Distribution and hydrogeochemical behavior of arsenic-enriched groundwater in the sedimentary aquifers: Comparison between Datong Basin, China and Kushtia District, Bangladesh

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ABSTRACT: High arsenic (As) groundwater has become a major global concern due to its widespread occurrence. A comparative hydrogeochemical study was performed on the As-enriched groundwater in Datong Basin, China and Kushtia District, Bangladesh. The elevated As concentrations of Datong Basin ranged from 0.31 to 452 $\mu\text{g L}^{-1}$. As enriched groundwater is mainly Na-HCO₃ type water, and characterized by higher pH value, high Na⁺, low Ca²⁺, SO₄²⁻ and NO₃⁻ along with moderate TDS. The release of As in aquifers resulted from the reduction of As-carrying crystalline iron (Fe) oxide/hydroxides and oxidation of organic matter. The aquifers of Kushtia District, Bangladesh are unconsolidated, alluvial in nature, and developed from Holocene floodplain and Pleistocene deposits. High As (6.04–590.7 $\mu\text{g L}^{-1}$) groundwater occurs mainly in shallow aquifers. The Ca-HCO₃ type groundwater is distinguished by circum-neutral pH, medium-high EC, high HCO₃⁻, and low content of NO₃⁻, SO₄²⁻, K⁺ and Cl⁻. The reductive suspension of MnOOH increased the dissolved As loads and redox responsive elements such as SO₄²⁻ and pyrite oxidation acted as the main mechanisms for As release in groundwater. The weak loading of Fe suggests that the release of Fe and As was decoupled in sedimentary aquifers of Kushtia District.

1 INTRODUCTION

High arsenic (As) groundwater is the most crucial natural hazard to public health and the largest environmental hazards in the world (Wang & Wai, 2004). Among the As polluted regions, India, Bangladesh, and China are mostly affected. Currently, millions of people are still consuming As contaminated water above the WHO recommended value (10 $\mu\text{g L}^{-1}$) for drinking water both in China and Bangladesh (Bibi *et al.*, 2008; Wang & Deng, 2009). Since last two decades, many studies have been conducted on the causes of As enrichment at Datong Basin, and Bangladesh but the different mechanisms of As release has remain poorly defined. The objectives of this study are (1) to compare the hydrogeochemical characteristics of the elevated As groundwater, and (2) to identify the different mechanisms of As release occurred in the sedimentary aquifers of these two areas.

2 METHODS

2.1 Study area

Datong Basin, one of the Cenozoic fault basins of the Shanxi rift system that is situated in the north of Shanxi Province, China and Kushtia District is located in the southwest of Bangladesh were selected as the study area.

2.2 Sampling and hydrogeochemical analysis

To investigate the hydrogeochemical properties of groundwater, a total of 132 groundwater samples (83

from Datong Basin and 49 from Kushtia District) were collected from tubewells. All of the water samples were filtered through 0.45 μm membranes and preserved in 50 ml polyethylene bottles for anion, cations and trace elements analysis. Hydride generation-atomic fluorescence spectrometry (HG-AFS) (Titan AFS 830) inductively coupled plasma atomic emission spectrometry (ICP-AES) and Ion Chromatography (IC) (Metrohm-761 Compact IC) were used to measure total dissolved As, major ions and concentration of trace elements respectively.

3 RESULTS AND DISCUSSION

Different mechanisms are responsible for As enrichment in the groundwater of each study area. Factor analysis (FA) was performed on the hydrochemical data to explore the causes of elevated As occurrence in the groundwater of these two study areas. In Datong Basin, China, *F1* explains 20% of total variance including strong positive loadings of Ca²⁺, Mg²⁺, Na⁺, Cl⁻, SO₄²⁻, Fe, Cu, Se, Li, B, Sr and V with the negative loading of EC, while others show weak loading (Table 1). The positive loading of Cl⁻ can be derived due to evaporation and may increase along the flow path from recharge to discharge area. *F2* accounts for 19.79% of total variance, with strong positive loading of pH, B, and Ba and negative loading of Ca²⁺. In Datong Basin, high As mostly occurs with the pH value exceeding 7.95. *F3* covers 16.28% of the total variance. Ba is an indicator for a marine groundwater environment, so this factor may be associated with a marine groundwater environment due

Table 1. Factor loadings of different chemical parameters of groundwater from the two study sites.

Components	Datong Basin, China			Kushtia, Bangladesh		
	F1	F2	F3	F1	F2	F3
As	-.311	.318	-.141	-.055	.146	.108
Ca ²⁺	.602	-.659	.245	.161	.075	.884
K ⁺	.441	-.225	.234	.680	.085	-.049
Mg ²⁺	.760	-.468	-.097	.507	.548	.253
Na ⁺	.798	-.077	-.521	.817	-.138	.217
Cl ⁻	.798	-.077	-.521	.888	.197	.105
NO ₃ ⁻	.407	-.149	-.049	-.004	.048	-.104
SO ₄ ²⁻	.806	-.365	-.271	.848	.048	-.002
pH	-.308	.504	-.484	-.017	-.557	-.049
EC	-.590	.103	.243	.347	.675	.215
T	-.148	.112	.024	.092	.181	.217
Depth	-.480	.147	.032	-.167	-.090	-.188
HCO ₃ ⁻	.477	.409	-.593	.193	.213	.806
TDS	.370	-.254	-.382	.487	.207	.683
Cr	.026	-.025	.250	.008	-.185	.050
Mn	.481	.264	.343	-.169	-.129	.783
Fe	.751	-.276	.407	-.197	.205	-.191
Ni	.370	.117	.398	-.008	-.010	.039
Cu	.822	.308	.133	-.203	-.097	.012
Pb	.467	.365	-.245	.042	-.785	-.122
Se	.710	.425	.241	-.321	.100	.304
Li	.876	.182	.159	-.010	-.778	.063
B	.684	.619	-.148	.362	.580	.075
Ba	.282	.612	.570	.552	.018	.269
Sr	.851	-.092	.258	.415	.307	.575
V	.641	.416	.121	-.015	-.736	-.104
Variability (%)	20.00	19.79	16.28	16.06	13.8	12.9
Cumulative (%)	20.00	39.79	56.07	16.06	29.9	42.8

to the strong positive loading of Ba (0.57). In Kushtia District, Bangladesh in F1 strong loading of Mg²⁺ suggests that it may come from the weathering of silicate or carbonate suspension. The excessive alkalinity may serve as an extra source of Mg²⁺ and the balance of Cl⁻ and SO₄²⁻. In F2, the strong negative loading of pH suggests that the pH-dependent reactions are not the predominant features of As enrichment in groundwater. F3 shows a positive loading of Ca²⁺ that is favorable to the development of the alkali-type soil. Regular and long-term irrigation with Ca²⁺ enriched water in this area makes the soil plastic as well as humid in the wet season and form clod soil crust in the dry season. HCO₃⁻ also shows a positive strong loading indicating three main weathering factors (evaporation, silicate, and carbonate) which increase the As concentration (Table 1).

The negative loading of HCO₃⁻ in Datong Basin, indicates that the alkaline environment is associated with the As mobilization (Table 1 and Figure 1a). The comparison of HCO₃⁻ and dissolved Fe suggests that Fe reduction plays a vital role in As release (Figure 1c). HCO₃⁻ and SO₄²⁻ are considered to induce As desorption from the Fe mineral shells, and their interactions in the present study reveal that the reactions of redox are the major controlling mechanisms to release As in aquifer system.

The weak loading of Fe (Table 1) of Kushtia District, Bangladesh may be ascribed to the re-precipitation of suspended Fe, suggesting that the release of Fe and As is decoupled or influenced by other geochemical

Figure 1. Correlation plots of As with (a) HCO₃⁻ for Datong Basin (b) HCO₃⁻ for Kushtia (c) Fe for Datong Basin and (d) SO₄²⁻ for Kushtia.

mechanisms (Radloff *et al.*, 2007). The strong positive loading of Mn (Table 1) indicates that the As is predominantly released with the bacterial reductive suspension of Mn oxy-hydroxides. The influence of HCO₃⁻ might dominate the As release of the aquifers of Kushtia District in reduction dissolution (Figure 1b). The significant loading of SO₄²⁻ demonstrates that the pyrite oxidation is the key mechanism to As discharge in groundwater (Figure 1d).

4 CONCLUSIONS

The association of As and Fe indicates that the iron-bearing minerals might be the key carrier of elevated As in aquifers. High alkaline situation, As release from sediments with high pH environment, and As enrichment under reducing condition are the responsible factors that control and mobilize As in groundwater from sedimentary aquifers of Datong Basin China. In Kushtia district high As mainly occurred in the shallow aquifer indicating that the shallow parts are recharged/infiltrated by the surface water supply. Low concentrations of NO₃⁻ and SO₄²⁻ reveal an absolute reducing situation. The high concentration of HCO₃⁻ maybe related to microbial mediated Fe(III) reduction of organic matter, which creates the reducing condition and releases As in the aquifers. The strong positive loading of Mn suggests that the suspension of MnOOH might be responsible for dissolved As loads.

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