas River Basin in Xinjiang as the research area, analyzing the hydrochemical and spatial distribution characteristics of groundwater with high As and F content. The results demonstrate that the groundwater in the study area is generally weakly alkaline and exhibits a reducing environment. The mass concentrations of As and F in groundwater range from 1.13 to 41.35 $g \cdot L^{-1}$ and from 0.06 to 8.02 $mg \cdot L^{-1}$, respectively. Groundwater samples exceeding the standard for As and F constitute 62.9% and 45.7% of the total samples, respectively, while samples showing co-enrichment of As and F account for 37.1% of the total. As-F coenriched groundwater is primarily distributed in the northern plain region near the desert edge, with the dominant hydrochemical types being $HCO_3 \cdot Cl$ and $HCO_3 \cdot SO_4 \cdot Cl$. The primary sources of As and F in groundwater are high-content rock strata in the southern mountainous area of the Manas River Basin. Regional geological structure and hydrogeological conditions represent important factors influencing As and F enrichment in groundwater. The high As groundwater in the Manas River Basin is characterized by high pH values, high HCO_3^-/Cl^- molar ratios, and low Eh values, indicating that a weakly alkaline reducing environment facilitates As enrichment. Conversely, high F groundwater is characterized by high pH, high Na^+ content, and low Ca^{2+} content, demonstrating that an alkaline environment with high Na^+ and low Ca^{2+} favors F enrichment.

Keywords: arsenic; fluoride; coenrichment; water chemistry; geological conditions; Manas River Basin

1.1 Study Area Overview

The Manas River Basin is located on the southern margin of the Junggar Basin, with geographical coordinates of 84°44 ~86°50 E and 43°4 ~46°0 N. The basin extends from the Taxi River in the east to the Kuitun River in the west, borders the Tianshan Mountains in the south, and reaches the Gurbantunggut Desert in the north. The overall topography is high in the south and low in the north, with landforms transitioning from mountains to plains to desert in sequence. The study area is situated in the piedmont depression zone on the northern slope of the Tianshan Mountains, where tectonic movement has caused folding of the Mesozoic and Cenozoic strata, forming a series of east-west trending folds and faults roughly parallel to the Tianshan Mountains. The anticline cores near the Tianshan Mountains consist of Mesozoic strata, the second row comprises Paleogene strata, and the third row at the northernmost edge consists of Neogene and early Quaternary strata. Major faults include the southern Junggar fault, the Huoerguosi-Manas-Turpan fault, the northern Manas buried fault, and the

Moguhu fault [14]. The lithology is primarily composed of Paleogene-Neogene and Quaternary sediments, with the Paleogene-Neogene sediments forming the basement of the mountainous and plain areas of the Manas River Basin, reaching a thickness of 1000 m. Quaternary sediments are widely distributed in the plain area, including piedmont plains (with sediment thickness of 400–1000 m) and alluvial plains [14,18].

The study area belongs to a mid-temperate continental arid climate zone, characterized by strong evaporation and low precipitation. Annual precipitation increases from north to south with rising elevation, while the average annual temperature ranges from 5.0 to 7.5°C. The highest elevation in the study area is 5246 m. Groundwater in the study area consists mainly of bedrock fissure water and Quaternary unconsolidated rock pore water. Bedrock fissure water is unevenly distributed in mountainous areas and is primarily recharged by glacial meltwater and snowmelt. Quaternary unconsolidated rock pore water is mainly distributed from alluvial-proluvial fans to alluvial plains (extending to the desert). The aquifer transitions from bedrock fissure water in mountainous areas to single-layer and multi-layer unconsolidated rock pore water in plain areas. The piedmont plain south of National Highway 312 consists of a single-layer unconfined aquifer composed of coarse-grained sand, gravel, and coarse sandstone with abundant pore structures and good permeability. The low plain area to the north features multi-layer aquifers, with depths less than 100 m comprising unconfined and shallow confined aquifers. Unconfined groundwater discharges as springs near reservoirs in Manas County and Shihezi City. Below 100 m depth lies the deep confined aquifer, primarily composed of gravel, pebble gravel, sandy gravel, or sand layers that become finer-grained and less permeable further north and deeper, resulting in poorer runoff conditions [14,18]. Groundwater is mainly recharged by precipitation infiltration, surface water leakage, farmland irrigation water, and spring return flow, and is discharged through artificial extraction, spring overflow, lateral runoff, and evapotranspiration [14,18].

1.2 Sample Collection and Analysis

Based on the China Geological Survey’ s groundwater pollution investigation project in the Junggar Basin, and following the analysis of groundwater distribution characteristics in the Manas River Basin, 95 groundwater samples were collected in July 2021. Sampling sites were selected along a north-south profile parallel to regional groundwater flow direction and an east-west profile perpendicular to it. The collection included 27 single-layer unconfined water samples, 24 unconfined water samples from the confined water area, and 44 confined water samples (excluding 10 samples from the same locations as 2019, totaling 95 samples). Sampling and analysis strictly followed the “Technical Specifications for Groundwater Environmental Monitoring (HJ/T 164-2004).” Before sampling, pumping was conducted (generally for more than 3 minutes) until groundwater hydrochemical conditions stabilized. Sampling bottles were rinsed three times with the collected water, and samples were filtered using a 0.22 m microporous membrane. Groundwater major cations and trace element samples were collected in 60 mL high-density polyethylene bottles acidified with 1:1 HNO_3, while anion samples were collected in 1.5 L plastic bottles without acidification. All samples were sealed with parafilm, stored in ice boxes, refrigerated at 4°C, and sent for analysis.

A multi-parameter analyzer (HANNA, HI9828) was used to measure water temperature, electrical conductivity (EC), dissolved oxygen (DO), and oxidation-reduction potential (Eh) on-site. Major cations (K^+, Na^+, Ca^{2+}, Mg^{2+}), dissolved total solids (TDS), major anions (Cl^-, ${SO_4}^{2-}$, ${HCO_3}^-$, ${CO_3}^{2-}$), and total hardness (TH) were analyzed according to the "Standard Examination Methods for Drinking Water" (GB/T 5750-2006). Major cations were determined using inductively coupled plasma atomic emission spectrometry (ICP-AES, Thermo iCAP6300) with analytical precision of 0.01 $mg \cdot L^{-1}$. Major anions were analyzed using ion chromatography (ICS 2000, Dionex). TDS was measured by gravimetric method with a detection limit of 4 $mg \cdot L^{-1}$. As concentration was determined by atomic fluorescence spectrometry (AFS) with a detection limit of 0.50 $g \cdot L^{-1}$, while F concentration was measured using ion-selective electrode method (PHS-3D) with a detection limit of 0.01 $mg \cdot L^{-1}$.

2.1 Groundwater Hydrochemical Characteristics

The pH values of groundwater in the study area range from 7.36 to 9.09, with an average of 8.11, indicating a weakly alkaline environment. TDS values range from 174.49 to 6803 $mg \cdot L^{-1}$, with an average of 772.93 $mg \cdot L^{-1}$. Groundwater with TDS less than 1000 $mg \cdot L^{-1}$ accounts for 82.86% of the samples, classifying it as freshwater. Eh values range from -125.10 to 81.20 mV, with an average of -86.95 mV, indicating a reducing environment. These characteristics demonstrate that the groundwater is generally weakly alkaline, reducing, and fresh (Table 1). According to the Shukarev classification and Durov diagram analysis (Fig. 3), cations are mainly distributed between the ${SO_4}^{2-}$ and ${HCO_3}^-$ axes, leaning more toward ${HCO_3}^-$, while anions are primarily distributed between the ${HCO_3}^-$ and Cl^- axes, leaning more toward Cl^-. The hydrochemical types show a pattern of transitioning from single-layer unconfined water to confined water. The single-layer unconfined water area is mainly weakly alkaline freshwater with $HCO_3 \cdot SO_4$ type; unconfined water in the confined water area is mainly weakly alkaline brackish water with $HCO_3 \cdot SO_4 \cdot Cl$ type; and confined water is mainly alkaline freshwater with $HCO_3 \cdot Cl$ (32.1%), $HCO_3 \cdot SO_4 \cdot Cl$ (25.0%), $SO_4 \cdot Cl$ (21.4%), and $HCO_3 \cdot SO_4$ (14.3%) types.

2.2.1 Horizontal Distribution Characteristics

Among the 95 groundwater samples, low As (\$ $10\mu g \cdot L^{\{-1\})andlowF(\ 1mg \cdot L}${-
1})$pointsaremainlydistributedaroundManasCountyandnearreservoirsinnorthernManasCounty, withnone$
$layerunconfinedwaterarea.Intheunconfinedwaterareaoftheconfinedwaterzone, allpointsexceptonemetthes$
$Fcoenrichedgroundwaterismainlydistributedinthenorthernplainareanearthedesertedgeandsporadicallyinth$

Cl (Fig. 4).

2.2.2 Vertical Distribution Characteristics

High As groundwater was detected in 59 of 95 samples, with an exceedance rate of 62.9%. No exceedances were found in single-layer unconfined aquifers or unconfined aquifers in the confined water area. In confined aquifers, 31 of 44 samples exceeded standards (78.6% exceedance rate), with average As concentrations of 18.95 $\mathrm{g \cdot L^{-1}}$. High F groundwater was detected in 43 of 95 samples, with an exceedance rate of 45.7%. In single-layer unconfined aquifers, 3 of 27 samples exceeded standards (11.1% exceedance rate) with an average concentration of 0.21 $\mathrm{mg \cdot L^{-1}}$. In unconfined aquifers of the confined water area, 6 of 24 samples exceeded standards (25.0% exceedance rate) with an average concentration of 0.57 $\mathrm{mg \cdot L^{-1}}$. In confined aquifers, 34 of 44 samples exceeded standards (53.6% exceedance rate) with an average concentration of 1.92 $\mathrm{mg \cdot L^{-1}}$ (Table 2). These results indicate that As-F coenrichment occurs only in confined aquifers.

2.3.1 Geological Conditions

The Quaternary genetic types in the study area are primarily aeolian, proluvial-alluvial, alluvial-proluvial, lacustrine, glacial-proluvial, proluvial-lacustrine, alluvial-lacustrine, slope, and chemical precipitation deposits. High As groundwater is mainly distributed in proluvial-alluvial deposits (mean value 23.46 $\mathrm{g \cdot L^{-1}}$, accounting for 81.8% of high As sampling points), followed by aeolian and alluvial deposits (mean value 10.34 $\mathrm{g \cdot L^{-1}}$, accounting for 13.6%). High F groundwater is mainly distributed in proluvial-alluvial deposits (mean value 2.57 $\mathrm{mg \cdot L^{-1}}$, accounting for 81.2% of high F sampling points), followed by aeolian deposits (mean value 1.44 $\mathrm{mg \cdot L^{-1}}$, accounting for 18.8%). As-F coenriched groundwater is distributed in proluvial-alluvial deposits (accounting for 76.9% of As-F coenriched sampling points) and aeolian deposits (23.1%). Therefore, proluvial-alluvial deposits significantly influence groundwater As and F concentrations.

2.3.2 Human Activities

Both natural factors and human activities influence As and F exceedances in the Manas River Basin. For example, mining of As and F mineral deposits and application of As/F-containing pesticides in agriculture can introduce these elements into surface water and groundwater [28]. The northern slope of the Yilianhabierga Mountains on the northern Tianshan piedmont contains numerous gold, copper, and pyrite deposits, while coal seams in the southern mountainous area are rich in As and F minerals. Mining activities can release these elements into the water system [29]. As the largest oasis agricultural area in Xinjiang, the Manas River Basin uses various pesticides containing As, such as lead arsenate, calcium arsenate, rice foot green, rice 宁, sodium arsenite, and Paris green.

Long-term application of these agents leads to soil As accumulation. Notably, from 2019 to 2021, TDS concentrations increased by 415.24–1762.87 $\mathrm{mg \cdot L^{-1}}$, while As and F concentrations increased by 0.03-0.26 $\mathrm{mg \cdot L^{-1}}$ and 0.06-0.21 $\mathrm{mg \cdot L^{-1}}$, respectively. These changes may result from human activities affecting local groundwater or from reduced groundwater extraction leading to rising water tables and weakened runoff intensity. Further investigation is needed to clarify these mechanisms.

2.3.3 Groundwater pH

High As and F groundwater is mainly distributed in confined aquifers, where pH values range from 8.11 to 9.09, indicating a weakly alkaline to alkaline environment. Statistical analysis shows significant positive correlations between pH and As concentration ($P < 0.001$) and between pH and F concentration ($P < 0.001$). As concentration increases gradually with rising pH, and when pH $>$ 8.0, As concentrations are generally > 10 $\mathrm{g \cdot L^{-1}}$. Similarly, when pH $>$ 8.0, F concentrations are generally > 1 $\mathrm{mg \cdot L^{-1}}$. Under alkaline conditions, adsorption of As and F decreases while desorption increases, facilitating their coexistence in groundwater (Fig. 6, Table 3).

2.3.5 Mineral Dissolution and Precipitation

The hydrogeochemical simulation software Phreeqc was used to calculate saturation indices (SI) of calcite, dolomite, gypsum, halite, and fluorite in groundwater. When SI > 0, minerals are saturated and cannot dissolve further; when SI $=$ 0, minerals are in equilibrium; when SI < 0, minerals are undersaturated and continue to dissolve. Results show that SI(calcite) and SI(dolomite) are > 0 (saturated), while SI(gypsum), SI(halite), and SI(fluorite) are < 0 (dissolved) (Fig. 8a). Precipitation of calcite [$CaCO_3$] and dolomite [$CaMg(CO_3)_2$] reduces Ca^{2+} content, creating favorable conditions for As and F enrichment. A significant positive correlation exists between F concentration and fluorite SI ($P < 0.001$), indicating that fluorite dissolution is a source of F in groundwater (Fig. 8b). The Gibbs diagram reveals that groundwater samples plot in the rock weathering and evaporation concentration zones, indicating that groundwater chemistry is influenced by rock weathering and evaporation, with minimal impact from precipitation (Fig. 7). Confined water samples mainly plot in the rock weathering zone, suggesting that As-F coenrichment is primarily controlled by rock weathering rather than evaporation.

2.4.1 Relationship between Groundwater As Concentration and Hydrochemical Parameters

The Cl^-/Br^- ratio ranges from 0.34 to 3.0, unaffected by mineral dissolution or evaporation, thus reflecting redox processes. High As groundwater has Cl^-/Br^- ratios of 0.77-3.0, indicating lower oxidation states. The reduction process may decrease ${SO_4}^{2-}$ content while increasing As content. Groundwater Eh ranges from -125.10 to 81.20 mV (average -86.95 mV), indicating a reducing environment favorable for As enrichment. A significant negative correlation exists between As concentration and Eh ($P < 0.001$), confirming that reducing conditions promote As release from rocks into groundwater (Fig. 9).

2.4.2 Relationship between Groundwater F Concentration and Hydrochemical Parameters

High F groundwater has TDS concentrations ranging from 174.49 to 1102.42 mg·L^{-1} and HCO_3^- concentrations of 95.23-6803 mg·L^{-1}. No correlation exists between F and TDS, but some samples show significantly increased F with little TDS change, suggesting that evaporation has minimal effect on F enrichment. In confined aquifers, F concentration shows a positive correlation with Na^+ concentration and a negative correlation with Ca^{2+} concentration. Under alkaline conditions, Ca^{2+} precipitates as calcite, promoting fluorite dissolution and increasing F concentration. The Na^+/Ca^{2+} ratio ranges from 0.34 to 191, with high F samples showing ratios of 2.00-191, indicating that high Na^+ and low Ca^{2+} alkaline environments favor F enrichment (Fig. 10).

2.5 Relationship between As and F in Groundwater

As concentrations range from 1.13 to 41.35 g·L^{-1} and F concentrations from 0.06 to 8.02 mg·L^{-1}. As-F coenrichment occurs only in confined aquifers, with 35 of 95 samples (37.1%) exceeding both standards. As concentration increases with F concentration when $F < 2$ mg·L^{-1} ($P < 0.001$), but the relationship becomes unclear when $F > 2$ mg·L^{-1} (Fig. 11). The average elevation of As-F coenriched sampling points is 340.77 m, lower than the overall average of 371.23 m, showing a negative correlation with elevation. Higher elevation areas have steeper slopes, coarser aquifer particles, greater hydraulic gradients, and stronger groundwater runoff, which are unfavorable for As and F enrichment. Lower elevation areas have gentler slopes, finer aquifer particles, smaller hydraulic gradients, and weaker runoff, creating favorable conditions for As and F accumulation.

3 Conclusions

(1) Groundwater As and F concentrations in the Manas River Basin range from 1.13-41.35 g·L^{-1} and 0.06-8.02 mg·L^{-1}, respectively. High As and high F groundwater samples account for 62.9% and 45.7% of the total samples, respectively, with As-F coenriched samples representing 37.1%. As-F coenriched groundwater is mainly distributed in the northern plain area near the desert edge in confined aquifers, with hydrochemical types of $HCO_3 \cdot Cl$ and $HCO_3 \cdot SO_4 \cdot Cl$.

(2) The enrichment of As and F in groundwater is primarily controlled by geological conditions, hydrogeological settings, and water-rock interactions.

The high As and F rock strata in the southern mountainous area represent the primary natural source. Under reducing conditions, microbially mediated mineral dissolution during organic matter degradation transfers adsorbed As and F from solid to liquid phases. Increased HCO_3^- promotes competitive adsorption and ion exchange, releasing As and F into groundwater. The well-sealed confined environment further facilitates As-F coenrichment.

(3) High As groundwater is characterized by high pH, high HCO_3^-/Cl^- ratios, and low Eh values, indicating that weakly alkaline reducing environments promote As enrichment. High F groundwater exhibits high pH, high Na^+ content, and low Ca^{2+} content, demonstrating that alkaline environments with high Na^+ and low Ca^{2+} favor F enrichment. Along the groundwater flow path from single-layer unconfined areas to confined areas, As and F concentrations generally increase.

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Note: Figure translations are in progress. See original paper for figures.