Global Distribution of Geogenic High-Arsenic Groundwater

Subjects: Water Resources

Contributor: Jiju Guo, Wengeng Cao, Guohui Lang, Qifa Sun, Tian Nan, Xiangzhi Li, Yu Ren, Zeyan Li

Groundwater constitutes a vital source of freshwater, accounting for roughly 95% of the total available freshwater resources on Earth. It is utilized not only for daily water needs but also for agricultural irrigation, industrial purposes, ecological recharge, and power generation.

Keywords: high-arsenic groundwater ; worldwide scale ; in situ remediation of arsenic ; human health risk assessment

1. Introduction

Groundwater constitutes a vital source of freshwater, accounting for roughly 95% of the total available freshwater resources on Earth [1]. It is utilized not only for daily water needs but also for agricultural irrigation, industrial purposes, ecological recharge, and power generation [2]. Therefore, groundwater holds significant value as a resource and plays a critical role in the environment. The degradation of groundwater quality represents a significant issue within the context of global environmental and climate change today. Since the Industrial Revolution, there has been widespread concern over the deterioration of groundwater quality [3]. Among the various groundwater quality issues, the release of high concentrations of heavy metals has had a significant impact on groundwater quality, and serious consideration must be given to its potential risks and hazards to human health. In particular, As is considered by the United States Agency for Toxic Substances and Disease Registry (ATSDR) to be the pollutant that poses the highest potential risk to human health due to the release from natural sources and the resulting high geogenic concentrations in groundwater [4]. The sources of As in groundwater primarily include natural origins such as geological formations, volcanic activity, and hydrothermal processes, as well as anthropogenic activities including mining, coal combustion, and petroleum extraction [5]. The majority of global health issues caused by As are linked to the consumption of water with high As concentrations. Due to the wide range of negative effects of high As concentrations on human health, the World Health Organization (WHO), the United States, and the European Union (EU) have lowered the Maximum Contaminant Level (MCL) of As in drinking water from 50 μg/L to 10 μg/L as a safe limit for As concentration in drinking water [6][7]. High-As groundwater is defined as groundwater with As concentrations above the WHO drinking water standard. The enrichment of high-As groundwater is primarily influenced by a combination of natural sources and hydrogeochemical conditions, with the majority of natural high-As groundwater primarily being a result of geological arsenic contamination [5]. Despite the established risks, many countries, such as Bangladesh, Nepal, Pakistan, Mexico, and Argentina, continue to adopt the 50 μg/L standard for arsenic concentration in their national drinking water guidelines, due to a lack of professional expertise, economic considerations, and the low-level arsenic detection technology [8].

Human exposure to As occurs through direct and indirect pathways. Direct exposure involves drinking water with a high As concentration, contact with skin, and inhalation of gasses with a high As concentration. Indirect exposure mainly occurs through the food chain; this includes eating crops, vegetables, and fruits cultivated in As-contaminated soil or irrigated with As-rich groundwater, as well as consuming meat products from animals raised in such environments. Prolonged exposure to As, regardless of the route, can result in serious health disorders affecting the skin, blood vessels, and nervous system. Extended periods of high As exposure also notably increase the risk of developing cancers in organs like the lungs, liver, kidneys, and skin [9][10] (**Figure 1**).

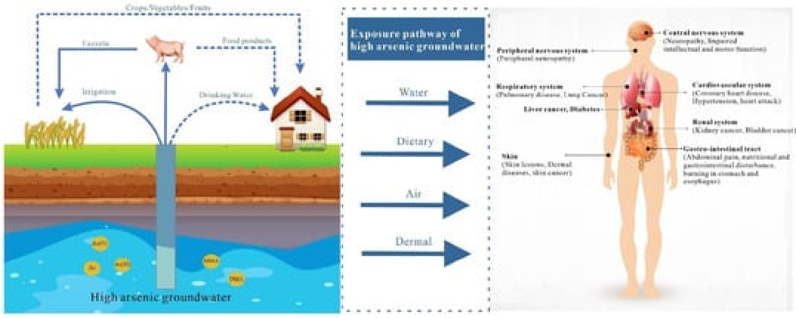

Figure 1. Different pathways of arsenic exposure in groundwater and effects on humans (MMA-Monomethylarsenite; DMA-Dimethylarsenite).

Environmental As exists in groundwater in both organic and inorganic forms, with varying levels of toxicity associated with different forms. The three primary forms of inorganic arsenic are as follows: pentavalent arsenate [As(V)], trivalent arsenite [As(III)], and metallic arsenic. Arsenic in organic form often occurs as various organic arsenic compounds such as Monomethylarsenite (MMA) and Dimethylarsenite (DMA) [11]. Among these, inorganic arsenic is more toxic to humans, and the toxicity significantly differs between the oxidation states of As(III) and As(V). The toxicity of As(III) is more than 60 times higher than that of As(V) and 70 times higher than that of methylated arsenic [12]. The heightened toxicity of As(III) is partially because of its reactivity towards biologically relevant molecules [13]. The methylated arsenic forms, including MMA and DMA, exhibit moderate toxicity, while other organic forms, such as arsenobetaine (AsB) and arsenocholine (AsC), are generally considered non-toxic [12]. In aqueous solutions, As(III) and As(V) primarily exist as oxyanions due to the high charge and small ion radius of As^{3+} and As^{5+}. The presence and dispersion of distinct arsenic compounds within hydrological systems are markedly influenced by both the redox potential and pH levels prevailing in aquatic environments [14]. Under circumstances characterized by moderate-to-high redox potentials, As tends to stabilize into the As(V) form (H_3AsO_4, $H_2AsO_4^-$, $HAsO_4^{2-}$, or AsO_4^{3-}). Conversely, in environments featuring predominantly acidic or weakly alkaline reducing conditions, and lower redox potentials, As(III) tends to be prevalent as the uncharged H_3AsO_3 molecule [15].

2. Global Distribution of Geogenic High-Arsenic Groundwater

High-As groundwater is widespread worldwide. According to statistics, 107 countries are affected by high-As groundwater, with the highest number in Asia (32) and Europe (31), followed by Africa (20), North America (11), South America (9), and Australia (4) [16]. The most affected countries are Bangladesh, India, Pakistan, China, Nepal [7], Laos [17], Cambodia [18], Myanmar [19], Vietnam [20], and the United States. The world map (**Figure 2**) displays the global distribution of geogenic high-As groundwater, predominantly found in inland basins and river deltas in South Asian, East Asian, and South American countries. Major countries are shown in **Table 1**. Generally, the river-marine sedimentary shallow (Holocene) aquifers in the river deltas are the main areas where high-As groundwater occurs naturally, and it occurs mainly under reducing aquifer conditions [21].

Figure 2. Main countries worldwide affected by geogenic high-arsenic groundwater (≥10 μg/L) (taking the maximum arsenic concentration), see **Table 1** for specific data.

Globally, the problem of geogenic high-As groundwater is particularly prominent in South and Southeast Asia, especially in Bangladesh and India [22]. In Bangladesh, 61 areas have been identified as having high-As groundwater. The potential population at risk is approximately 20 million people [23]. According to the National Drinking Water Survey of Bangladesh, around 8% of the water samples had As levels exceeding the Bangladesh standard of 50 μg/L, while around 18% of the samples were above the WHO guideline of 10 μg/L [24]. The concentration of As in groundwater is higher in Bangladesh compared to other countries, and some tube wells even contain As concentrations as high as 4730 μg/L [25][26]. In India, high-As groundwater has already affected twenty states and four union territories, and about 100 million people are under threat from the toxicity of high-As groundwater [16][27][28][29]. The impact of high-As groundwater in India is concentrated on the Ganges–Yarlung Tsangpo Plain, seen on the neo-alluvial (Holocene) floodplains of the rivers in the Himalayas [30][31]. Approximately 50–60 million individuals in Pakistan consume high-As groundwater (>50 μg/L) in vulnerable areas [32]. A meta-analysis of groundwater affected by As in Pakistan showed that 73% of these groundwater samples contained arsenic above 10 μg/L [33]. China is also one of the world's most representative areas of high-As groundwater, with more than 20 provinces/autonomous regions having high-As groundwater problems. These high-As groundwater provinces are mainly located in the fluvial/alluvial-lacustrine plains and basins (Yinchuan Plain, Hetao Plain, Guide Basin, Hohhot Basin, Junggar Basin, Datong Basin, etc.) located in arid/semi-arid regions and alluvial plains/basins and river deltas in humid/semi-humid regions (Yangtze River, Yellow River Delta, Pearl River Delta, Delta, Huaihe River, Alluvial Plain, Yellow River, Yuncheng Basin, Taiyuan Basin, Songnen Plain, etc.) [34][35][36][37]. The population affected by high-As groundwater contamination in China was estimated to be about 19.6 million according to a statistical risk assessment model developed by Rodríguez-Lado et al. [38].

Table 1. The occurrence of high-As groundwater reported by major countries in the world.

Country	Study Area	Max As conc. (μg/L)	Samples	Environmental Condition and/or Enrichment Mechanism	References
Afghanistan	**Ghazni and maidan Wardak provinces**	**990**	**746**	**The weathering and leaching action**	[39]
Argentina	**Santiago del Estero Province**	**14,969**	**40**	**Volcanic ash sedimentary environment; agricultural irrigation**	[40]
	La Pampa	**5300**	**44**	**The geological factors; weathering of volcanic ash and loess; oxidizing condition**	[41]
Australia	**Stuarts Point coastal**	**85**	**140**	**Desorption of As from Al-hydroxides and As-enriched Fe-oxyhydroxides; high concentrations of HCO_3^- and PO_4^-**	[42]
Bangladesh	**Noakhali**	**4730**	**52,202**	**Eroded by flood plain rivers**	[25]
Bolivia		**364**	**24**	**The alteration of volcanic rocks; evaporation and redox reactions**	[43]
Botswana	**Botswana**	**116**	**20**	**Delta; evaporation concentration; weakly alkaline environment; pH 6.29–8.60**	[44]
Brazil		**2980**		**Anthropogenic; volcanic activity and weathering of rocks**	[43]
Burkina Faso		**1630**	**45**	**Zones of gold mineralization in volcano-sedimentary rocks**	[45]

Country	Study Area	Max As conc. (μg/L)	Samples	Environmental Condition and/or Enrichment Mechanism	References
China	Datong Basin	1932	1022	The weak alkaline reductive environment; high HCO_3^- concentration; water–rock interactions	[46]
	Hetao Basin	572	63	The reducing conditions; the dissolved organic; the competitive effects of other anions	[47]
	Jianghan Basin	2330	34	The high HCO_3^- concentrations; microorganisms and exogenous substances; the seasonal variation; strongly reducing environment; reducing environment	[48]
	Taiwan (Lanyang and Chianan Plain)	1010		Alluvial plain; high DOC; strong reducing conditions	[49]
	Tarim Basin	91.2	233	Reducing environment; the dissolved organic; reductive dissolution release;	[50]
	Yinchuan	177	92	Agricultural irrigation; the reductive dissolution of Fe oxides; the high PO_4^- concentrations	[51]
	Pearl River Delta	161	18	Reductive environment; the high NH_4^+ concentrations; high concentrations of NH_4^+ and organic matter	[52]
Cambodian		1610	207	Holocene alluvial sediments; reducing environment	[53]
Costa Rica	Northern Costa Rica	29,100	35	Associated with the volcanic rock	[43]
Czech Republic	Mokrsko	1690	62	pH > 9	[54]
Ecuador		969	67	In hot springs	[43]
Ethiopia	Southwestern Ethiopia	184.5	44	pH < 7	[55]
Ghana		1760	357	Spillages of the mines; pH 4.8–6.99	[56]
Hungary	Southern Hungary	260	73	At a depth of 0.8–2.4 km and containing CH_4	[57]
India	Bhair	1466	1365	Ganga Plain; Holocene newer alluvium and the Pleistocene older alluvium	[58]
	Shahpur block, Bhojpur district, Bihar state	1805	4704	Ganges plain	[28]
	Punjab	3192	4780	Alluvial aquifers	[58]
Iran	Kurdistan Some villages	1500	27	Mining and sedimentary environment	[59]
	East Azarbaijan-Tabriz Plain	2000	18	Hydrogeological and environmental reducing conditions	
	Ardabil-A city	5834	163	Interaction of hydrothermal fluids with the rocks and geogenic source-geological structure	
	Mazandar an-Haraz River	110	20	Geogenic source and mining	
	Tabas South Khorasan	53	29	Weathering	
	Razavi Khorasan Chelpu Kashmar	606	12	Geogenic Origin sedimentary environment	
	Isfahan Mutehgold mining district	1061	17	Weathering and mining	
Japan		38	136	Reducing environment and factory blowdown	[26]

Country	Study Area	Max As conc. (μg/L)	Samples	Environmental Condition and/or Enrichment Mechanism	References
Korea	Geumsan County	113	150	Oxidation reaction of sulfide minerals in metasedimentary rocks and desorption process under high pH conditions	[60]
Nigeria	Warri-Port Harcourt, Ogun State, Kaduna	750	20	Alluvial sediments, reducing environment, slightly acidic	[16]
Pakistan	Kasur, Shhiwal, Bahawalpur, and Rahim Yar Khan	3090	395	Irrigation and factory sewage	[61]
	Lahore municipality	85	41	Topsoil and extensive irrigation of unconfined aquifers, reductive dissolution	[32]
	Mailsi	812	44	Human activity	[49]
Paraguay		120	37	Human activity and volcanic ash deposition environment	[43]
Lao PDR	Vientiane	24.4	3	Reducing environment	[17]
	Borikhamxay	30	7	Reducing environment	
	Champasack	25.6	27	Reducing environment	
	Attapeu	31.6	10	Reducing environment	
Myanmar	Ayeyarwady	630	55	Reductive dissolution of Fe oxyhydroxides	[49]
Mexico	La Laguna Region	5000	29	Adsorption or coprecipitation on iron oxides, clay-mineral surfaces, and organic carbon	
	Zacatecas	75.4	182	Geological origin, water–rock interaction	[49]
Nepal	Nawalparasi	2620	18,000	Seasons and climate change, water–rock interaction	
Pakistan	Larkana Sindh,	318	58	pH 6.8–8.1	[62]
	Punjab	655	141	pH 7.0–9.3	[63]
Spain	Duero Cenozoic Basin	613	514	pH 5.87–1.58	[64]
Thailand	Suphan Buri	5000	21	pH 5.20–5.90; Eh 250–370 mV	[16]
USA	San Joaquin Valley, California	148.5	4983	Arid and semi-arid basins; alluvial, fluvial, and lacustrine deposits; pH > 7.8; reducing conditions	[65]
	Lahontan Valley, in Churchill County, Nevada	4100	59	Lacustrine sediments	[66]
Vietnam	Mekong Delta	850	109	pH 7.22–8.63	[49]

In Europe, As contamination in groundwater is attributed to geothermal and hydrothermal systems, dominated by bedrock and volcanic deposits [67]. The situation in the Pannonian Basin (Romania, Serbia, and Hungary) is particularly noteworthy, as over 600,000 residents may be exposed to high-As groundwater [57]. Additionally, the maximum concentration of arsenic found in bedrock groundwater in Finland is 1040 μg/L. The highest concentration of As recorded in the Ischia Island area, southern Italy, was 1479 μg/L, which was 148 times higher than the MCL. Hydrothermal activity and thermal control seem to be the main factors responsible for the liberation of As from minerals [68].

The United States and Canada have also experienced extensive geogenic high-As groundwater contamination, although the concentration is lower than that of Asian countries [69]. In Latin America, arsenic compounds in groundwater are mainly derived from geothermal fluids as well as volcanic activity [43]. The As levels of drinking water are too high in 13 of Mexico's 31 states [70]. In particular, As concentrations of 5000 μg/L were discovered in pore weakly permeable layers in the La Laguna area [71]. Groundwater As sources of geothermal origin have been identified at Juventino Rosas in the State of Guanajuato and Ixtapan de la Sal and Tonatico in the State of México [72]. The area of Argentina most affected by

As in groundwater is the Chaco-Pampean Plain, with approximately 88% of the 86 collected groundwater samples surpassing the WHO guideline values, and the population at risk in Argentina is about 4 million people [54].

In Africa, high-As groundwater has been found in only a few areas across the continent, primarily in the western and southern regions, more due to insufficient research rather than a shortage of problems [67]. Twenty countries in Africa have recorded high concentrations of arsenic in groundwater, including Botswana, Burkina Faso, Ethiopia, and Ghana [67]. The maximum concentration of As in groundwater in Burkina Faso was 1630 μg/L, while an analogous maximum concentration of 1760 μg/L was detected in groundwater in Ghana [56][73].

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