Groundwater Quality Assessment and Fluoride Enrichment Characteristics in the Western Tarim Basin: A Case Study of Aketao County (Postprint)

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Abstract

To investigate groundwater quality conditions and fluoride enrichment characteristics in the western Tarim Basin, groundwater samples from three aquifers in Aketao County were collected and analyzed. The improved Nemerow evaluation method and health risk assessment model were employed to evaluate the regional groundwater, followed by an investigation of fluoride enrichment mechanisms. The results demonstrate that: according to the improved Nemerow water quality assessment, water quality at numerous groundwater aquifer sampling points in the study area falls between Class II and Class III water categories, although some individual aquifer samples are classified as Class IV water; analysis via the health risk assessment model reveals that the overall health risks posed by fluoride and chloride in shallow, middle, and deep groundwater (adults: 1.28\$×10^{-6}\$ a^{-1}, 1.02\$×10^{-6}\$ a^{-1}, and 9.36\$×10^{-7}\$ a^{-1}; children: 1.63\$×10^{-6}\$ a^{-1}, 1.30\$×10^{-6}\$ a^{-1}, and 1.19\$×10^{-6}\$ a^{-1}) are below the maximum acceptable risk level recommended by the U.S. Environmental Protection Agency (US EPA). The fluoride exceedance rates for shallow, middle, and deep groundwater samples are 57.14%, 40.00%, and 60.00%, respectively, with mean concentrations exceeding the standard limit ($1.0\ \mathrm{mg \cdot L^{-1}}$). Groundwater fluoride enrichment in the study area exhibits characteristics of alkaline tendency, Na^+ richness, and Ca^{2+} deficiency, with rock weathering, silicate mineral dissolution, and ion exchange acting as promoting factors for fluoride accumulation.

Full Text

Preamble

Evaluation of Groundwater Quality and Fluoride Enrichment Characteristics in the Western Tarim Basin: A Case Study of Akto County

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Abstract: To investigate groundwater quality conditions and fluoride enrichment characteristics in the western Tarim Basin, this study collected and analyzed groundwater samples from multiple aquifers in Akto County as a representative case. An improved Nemerow evaluation method and health risk assessment model were applied to evaluate regional groundwater quality and examine fluoride enrichment mechanisms. The results indicate that according to the improved Nemerow water quality assessment, water quality at many aquifer sampling points in the study area falls between Class II and Class III, though some aquifer samples are classified as Class IV. Analysis using the health risk assessment model reveals that the overall health risks posed by fluoride and chloride in shallow, middle, and deep groundwater (adults: 1.28$×10^{-6}$ a^{-1}, 1.02$×10^{-6}$ a^{-1}, and 9.36$×10^{-7}$ a^{-1}; children: 1.63$×10^{-6}$ a^{-1}, 1.30$×10^{-6}$ a^{-1}, and 1.19$×10^{-6}$ a^{-1}) are below the maximum acceptable risk values recommended by the US Environmental Protection Agency (US EPA). The fluoride exceedance rates in shallow, middle, and deep groundwater samples are 57.14%, 40.00%, and 60.00%, respectively, with mean concentrations all exceeding the limit of 1.0 mg·L^{-1}. Fluoride enrichment in the study area' s groundwater exhibits alkaline characteristics, with high Na^{+} and low Ca^{2+} content. Rock weathering, dissolution of silicate minerals, and ion exchange processes promote fluoride enrichment.

Keywords: hydrogeochemistry; fluoride; water quality assessment; western Tarim Basin

With urbanization and industrial development, groundwater has become a crucial drinking water resource in northern China. Geological conditions, hydrodynamic factors, and human activities often cause enrichment of hydrochemical elements, posing serious threats to human health. Fluoride contamination in groundwater has emerged as a key research priority in groundwater resource development. Long-term consumption of low-fluoride water can cause adverse physiological reactions such as dental caries and osteoporosis, while prolonged intake of high-fluoride water severely threatens human health, causing dental fluorosis and skeletal diseases. Groundwater fluoride pollution events have been reported in many countries, including India, Ghana, Pakistan, and Italy. Statistics show that over 200 million people worldwide consume water with fluoride levels exceeding World Health Organization (WHO) guidelines, with approximately 26 million people in China affected by high-fluoride groundwater.

Fluoride enrichment in groundwater is primarily controlled by hydrogeological conditions, dissolution of fluoride-bearing minerals (fluorite, mica, etc.), ion exchange processes, and human activities. Most fluoride originates from dissolution of rock minerals at the Earth's surface and corresponding hydrogeochemical reactions, while human activities and atmospheric precipitation contribute relatively minor proportions. Consequently, groundwater fluoride enrichment is considered the result of long-term exposure to fluoride-bearing minerals and hydrogeochemical reactions. Studies have demonstrated that arid and semi-arid climates favor fluoride enrichment. Research in the Tarim Basin has reported fluoride concentrations reaching 9.1 $\mathrm{mg \cdot L^{-1}}$, with exceedance rates reaching 47.8% in the Kashgar region and 22.2% in the Yarkant River Basin. However, few studies have investigated fluoride sources, genesis, and enrichment processes in the western Tarim Basin, and it remains unclear whether fluoride levels in this region pose health risks, necessitating investigation and analysis of fluoride content across different aquifers.

Based on fluoride survey results, this study employs an improved Nemerow pollution index and the US EPA health risk assessment model to evaluate water quality and fluoride health risks across different aquifers in the western Tarim Basin. Taking groundwater from shallow (0–50 m), middle (50-100 m), and deep (100–200 m) aquifers in Akto County, western Tarim Basin as the research object, this paper identifies fluoride content in different aquifers, evaluates regional groundwater quality, and uses fundamental hydrogeochemical theory to analyze fluoride enrichment processes. These findings provide reference for strengthening water resource protection, optimizing water quality monitoring networks, and addressing safe drinking water issues in the western Tarim Basin.

1.1 Study Area Overview

[Figure 1: see original paper] Schematic diagram of the study area

The Tarim Basin, located in southern Xinjiang Uygur Autonomous Region, is China' s largest inland basin, situated between the Tianshan, Kunlun, and Altun Mountains. The western Tarim Basin is rich in mineral resources and features unique geological structures (fault-bend folds and fault-propagation folds). The study area, Akto County, lies in the western Tarim Basin (75°08 –76°16 E, 38°56 –39°33 N), covering a total area of 6,392 $\mathrm{km^2}$. The region has a warm temperate continental arid to semi-arid climate with large temperature variations, a multi-year average temperature of 12.99°C, strong evaporation (annual evaporation of 2,700 mm), and low precipitation (annual rainfall of 64 mm). The terrain slopes from high in the southwest to low in the northeast, with well-developed dendritic river systems. Soil types include oasis loess, brown calcic soil, alpine desert soil, saline soil, and aeolian sandy soil. Groundwater originates primarily from surface water conversion, and the thick Quaternary loose sediments in plain areas provide favorable conditions for groundwater storage and migration. Differences in topography, lithology, and drainage distribution result in varying groundwater hydrochemistry.

The study area is dominated by Silurian, Carboniferous, Permian, Lower Cretaceous, Paleogene, Neogene, and Quaternary strata. The shallow aquifer lithology is primarily limestone, the middle aquifer mainly biotite quartz schist, and the deep aquifer predominantly biotite granulite.

1.2 Sample Measurement Methods

Sampling was conducted following the *Technical Specification for Groundwater Environmental Monitoring* (HJ/T 164-2004). Sampling points were spaced at least 10 km apart, with 15 sampling points established (Fig. 1). Before sampling, each well was pumped for 15 minutes to remove stagnant water. Sampling depths were divided into three categories: shallow water (0–50 m), middle water (50–100 m), and deep water (100–200 m). Shallow water ($n = 15$), middle water ($n = 14$), and deep water ($n = 10$) samples were collected, with some sampling points providing samples from all three aquifers. A portable sampling pump was used to collect water from different aquifers at the same point to prevent cross-contamination. Field measurements of pH, dissolved oxygen, and temperature were conducted using a Multi340i portable water quality multi-parameter analyzer.

Polyethylene bottles were used for sample collection. Before sampling, polyethylene bottles were rinsed with ultrapure water and then conditioned with the sample water three times. Water samples were filtered through 0.45 m membranes and stored in polyethylene bottles. For cation analysis (K^+, Na^+, Ca^{2+}, Mg^{2+}), samples were acidified with nitric acid (analytical grade) to $pH < 2$. For anion analysis (F^-, Cl^-, ${NO_3}^-$, ${SO_4}^{2-}$), samples were only filtered. All samples were stored in a 4°C refrigerator and transported to the laboratory within 7 days for analysis. Anions were measured by ion chromatography (Thermo Scientific ICS-600), cations by atomic absorption spectrophotometry (Thermo Scientific ICE3500), and total dissolved solids (TDS) by gravimetric method.

1.3 Improved Nemerow Water Quality Assessment

The improved Nemerow pollution index method was used for water quality evaluation. The calculation formulas are as follows:

$$F_i = \frac{C_i}{S_i}$$

$$P = \sqrt{\frac{\max(F_i^2) + \bar{F}_i^2}{2}}$$

$$\bar{F}_i = \frac{\sum_{i=1}^{n} R_i F_i}{\sum_{i=1}^{n} R_i}$$

$$R_i = \frac{r_i}{\sum_{i=1}^{n} r_i}$$

$$r_i = \frac{S_{\max}}{S_i}$$

$$P_{\text{improved}} = \sqrt{\frac{F_{\max}^2 + \bar{F}_i^2}{2}}$$

Where:
F_i is the single-factor pollution index; C_i is the measured concentration of the evaluation factor ($\text{mg}\cdot\text{L}^{-1}$); S_i is the background concentration of the evaluation factor ($\text{mg}\cdot\text{L}^{-1}$); P is the Nemerow pollution index; R_i is the weight value of each evaluation factor; r_i is the correlation ratio of each factor; $S_{\max}$ is the maximum background concentration of evaluation factors; $\bar{F}_i$ is the weighted average of single pollution indices; $F_{\max}$ is the maximum pollution index among F_i; F_{avg} is the mean pollution index of the top m weighted components; and m is the number of items among the top n items.

As industrial and domestic water in the study area is primarily sourced from groundwater, environmental background values were referenced against the *Groundwater Quality Standard* (GB/T 14848-2017).

1.4 Health Risk Assessment Method

Humans are exposed to fluoride and chloride through multiple pathways, including drinking water and skin contact. Based on behavioral and physiological differences, the population was divided into adults and children. The US EPA health risk assessment model was used to evaluate hazards from fluoride and chloride in western Tarim Basin groundwater. The primary exposure pathways studied were ingestion and dermal contact.

Health Risk Model for Drinking Water Pathway:

$$R_i^d = \frac{D_i \times 10^{-6}}{RfD_i \times 72}$$

$$D_i = \frac{C_i \times IR \times EF \times ED}{BW \times AT}$$

Where:
R_i^d is the annual individual health risk from drinking water (a^{-1}); D_i is the average daily exposure dose through drinking water contact ($\mathrm{mg} \cdot (\mathrm{kg} \cdot \mathrm{d})^{-1}$); RfD_i is the daily exposure dose per unit body weight for fluoride and chloride through drinking water ingestion ($\mathrm{mg} \cdot (\mathrm{kg} \cdot \mathrm{d})^{-1}$), with values of 0.06 $\mathrm{mg} \cdot (\mathrm{kg} \cdot \mathrm{d})^{-1}$ for fluoride and 0.10 $\mathrm{mg} \cdot (\mathrm{kg} \cdot \mathrm{d})^{-1}$ for chloride; IR is daily water intake (2.2 $\mathrm{L} \cdot \mathrm{d}^{-1}$ for adults, 1.0 $\mathrm{L} \cdot \mathrm{d}^{-1}$ for children); EF is exposure frequency (350 $\mathrm{d} \cdot \mathrm{a}^{-1}$); ED is exposure duration (30 years for adults, 6 years for children); BW is average body weight (64.3 kg for adults, 22.9 kg for children); AT is average exposure time (25,550 days for adults, 2,190 days for children); and C_i is the mass concentration of fluoride and chloride ($\mathrm{mg} \cdot \mathrm{L}^{-1}$).

Health Risk Model for Dermal Contact Pathway:

$$R_i^j = \frac{D_j \times 10^{-6}}{RfD_i \times 72}$$

$$D_j = \frac{C_i \times SA \times K_p \times FE \times EF \times ED \times f}{BW \times AT \times \tau}$$

Where:
R_i^j is the annual individual health risk from dermal contact (a^{-1}); D_j is the average daily exposure dose through dermal contact ($\mathrm{mg} \cdot (\mathrm{kg} \cdot \mathrm{d})^{-1}$); SA is body surface area (16,000 cm^2 for adults, 6,660 cm^2 for children); K_p is dermal adsorption parameter (2×10^{-3} $\mathrm{cm} \cdot \mathrm{h}^{-1}$); FE is bathing frequency (1 $\mathrm{time} \cdot \mathrm{d}^{-1}$); f is intestinal adsorption ratio (1); and τ is lag time (0.56 h).

Overall Health Risk:

$$R_{\text{total}} = R_i^d + R_i^j$$

2.1 Groundwater Risk Assessment

The hydrochemical characteristics of the study area are presented in Table 7. The pH ranges from 7.20-7.70 for shallow groundwater, 7.30-8.00 for middle groundwater, and 7.60-8.40 for deep groundwater, indicating weakly alkaline conditions. Total dissolved solids (TDS) show a trend of shallow > middle > deep groundwater, which is related to weathering intensity. Compared to the standard values, the mean fluoride concentrations in shallow (1.77 $\mathrm{mg} \cdot \mathrm{L}^{-1}$), middle (1.18 $\mathrm{mg} \cdot \mathrm{L}^{-1}$), and deep (1.21 $\mathrm{mg} \cdot \mathrm{L}^{-1}$) aquifers all exceed the limit specified in *Groundwater Quality Standard* (GB/T 14848-2017) (1.0 $\mathrm{mg} \cdot \mathrm{L}^{-1}$).

The fluoride exceedance rates are 57.14% for shallow, 40.00% for middle, and 60.00% for deep groundwater.

To determine whether current groundwater quality meets drinking water standards, the improved Nemerow pollution index was applied to evaluate water quality across different aquifers in the western Tarim Basin (Table 1). The proportions of water quality grades are shown in Table 2. In shallow groundwater, 60.00% of samples are Class II water and 20.00% are Class III water, with 20.00% classified as Class IV water. In middle groundwater, 57.14% are Class II water, 28.57% are Class III water, and 14.29% are Class IV water. Deep groundwater shows 90.00% Class II water and 10.00% Class IV water, with no Class III water present. Most sampling points across the three aquifers fall between Class II and Class III, meeting standards for centralized drinking water sources. However, individual sampling points in each aquifer do not meet drinking water standards (Class IV water), requiring attention to drinking water safety. Comparatively, shallow groundwater has the largest proportion of Class IV water, likely due to greater susceptibility to external factors (climate, human activities). The shallow aquifer lithology is primarily limestone ($CaCO_3$), which dissolves to increase Ca^{2+} and HCO_3^- concentrations, creating more favorable conditions for fluoride enrichment compared to middle and deep aquifer lithologies.

To further investigate potential health risks and considering the arid to semi-arid climate conditions that significantly impact fluoride and chloride contamination, the US EPA-recommended health risk assessment model was used to evaluate fluoride and chloride across different aquifers. Comprehensive health risk assessments were conducted for both drinking water and dermal pathways (Table 4). Drawing on research by Ding et al. regarding fuzzy analysis of human health risks, the average health risks in the study area fall within the low-risk zone (1.0\$×10^{-6}\$ to 1.0\$×10^{-4}\$) (Fig. 2). The results align with those from the improved Nemerow pollution index method.

Across different populations, health risks from drinking water pathways exceed those from dermal pathways, related to the degree and frequency of water contact. Among different aquifers, the average health risk follows the trend: shallow > deep > middle groundwater, with health risks decreasing with increasing burial depth. The overall health risks from all aquifers are below the maximum acceptable risk values of US EPA (1\$×10^{-4}\$ a^{-1}) and the International Commission on Radiological Protection (5\$×10^{-5}\$ a^{-1}). However, children' s health risks exceed those of adults, likely due to differences in immunity and respiration rates. In arid climates, evaporation favors fluoride enrichment in groundwater. Shallow groundwater tends toward evaporation-crystallization mechanisms, and combined with soil capillary action, soluble ions move upward, forming the dominant factor for relatively high fluoride concentrations in shallow water. The positive correlation between TDS and fluoride enrichment indicates that high evapotranspiration in arid regions lowers the water table and increases TDS, indirectly promoting fluoride enrichment. Middle and deep groundwater tend toward rock weathering mechanisms, with fluoride enrichment related to burial depth—greater depth results in less evaporation-crystallization and more rock weathering, indicating that rock weathering is the main geochemical process controlling fluoride in this region.

2.2 Groundwater Hydrochemical Zoning

High fluoride concentrations in groundwater are related to groundwater type. As shown in Fig. 3, the major ions with significant mass proportions include Na^+, Ca^{2+}, Mg^{2+}, HCO_3^-, SO_4^{2-}, and Cl^-. The hydrochemical types are: $Cl \cdot SO_4$-$Na \cdot Ca$ type for shallow water, $SO_4 \cdot HCO_3$-$Na \cdot Mg$ type for middle water, and $SO_4 \cdot HCO_3$-$Ca \cdot Na$ type for deep water. These results are consistent with findings by Dehbandi et al. and Hossain et al. Based on endmember values, the weathering of dolomite, basalt, and granite primarily involves sulfate dissolution of granite and carbonate rocks, which explains the high SO_4^{2-} concentrations in the water.

2.3 Rock Weathering

Gibbs diagrams illustrate two important natural mechanisms controlling major ion chemistry in groundwater: rock weathering and evaporation-crystallization. The scatter plot of TDS versus the weight ratio of $Na^+/(Na^+ + Ca^{2+})$ and the weight ratio of $Cl^-/(Cl^- + HCO_3^-)$ shows that shallow, middle, and deep groundwater samples in the study area plot between the rock weathering and evaporation-crystallization mechanisms, with most sampling points 偏向 ing toward rock weathering chemical processes. This is consistent with the endmember analysis above and aligns with research results from other aquifer studies in the region. In arid climate zones, evaporation processes favor fluoride enrichment in groundwater. Shallow groundwater tends toward evaporation-crystallization mechanisms, and combined with soil capillary action, soluble ions move upward, forming the main controlling factor for relatively high fluoride concentrations in shallow water.

2.4 Dissolution and Precipitation

The scatter plot of molar concentration ratios shows that major ion concentration ratios in groundwater are primarily distributed between silicate mineral dissolution and carbonate mineral dissolution, 偏向 ing toward silicate mineral dissolution. This indicates that groundwater chemistry in the study area is mainly related to silicate and carbonate mineral dissolution, though contributions from evaporite mineral dissolution cannot be excluded.

Fluorite (CaF_2) is the most common fluoride-bearing mineral. Changes in Ca^{2+} concentration affect fluorite dissolution or precipitation [Eq. (9)], thereby controlling fluoride variation in groundwater. In hydrogeochemistry, mineral dissolution and precipitation are also related to the saturation index (SI) of minerals in groundwater. Geochemical modeling software PHREEQC was used to calculate SI values for fluorite, calcite, and dolomite. When $SI < 0$, minerals tend to dissolve; when SI > 0, minerals tend to precipitate; when SI = 0, minerals are in equilibrium.

The results show that most sampling points have calcite in an unsaturated state. A few fluorite samples in shallow water are saturated, while most are unsaturated. The formation of high fluoride water may be due to Eq. (8) proceeding to the right, reducing Ca^{2+} concentration and indirectly promoting fluorite dissolution. Some dolomite samples in shallow groundwater are saturated, while dolomite in middle and deep groundwater is unsaturated, promoting Eq. (8) to the right, reducing Ca^{2+} concentration, and facilitating Eq. (9) to increase fluoride concentration. This demonstrates that water bodies unsaturated with respect to calcite and dolomite drive fluorite dissolution in strata, providing favorable conditions for further increasing groundwater fluoride concentrations.

2.5 Cation Exchange

Cation exchange processes in high-fluoride areas were investigated to further explain fluoride enrichment causes. The chlor-alkali index (CAI) proposed by Schoeller was used for analysis:

$$CAI_1 = \frac{Cl^- - (Na^+ + K^+)}{Cl^-}$$

$$CAI_2 = \frac{SO_4^{2-} + HCO_3^- + CO_3^{2-} - (Na^+ + K^+)}{Cl^-}$$

When both CAI_1 and CAI_2 are positive, ion exchange occurs between Na^+ in groundwater and Ca^{2+} in surrounding rocks; when negative, Ca^{2+} in groundwater exchanges with Na^+ in surrounding rocks. The larger the absolute value, the stronger the cation exchange in the groundwater environment. Fig. 7 shows that both CAI_1 and CAI_2 are negative for all three aquifers, indicating that Ca^{2+} in groundwater exchanges with Na^+ in surrounding rocks. This indirectly promotes fluorite dissolution and increases fluoride concentration, demonstrating that high Ca^{2+} inhibits fluoride enrichment, consistent with previous analysis. Cation exchange indirectly controls fluoride concentration by altering cation concentrations (particularly Ca^{2+}) in groundwater. Low Ca^{2+} is more favorable for fluoride enrichment, as shown in Eqs. (10) and (11):

$$2NaX + Ca^{2+} \rightarrow 2Na^+ + CaX_2$$

$$CaF_2 + 2NaX \rightarrow 2Na^+ + 2F^- + CaX_2$$

3 Conclusions

Based on analysis of hydrochemical characteristics of shallow (0–50 m), middle (50–100 m), and deep (100–200 m) groundwater in the study area, this paper identified fluoride content in different aquifers, evaluated groundwater quality, and investigated fluoride enrichment processes. The main conclusions are:

1) Water quality at many aquifer sampling points falls between Class II and Class III, though Class IV water exists in each aquifer. Using the US EPA health risk assessment model, the overall health risks from fluoride and chloride in shallow, middle, and deep water are 1.28$\times10^{-6}$ a^{-1}, 1.02$\times10^{-6}$ a^{-1}, and 9.36$\times10^{-7}$ a^{-1} for adults, and 1.63$\times10^{-6}$ a^{-1}, 1.30$\times10^{-6}$ a^{-1}, and 1.19$\times10^{-6}$ a^{-1} for children, respectively—all below US EPA maximum acceptable risk values. However, fluoride exceedance occurs at sampling points across all aquifers. Therefore, attention must be paid not only to further deterioration of shallow groundwater quality but also to fluoride exceedance to avoid increased health risks.

2) Hydrochemical types in different aquifers are: $Cl \cdot SO_4$-$Na \cdot Ca$ type (shallow water), $SO_4 \cdot HCO_3$-$Na \cdot Mg$ type (middle water), and $SO_4 \cdot HCO_3$-$Ca \cdot Na$ type (deep water). Mean fluoride concentrations are 1.77 $mg \cdot L^{-1}$, 1.18 $mg \cdot L^{-1}$, and 1.21 $mg \cdot L^{-1}$, respectively, with shallow water > deep water > middle water. All aquifers exceed the *Groundwater Quality Standard* (GB/T 14848-2017) limit of 1.0 $mg \cdot L^{-1}$.

3) High-fluoride groundwater in the study area is characterized by alkaline conditions, high Na^+, and low Ca^{2+}. Alkaline conditions favor Ca^{2+} precipitation, promoting fluorite dissolution and F^- release. Silicate mineral dissolution, rock weathering, and ion exchange are important factors affecting groundwater fluoride concentration. Extensive livestock grazing and random grassland reclamation have caused grassland degradation and desertification, which also influence fluoride enrichment in groundwater.

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

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