Geochemical Characteristics and Enrichment Mechanism of High-Arsenic Groundwater in the Aksu River Basin, Xinjiang (Postprint)

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Abstract

Twenty percent of domestic and irrigation water in the Aksu River Basin is sourced from groundwater, making it crucial to understand the distribution and formation of high-arsenic groundwater in this region. Based on hydrochemical and isotopic results, statistical and graphical methods were employed to analyze groundwater hydrochemical characteristics, spatial distribution and speciation of arsenic, combined with geological and geomorphological conditions and hydrogeochemical processes to reveal the occurrence conditions of arsenic, and to investigate the influencing factors and processes of high-arsenic groundwater enrichment in typical profiles. The results indicate that single-structure unconfined groundwater in the study area is alkaline, oxidizing, and slightly brackish; unconfined groundwater in confined water areas is alkaline, weakly oxidizing, and saline; and confined groundwater is alkaline, reducing, and fresh. Arsenic content ranges from 0.05 to 160 $\mu g \cdot L^{-1}$, with an exceedance rate of 19.5%. The hydrochemical types of high-arsenic groundwater are primarily $Cl \cdot SO_4$-Na, $SO_4 \cdot Cl$-$Na \cdot Ca$, and Cl-Na, mainly distributed in unconfined and confined groundwater at depths of 10–42 m in the middle and lower reaches of the Aksu River and Tailan River within confined water areas. From the piedmont inclined plain to the fine-soil plain, the occurrence environment of unconfined groundwater transitions from oxidizing to weakly oxidizing to weakly oxidizing and partially reducing, while confined groundwater remains in a reducing environment. Groundwater As content and saturation index exhibit a general trend of gradual increase. Single-structure unconfined groundwater and confined groundwater are primarily controlled by water-rock interaction, whereas unconfined groundwater in confined water areas is mainly controlled by evaporation concentration. The distribution of groundwater As is primarily related to structural factors, with natural sources mainly derived from the dissolution of orpiment. Arid climate, geological and geomorphological conditions, and sedimentary environment create external conditions for the enrichment of high-arsenic groundwater, while pH, desulfurization, redox potential (Eh), and groundwater circulation patterns influence the release and migration of As. This study contributes to understanding the formation and evolution patterns of high-arsenic groundwater in the Aksu River Basin and is of great significance for ensuring water supply safety.

Full Text

Abstract

In the Aksu River Basin, where domestic and irrigation water is sourced from groundwater, understanding the distribution and formation of high-arsenic groundwater is critical. Based on hydrochemical and isotopic data, statistical and graphical methods were employed to analyze groundwater hydrochemical characteristics, arsenic spatial distribution and speciation, and to reveal arsenic occurrence conditions in conjunction with geological, geomorphological, and hydrogeochemical processes. The study investigated influencing factors and enrichment processes of high-arsenic groundwater in typical profiles. Results show that single-structure phreatic water in the study area is alkaline, oxidizing, and brackish, whereas phreatic water in the confined aquifer zone is alkaline, weakly oxidizing, and saline, and confined water is alkaline, reducing, and fresh. Arsenic concentrations ranged from 0.05 to 160 $g \cdot L^{-1}$, with 19.5% of samples exceeding the standard. High-arsenic groundwater is primarily of the $Cl \cdot SO_4$-Na and $SO_4 \cdot$ Cl-Na $\cdot$ Ca types, mainly distributed in phreatic and confined waters at depths of 10–42 m in the middle and lower reaches of the Aksu and Tailan Rivers. From the piedmont alluvial plain to the fine-soil plain, the phreatic water occurrence environment transitions from oxidizing to weakly oxidizing and partially reducing, while confined water remains in a reducing environment. Groundwater arsenic content and saturation indices generally show a gradual increasing trend. Single-structure phreatic water and confined water are primarily controlled by water-rock interactions, whereas phreatic water in the confined zone is mainly controlled by evaporation concentration. Groundwater arsenic distribution is primarily related to structural factors, with natural sources mainly from realgar dissolution. The arid climate, geological setting, geomorphology, and sedimentary environment create external conditions for high-arsenic groundwater enrichment, while pH, desulfurization processes, redox potential (Eh), and groundwater circulation patterns influence arsenic release and migration. This study contributes to understanding the formation and evolution of high-arsenic groundwater in the Aksu River Basin and is significant for ensuring water supply safety.

Keywords: Aksu River Basin; high-arsenic groundwater; arid region; genesis analysis; occurrence environment

1. Study Area Overview

The Aksu River Basin is located at the southern foothills of the central Tianshan Mountains and the northwestern margin of the Tarim Basin, with geographical coordinates between 79°34 -81°03 E and 40°16 -41°30 N. The terrain is high in the north and low in the south, bordered by the Hantengri Mountains to the north and facing the Taklamakan Desert to the south. Administratively, it includes Wushi County, Wensu County, Aksu City, Awat County, and Alar City [Figure 1: see original paper]. The study area experiences a warm temperate continental arid climate with abundant sunshine, little rainfall, and dry air. Four independent rivers—the Aksu, Keya, Tailan, and Kalayuergun Rivers—flow from west to east, primarily fed by alpine snowmelt and mountain precipitation.

Quaternary strata are distributed throughout the region, from the northern piedmont alluvial plain to the southern alluvial-fine soil plain, with sedimentary facies transitioning from proluvial-alluvial to swamp-lacustrine and chemical deposits, showing clear zonation. Basin groundwater consists of Quaternary pore water. In the piedmont alluvial plain, it forms a single-structure phreatic aquifer composed of sandy gravel and coarse sand with rapid flow, representing the groundwater recharge zone. In the upper fine-soil plain, a continuous aquitard appears, creating a dual-structure of phreatic and confined aquifers. The aquifer material gradually changes to medium-coarse and medium-fine sand, with weaker hydrodynamic conditions, forming the groundwater runoff zone. In the lower fine-soil plain, the aquifer consists of medium-fine and fine sand with shallow burial and slow flow, constituting the groundwater discharge zone [Figure 2: see original paper]. Groundwater flow direction generally follows the river course (north to south). The recharge-runoff-discharge relationships are controlled by both geological conditions and human activities. In the piedmont alluvial plain, groundwater receives lateral inflow from mountain valleys, discharging mainly through lateral runoff. In the upper fine-soil plain, a densely populated agricultural area, groundwater and surface water interact frequently, receiving lateral runoff from upstream and vertical recharge from rivers, canals, and irrigation, with discharge through evaporation, runoff, and river outflow. The lower fine-soil plain receives lateral runoff from upstream and minor canal leakage, discharging primarily through evaporation [24,26].

2. Sample Collection and Testing

Based on previously collected hydrogeological data, the research team conducted field investigations and sampling in the Aksu River Basin during May–June 2021. Considering groundwater burial depth, hydrodynamic conditions, and well distribution, samples were collected along a north-south profile following the groundwater flow direction, adopting a principle of "points to lines, lines to area" to study the formation and evolution of high-arsenic groundwater. A total of 102 hydrochemical samples were collected (sampling depth 1.7–130 m), including 28 single-structure phreatic water samples (depth 1.7–130 m), 30 phreatic water samples from the confined zone (depth 1.3–45 m), and 44 confined water samples (depth 8-90 m). Additionally, 30 isotope samples were collected. All groundwater samples were used for drinking and irrigation.

Before sampling, wells were pumped continuously for approximately 5 minutes until physicochemical parameters stabilized. Portable meters were rinsed with deionized water before measuring pH, water temperature, electrical conductivity (EC), and oxidation-reduction potential (Eh). Samples were then numbered, sealed, and stored in a 4°C cooler for transport. Bicarbonate ($HCO_3{}^-$) was measured on-site using acid-base titration, while other hydrochemical parameters (K^+, Na^+, Ca^{2+}, Mg^{2+}, Cl^-, $SO_4{}^{2-}$, $NO_3{}^-$, F^-, Fe, Mn, As) were analyzed at a certified laboratory of the Xinjiang Bureau of Geology and Mineral Resources. Isotope samples were sent to the United States for testing. Data reliability was verified using ion balance calculations, with errors within \$±\$5%, confirming data quality.

3. Results

3.1 Regional Groundwater Chemistry Characteristics

Statistical results show that single-structure phreatic water had TDS values of 248-16,500 $mg \cdot L^{-1}$ (mean 1,281 $mg \cdot L^{-1}$), pH 7.47-8.76 (mean 7.75), and Eh 29.3-652.61 mV. Phreatic water in the confined zone had TDS 468-59,336 $mg \cdot L^{-1}$ (mean 5,689 $mg \cdot L^{-1}$), pH 6.59-8.88 (mean 7.81), and Eh -136.07 to 383.07 mV. Confined water had TDS 200-1,920 $mg \cdot L^{-1}$ (mean 769 $mg \cdot L^{-1}$), pH 7.47-8.76 (mean 7.75), and Eh -189.13 to 113.87 mV. These data indicate that regional groundwater is weakly alkaline, with confined water having lower TDS than phreatic water, and phreatic water TDS gradually increasing from piedmont to fine-soil plain.

The Piper diagram [Figure 3: see original paper] shows most sample points clustered in the $Cl \cdot SO_4$-Na and $SO_4 \cdot$ Cl-Na $\cdot$ Ca regions. In single-structure phreatic water, high-arsenic groundwater ($As > 10 \;\; g \cdot L^{-1}$) is primarily of the $Cl \cdot SO_4$-Na $\cdot$ Ca type. In phreatic water from the confined zone, high-arsenic groundwater is mainly $SO_4 \cdot$ Cl-Na type. In confined water, high-arsenic groundwater is primarily $SO_4 \cdot$ Cl-Na $\cdot$ Mg type. Low $NO_3{}^-$ concentrations (data not shown) indicate minimal contribution from anthropogenic activities (domestic sewage and industrial wastewater), suggesting water chemistry originates from mineral weathering of evaporites, silicates, and carbonates in the strata. Previous studies have linked the $Cl \cdot SO_4$-Na type to high arsenic and fluoride concentrations in arid regions [29], consistent with this study [Figure 3: see original paper]. The proportion of arsenic exceeding standards in confined water is significantly higher than in phreatic water, likely related to redox conditions.

3.2 Speciation Characteristics

pH and Eh are the main factors controlling arsenic speciation. Generally, arsenic exists as H_3AsO_3 under anaerobic conditions with pH < 9.2, as $HAsO_4{}^{2-}$

under alkaline oxidizing conditions, as $H_2AsO_4^-$ under near-neutral oxidizing conditions, and as H_3AsO_3 under oxidizing acidic conditions [15,28]. Based on pH-Eh data, arsenic speciation in basin groundwater was predicted. The pH-Eh diagram [Figure 4: see original paper] shows all samples fall within the $HAsO_4^{2-}$ stability field, existing as inorganic $HAsO_4^{2-}$ under alkaline conditions and $H_2AsO_4^-$ under acidic conditions.

3.3 Distribution Characteristics

3.3.1 Vertical Distribution Among the 102 tested samples, 19.5% exceeded the arsenic standard of 10 $\mathrm{g \cdot L^{-1}}$, including 10.3% of single-structure phreatic water, 21.1% of phreatic water in the confined zone, and 31.8% of confined water. Arsenic concentration shows a trend of first increasing then decreasing with depth, with high-arsenic groundwater concentrated at 10–42 m depth in phreatic and confined waters of the confined zone, where phreatic water arsenic content is higher than confined water. pH decreases with increasing depth, while high-arsenic groundwater is mainly distributed in phreatic water at 0–17 m depth. Eh shows clear zonation, with confined water Eh primarily between -100 and 85 mV, while single-structure phreatic water and phreatic water in the confined zone mainly range from 85 to 300 mV [Figure 5: see original paper].

3.3.2 Horizontal Distribution Based on the principle that larger determination coefficients and smaller residuals indicate better fit, semi-variogram models for groundwater arsenic were calculated using GS+ software . The nugget-to-sill ratios for phreatic and confined water are 0.43 and 0.38, respectively, indicating strong spatial correlation and that arsenic heterogeneity is primarily controlled by structural factors (hydrogeology, topography, occurrence environment) rather than random factors. The range for phreatic water is much larger than for confined water, suggesting phreatic water arsenic is influenced by structural factors over larger areas.

Comparing arsenic concentrations across different counties, Wushi County shows no exceedance (mean 0.43 $\mathrm{g \cdot L^{-1}}$), while Alar City, Awat County, Aksu City, and Wensu County all show varying degrees of exceedance, with Alar City being the most severe (mean 13.94 $\mathrm{g \cdot L^{-1}}$) . From a geomorphological perspective, groundwater arsenic generally increases from piedmont to fine-soil plain. In phreatic water, exceedance areas are concentrated along the lower Aksu River and middle-lower Tailan River, primarily in Kezile Town and Gule Awati Township of Wensu County, Awat County, Aksu City, and the 7th and 8th Regiments of Alar City [Figure 6: see original paper]. Confined water exceedance is mainly distributed in the 7th and 8th Regiments, where arsenic concentrations commonly exceed 30 $\mathrm{g \cdot L^{-1}}$, with exceedance rates of 31.25–66.67% [Figure 6: see original paper].

3.3.3 Profile Distribution Characteristics To understand variations in groundwater components at the watershed scale, a typical profile was analyzed

[Figure 7: see original paper]. Along the river course from piedmont to fine-soil plain, arsenic, TDS, and pH show similar trends, with maximum values at point TJ051, decreasing sharply to 0.7 $g \cdot L^{-1}$ at MJC007 in the desert zone. SO_4^{2-} is low in the piedmont alluvial plain with small variation range, increasing sharply in the fine-soil plain and gradually decreasing in the desert zone. The desert zone' s sparse vegetation, large soil pores, and good permeability facilitate surface water-groundwater dissipation, which is unfavorable for arsenic enrichment [29,31]. Overall, downstream groundwater shows enrichment of arsenic, TDS, and pH, with hydrochemical types transitioning from SC/LS to SL/LS.

4. Discussion

4.1 Main Hydrogeochemical Processes Controlling High-Arsenic Groundwater

Principal component analysis was used to identify representative characteristic factors controlling high-arsenic groundwater hydrogeochemistry. Three principal components were extracted with eigenvalues >1, explaining 56.25%, 18.37%, and 11.38% of variance, respectively, with a cumulative contribution of 86.00% . Factor 1 includes TDS, Na^+, Cl^-, SO_4^{2-}, F^-, and As, representing dissolution of gypsum, evaporites, and calcite. Most samples plot in the fine-soil plain where poor hydrodynamic conditions and strong evaporation lead to high TDS. Factor 2 includes Eh, Ca^{2+}, and Mg^{2+}, indicating silicate dissolution that promotes calcite and dolomite precipitation. Factor 3 includes HCO_3^- and pH, reflecting the relatively closed environment of confined zones where desulfurization promotes iron oxide reduction dissolution, increasing arsenic concentration.

Gibbs diagrams [Figure 11: see original paper] show that single-structure phreatic water and confined water samples plot in the water-rock interaction zone, while phreatic water in the confined zone plots in the evaporation concentration zone. This indicates that single-structure phreatic water and confined water are controlled by water-rock interactions, whereas phreatic water in the confined zone is controlled by evaporation concentration. The Aksu River Basin' s typical alluvial-proluvial geomorphology, with its independent and unified flow system, long groundwater flow paths, and distinct zonation of renewal rates and water quality, creates these different controlling processes.

4.2 Regional Geomorphology and Sedimentary Environment

Neotectonic movements have controlled the Quaternary geological evolution of the study area. The Himalayan orogeny caused continuous uplift of surrounding mountains, forming the prototype of an arid inland basin [31,35]. Climate warming led to alpine snowmelt, creating massive floods that transported detrital materials southward, forming the Quaternary alluvial-proluvial plain [24,36]. High-arsenic groundwater occurs mainly in Quaternary alluvial deposits (pal) and swamp-chemical deposits (ch+fl) along the lower Aksu River and middle-lower Tailan River, with lithology dominated by silty clay and sub-sandy soil

[Figure 8: see original paper].

The Ca^{2+}/Cl^- ratio reflects hydrodynamic conditions [31,33], with larger ratios indicating better conditions. Arsenic content increases as Ca^{2+}/Cl^- decreases [Figure 9: see original paper], indicating that confined zone phreatic and confined waters have poorer runoff conditions than single-structure phreatic water, favoring arsenic enrichment. From piedmont to fine-soil plain, terrain becomes flatter, aquifer materials finer, and recharge conditions poorer, creating favorable external conditions for high-arsenic groundwater enrichment.

4.3 Climate and Hydrogeological Conditions

From Early Pleistocene to Holocene, the Aksu River Basin experienced warm-humid to warm-dry climate cycles, causing lake and swamp shrinkage and forming discontinuous swamp-chemical deposits [24,36]. Gypsum, calcite, halite, arsenic, and fluorine were deposited in sediments at certain depths. These deposits, rich in organic matter, release salts, arsenic, and fluorine to groundwater through biogeochemical processes [31]. These swamp-chemical deposits are typically located 10–40 m deep in the eastern study area [37], consistent with the depth distribution of high-arsenic groundwater [Figure 5: see original paper].

4.4 Dissolution Processes

Saturation indices (SI) of major mineral phases were calculated using PHREEQC [Figure 12: see original paper]. Calcite and dolomite are supersaturated, gypsum is near equilibrium, and rock salt and realgar are undersaturated. Along the profile, calcite and dolomite remain supersaturated, with trends opposite to Ca^{2+} and Mg^{2+}, possibly related to silicate dissolution. Gypsum, rock salt, and realgar SI increase from upstream to downstream, with gypsum reaching equilibrium at MJC007. Realgar shows two peaks (JJA095-JJC056 and after JJC102), corresponding to areas rich in realgar and hematite in Kezile Town, Gule Awati Township, and the 7th–8th Regiments. Although realgar SI is low, mineral dissolution provides the initial arsenic source, which migrates toward evaporation zones through subsequent evaporation concentration [Figure 10: see original paper].

4.5 Occurrence Environment

In the confined water system, arsenic content shows positive correlation with SO_4^{2-}/Cl^- ($P < 0.05$) and negative correlation with Eh ($P < 0.05$), reflecting the relatively closed environment where desulfurization promotes iron oxide reduction dissolution [Figure 13: see original paper]. Arsenic content also shows negative correlation with HCO_3^- ($P < 0.05$). In alkaline conditions ($pH > pHzpc$), mineral surfaces become negatively charged, inhibiting arsenic adsorption [13,15]. Under oxidizing conditions, arsenic release is mainly from pyrite and sulfide dissolution; under reducing conditions, iron oxide dissolution releases adsorbed arsenic [5,12,39].

4.6 Human Activity Impacts on Occurrence Environment

Tritium values in surface water and groundwater indicate the degree of water exchange and occurrence environment [Figure 14: see original paper]. In the recharge zone, tritium values are close to modern surface water, indicating oxidizing conditions (Eh = 293 mV). In the runoff zone, tritium values are lower (181 mV), indicating weakly oxidizing conditions with multiple surface water-groundwater interactions. In the discharge zone, tritium is absent, theoretically indicating reducing conditions, but wells show microconnections with surface water. Confined water samples show no tritium, indicating a closed reducing environment (Eh = -94 mV).

4.7 Formation Process in Typical Profile

A conceptual model [Figure 15: see original paper] illustrates high-arsenic groundwater formation. In the piedmont alluvial plain, snowmelt and storm floods recharge groundwater in an alkaline oxidizing environment where arsenic is adsorbed onto Fe-Mn oxides. In the runoff zone, fine aquifer materials, weak hydrodynamic conditions, and agricultural activities create a weakly oxidizing environment where realgar dissolves and Fe-Mn oxides undergo microbially-driven reductive dissolution, releasing arsenic. Evaporation increases ion concentrations and shifts hydrochemical types from SC-NC to SL/LS. In the discharge zone, extremely weak hydrodynamic conditions, enhanced ion exchange, and evaporation further increase ion concentrations, with HCO_3^- and Na^+ becoming dominant.

5. Conclusions

Through statistical and geostatistical analysis of high-arsenic groundwater in the Aksu River Basin, the formation mechanisms, migration, spatial distribution, and water-rock interaction types were systematically studied:

1) Arsenic concentrations in single-structure phreatic water, phreatic water in the confined zone, and confined water were 0.05-160 $\mathrm{g}\cdot\mathrm{L}^{-1}$ (mean 5.62 $\mathrm{g}\cdot\mathrm{L}^{-1}$), 0.10-151.08 $\mathrm{g}\cdot\mathrm{L}^{-1}$ (mean 8.99 $\mathrm{g}\cdot\mathrm{L}^{-1}$), and 0.8-75.39 $\mathrm{g}\cdot\mathrm{L}^{-1}$ (mean 12.37 $\mathrm{g}\cdot\mathrm{L}^{-1}$), respectively. Exceedance rates were 10.3%, 21.1%, and 31.8%, respectively. High-arsenic groundwater ($\mathrm{As} \geq 10\ \mathrm{g}\cdot\mathrm{L}^{-1}$) is mainly of the $Cl\cdot SO_4$-Na and $SO_4\cdot Cl$-Na$\cdot$Ca types. Arsenic speciation is dominated by $HAsO_4^{2-}$ in phreatic water and H_3AsO_3 in confined water.
2) Vertically, high-arsenic groundwater occurs mainly at 10–42 m depth. Horizontally, it does not gradually increase simply as a concentration gradient from piedmont to fine-soil plain. High-arsenic phreatic water is mainly distributed in Kezile Town and Gule Awati Township of Wensu County, Awat County, and the 7th–8th Regiments of Alar City. High-arsenic confined water is mainly near the 7th–8th Regiments.
3) Single-structure phreatic water and confined water are controlled by water-

rock interactions, while phreatic water in the confined zone is controlled by evaporation concentration. Arsenic content is inversely proportional to SO_4^{2-}/Cl^- and Eh, but poorly correlated with HCO_3^-.

4) The natural source of arsenic is realgar dissolution. The arid climate, flat terrain, weak hydrodynamic conditions, and hydrogeological setting combining orogenic belts with chemical and swamp deposits create external conditions for arsenic enrichment. Occurrence environment factors including pH, Eh, desulfurization, and groundwater circulation patterns, along with human activities, affect arsenic release and migration.

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