

Review

Worldwide Distribution, Health Risk, Treatment Technology, and Development Tendency of Geogenic High-Arsenic Groundwater

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Abstract: The presence of high concentrations of geogenic arsenic (As) in groundwater poses a serious threat to the health of millions of individuals globally. This paper examines the research progress of groundwater with high concentrations of geogenic As through a comprehensive literature review and analysis, covering distribution, health risks, in situ remediation, regulatory technologies, and development trends, to establish a reference for future research. The global distribution of geogenic high-As groundwater is mainly in inland basins and river deltas of countries in South Asia, East Asia, and South America. High-As risk areas can be modeled using hydrogeologic data and field measurements. This modeling approach allows for assessing and measuring potential areas of high-As groundwater. In order to provide safe drinking water promptly and effectively to areas affected by high-As groundwater, in situ rapid detection and remediation techniques have been given significant attention. This paper introduces household- or community-scale As removal technologies, including flocculant–disinfectant, bucket treatment units, use of activated alumina, use of nano zero-valent iron, aquifer iron coating technology, and bioremediation, summarizing the basic mechanisms of arsenic removal for each technology. Guaranteeing the sustainability of site-scale remediation technologies, reasonable aquifer management, and exploring alternative water sources are crucial for combating high-As groundwater contamination. Future studies should aim to elucidate the mechanisms of As's coexistence with other pollutants in groundwater, effectively treating As-containing wastes or sludge produced during the treatment process and exploring better treatment options.

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



Citation: Guo, J.; Cao, W.; Lang, G.; Sun, Q.; Nan, T.; Li, X.; Ren, Y.; Li, Z. Worldwide Distribution, Health Risk, Treatment Technology, and Development Tendency of Geogenic High-Arsenic Groundwater. *Water* **2024**, *16*, 478. <https://doi.org/10.3390/w16030478>

Academic Editors: Peiyue Li, Jianhua Wu and Vetrinuragan Elumala

Received: 26 December 2023

Revised: 25 January 2024

Accepted: 29 January 2024

Published: 31 January 2024



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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 $\mu\text{g/L}$ to 10 $\mu\text{g/L}$ 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 $\mu\text{g/L}$ 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).

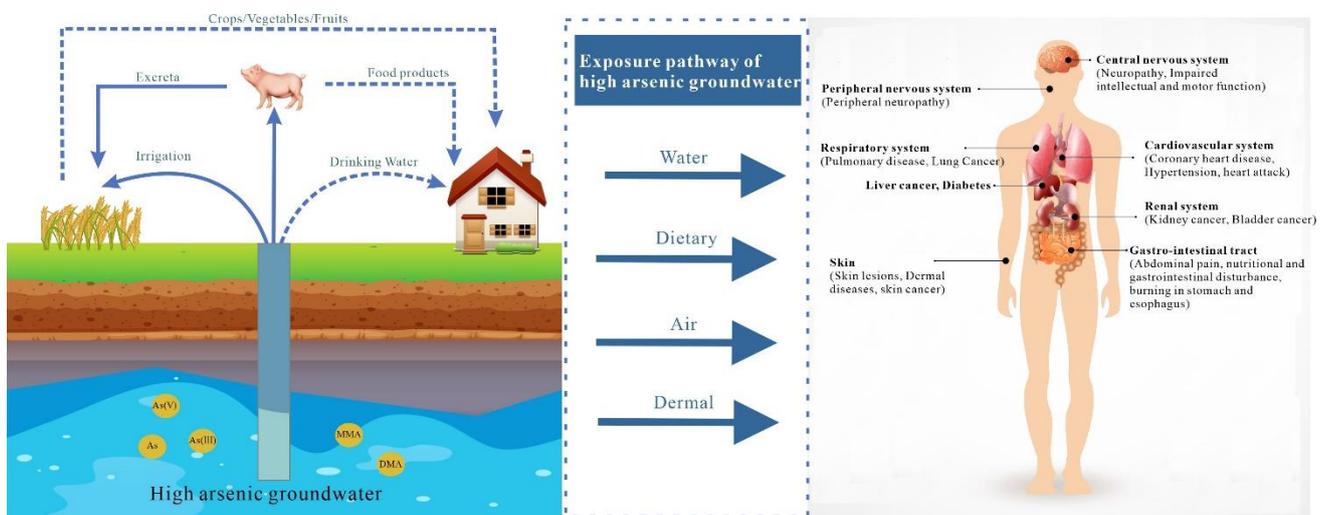


Figure 1. Different pathways of arsenic exposure in groundwater and effects on humans (MMA—Monomethylarsenite; DMA—Dimethylarsenite).

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].

Based on a thorough review of the literature, this paper comprehensively summarizes and maps the distribution of geogenic high-As groundwater worldwide. In addition, health risk assessment methods for high-As groundwater across the globe are summarized. Several remediation technologies are also condensed, including in situ monitoring and detection, aquifer remediation, and technologies for preventing and controlling regional As pollution. Furthermore, the paper discusses the development trend of high-As groundwater research.

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].

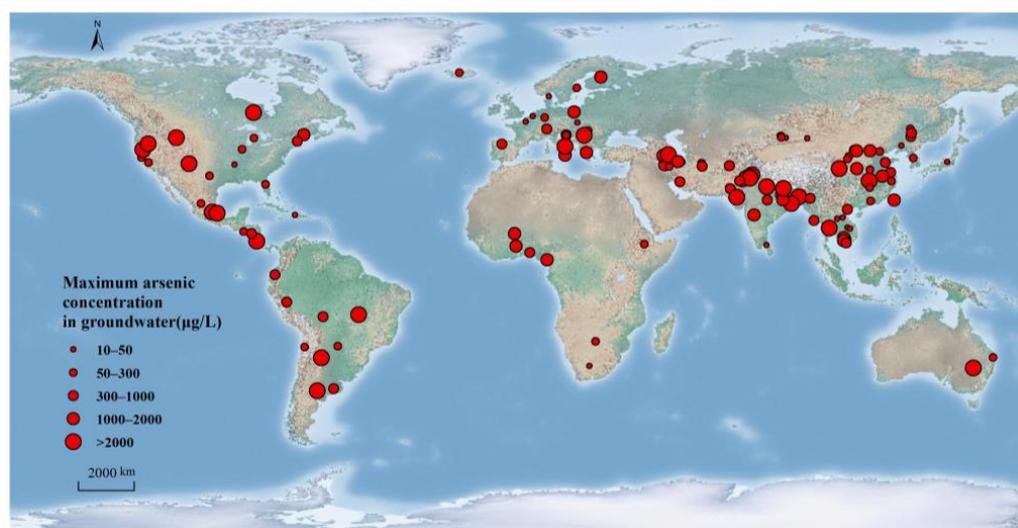


Figure 2. Main countries worldwide affected by geogenic high-arsenic groundwater ($\geq 10 \mu\text{g/L}$) (taking the maximum arsenic concentration), see Table 1 for specific data.

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 [16,27–29]. 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.) [34–37]. 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]

Table 1. Cont.

Country	Study Area	Max As conc. ($\mu\text{g/L}$)	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]
	Jiangnan 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	
	East Azarbaijan-Tabriz Plain	2000	18	Hydrogeological and environmental reducing conditions	[59]
	Ardabil-A city	5834	163	Interaction of hydrothermal fluids with the rocks and geogenic source-geological structure	

Table 1. Cont.

Country	Study Area	Max As conc. ($\mu\text{g/L}$)	Samples	Environmental Condition and/or Enrichment Mechanism	References
Iran	Mazandar an-Haraz River	110	20	Geogenic source and mining	[59]
	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]
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	[49]
	Zacatecas	75.4	182	Geological origin, water-rock interaction	
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]

Table 1. Cont.

Country	Study Area	Max As conc. ($\mu\text{g/L}$)	Samples	Environmental Condition and/or Enrichment Mechanism	References
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 $\mu\text{g/L}$. The highest concentration of As recorded in the Ischia Island area, southern Italy, was 1479 $\mu\text{g/L}$, 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 $\mu\text{g/L}$ 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 $\mu\text{g/L}$, while an analogous maximum concentration of 1760 $\mu\text{g/L}$ was detected in groundwater in Ghana [56,73].

3. Health Risks

High-As groundwater is directly or indirectly exposed to humans through ingestion, inhalation, and dermal exposure. By conducting health risk assessments and potential exposure assessments for areas with elevated levels of As in groundwater, it is possible to quantify the potential hazards to human health. This will also enable effective measures to be developed to control and protect public health.

3.1. Health Risk Assessment

The United States Environmental Protection Agency (USEPA) defines Human Health Risk Assessment as a procedure for estimating the nature and likelihood of adverse health effects in people likely to be exposed to chemicals in polluted environments [74]. There are generally four steps: hazard identification, dose–response assessment, exposure assessment, and risk characterization [75,76] (Figure 3). Currently, two human health risk assessment methods are commonly used, namely deterministic risk assessment and probabilistic risk assessment.

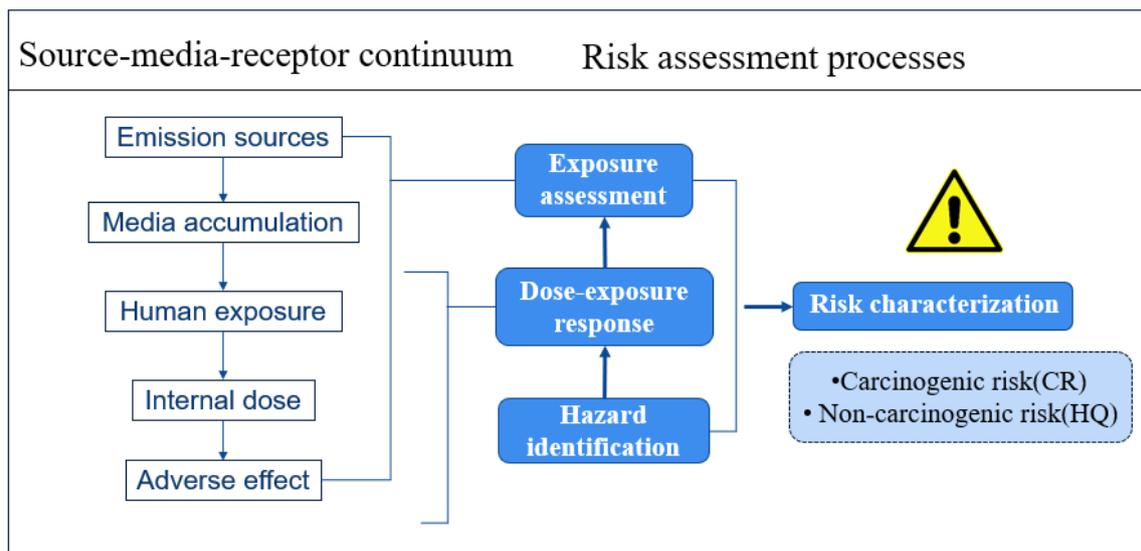


Figure 3. The generic conceptual model and steps of health risk assessment (modified after [75]).

An adoption of the health risk assessment model for the water environment was proposed by the USEPA [77]. Exposure to As in drinking water is mainly via both drinking water and skin contact [78]. The equations for long-term average daily exposure to groundwater for both exposure routes are given in Equations (1) and (2):

$$ADD_{oral} = \frac{C \times IR \times EF \times ED}{BW \times AT} \quad (1)$$

$$ADD_{dermal} = \frac{C \times SA \times K_p \times ET \times EF \times ED \times CF}{BW \times AT} \quad (2)$$

where ADD_{oral} and ADD_{dermal} are the average daily exposure dose (mg/kg/day) for drinking water and dermal routes of exposure, respectively; C is the concentration of As in groundwater (mg/L); IR is the intake rate (L/day); EF is the exposure frequency, which is the duration of exposure in one year (day/year); ED is duration of exposure, which represents the number of years in a lifetime that the substance is ingested into the body (years); BW is the average body weight (kg); AT is the average exposure time (days); SA is the surface area of skin in contact with groundwater (cm²); K_p is the skin permeability coefficient of different indicators (cm/h); ET is the exposure time (h/day); and CF is the conversion factor (L/cm³).

According to the health risk assessment model, the potential risk to human health from chemical carcinogens and non-carcinogens under the same exposure pathway is calculated differently. The hazard quotient (HQ) was used to calculate the non-carcinogenic risk to consumers of high-As groundwater in the study area, where $HQ > 1$ indicates that it may have a potential non-carcinogenic but harmful effect, and $HQ < 1$ indicates that the substance is considered safe for drinking water consumers [79]. The formula is as follows:

$$HQ = \frac{ADD}{RfD} \quad (3)$$

where RfD is the reference dose (mg/kg/day) and the reference dose for both oral and dermal exposure to As is 0.3.

The sum of the HQ s calculated for each individual element is the overall likelihood of non-cancer effects for all elements, expressed as a combined hazard index (HI). $HI > 1$ indicates a potential for an adverse effect on human health and the necessity for further

study. The higher the value of the *HI*, the higher the level of adverse health effects and non-cancer health risks [80]. The calculation formula is shown below:

$$HI = \sum_{i=1}^n HQ_i \quad (4)$$

The risk of developing cancer due to the different routes of exposure to carcinogens is given in Equations (5)–(7):

$$CR_j = \sum_{j=1}^n (ADD_j \times SF_j) (CR < 0.01) \quad (5)$$

$$CR_j = \sum_{j=1}^n [1 - \exp(-ADD_j \times SF_j)] (CR \geq 0.01) \quad (6)$$

$$CR_{total} = \sum_{j=1}^n CR_j \quad (7)$$

where *CR* is the individual carcinogenic risk index, and *SF_j* is the cancer slope factor for As (kg·day/mg). *CR_{total}* is the overall carcinogenic risk index. If *CR* or *CR_{total}* < 10^{−6}, the level of carcinogenic risk can be ignored; if *CR* or *CR_{total}* > 10^{−4}, it indicates the presence of an unacceptable human carcinogenic risk [81–84].

3.1.1. Deterministic Risk Assessment

Through determined parameters and known condition values, risk assessment findings in the Rupnagar district of Punjab, India, and the Salda Lake Basin in Burdur province, Turkey, have confirmed the potential for high non-carcinogenic and carcinogenic effects of As [79,85]. Similarly, researchers conducted monitoring and data collection in the Kuitun River Basin in the southwestern part of the Junggar Basin, China. The health risk assessment indicates that the risk of developing cancer from As in groundwater is higher in adults than in children. Particularly, the average *CR* for adults who consume groundwater was much greater than that for children, with a difference of one magnitude order between them [84]. As poses a greater cancer risk to adults than to children. Conversely, adults are at lower risk for non-cancerous health effects from As exposure than children [86]. Recently, Kumar et al. conducted a study on a floodplain village situated near the Ganga Gyaspur Mahaji River in the Patna district. Their findings suggest that the local As concentration was as high as 826.4 µg/L. Furthermore, 86% of the samples collected were found to be highly contaminated (>50 µg/L). Residents of the village exhibit obvious symptoms of arsenic poisoning, and many suffer from skin problems (hyperkeratosis of the palms and soles, melanosis), gastrointestinal problems (gastritis, flatulence, and constipation), anemia, loss of appetite, respiratory distress, mental retardation, and other related illnesses, and they are at high risk for cancer [87].

3.1.2. Probabilistic Risk Assessment

Traditional deterministic health risk assessment methods are subject to variability and uncertainty in contaminant concentrations, exposure parameters, and environmental media. These factors can potentially overestimate the risks posed by toxic pollutants [88,89]. USEPA (2001) proposed Monte Carlo simulation (MCS) for uncertainty analysis; MCS minimizes uncertainty by calculating the probability that a risk will exceed safety standards using repeated samples from a probability distribution and identifying key factors that should be prioritized in risk management [90]. Some studies identify sensitive model parameters based on analyses of correlation and variance properties while taking into account different exposure scenarios related to groundwater pathways. Using the MCS methodology to incorporate changes in the key parameters for health risk assessment will thereby greatly reduce the inherent uncertainty due to variability in environmental systems

and exposure patterns [91–93]. A health risk assessment conducted in the northwest of Iran indicated that the primary pathway of adult exposure to As was through the consumption of local groundwater, with *ED*, *C*, and *AT* being the most influential factors in affecting the probabilistic health risk assessment [94]. Zhu et al. used stochastic-triangular fuzzy numbers (a combination of MCS and triangular fuzzy numbers) to evaluate the probabilistic health risk of As contamination in groundwater [95]. Chen et al. synthesized the overall state of environmental pollution with demographic variability, significantly reducing the inaccuracies of the assessment outcomes, and demonstrated that the *CR* for adults was lessened compared to the deterministic risk assessment model that uses fixed parameters. The assessment results indicated that for the population aged between 3 and 79 years, except for the age group of 4–6 years, the CR_{total} far exceeded the safe limit, reaching 10^{-4} to 10^{-3} , and the non-carcinogenic risk for the population aged 6 months to 79 years was above the acceptable threshold [88].

Only the bioavailable portion of the pollutant can be absorbed by the body [96]. The bioaccessible concentration is the concentration of a contaminant that is capable of being dissolved in the gastrointestinal system and then absorbed into the bloodstream. It is usually less than the total concentration of the contaminant that significantly influences the assessment of the health risk of human exposure to the toxic element [97]. Health risk assessment based on bioavailability concentrations provides a reliable method to avoid risk overestimation and unnecessary soil remediation [97]. For the health risk assessment of As in groundwater, the bioaccessible concentration can be considered as a tool to determine the actual human risk of different forms of As in groundwater.

3.2. Potential Exposure Assessment

As enrichment in groundwater typically results from the synergistic effects of multiple environmental factors. Because of the variable hydrogeochemical conditions and complex contamination sources, it is not possible to fully constrain the fate of high-As groundwater due to limited groundwater samples, and the distribution and sources of As in groundwater are difficult to predict, posing a serious challenge to its control and risk management [98]. To investigate compositional patterns in high-As groundwater samples, statistical analysis methods such as Self-Organizing Maps (SOMs) and Principal Component Analysis (PCA) have been employed. By reducing dimensionality and classifying data, these methodologies facilitate an accurate interpretation of the relationship between dependent and predictor variables. They assist in understanding the complexity of groundwater systems and the factors contributing to arsenic contamination.

The SOM analysis is an unsupervised learning algorithm that effectively reduces computation through dimensionality reduction. It describes the similarity of datasets by dividing similar data [99]. This method has good prospects for application in groundwater quality evaluation. Jiang et al. combined SOMs and health risk assessment to demonstrate that the carcinogenic risk of As in groundwater exceeds unacceptable risk levels for both children and adults [100].

PCA is also a statistical approach that simplifies the structure of data by reducing the dimensionality of the data and simplifies more variables into a few unrelated comprehensive indicators. PCA is frequently utilized for comparing compositional patterns among water samples and determining the factors that affect the compositional patterns of each sample [101]. Hierarchical Cluster Analysis (HCA) and PCA were used in the Hetao Basin of Inner Mongolia. The PCA results indicate that geological, reductive, and oxidative factors control high concentrations of As in groundwater [102]. A study proposes that the microbial-mediated decay of organic matter and reduction-based dissolution of Fe-Mn hydroxyl oxides are the primary mechanisms responsible for the high levels of arsenic in a shallow aquifer in eastern Bangladesh through correlation matrix, PCA, and saturation index calculations [103].

Advanced predictive models, including machine learning algorithms, have been developed to identify areas at high risk of As contamination in groundwater. Machine learning models, such as Logistic Regression, Artificial Neural Network (ANN), and Random Forest

(RF), are increasingly being deployed for predictive mapping of the distribution of high-As groundwater at global, national, watershed, and regional scales [104]. These models are capable of processing large datasets, learning from data, and identifying patterns that may not be immediately apparent to human analysts.

The distribution of As in groundwater exhibits significant heterogeneity and is frequently modeled with binary target variables to create probabilities, including logistic regression. Wu et al. used a logistic regression-based distribution model for As in groundwater and also combined modeled As hazards with simple exposure pathways and dose-response modeling to estimate the health hazards of As in Gujarat's groundwater drinking water. The study findings indicated an estimated prevalence of 700 cases of skin cancer and approximately 10 cases of premature avoidable mortality per year from internal cancers (including lung, liver, and bladder) [105]. Around 44.1 million individuals in the United States utilize well water in their households. A logistic regression model based on As concentrations in 20,450 domestic wells estimated that approximately 2.1 million people use drinking water with As levels greater than 10 µg/L [106].

ANN is an adaptive system that utilizes interconnected nodes or neurons in a hierarchical structure, similar to the human brain, to learn from data. It enables the modeling of intricate connections between variables. Global ANN models have been developed to link As levels to environmental indicators. These models aim to identify areas of high-As groundwater worldwide and to recognize significant indicators through a combination of the classification regression tree method and the ranked importance method. The models also aim to identify and quantify interactions between critical indicators using the neural interaction detection algorithm framework [107].

In recent years, a machine learning method has gained rapid development—RF. It is a classification model that creates a group of decision trees. This can be utilized to forecast a binary class based on the related independent variables [108]. This method can aid in identifying and comprehending the intricate relationships between various factors, contributing to the assessment of potential risks to human health and the environment from high-As groundwater. Joel Podgorski and Michael Berg utilized an RF machine learning algorithm to construct a worldwide predictive map for groundwater As amounts surpassing 10 µg/L, based on 11 geospatial environmental parameters and over 50,000 aggregated data points in terms of measured groundwater As concentrations. It is estimated that approximately 94–220 million individuals could be exposed to groundwater containing high levels of As, with the majority of these individuals (94%) in Asia [109]. Connolly et al. implemented Random Forest modeling based on high-resolution, satellite remote sensing, long-term measurements of surface inundation duration and frequency to effectively predict nonhomogeneous groundwater As concentrations at fine spatial scales in Vietnam, Cambodia, and Bangladesh [110]. Sumdang et al. used three distinct machine learning algorithms (ANN, Support Vector Machine, and RF) to assess the risk of As contamination in the urbanized coastal aquifer within the Rayong Groundwater Basin in Thailand. Upon comparative analysis of the three machine learning algorithms, the RF approach demonstrated superior predictive accuracy and exhibited the least uncertainty in modeling, rendering it the most appropriate method for evaluating As contamination in the groundwater of the study area [111].

Probability maps generated using machine learning predictions can serve as a guide for targeted As testing in groundwater, visually representing areas with higher probabilities of As contamination as analyzed through these statistical and machine learning methods. While we acknowledge the robust capabilities of machine learning in predicting and assessing the risk of high-As groundwater, the heterogeneity of aquifers at a small scale (<1 km) cannot be adequately modeled using existing global datasets. Accurate risk assessment also necessitates the further development and refinement of these models [109].

4. In Situ Remediation and Regulation Technology of Arsenic

At present, a variety of ex situ remediation techniques have been widely used for As removal around the world [55]. However, many of these methods face financial limitations,

the need to treat sludge/waste containing As, high technical difficulties, and secondary pollution when applied under on-site conditions. Moreover, the selection of remediation technology for a specific region depends on various factors such as the pH of the groundwater, oxidation state of As, and redox potential [112]. Technical, economic, and social factors should be comprehensively considered. To provide safe drinking water in areas of high-As groundwater contamination while ensuring the immediacy and effectiveness of remediation, it is necessary to implement rapid in situ detection, in situ remediation, and regional aquifer regulation in the field.

4.1. In Situ Rapid Detection Technology

In situ remediation of high-As groundwater requires close monitoring and testing to provide timely and accurate data that are critical to the overall remediation process. These data help to determine remediation progress, identify issues, and mitigate potential environmental and health risks to ensure remediation effectiveness. Water quality monitoring is essential for maintaining sustainable groundwater quality, and adequate water quality monitoring data can provide a basis for policy development. It is recommended that a regular groundwater monitoring network be established to track changes in As levels and groundwater quality [113]. Further development is required for monitoring strategies and methods to effectively estimate and predict As contamination in groundwater. At present, there are a variety of methods that can be used to detect the presence of As in groundwater, either through laboratory analyses or field sampling. The analytical methods typically used in laboratories to detect As are hydrogenated atomic fluorescence spectrometry (HG-AFS), hydrogenated atomic absorption spectrometry (HG-AAS), inductively coupled plasma atomic emission spectrometry (ICP), neutron activation analysis (NAA), and X fluorescence spectrometry (XRF) [114,115]. All of these methods rely on bulky equipment, are not suitable for detecting As in the field or at a distance, and are complicated and expensive to maintain. Various laboratory techniques for As detection are summarized in Table 2.

Table 2. Summary of analytical methods for As testing (data source [11,116]).

Methods	Detection Limit (ppb)	Sample Size (mL)	Remarks	Advantage	Disadvantage
HG-AAS	0.05	50	Single element	High sensitivity, capable of distinguishing between As(III) and As(V), reduces interference from the sample matrix.	Transition metals may interfere with detection and inhibitors like L-cysteine may be required to prevent interference.
GF-AAS	1–5	1–2	Single element	Suitable for the analysis of non-volatile compounds and offers high reliability.	Preconcentration is needed and, in some cases, matrix modification is necessary to increase detection sensitivity.
ICP-AES	35–50	10–20	Multi-element	More accurate for multi-element samples.	Less frequently used, not as sensitive as ICP-MS.
ICP-MS	0.02–1	10–20	Multi-element	Combines powerful separation capability with a low detection limit. Capable of effectively achieving selective detection.	Sensitive to high chloride levels, which may cause interference.
HG-AFS	0.01	40–50	Single element	High sensitivity, capable of eliminating matrix scattering and interference from the sample matrix.	Potential interference issues similar to HG-AAS, may require specific mechanisms to eliminate interference.

The most important feature of the on-site detection method is that it can be tested and sampled on the field. The principle behind the use of kits for rapid on-site detection of As in groundwater is that when any metal arsenide reacts with a strong acid, arsenide gas is produced. Most As detection kits depend on the use of zinc metal and hydrochloric acid to convert inorganic arsenic into arsenical gas (AsH_3). The gas is allowed to pass through the indicator paper of mercury bromide (HgBr_2), and the intensity of the color represents the concentration of As [7]. In an evaluation of the performance of eight commercially available arsenic field test kits, a variance in accuracy and precision was identified. The LaMotte and Quick II kits were found to provide accurate and precise estimates of arsenic concentrations, whereas kits like Econo-Quick, Quick, Wagtech, and Merck were either accurate or precise but not both. The Hach and Econo-Quick II kits were neither accurate nor precise. Furthermore, the test strips from the underperforming kits often produced colors that were lighter than those on the arsenic color calibration charts provided by the manufacturers, suggesting a need for improved quality control of these kits [117]. There are many other on-site kits, including the AAN kit (Japan), AIIHPH kit (India), E-Merck kit (Germany), ENPHO kit (Nepal), NIPSOM kit (Bangladesh), Wagtech Arsenator (UK), and Modified AAN kit (Nepal) [116].

Some electrochemical techniques can also be applied in field conditions, primarily for the detection of As speciation in groundwater. A method for the detection of inorganic arsenic (i.e., the transformation of As(III) to As(V)) at a 25 μm diameter gold microwire electrode using anodic stripping voltammetry (ASV) under the conditions of low-concentration KMnO_4 has been developed. This method can be utilized over a wide pH range, does not require chemical reduction, offers good sensor repeatability, is easy to operate, and is suitable for the rapid and accurate measurement of total arsenic concentration in groundwater [118]. The determination of As(III) in water at a natural pH can also be conducted using cathodic stripping voltammetry (CSV) without the need for reagent addition or O_2 removal, making it applicable for the long-term monitoring of As(III) in well water or streams [119].

4.2. In Situ Remediation

With the development of various technologies for example chemical oxidation, adsorption, ion exchange, coagulation and precipitation, membrane technology, bioremediation, and microbial remediation, ex situ treatment methods have been widely studied [120,121]. Figure 4 illustrates the general flow of the As removal process. However, many technologies have been successful in the laboratory and their applicability at the field scale has been hampered by issues of technical feasibility, managing sludge, secondary contamination, and financial constraints in large-scale applications [8,112].

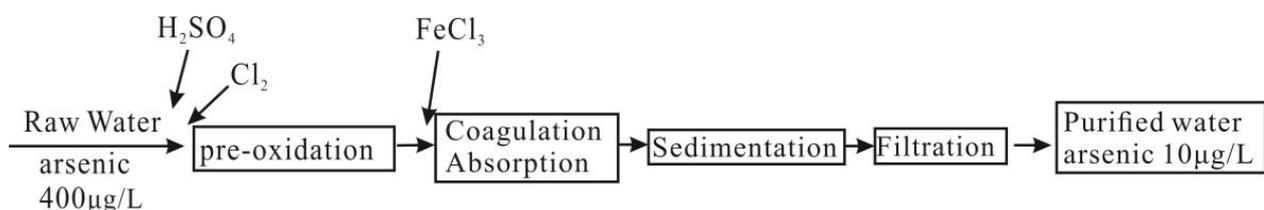


Figure 4. General schematic diagram of arsenic removal treatment process (after [112]).

Most in situ treatment methods work based on oxidation, flocculation, sedimentation, filtration, and adsorption. For example, flocculant–disinfectants, bucket treatment units, use of activated alumina, use of nano zero-valent iron, and aquifer iron coating techniques are As removal technologies that can be adopted by households or communities. Bioremediation is another proven method for achieving in situ remediation.

Flocculants–Disinfectants: When the flocculant–disinfectant powder is added to water, it employs precipitation, coagulation, and flocculation processes to remove As, other

heavy metals, organic material, and microorganisms from groundwater. Following the treatment, a residual amount of free chlorine is left in the water, which aids in further disinfection [122]. A 12-week study of 103 As-contaminated tube-well water households in Bangladesh showed that the median As concentration in tube-well water decreased from a baseline of 136 µg/L to 16 µg/L with the use of a flocculant–disinfectant, significantly reducing As in drinking water [123]. This is a technique for replicating municipal water treatment processes at the household level, providing options for rural residents at risk of waterborne diseases [124].

Bucket Treatment Unit (BTU): The BTU is a portable As removal unit based on the principles of adsorption, precipitation, and coagulation. This method uses two buckets with a capacity of 20 L, one of which is placed above the other, and can remove arsenic in nearly 2–3 h. The technology is low-cost, simple to install, and uses only basic chemicals like aluminum sulfate and potassium permanganate [60]. The DPHE-Danida project in Bangladesh distributed several thousands of BTU units in rural areas of Bangladesh [125]. It has good As removal performance under both field and laboratory conditions, but due to alterations in the parameters of water quality (especially pH) and inadequate mixing, the effect of rapid evaluation may be poor [126].

Aquifer Fe-coating technology: An in situ remediation study in the Datong Basin, China, showed a significant enhancement in arsenic removal efficiency through an in situ remediation approach. The method involved an alternating injection of Fe(II) (5.0 mM) and NaClO (2.6 mM) to induce the formation of Fe oxides/hydroxides within the aquifer. This process encompassed the oxidation of Fe(II) and As(III) using NaClO, as well as the adsorption/co-precipitation of As(V) in the targeted aquifer [127].

Activated Alumina (AA): The adsorption effect during As removal is related to the nature of the adsorbent material, including the density of the effective adsorption sites and the size of the specific surface area [114]. AA has been widely used in the adsorption of As because of its porosity, high dispersibility, and large specific surface area [128]. AA consisting of amorphous and γ -alumina oxides was obtained by a dehydration reaction under high-temperature conditions using $\text{Al}(\text{OH})_3$ as a raw material. AA adsorption is a physical/chemical process wherein the available adsorption sites on the oxide surface are utilized to remove ions from solution. The actual degree of As removal and the service life of the AA media depend on various factors, including pH, the oxidation state of arsenic, competing ions, the empty bed contact time (EBCT), and the regeneration process [129]. AA is more effective in removing As under acidic conditions, particularly for As(V). Ions such as sulfate and chloride may interfere with the removal of As; increasing EBCT can slightly enhance the effectiveness, while the life span and efficacy of the activated alumina beds may decline after regeneration with high-concentration NaOH solutions [129]. The Battelle Memorial Institute received funding from the USEPA, studied two AA plants employed in the treatment of As and showed that raw water As(III) concentrations ranged from 0.3 to 28.8 µg/L, and the finished water contained less-than-detectable levels of As(III), indicating that As(III) was almost completely removed [130]. Field trials of electrocoagulation followed by adsorption on AA in West Bengal, India, have shown that this technology can decrease As concentrations in drinking water to below 10 µg/L [131]. However, if groundwater contains high concentrations of iron, the precipitation of $\text{Fe}(\text{OH})_3$ in the presence of air may occur, potentially leading to the clogging of filtration columns and thus reducing the adsorption efficiency of AA [128]. When applied in field applications, it is essential to consider the multitude of factors influencing the removal of As by AA to ensure optimal performance.

Nano Zero-Valent Iron (NZVI): In field applications, researchers have employed NZVI as an adsorbent due to its core–shell structure—consisting of a highly reductive metallic core with a surrounding amorphous iron (oxy)hydroxide layer. Such a structure enables NZVI to simultaneously undergo reduction and oxidation mechanisms, facilitating the coordination and oxidation of As(III) [132]. As(III) is strongly adsorbed onto NZVI within a short timeframe (on the scale of minutes), and this effective adsorption occurs across a

variety of pH levels and anionic environments. The actual reactive site on the NZVI surface is likely a stable or metastable Fe(II), Fe(II)/(III) mixture, or an Fe(III) oxide, hydroxide, or oxyhydroxide corrosion product [133,134]. The efficacy of As(III) adsorption sites evolves over time; initially (within 0–24 h), amorphous Fe(II)/(III) species and magnetite (or maghemite) function as adsorption sites. As NZVI corrodes over time, the formation of more crystalline magnetite and lepidocrocite provides ongoing sites for As(III) adsorption. Moreover, As(III) in contact with corroded NZVI surfaces is oxidized to As(V), which is then adsorbed by Fe(III) oxides. Consequently, As(III) and As(V) get sequestered in layers formed atop NZVI corrosion product films, which are overlain by subsequent layers, becoming isolated from the surrounding solution [135]. NZVI is a highly efficient material for removing As(III), with the main advantages being that it is (a) non-toxic and cost-effective and (b) it is a strong reductant; hence, it is capable of effectively removing both inorganic and organic arsenic, and it is effective in removing arsenic in conditions of low pH and high sulfide content in water [136]. Neumann et al. investigated As removal in the field, employing SONO household filters based on zero-valent iron [137]. The removal process with NZVI involves several mechanisms (Figure 5). However, the application of this nanoadsorbent is complex in terms of material synthesis, with the potential interference in the adsorption of As(III) from competing anions such as HCO_3^- , H_4SiO_4^0 , and $\text{H}_2\text{PO}_4^{2-}$ [135,138].

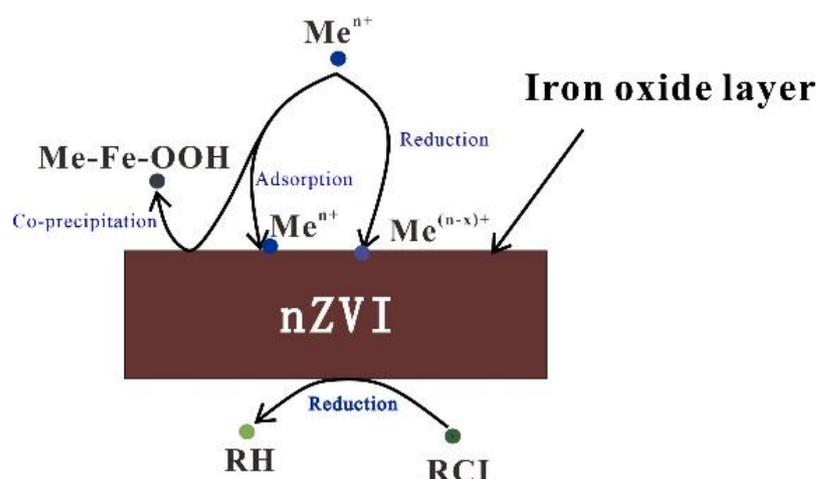


Figure 5. Schematic model of the removal mechanisms of NZVI (modified after [139]).

Bioremediation: Bioremediation mainly comprises microbial in situ remediation and phytoremediation. Microbial communities undertake the bioremediation of various forms of As through oxidation, reduction, methylation, and biosorption. Phosphate transporter proteins Pst and Pit mediate As(V) uptake, while glycerol transporter GlpF uptakes As(III) [140]. The detailed process of action is shown in Figure 6. Some microorganisms, for example, *Gallionella ferruginea* and *Leptothrix ochracea*, accelerate the biological oxidation of iron and provide a favorable environment for As adsorption [141]. The pathway of As(V) methylation initially involves the reduction of As(V) to As(III), followed by the methylation of As(III) to dimethylarsine via the coenzyme S-adenosylmethanethionine [142,143]. Multiple As transformation mechanisms exist in bacteria and other algae. For example, *Leptolyngbya boryana* has strong As tolerance, biotransformation, and accumulation capabilities, and it has multiple As biotransformation pathways that have attracted considerable attention as a promising alternative for As remediation [144]. Phytoremediation remediates heavy metal-contaminated water through several processes including phytoextraction, phytodegradation, inter-root filtration, phytostabilization, and phytovolatilization [121]. Researchers investigated the accumulation and removal of As from As-contaminated site soils by the As-hyperaccumulating plant, Chinese brake fern (*Pteris vittata* L.), in a two-year field experiment [145]. It has been reported in the literature that a method of the

PvACR3 transgene has been designed for the phytoremediation of engineered As-tolerant and -hyperaccumulating plants [146]. Bioremediation approaches exhibit commendable environmental compatibility and potential cost-effectiveness; however, extensive research is still requisite.

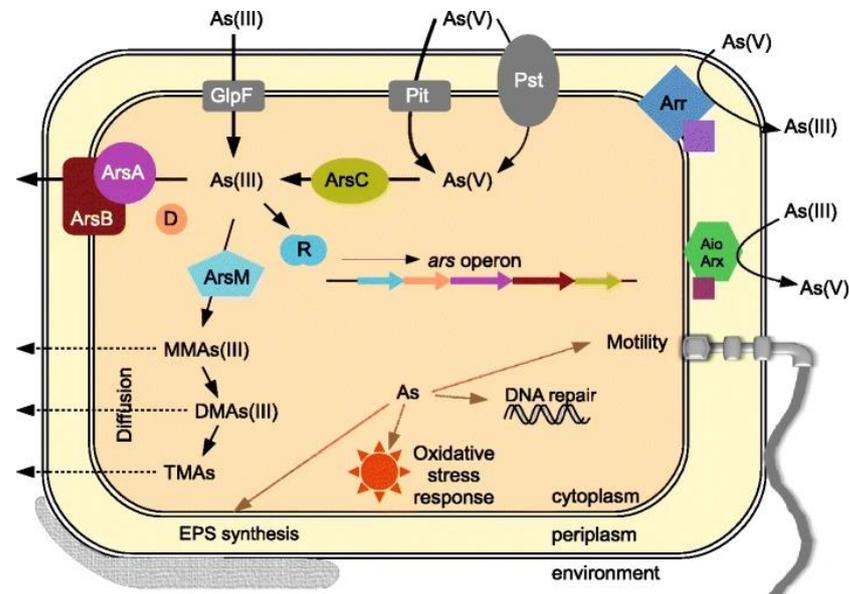


Figure 6. Overview of bacterial interactions with As (after [147]).

4.3. Regional Aquifer Arsenic Regulation

The sustainable management and use of groundwater are extremely crucial for safe drinking water [46]. The most direct approach to the problem of high-As groundwater is to eliminate the source of contamination. Various remediation techniques above are widely used. Furthermore, the use of centralized and decentralized drinking water treatment technologies can significantly reduce the exposure of As in drinking water.

Instead of expensively treating the reclaimed water or abandoning the existing managed aquifer recharge (MAR) sites, one management strategy involves pretreatment of the recharge water. This includes adjusting pH, controlling calcium concentration, adding chemical oxidants, and incorporating amendments for in situ treatment to minimize adverse geochemical interactions between recharge water and native groundwater. Subsequently, to prevent the initial release of geogenic As, oxidants (dissolved oxygen and/or nitrate) in the injection water can be eliminated using membrane contactors and/or the addition of sodium bisulfite [148]. In addition, the mobilization of As to the interface where mixing transpires between recharge water and native groundwater, known as the buffer zone, and trying to impede the restoration of compromised groundwater by restricting the pumping activities within the buffer zone can be conducted [148].

Effective aquifer management can also lead to a decrease in concentrations of As in groundwater (Figure 7). The majority of wells exhibiting arsenic trends are situated within regions characterized by dense networks of wells. Intensive pumping in these regions leads to an elevated vertical displacement of groundwater. The oxidation of groundwater moving downward may lead to a decrease in As. However, deeper or situated in the trough, the reduction conditions of the aquifer are conducive to the migration of As, which may further deteriorate the groundwater quality in the long term [65].

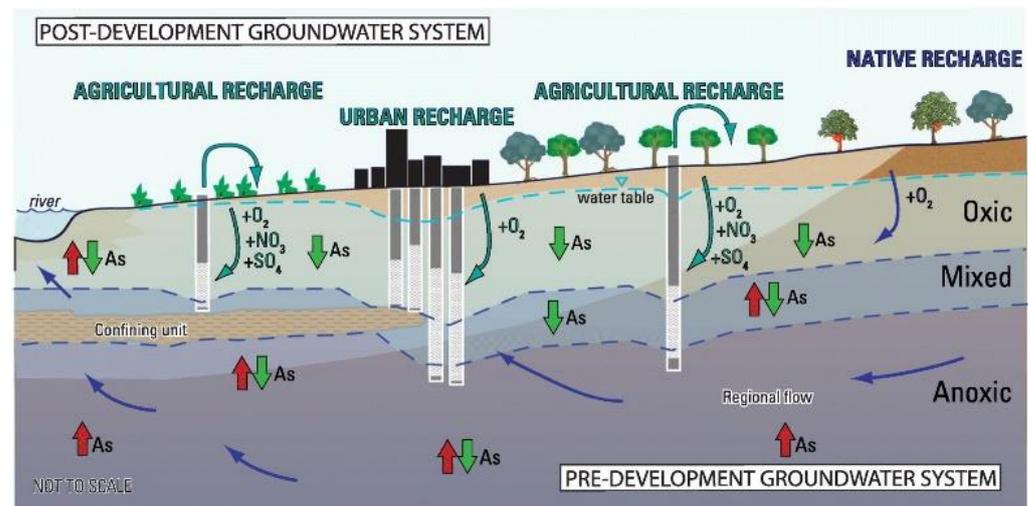


Figure 7. Effects of anthropogenically modified flow systems on arsenic (after [65]).

In addition, alternative water sources can be selected to reduce the consumption of high-As groundwater, which requires the identification and utilization of clean water sources, for example, collecting rainwater, treated surface water, and the combined use of groundwater and surface water as domestic water, or As pipe wells that take water from deep low-As aquifers. Compared with water-rich countries, rainwater harvesting is generally seen as a backup source of supply [149]. Rainwater with relatively good water quality after filtration is a viable way to ensure a safe water supply [150]. Combining low-As water with high-As water may be an option to reduce As in groundwater to acceptable levels [36]. In some areas, shallow pipe wells are generally contaminated, and deep pipe wells are characterized by low As; deep tube wells may provide higher quality water and lower overall burden. Despite ultimately having problems with microbial quality, it is effective in reducing As exposure [151,152].

5. Development Tendency

The sources, distribution, and behavior of As in groundwater under geological and hydrogeological conditions and treatment methods have been extensively studied worldwide. To mitigate the health and environmental risks caused by geogenic high-As groundwater and ensure sustainable water resources management, further research and optimization are needed in the following areas [10,55,114,153,154].

1. Although there are more remediation technologies for high-As groundwater contamination, all of them have certain limitations, which is a relatively complex process. At present, no remedial technique can attain complete remediation, and a single method is inevitably constrained by factors, for example, the environmental conditions of the groundwater and the economic status of the area. The development of biological–plant combination techniques, chemical/physical–chemical–biological combined combination techniques, and physical–chemical combination remediation techniques is a new direction in the future.
2. The main challenges in managing water with high levels of As include applying in situ remediation technologies, operating and maintaining large water treatment plants, and managing As-containing sludge and waste. The management of high-As groundwater involves several fields, including technology, engineering, finance, and environmental management, and it requires an integrated approach. As-containing sludge or waste generated during groundwater treatment must be properly handled to prevent secondary contamination and environmental hazards.
3. Based on an extensive review of the literature, it has been found that in many countries, natural groundwater contains arsenic coexisting to varying degrees with additional

contaminants, such as fluoride (F), nickel (Ni), molybdenum (Mo), and antimony (Sb), among others. There is a potential for interaction occurring among these elements. There is a lack of studies on the simultaneous presence of As with other emerging contaminants in groundwater. Further laboratory simulations are necessary to enhance the comprehension of how environmental, hydrological, geological, and anthropogenic factors impact the mechanisms of arsenic coexistence with other contaminants. In addition, more research is needed to explore the migration of As in groundwater and its interaction with other organic and inorganic components during the transformation process. An in-depth study of As migration interaction with other elements, organic chemical transformations, and biochemical processes can provide better prediction and simulation tools, which can help to develop more effective strategies for the management of high-As groundwater pollution.

6. Conclusions

This paper highlights the distribution of geogenic high-As groundwater worldwide, health risks, and management techniques and discusses future trends. There are 107 countries affected by geogenic high-As groundwater pollution, mainly in Asia, Europe, and Africa. The countries suffering the most severe effects include India, Bangladesh, China, Nepal, and Pakistan.

The health risk assessment model can link high-As groundwater with the health hazards of the public and quantitatively describe the harm of high-As groundwater to human health. The main exposure pathways for As in groundwater are the ingestion of drinking water and skin contact. Risk assessment results indicate that As in groundwater poses a serious threat with high carcinogenic and non-carcinogenic effects. In addition, modeling methods for predicting As pollution using hydrogeological and field measurements have been utilized to assess and measure potential areas of As contamination. In particular, machine learning models can be used at global, national, watershed, and regional scales to provide essential data for improving public health and water management.

Various in situ remediation technologies for groundwater have received great attention, usually involving three aspects: in situ monitoring and detection, in situ remediation, and regional aquifer regulation. In situ remediation technologies include but are not limited to redox, adsorption, biosorption, and other technologies, such as flocculant–disinfectant, barrel treatment unit, use of activated alumina, use of nano zero-valent iron, and aquifer iron coating technology. The successful application of these remediation materials and technologies in the field will help reduce the treatment cost of high-As groundwater and reduce environmental impacts.

Further studies are necessary to uncover the mechanisms behind the coexistence of As with other contaminants in groundwater and to determine the correct disposal of waste containing As produced during the treatment process. Additionally, better treatment options must be explored. The worldwide issue of high levels of As in groundwater necessitates cross-border collaboration and integrated solutions to protect both public health and environmental sustainability.

Author Contributions: Writing—original draft; J.G.; conceptualization; W.C. and G.L.; methodology; J.G., W.C. and Q.S.; software; formal analysis; T.N.; review and editing; W.C., G.L., Q.S., Y.R. and Z.L.; visualization; supervision; J.G., T.N. and X.L. All authors have read and agreed to the published version of the manuscript.

Funding: This research was financially supported by Hebei Natural Science Fund for Distinguished Young Scholars (D2023504030), National Key R&D Program of China (2022YFC3703701), and the National Natural Science Foundation of China (41972262).

Data Availability Statement: The data will be provided on request to the corresponding author.

Acknowledgments: The authors would like to appreciate the reviewers and editors for their helpful comments and suggestions.

Conflicts of Interest: The authors declare no conflict of interest.

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