Review

Arsenic in Water: Understanding the Chemistry, Health Implications, Quantification and Removal Strategies

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Abstract: Arsenic, the 20th most common element in Earth's crust and historically regarded as the King of Poisons, occurs naturally in two oxidation states, Arsenate (V) and Arsenite (III), and is prevalent worldwide through natural and anthropogenic means. The cations of the metalloid exhibit unique chemical behaviour in water and are found to be components of approximately 245 natural minerals, making its occurrence in drinking water a compelling challenge, especially in groundwater. This comprehensive review collates information regarding the prevalence of arsenic contamination in water worldwide and its impact on human health, its chemical behaviour, methods for detection and quantification, and treatment strategies. A comprehensive search was conducted, and the selection of eligible studies was carried out using the PRISMA (the preferred reporting items for systematic reviews and meta-analyses) guidelines. Essential characteristics of eligible research studies were extracted based on geographical areas, origins, concentration levels and the magnitude of populations vulnerable to arsenic contamination in groundwater sources. Arsenic contamination of water affects over 100 countries including Canada, the United States, Pakistan, China, India, Brazil and Bangladesh, where hydrogeological conditions favour prevalence and groundwater is the primary water source for food preparation, irrigation of food crops and drinking water. This leads to human exposure through absorption, ingestion and inhalation, causing numerous health disorders affecting nearly all systems within the human body, with acute and chronic toxicity including cancers. The presence of arsenic in water poses a considerable challenge to humanity, prompting scientists to devise diverse mitigation approaches categorized as (a) oxidation processes, (b) precipitation methods, (c) membrane technologies, (d) adsorption and ion exchange methods, and (e) social interventions. This comprehensive review is expected to be a valuable source for professionals in the water industry, public management, and policymaking, aiding their ongoing and future research and development efforts.

Keywords: arsenic; drinking water; epidemiology; public health; detection; quantification; mitigation

1. Introduction

Arsenic (As) occurs naturally, ranking as the 20th most abundant non-essential trace element in the Earth's crust, where it is a component of 245 minerals. It is the 12th most abundant element in the human body and the 14th most prevalent in seawater [1]. With an atomic number of 33 and an atomic mass of 74.9216 g/mol, arsenic exhibits physical and chemical properties that lie between those of metals and non-metals, categorizing it as a metalloid [2]. Arsenic exists in three allotropic forms: metallic grey, which has a density of 5.73 g/cm^3 and is the most commonly occurring; yellow, with a density of 2.03 g/cm^3; and black, with a density of 5.72 g/cm^3 [1]. Since its discovery by Albertus Magnus

in 1250 A.D., arsenic has been a significant and often controversial element in human history. It holds potential applications in a wide range of fields, including agriculture, chemical engineering, metallurgy, pharmaceuticals, livestock management, and electronics. In the environment, arsenic is found in both organic and inorganic forms [3]. It exists in four oxidation states: arsine (As(-III)), arsenite (As(+III)), elemental arsenic (As(0)), and arsenate (As(+V)). Among these, the more dominant forms of inorganic arsenic (iAs) in the environment are arsenate and arsenite. Organic arsenic (oAs) compounds, such as dimethylarsinous acid (DMA) and monomethylarsonic acid (MMA), can dissolve in water, which increases their relative abundance [4].

Arsenic is classified in group 15/VA of the periodic table, also known as the nitrogen group, which includes essential elements like nitrogen and phosphorus. While it is not an essential nutrient, living organisms tend to accumulate arsenic. This element has profoundly impacted human lives, particularly due to its presence in drinking water, which poses a significant health challenge [3]. The impact of arsenic contamination on public health is severe and multifaceted. Chronic exposure to arsenic, primarily through drinking contaminated water, can lead to a plethora of adverse health effects. These include skin lesions, cardiovascular diseases, neurotoxicity, diabetes and various forms of cancer, particularly skin, lung and bladder cancers. Arsenic's ability to interfere with cellular and genetic mechanisms underlies its toxicity. It can induce oxidative stress, disrupt enzyme function and alter DNA repair processes, leading to carcinogenesis and other serious health conditions [5].

In many parts of the world, particularly in regions such as South Asia, arsenic contamination of groundwater is a critical issue. Millions of people are at risk, and the problem is compounded by the lack of alternative safe water sources and inadequate public health infrastructure. The ingestion of arsenic-contaminated water and food crops irrigated with such water leads to bioaccumulation, posing long-term health risks to entire communities [6,7]. The current review aims to collate comprehensive information and evidence regarding the global prevalence of arsenic contamination. It will examine the chemical behaviour of arsenic in water, its health implications, methods for its detection and quantification, and various mitigation strategies. A comprehensive search was conducted from inception to February 2022. The selection of eligible studies was carried out using the PRISMA (the preferred reporting items for systematic reviews and meta-analyses) guidelines. This review seeks to provide a detailed understanding of arsenic's environmental impact and offer insights into effective solutions for managing its presence in drinking water and other critical contexts. By addressing the multifaceted challenges posed by arsenic contamination, this review aims to contribute to global efforts to mitigate its impact on public health and ensure safe water access for affected populations.

2. Prevalence of Arsenic Contamination

In 2011, As exposure was estimated to affect 150 million people worldwide [8]. A report from 2007 indicated that approximately 137 million individuals were exposed to arsenic through contaminated drinking water in 70 countries [9]. However, as of 2021, global information revealed that arsenic contamination in groundwater had impacted 107 countries, affecting a total population of 230 million [10]. The presence of arsenic beyond its safe limit (0.01 mg per liter stated by the World Health Organization (WHO)), especially in groundwater, is the focus of global attention, particularly in countries like China, Pakistan, Bangladesh, the United States, Canada, Brazil, India, Nepal and Thailand (totalling 49.84% of the world's population), where groundwater is the primary source of drinking water and is used for irrigation of food crops and food preparation [11,12]. As contamination in groundwater is commonly attributable to natural sources, whereas arsenic's environmental existence (in air, water and soil) comes from both anthropogenic and natural sources. Anthropogenic sources of arsenic include smelting, waste incineration, pesticide application, nonferrous metals mining, wood combustion and coal combustion. Many natural phenomena also contribute to environmental concentrations of arsenic, including

hydrothermal/geothermal activity, dust storms, forest fires, pedogenesis and volcanic eruptions [13]. Various biological and physicochemical reactions govern the dissolution of As in groundwater. Pathways responsible for As contamination in groundwater are the deposition of organic deposits in river basins and floodplains, erosion of mineral rocks in mountain ranges containing As and anoxic conditions in aquifers. The anoxic conditions develop in aquifers due to flat geology, a humid environment and long residence times of water in the aquifer, which cause adsorbed As to be released into the water and dissolved. Scientists have observed another pathway of mobilization of As in arid areas of the world, such as in central Asia, North America and Australia, where high pH conditions cause the reaction and dissolution of As in oxygen-rich groundwater. The risk assessment under different hydrogeological conditions of As contamination in groundwater has been illustrated at a global level (Figure 1) [14].
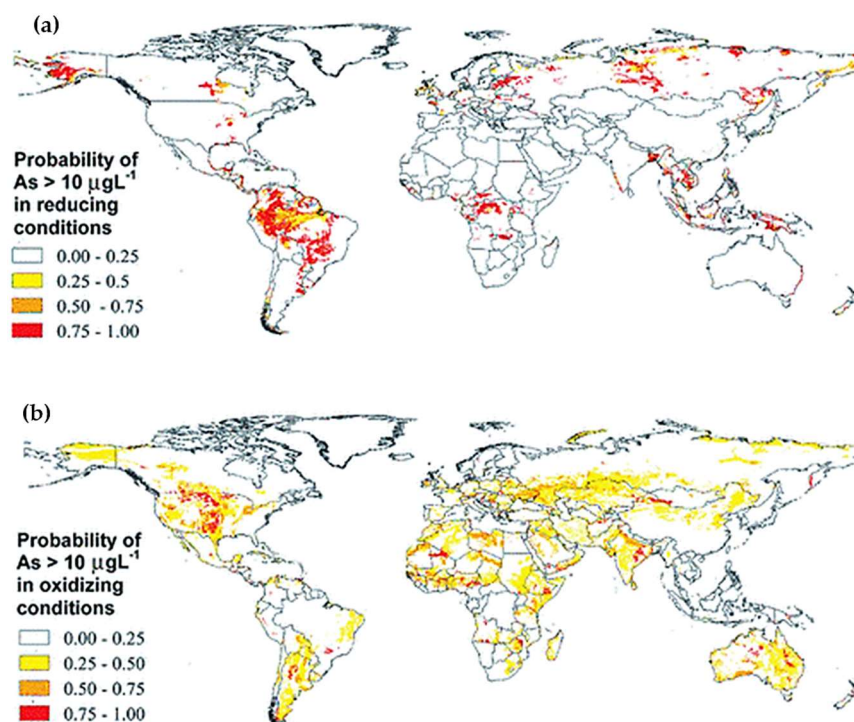

Figure 1. Chemical mapped risk assessment of arsenic in drinking water/global probability map of arsenic contamination under different hydrogeological conditions in groundwater, (**a**) reducing groundwater conditions, and (**b**) high pH/oxidizing conditions, adapted with permission from [14], publisher ACS Publications.

High concentrations of arsenic (As) in groundwater, higher than the permissible limits set by WHO (0.01 mg/L) [11,12], have been reported worldwide, rendering populations in these areas vulnerable to exposure to arsenic and its harmful effects. Most of the arsenic-contaminated zones are situated in sedimentary basins near deltaic areas and mountain belts [15]. Regions with a tropical climate are more susceptible to arsenic pollution, as this environment fosters the release of arsenic from mineral sources [16]. Table 1 summarizes the range of arsenic concentrations (μg/L) in groundwater across different parts of the world, the sources of contamination and the affected populations, providing an overview of the global prevalence of this issue. To construct this, a comprehensive search was conducted across multiple databases, including PubMed, Scopus and Google Scholar, from inception to February 2022, using keywords such as "*arsenic contamination*", "*arsenic in ground water*", "*arsenic prevalence*", "*arsenic in drinking water*". The preferred reporting items for systematic reviews and meta-analyses (PRISMA) guidelines [17] were followed for the search and selection of eligible studies. Essential characteristics of eligible research

studies were extracted using a previously published method [18–23], based on geographical areas, origins, concentration levels, and the magnitude of populations vulnerable to arsenic contamination in groundwater sources.

Table 1. Description of the geographical areas, origins, levels of concentration, and the magnitude of the population vulnerable to arsenic (As) contamination in global groundwater sources.

Country/Region	Districts/Provinces	Contamination Source	Concentration [As] μg/L	Population at Risk (Million)	References
Bangladesh	61 *	Flood plain, deltaic sediments, alluvial aquifers and organic matter	2–4730	85	[24–28]
Japan	3 *	[As] sulfide from industrial activities, holocene coastal sands, volcanic activity and quaternary alluvium aquifer	1–25,700	***	[29,30]
Taiwan	4 *	Fine-grained fluvial deposits, aquifers and aquitards and black shales	2.5–1820	0.1–0.2	[31–34]
Pakistan	27 *	Dissolution of sedimentary deposits and minerals, especially iron oxyhydroxides	3.0–2580	13	[33,35–38]
Cambodia	1 **	Ferrous sediments	1–1610	2.4	[39]
Australia	4 *	Gold mining, iron oxyhydroxides and iron hydro-oxides	5–300,000	0.001976	[10,40]
Eastern Croatia	1 *	Sediments	27	0.12	[41]
India	20 *	Alluvial, deltaic and igneous sediments	1–3880	50	[10,42]
Canada	5 **	Thermal springs, volcanic rocks, uranium mines and sediments	1.5–100,000	minimal	[11,43,44]
Ghana	5 *	Gold mining	<1–4500	<0.1	[45]
Thailand	1 *	Mining and dredged alluvium	1–5000	0.0015	[46–49]
Argentina	4 *	Volcanic ash and thermal springs	10–2000	2	[42,46]
Slovakia	2 *	Aquifer sediments	37–39	1.1 ****	[50,51]
Mexico	3 *	Volcanic sediments and mining	8–620	0.4	[11,46]
New Zealand	4 *	Volcanic activities	21–8500	minimal	[10]
Romania and Hungary	2 *	Sediments	0.5–240	0.4	[11,46,52]
Germany	1 *	Sandstone and alluvium sediments	10–150	minimal	[53,54]
Afghanistan	1 *	Sediments	10–500	0.5	[29,55]
Vietnam	2 *	Pleistocene and Holocene sediments	1–3050	0.5–10	[55,56]
Italy	3 *	Volcanic aquifers	0.1–6940	1 *****	[57]
UK	4 **	Mining, limestone, sandstone, estuarine alluvium, alluvial or glacial aquifers	<1–355	0.56	[58,59]
Greece	1 *	Mine tailings	10,000	***	[55,60]
Brazil	1 *	Gold mining	0.4–350	***	[29,55,61]
Spain	1 *	Alluvial sediments	<1–100	>0.05	[11,62,63]
Chile	1 *	Volcanogenic sediments, thermal springs and mining	100–1000	0.5	[64]

* Number of districts. ** Number of provinces. *** No data available. **** During 30 years. ***** District of Sicily only.

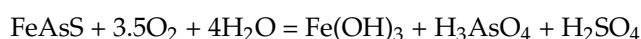## 3. Behaviour of Arsenic in Water

A potential reason for the global prevalence of arsenic (As) in water is its sensitivity to mobilization under conditions (oxidizing, reducing, and pH range of 6.5–8.5) most typically found in groundwater aquifers [65,66]. This is distinct from other heavy semi-metals

and oxo-anion generation elements (such as Selenium (Se), Antimony (Sb), Molybdenum (Mo), Vanadium (V), Chromium (Cr), Uranium (U) and Rhenium (Re)). Groundwater, about half of all drinking water worldwide, is particularly vulnerable in comparison with surface water due to contact with arsenical minerals in groundwater aquifers [67]. Estimates suggest that 99% of the As on earth is associated with mineral ores [68]. There are 245 minerals rich in arsenic compounds, out of which 20% are arsenides, 20% are sulfosalts and sulfides, and 60% are elemental As, arsenides, oxides, arsenites and silicates. Manganese nodules, iron deposits and sedimentary rocks are minerals considered rich in arsenic [1]. Table 2 details the concentration of arsenic in As-rich minerals, and the chemical formulation of the minerals along with the type of mineral ore is presented [69].

Table 2. Arsenic concentration ranges in various ores, mineral types and chemical formulas. Reproduced with permission from [69], publisher Natural Environment Research Council (NERC), UK.

Ores	Mineral Type	Chemical Formula	[As] Concentration Range (g/kg)
Marcasite	Sulfide minerals	FeS_2	0.02–276
Pyrite	Sulfide minerals	FeS_2	0.1–120
Maghemite	Oxide Minerals	$\gamma\text{-}Fe_2O_3$	Up to 186
Iron (III) oxyhydroxide	Oxide Minerals	$FeO(OH)$	Up to 76
Haematite	Oxide Minerals	Fe_2O_3	Up to 29
Sphalerite	Sulfide minerals	$(Zn,Fe)S$	0.005–17
Galena	Sulfide minerals	PbS	0.005–10
Chalcopyrite	Sulfide minerals	$CuFeS_2$	0.010–5
Jarosite	Sulphate minerals	$KFe_3(SO_4)_2(OH)_6$	0.034–1
Apatite	Phosphate minerals	$Ca_{10}(PO_4)_6(OH,F,Cl)_2$	<0.001–1
Pyrrhotite	Sulfide minerals	FeS	0.005–0.1
Halite	Halide Minerals	$NaCl$	<0.003–0.03
Fluorite	Halide Minerals	CaF_2	<0.02
Magnetite	Oxide Minerals	Fe_3O_4	0.027–0.041
Barite	Sulphate minerals	$BaSO_4$	<0.001–0.012
Dolomite	Carbonate minerals	$CaMg(CO_3)_2$	<0.003
Siderite	Carbonate minerals	$FeCO_3$	<0.003
Amphibole	Silicate minerals	$X_7Si_8O_{22}(OH)_2$	0.0011–0.0023
Feldspar	Silicate minerals	$KAlSi_3O_8\text{–}NaAlSi_3O_8\text{–}CaAl_2Si_2O_8$	<0.00001–0.0021
Biotite	Silicate minerals	$K_3AlSi_3O_{10\ 2}$	0.0014
Quartz	Silicate minerals	SiO_2	0.0004–0.0013
Calcite	Carbonate minerals	$CaCO_3$	0.001–0.008
Gypsum/anhydrite	Sulphate minerals	$CaCO_4$	<0.001–0.006
Pyroxene	Silicate minerals	$XYZrOu$	0.00005–0.0008

Oxidation of sulfide minerals is considered the primary source of release of As into water with NO_3^-, Fe^{3+} and O_2 acting as oxidizing agents [70]. The reaction rate of arsenopyrite with Fe^{3+} as an oxidizing agent is nearly ten times that of pyrite oxidation. For instance, the overall reaction of arsenopyrite oxidation with O_2 as the oxidizing agent can be expressed as follows: [71]

$$FeAsS + 3.5O_2 + 4H_2O = Fe(OH)_3 + H_3AsO_4 + H_2SO_4$$

All the constituents of such reactions are naturally occurring except NO_3^-, which is human induced using pesticides.

Arsenic may occur in the environment in different oxidation states, whereas in natural waters, inorganic pentavalent arsenate (As(V)) and inorganic oxyanions of trivalent arsenite (As(III)) are more commonly found [72]. Biological activities in surface waters may produce organic arsenic but are not quantitatively significant; however, their significance becomes evident in cases where waters are substantially influenced by anthropogenic activities [48]. Organic arsenicals such as lipids containing arsenic, arsenobetaine, trimethylarsine oxide, arsenosugars, tetramethylarsonium and arsenocholine are mainly found in marine life and can also become part of other non-marine organisms [73].

The distribution of arsenic (organic and inorganic) in drinking water is a function of the water's pH. Many of the toxic trace elements (Zn^{2+}, Ni^{2+}, Cu^{2+}, Pb^{2+}, Cd^{2+} and Co^{2+}) exist as cations, and their solubility in water increases with the increase in pH; however, the point of minimum solubility occurs at a different pH value for every metal [35]. Typically, in water, coprecipitation or precipitation (with phosphate mineral, oxide, carbonate or hydroxide) or adsorption (to metal oxide (hydrous), organic matter or clay) limits the solubility of mostly all the cationic trace elements [74]. However, the solubility of oxo-anions, including As(V), decreases with increasing pH [75]. Therefore, ions with a negative charge (oxyanion/anion) may remain chemically dissolved in solution with typical values in the tens of mg/L (high concentration) at a pH value of nearly 7 (neutral), and subsequently, arsenic and other elements with the same properties are abundant trace pollutants, especially in groundwaters, due to more favourable hydrogeological conditions [76,77].

In comparison with other anion-producing metals (such as Cr, U, and Se), arsenic is the most complex and problematic in an aqueous environment due to its wide mobility range in reduction–oxidation conditions [78]. In sulphatic waters and reducing aqueous conditions, most trace elements form insoluble sulfides, which reduce the risk of contamination by reducing the mobility of toxic elements to negligible amounts. However, arsenic distinguishes itself by remaining mobile even under reducing environments. Therefore, arsenic is typically found in concentration value ranges in mg/L where other anion-forming elements are one thousand times less in comparison (in waters under reducing conditions). Abundantly found forms of inorganic arsenic (As(III) and As(V)) in drinking waters exist as AsO_3^{3-} (arsenite) and AsO_4^{3-} (arsenate). $As(OH)_3$, $As(OH)_2^+$, $As(OH)_4^-$ and AsO_3^{3-} are trivalent forms (arsenite) of [As], whereas $H_2AsO_4^-$, AsO_4^{3-} and $HAsO_4^{2-}$ are pentavalent forms (arsenate) of arsenic, found in natural waters. Names of arsenicals, chemical formulae and structure (two-dimensional and three-dimensional) of environmentally problematic arsenic compounds (organic and inorganic) are summarized in Table 3 [79,80].

Table 3. List of important arsenicals, their type, synonyms and structural (2D and 3D) and chemical formulas [79,80].
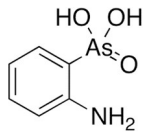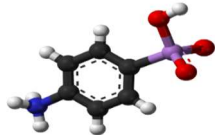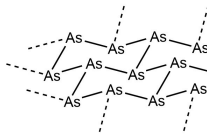
Name	Family	Synonyms	Structural Formula	Chemical Formula	Solid State Structure	Comments
Arsenilic acid	Organic	Arsonic acid, (4-aminophenyl)-		$C_6H_8AsNO_3$		a, b, c, d and n are applicable.
Arsenic	Inorganic	Metallic arsenic		[As]		c is applicable.

<div align="center">**Table 3.** *Cont.*</div>

Name	Family	Synonyms	Structural Formula	Chemical Formula	Solid State Structure	Comments
Arsenic(V) pentoxide	Inorganic	Arsenic oxide [As_2O_5]		As_2O_5		a and c are applicable.
Arsenic(III) sulfide	Inorganic	Arsenic sulfide [As_2S_3]		As_2S_3		c and e are applicable.
Arsenic(III) trichloride	Inorganic	Arsenic chloride [$AsCl_3$]		$AsCl_3$		c and f are applicable.
Arsenic(III) trioxide	Inorganic	Arsenic oxide [As_2O_3]		As_2O_3		a and b are applicable.
Arsenobetaine	Organic	Arsonium, (carboxymethyl) trimethyl-, hydroxide, inner salt; 2-(trimethylarsonio)acetate		$C_5H_{11}AsO_2$		d, b, c and a are applicable.
Arsine	Inorganic	Arsenic hydride		AsH_3		g is applicable.
Calcium arsenate	Inorganic	Arsenic acid [H_3AsO_4] calcium salt (2:3)		$(AsO_4)_2.3Ca$		a, c and h are applicable.
Dimethylarsinic acid	Organic	Cacodylic acid		$C_2H_7AsO_2$		b, a, c and d are applicable.
Metal (ME)/Lead arsen-ate/Me = Na, Ca, K, Mg, Al, Mn, Cu, Co, Cd, Fe, Ni, Pb, Sr, Zn	Inorganic	Arsenic acid [H_3AsO_4], Me/Pb (2+) salt (1:1)		$HAsO_4.Pb/ME$		j, a, c and b are applicable.
Methanearsonic acid, disodium salt	Organic	Arsonic acid, methyl-, disodium salt		$CH_3AsO_3.Na_2$		I, k, d, a and c are applicable.

Table 3. *Cont.*

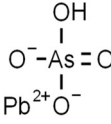| Name	Family	Synonyms	Structural Formula	Chemical Formula	Solid State Structure	Comments
Methanearsonic acid, monosodium salt	Organic	Arsonic acid, methyl-, monosodium salt		$CH_4AsO_3.Na$		I, k, d, a and c are applicable.
Potassium arsenate	Inorganic	Arsenic acid [H_3AsO_4], dipotassium salt		$H_2AsO_4.K$		l, a and c are applicable.
Potassium arsenite	Inorganic	Arsenous acid, potassium salt		$AsO_2.K$		l, a and c are applicable.
Sodium arsenate	Inorganic	Arsenic acid, [H_3AsO_4], monosodium salt		$H_2AsO_4.Na$		m, a, and c are applicable.
Sodium arsenite	Inorganic	Arsenous acid, sodium salt		$AsO_3.Na$		a, c and k are applicable.
Sodium cacodylate	Organic	Arsenic acid, dimethyl-, sodium salt		$C_2H_6AsO_2.Na$		a, b, c, d and k are applicable.

(a) Red = oxygen, (b) white = hydrogen, (c) purple = arsenic, (d) black = carbon, (e) yellow = sulphur, (f) green = chlorine, (g) grey and light gey are arsenic and hydrogen, respectively, (h) white = calcium, (I) white = hydrogen, (J) grey = metals, (k) dark purple = sodium, (l) dark purple = potassium, (m) hydrogen and sodium are dark purple and white, respectively, and (n) blue = nitrogen.

pH and redox potential (Eh) are considered the most important factors controlling arsenic speciation in water [81]. The reduced, trivalent form, As(III), is normally found in groundwater (anaerobic conditions) and the oxidized, pentavalent form, As(V), is found in surface water (aerobic conditions). The primary forms of arsenic typically discovered in environmental samples include As(III), As(V), various arsenious acids (H_3AsO_3, $H_2AsO_3^-$ and $HAsO_3^{2-}$) and arsenic acids (H_3AsO_4, $H_2AsO_4^-$ and $HAsO_4^{2-}$), with arsenic demonstrating anionic behavior in aqueous systems. Arsenate tends to prevail under oxidizing conditions, manifesting as the $H_2AsO_4^-$ type at lower pH levels (below roughly 6.9) or as the $HAsO_4^{2-}$ form at higher pH levels. Under reducing conditions with a pH less than about 9.2, the uncharged arsenite species H_3AsO_3 becomes predominant. At moderate or high redox potentials, arsenic can be present as pentavalent oxyanions (arsenate): H_3AsO_4, $H_2AsO_4^-$, $HAsO_4^{2-}$ and AsO_4^{-3} [82]. However, under more reducing conditions (acidic and mildly alkaline) and lower redox potential, the trivalent arsenic species (H_3AsO_3) are prevalent. At low pH levels in the presence of sulfide, $HAsS_2$ can form; in extreme reducing conditions, arsine, arsine derivatives and arsenic metal can occur. The distributions of these species in relation to pH and redox conditions (Eh) are illustrated in Figure 2 [9,83,84].

Figure 2. Eh–pH diagram of [As]–H$_2$O at 1 ATM pressure and 25 °C temperature, adapted with permission from [9], published by WHO.

The speciation of arsenic (arsenate and arsenite) in water under reducing conditions (as an example) as a function of pH is presented in Figure 3 [10]. Saxena et al., 2004 [85], reported the relationship of Eh and pH under other hydrogeological conditions with arsenic speciation in India, USA, South Korea and Bangladesh. The scientific analysis of the arsenic levels in the groundwater and pH shows a linear correlation. Overall, it is observed that [As] remained mobile in pH ranges of waters starting from >3.5 up to <9.0, and in the case of redox potential, [As] is found mobilizing in both oxidizing and reducing conditions. The behaviour of arsenic in water with high concentrations of sulphur is important. It has been shown that high concentrations of sulfides negatively impact dissolution of As. Dissolved arsenic-sulphur compounds are more regularly found in the presence of reduced sulphur in waters. The reducing condition causes the precipitation of sulphur compounds bearing arsenic, which reduces the levels of dissolved arsenic in the water [83,85].

Figure 3. *Cont.*

Figure 3. Distribution of speciation of arsenite, arsenate and organic arsenical in NaCl as a function of pH under experimental conditions (temperature: 25 °C, pressure: 1 ATM). Reproduced with permission from [10], publisher Elsevier B.V.

4. Arsenic Implications on Human Health

Arsenic has been termed the "*Poison of Kings*" having a significant role in the murder of several monarchs [86]. Furthermore, based on epidemiological evidence, the International Agency for Research in Cancer (IARC) has classified As and its compounds as carcinogenic [87]. Consumption of As contaminated water and food and occupational exposure through the air are the sources of arsenic poisoning by iAs. The primary exposure route for humans of oAs is by consumption of seafood (mussels, fish, prawns, oysters, etc.) which leads to severe impacts on human health [88]. The term "*Arsenicosis*" refers to medical complications resulting from low-dose long-term exposure to As [89]. As presents in the environment because of both natural and anthropogenic sources, resulting in As exposure. Recognised routes of As entering the human body are absorption through the skin, ingestion through the mouth and inhalation through the nose. The gastrointestinal tract of the human body absorbs both valences of As found in the environment, i.e., As(III) and As(V) [90]. However, the absorption capacity through the gastrointestinal tract changes for the various arsenicals; for example, $AsHO_4Pb$ and As_2S_3 have the l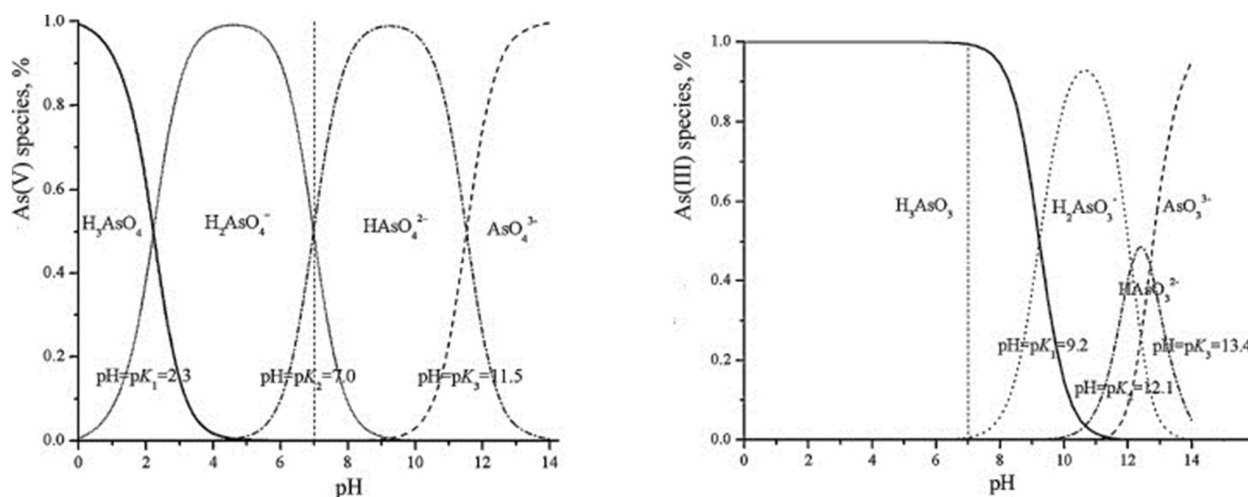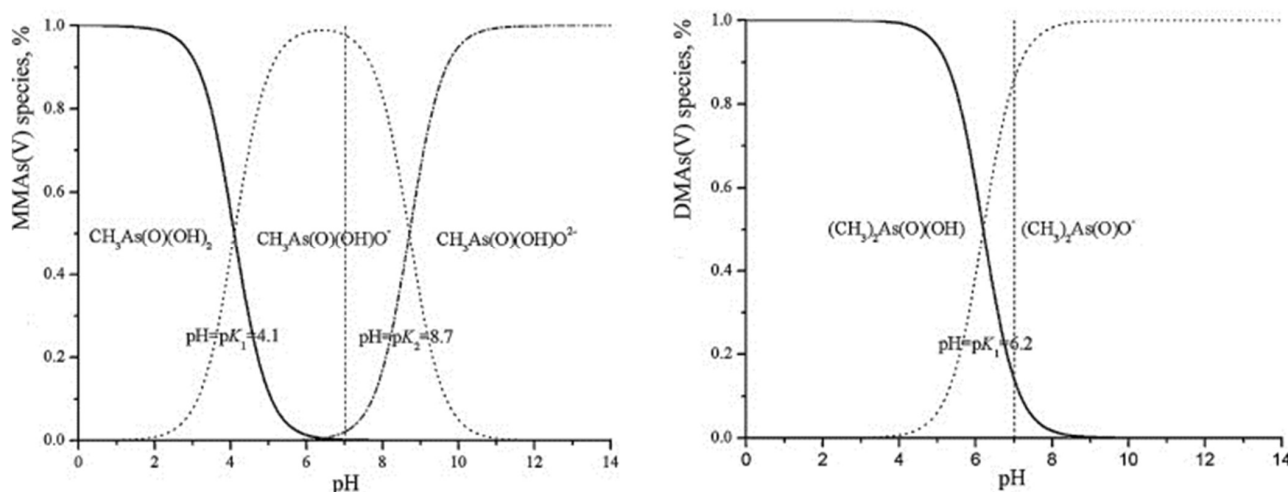owest rates of absorption through the mouth among As compounds, whereas, $AsHO_4Na$ has higher absorption rates in comparison. The inhalation of As depends upon the molecular size of the As compound; hence, in this case, $AsHO_4Na$, $NaAsO_3$ and As_2S_3 have faster absorption rates than $AsHO_4Pb$ and AsS [91]. The As(V) type of arsenics is less toxic in comparison with As(III) because trivalent arsenic compounds are more soluble in water. Both trivalent and pentavalent forms of As are reported to accumulate in human body fluids and are absorbed into human tissues [92]. The As in the human body distributes among the organs such as kidneys, skin, liver and lungs. The human liver subjects the As to non-enzymatic and enzymatic methylation. During methylation, iAs is reduced from pentavalent to trivalent forms of As, which are more toxic and mobile [92]. Urine, (renal system), transports about 70% of iAs and oAs. As the excretion process of iAs is longer, the retention time of iAs in the human body is greater than that of oAs [93]. Absorbed As can be found in human urine, hair and nail samples. The total As quantities in nails and hair are used as past As exposure indicators as As stockpiles in keratin-high tissues of the human body. Conversely, As quantities in human blood and urine are taken as indicators of recent exposure to arsenic due to its rapid metabolism [94]. A survey involving the collection of skin scales, urine, and hair samples from 10,000 residents of arsenic-affected villages in Bangladesh (one of the most severely affected countries) indicated that 95.1% and 93.8% of urine and nail samples, respectively, accumulated higher than normal levels of arsenic [95]. Another test (conducted in Pakistan) reported accumulation of As to levels higher than normal (Table 4)

in urine, hair and nails in the human body (sample size of 395 villagers) [96–98]. Low to moderate water levels (10–300 µg/L) can have adverse implications on human health, causing lung disease, effects on the neurological system, dermatological disorders, diabetes, cardiac diseases, chronic liver diseases and kidney diseases. The human body systems affected by As are illustrated in Figure 4 [99].

Table 4. Presentation of acceptable values/ranges of [As] in humans' nails, hair and urine.

Sample Type	Acceptable Range [As]	Units	Reference
Urine	0.005–0.04	mg/day	[90]
Nails	0.43–1.08	mg/kg	[88]
Hair	1	mg/kg	[91]

Figure 4. The figure depicts the human body systems affected by health implications from As absorption and accumulation.

Acute (short-term exposure) and prolonged (chronic) exposure of As poisoning have a diverse range of symptoms and can lead to multisystemic diseases (Figure 5) [100,101]. The development of health implications of acute exposure to As appears in less time in comparison with chronic exposure to As, which develops over a long period.

A recently published comprehensive review reports the critical health issues associated with As poisoning [102]. Carcinogenetic effects of As poisoning are the most common and prevalent. The results of epidemiological and animal studies have shown that iAs can be categorized as group 1 (IRAC) carcinogens. In contrast, DMA and MMA oAs can be classified as potential group 2b (IRAC) carcinogens. Arsenobetaine and other organic arsenicals have not been declared carcinogens and are placed in group 3 (IARC) [103]. It is observed that chronic exposure to As causing kidney, lung and bladder cancers has led to the death of the patients even after forty years of reduction in exposure to As [104]. Key epidemiological corroboration relating the carcinogenicity to As has been gained from case studies in Argentina [105], Bangladesh [106], Taiwan [107], and Chile [108], where residents consume drinking water containing high (e.g., 150 µg/L) concentrations of As. Other associated health effects from exposure to As include neurobehavioral disorders and neuritis. As toxicity is also linked to effects on the sensory system of nerves in the brain [109]. It has been observed during a study on Mexican children that the effects of toxicity of As in the form of neurobehavioral disorder, neuritis and disorder of the sensory system of the brain varies with the (factor of) time of exposure of As, nutritional factors and dose of

As [110]. Another health implication of As exposure is diabetes, a relationship between type 2 diabetes, obesity and arsenic exposure has been reported, with a higher incidence of diabetes where As concentration in drinking water is two to five times higher than the permissible limit of WHO [111]. As also has the potential to cause cardiovascular disease as it can affect thrombocytes (i.e., agglutination). Exposure to inorganic trivalent arsenicals may be the leading cause of this disease [112]. Skin diseases are also widely associated with exposure to As. According to a survey conducted in Bangladesh, melanosis and keratosis are found in 13.9% of the total 167 people who were exposed to As contamination in drinking water within the permissible value. In addition, Mee's lines, vesiculation and dermatitis (skin diseases) are also associated with the intake of water contaminated with As [113]. Complications may also arise during pregnancy [114], with foetus mortality and premature birth cases increasing with an increase in exposure to As [115].

Figure 5. Chronic and acute [As]-poisoning-implicated diseases, adapted with permission from [101], publisher Springer-Verlag GmbH Germany.

5. Techniques Used for Arsenic Quantification in Water

The detection and quantification of As and its species in water is vital to understand the complex biological and environmental chemistry of each single arsenical, as each As compound poses a different toxicity level [116]. Therefore studies (environmental and health) of As require the detection and quantification of As species of diverse properties and toxicities. Hence, quantification of total As is not adequate and must be accompanied by arsenic speciation, defined as the analytical techniques (classical and contemporary) of recognising and/or quantifying the amount of one or more individual arsenical in a sample [117]. Recent comprehensive scientific publications have reported the determination of total As and speciation in water samples [118–124]. Many analytical techniques are used to quantify As levels in natural waters, such as atomic fluorescence spectroscopy (AFS) [125], inductively coupled plasma mass spectroscopy (ICP–MS) [126], Raman spectroscopy (RS) [127], graphite furnace atomic absorption spectroscopy (GF-AAS) [128], total reflection X-ray fluorescence (TXRF) spectrometry [129], the frontal chromatography–ICP–MS method (FC–ICP–MS) [130], electrothermal atomic absorption spectrometry (ETAAS) [131], high-performance liquid chromatography coupled with inductively coupled plasma-mass spectrometry (HPLC-ICP-MS) [132], hydride

generation–atomic absorption spectrometry (HG-AAS) [133], surface-enhanced Raman spectroscopy (SERS) [134] and laser-induced breakdown spectroscopy (LIBS) [135].

The total As concentration in water samples, usually in units of µg/L, is only detectable by refined analytical techniques such as GF-AAS, ICP-MS and HG-AAS [126,128,133]. Coupling techniques are adopted alongside for the speciation of As, which involves the combination of a sensitive detector such as ICP-MS with HPLC [136]. The selection of the analytical technique depends upon factors such as the detection limit (LOD), the limit of quantification (LOQ), the cost of the method/apparatus, the time required for the technique, the pre-treatment requirement of the sample, the availability of the device and predetermined information as to whether the method is suitable for total [As] determination only or in addition, to quantify As species in the water sample [137–139]. Historically, spectrometric or colorimetric methods of total As determination were used. In the process, species of As in the test sample are reduced to As (mirror) or AsH_3, which then passes through a filter (can be modified but is usually a $HgBr^{2-}$ impregnated filter), which turns the contaminant into a yellow to brown colour. These methods are popular in forensic toxicology as they present visible proof of As determination. Such plans are cheap, easy to perform and applicable for the determination of high values of As in natural waters [124].

Electrochemical methods, particularly volumetric methods, are cheap, accurate and come with easy-to-use apparatus, and these methods are widely used for As determination. Much work has been conducted regarding modifying the method for better results. Ag and Pt electrodes are used in anodic stripping voltammetry (ASV) methods, and glassy-carbon electrodes are used in cathodic stripping voltammetry (CSV) methods with a low LOD (around 0.02 µg L^{-1}) for arsenic detection. In the procedure, As(V) is reduced to As(III) with the use of various chemicals, and a low limit of detections is achieved to calculate total As [140]. For the determination of total As, more sensitive detections such as AFS, AAS, MS and AES are used. The speciation analysis follows the determination of total As when required. To perform speciation analysis, the best methodology is coupling two analytical systems.

All the chemical forms of As in water are separated with one technique, and the second method is used to detect the separated species. In addition to coupling two analytical techniques and their deployment, there are other important steps for completing arsenic speciation analysis. The first step involves extraction of As, following this is the separation of different arsenicals, and the final stage is the quantification of each As compound [7]. The commonly used method for As speciation is HPLC coupled with ICP-MS. HPLC has produced effective results by separating arsenic species and can be quantified with a detector. A detector with high ionization efficiency, like ICP-MS, is commonly used. Other essential qualities of a sensor for As speciation are required, and characteristics of ICP-MS high selectivity include a wide range for sensitive detection and low matrix interference. However, HPLC-ICP-MS is only able to determine the identities of compounds with the use of reference standards. Therefore, in recent years, ESI-MS has been used along with ICP-MS to detect new species of As [141]. A review of contemporary analytical techniques for quantifying As species is presented in Table 5.

Table 5. A review of contemporary analytical techniques for quantification of arsenic species.

[As] Species	Methodology	Technique Description	Detection Limit (µg/L)	Reference
DMA, As(III) and As(V)	HG-ASS	$NaBH_4$ was used as a reductant in a separate reduction media. River water samples were analysed using HG-ASS.	600, 1100 and 500	[142]
Total As and As(III)	GF-AAS	A sample preconcentration method for As(III) by APDC and ion exchange resin followed with detection by GF-AAS.	0.001	[143]

Table 5. *Cont.*

[As] Species	Methodology	Technique Description	Detection Limit (µg/L)	Reference
As(III)	SERS	Fe$_3$O$_4$@Ag was used as a magnetic substrate. EXAFS [T] spectroscopy was used to characterise the molecular structure of As on Fe$_3$O$_4$@Ag followed by SERS.	10	[144]
As(V)	SERS [S]	SERS quantification analysis technique deployed for [As(V)] using multi-layer silver nanofilms deposited on glass slides as SERS-active substrates by an electroless deposition process.	~5	[145]
As(V)	UV/vis Spectrometry	S–layer protein functionalized AuNPs reacted with As species, which changed the colour of AuNPs, detected by UV/vis spectrometry.	240	[146]
As(III)	UV/vis Spectrometry	Spectrometry method deployed with receptor as glucose in AuNPs platform.	0.53	[147]
Total As	SPRS [B]	Sensor probes of thiol-containing organic compounds using a SPRS method to detect As.	10	[148]
As(III), As(V), DMAA, MMAA and AsBet	HPLC-ICP-MS	The method involved Ion Chromatography hyphenated to an inductively coupled plasma mass spectrometer.	0.02	[132]
Total As	FP-XRF [C]	X-ray tube-based FP-XRF methodology was used to determine low concentrations of As.	<0.02	[149]
Total As	ICP-AES	Electrolytic reduction was used as an alternate hydride generation method in atomic spectrometry for sample introduction. As detected by Flame atomic absorption spectrometry.	0.7	[150]
Total As	HPLC-ICP-MS	Species of As separated using an isocratic elute and identified by ESI-MS [U].	~0.1	[122,151]
Total As	ICP-SF-MS	SPE [F] cartridges packed with anion exchange resin (modified conventional sorbent) before detection by ICP-SF-MS.	0.06	[152,153]
Total As, As(III) and As(V)	HG-AAS	SPE protocol with dual sorbent followed. SBAE [G] resin and hydrate iron (III) oxide particle-integrated HY resin cartridges were used before detection by HG-AAS.	0.24 [I]	[154]
As(III)	ETAAS [J]	Based on IL [V] dispersive microextraction technique implemented in a flow analysis system followed by ETAAS detection.	0.05	[155]
As(V)	ICP-OES [K]	After pre-treatment of the sample with mesoporous silica, total iAs is quantified by oxidation (by KMnO$_4$) of As(III) to As(V) and followed by ICP-OES and the As(III) measurement is found by the subtraction of As(V) from total As.	0.05	[156]

Table 5. *Cont.*

[As] Species	Methodology	Technique Description	Detection Limit (µg/L)	Reference
As(III) and As(V)	ICP-MS	Pre-treatment of water sample by SPE procedure with CNFs [L] modified with APDC [M] and then analysed with ICP-MS.	0.0045 and 0.24	[157]
Total As	HG-AAS	HG-AAS was adopted after pre-treatment of the sample with cellulose fibre coated with $Y(OH)_3$ precipitate layer as adsorbent.	0.012	[158]
As(V)	AFS [P]	MWCNTs [N] modified with BPEI [O] used as a sorbent for pre-treatment of water sample followed by sequential injection technique and then analysed by AFS.	0.014	[159]
As (Ultra Trace)	AFS	Nano-TiO_2 colloid was used to pre-treat the sample. A slurry sampling technique was adopted, followed by AFS detection.	0.01	[160]
As(III)	HG-AFS	Pre-treatment of the sample using Fe_3O_4 nanoparticles and SPE procedure was achieved, followed by HG-AFS method for detection.	0.0135	[161]
Total As and As(V)	HG-AFS	Methyl esterified egg-shell membrane was used to pre-treat the sample, followed by detection using a HG-AFS method.	0.015	[162]
	Colorimetric Chemo-sensor	Mn_3O_4 nanoparticles were used as absorbents. Adsorption of [As] changed the surface morphology of Mn_3O_4 and the colour to yellow. This property led to the development of a novel colourimetric chemosensor method for [As] detection.	1.32	[163]
Total As	ASV [W]	ASV involving the reduction of As^{3+} to As0, followed by stripping or oxidation to As^{3+} or As^{5+} species using electrodes modified by nanoparticles (carbonaceous nanomaterials).	0.05	[119]
As(III), As(V), MMA and DMA	HPLC-GHG-AAS [A1]	As species analysis by interfacing solid phase preconcentration-LC [X] separation-GHG [Y]-QFAAS [Z].	0.019, 0.33, 0.39, 0.62	[164]

A = AuNPs = gold nanoparticles; B = SPRS = surface plasmon resonance sensor; C = FP-XRF = field portable energy dispersive X-ray fluorescence; D = ICP-AES = inductively coupled plasma optical emission spectrometry; E = inductively coupled plasma sector field mass spectrometric; F = SPE = solid phase extraction; G = SBAE = strong basic anion exchange; I = total [As]; J = ETAAS = electrothermal atomic absorption spectrometry; K = ICP-OES = inductively coupled plasma-optical emission spectrometry; L = CNFs = carbon nanofibers; M = APDC = ammonium pyrroine-dithio carbarnate; N = MWCNTs = multi-wall carbon nanotubes; O = BPEI = cationic polyethyleneimine; P = AFS = atomic fluorescence spectrometer; Q = MESM = methyl esterified egg-shell membrane; R = NaBH4 = sodium tetra hydroborate (III); S = SERS = surface-enhanced Raman scattering; T = EXAFS = extended X-ray absorption fine structure; U = ESI-MS = electrospray ionization-mass spectrometry; V = IL = ionic liquid; W = ASV = anodic stripping voltammetry; X = LC = liquid chromatography; Y = GHG = gradient hydride generation; Z = QFAAS = quartz flame atomic absorption spectrometry; A1 = HPLC-GHG-AAS = HPLC-gradient hydride generation atomic absorption spectrometry.

As field detection kits are used to quantify As levels in water sources in areas of the world where water supply is decentralised and infrastructure of water testing is limited. From a public health standpoint, the utmost importance lies in the capability of a

measurement method to differentiate samples that exceed and fall below pertinent and actionable benchmarks for drinking water quality. Eight field test kits, currently accessible in Bangladesh, were evaluated by contrasting the estimates provided by these kits with analyses utilising hydride generation (HG-AAS) techniques. The outcomes of examinations designed to mitigate colour-matching errors influenced by user interpretation indicated that among the kits evaluated, the LaMotte and Quick II kits yielded accurate and precise arsenic estimations. Regarding four kits (Econo-Quick, Quick, Wagtech and Merck), accuracy or precision was achieved, but not both [165]. Conversely, the Hach and Econo-Quick II kits displayed neither accuracy nor precision. Considering these findings, the investigators proposed that stakeholders exercise caution when utilising field test kits with suboptimal performance. It is advised that such kits be reevaluated once enhanced quality control measures are introduced for their constituent components. Furthermore, the researchers advocate for field test kit manufacturers to include appropriate internal standards within each kit package, enabling users to validate the accuracy of the manufacturer's colour charts. A comparison of the test results and credentials of various test kits available in the market are presented in Table 6 [165].

Table 6. Presentation of the test results and other characteristics of [As] field detection kits, modified from [165].

Test Kit	Manufacturer	No. of Test in Box	Reaction Time per Test (Hr)	Methodology	Concentration Intervals on Colour Chart (µ/L)	Results
Merck	Merck, Darmstadt, Germany	100	0.35	Colorimetric	5, 10, 25, 50, 100, 250, 500	Either accurate or precise, but not both.
Quick II	Industrial Test Systems, Rock Hill, USA	200	0.25	Colorimetric	<1, 2, 3, 4, 5, 6, 7, 8, 10, 13, 20, 25, 30, 40, >50, >80, >120, >160	Accurate and precise estimates of [As].
Econo-Quick II	Industrial Test Systems, Rock Hill, USA	50	0.25	Colorimetric	<2, 4, 10, 15, 20, 25, 30, 40, 50, 60, 70, 80, 100, >150, >300	Neither accurate nor precise estimate of [As].
LaMotte	LaMotte, Chestertown, USA	50	0.25	Colorimetric	<4, 4, 8, 10, 12, 14, 16, 20, 25, 30, 50, 85, 100, 150, 175, 200, 300, 400	Accurate and precise estimates of As.
Wagtech	Palintest, Gateshead, UK	200	0.35	Colorimetric	<10, 20–40, 50, 60–80, 100, 100–200, 200–300, 300–400, 400–500	Either accurate or precise, but not both.
Quick	Industrial Test Systems, Rock Hill, USA	100	0.25	Colorimetric	0, 5, 10, 20, 30, 40, 50, 60, 80, 100, 150, 200, 250, 300, 400, 500, >500	Either accurate or precise, but not both.
Econo-Quick	Industrial Test Systems, Rock Hill, USA	300	0.25	Colorimetric	0, 10, 25, 50, 100, 200, 300, 500, 1000	Either accurate or precise, but not both.
Hach EZ	Hach, Loveland, USA	100	0.35	Colorimetric	0, 10, 25, 50, 250, 500	Neither accurate nor precise estimate of [As].

6. Techniques Used for Arsenic Mitigation from Water

The underlying chemistry of arsenic-contaminated water stands as the primary influencers in As removal. Removal methods are generally more proficient in eliminating arsenate, as arsenite remains mostly uncharged at pH levels below 9.2. Consequently,

the trivalent form of arsenic, being less charged, becomes less amenable to precipitation, adsorption or ion exchange. As a result, treatment strategies are deemed more effective when employing a two-step process: initially oxidizing arsenite into arsenate, followed by employing a technique to eliminate the arsenate [78,166–170]. Figure 6 provides an overview of existing technologies currently applicable for purging arsenic from water sources (Figure 6).

Figure 6. Overview of As removal strategies from water.

6.1. Oxidation Method

Oxidation, which transforms soluble arsenite into arsenate, does not inherently eliminate arsenic from the solution. Therefore, a subsequent removal method like adsorption, coagulation or ion exchange becomes necessary to extract arsenic from the solution [171]. The oxidation process plays a key role in improving the efficiency of removal of As; therefore, in most situations of such contamination removal, pre-oxidation of As(III) to As(V) is deemed necessary and a sample redox reaction of conversion is given as follows: [172]

$$H_3AsO_4 + 2H+ 2e^- \rightarrow H_3AsO_3 + H_2O \quad E0 = +0.56 \text{ V}$$

Selection of the oxidant depends upon factors such as the characterization of other water-soluble containments or interfering substances (such as S^{2-} and TOC, as these greatly affect the performance (increase in time) of ozone as an oxidant) [173]. The by-products are a concern, e.g., TiO_2 as an oxidant when used for oxidation of As-bearing water, produces a by-product which is costly and difficult to dispose of [174]. In addition, if the water used for As removal contains highly dissolved organic carbon (DOC) and is oxidized using chlorine as an oxidizing agent, the by-product generated may contain toxic dibutyl phthalate (DBP) [175]. Other factors to consider are the pH value on which the oxidant reacts, the reaction rate and the concentration of the oxidizing agent required. Oxidation by ClO^-, O_2 and manganate (VII) is commonly adopted in developing countries due to cost concerns. However, it is observed that using atmospheric oxygen is a lengthier process, and manganate (VII), ClO− and ozone readily oxidize the arsenite to arsenate [176,177]. A review of contemporary oxidization

agents (chemical formula, pH value, initial concentration of system and the brief result of the method) for the oxidation of As(III) to As(V) is presented in Table 7.

Table 7. Various chemical oxidants are in use for the oxidation of As(III) to As(V) along with the result of the method, pH value, initial concentration of system and chemical formulas.

Oxidizing Agent	Chemical Formula	pH	Initial Conc. (μg/L)	Results	Reference
Ozone and Oxygen	O_3 and O_2	7.6–8.5	46–62	Oxidation of As(III) to As(V) was fast (4 min) with O_3 and slow (2–5 days and 4–9 days, respectively) with pure O_2 and air.	[178]
Chlorine dioxide	ClO_2	5.7–6–7 and 8	300	The 90% oxidation (maximum) of As(III) to As(V) yield was achieved with ClO_2 to As(III) concentration ratio of 3 at a contact time of 6 days.	[179]
Hydrogen peroxide	H_2O_2	7.5–10.3	50	Oxidation efficiency was found to increase with an increase in pH from 7.5 to 10.3.	[180]
Monochloramine	NH_2Cl	5.7–6–7 and 8	50	A 100% oxidation yield was achieved using an oxidant-to-concentration ratio of 3 and with contact times of 2 days.	[179]
Hypochlorite	ClO^-	5.7–6–7 and 8	50 and 300	An oxidant to As(III) ratio of 3 resulted in 100% oxidation of As(III) to As(V) after 5 min of contact time for all the pH conditions.	[179]
Chlorine	Cl_2	8.3	300	As(III) was oxidized to As(V) with a stoichiometric ratio of 0.99 at the fastest rate.	[181]
Potassium permanganate	$KMnO_4$	5.7–6–7 and 8	50	An oxidant to As(III) ratio of 1–2 resulted in 100% oxidation of As(III) to As(V) after 1 min for all the pH conditions studied.	[179]

Recently, in situ oxidation of arsenic (As) has been adopted in groundwater aquifers to control the mobilization of As in the subsurface. Aerated water is periodically pumped into the groundwater anoxic aquifer through a well or multiple wells. When dissolved Fe(II) reacts with aerated water, it oxidizes and results in the formation of precipitate on the surface of the aquifer, which is soil; this creates a surface capable of adsorption of As and restricts its mobility. The process is pH sensitive and affected by interfering ions [182]. Photochemical oxidation is another method of oxidizing As in water, evolving into a different and new range of oxidation methods. In these methods, ultraviolet (UV) radiation is used to catalyse the oxidation of As(III) to As(V) in the presence of iron ions [183]. Some other compounds, such as H_2O_2, can also be combined with UV radiation for the process [184]. Deriving a sustainable source of UV radiation from solar energy has also been reported, and a portable device for household As mitigation from water has been developed [185]. Microorganisms such as chemoautotrophic bacteria and heterotrophic bacteria are used to facilitate the oxidation of As(III) to As(V), which is called biological oxidation. These bacteria reduce nitrate or oxygen by acting as electron donors. The biological oxidation of manganese and iron, which are established absorbents of As, is an indirect method of determining As mitigation in water [186]. Other bacteria, such as Thermus thermophilus and Thermos aquaticus, can also oxidize arsenite to arsenate [187]. The efficiency of oxidation of As(III) to As(V) has been continuously improved with the advancement of materials and methods used. Table 8 presents a review of some of the

latest developments in the field where oxidation processes have been employed for arsenic mitigation from water.

Table 8. Case studies using the oxidation process for arsenic mitigation from water.

Water Type	Materials	Method	Study Characteristics	Reference
Deionized water	Fe–Cu–Mn composite oxide (FCMO$_x$)	Synergistic oxidation and ultrasonic coprecipitation method	FCMOx adsorbent was used to separate As from aqueous solutions. FCMOx showed efficient results for photocatalytic oxidation with the use of UV irradiation. The combination of adsorption and synergistic oxidation improved the removal efficiency of As(III).	[188]
Deionized water	Fe-biochar fibres, H$_2$O$_2$ and Hydroxylamine (HA).	Oxidation and adsorption	Fe-biochar fibres integrated with H$_2$O$_2$ and Hydroxylamine (HA) were used to prepare a heterogenous material which showed efficient and rapid result for the chemical oxidation of As(III) to As(V) followed by absorption of As(V) by Fe-biochar.	[189]
Distilled water	Santa Barbara Amorphous-15 (SBA-15) and TiO$_2$.	Photocatalytic oxidation and adsorption	Impregnation was performed to produce Ti-SBA-15. This novel material was an excellent photocatalyst at all pHs with 98% efficiency.	[190]
Distilled water	*Ancylobacter* sp. TS-1, zeolite, polypropylene, graphite and sand.	Biological oxidation	*Ancylobacter* sp. TS-1 (biomaterial) was used to make films on all materials mentioned excluding polypropylene. Film of *Ancylobacter* sp. TS-1 on graphite material was the most rapid oxidant of As(III) to As(V).	[191]
Milli-Q water	Fe-Mn-incorporated titanosilicate material (ETFMS-10).	Adsorption and oxidation	ETFMS-10 was prepared by hydrothermal method. The material was used to oxidize and adsorb As with efficiency of 70%.	[192]
Gold mine effluent.	*Alishewanella agri* strain (PMS5).	Bio-Oxidation	Microorganisms (PMS5) showed an ability to reduce As(V) and oxidize As(III) with efficiency of 75.5–94.7% and 8%, respectively.	[193]
Ultrapure water.	Manganese-doped Lanthanum oxycarbonate (MnL) and H$_2$O$_2$.	Oxidation and adsorption	MnL catalysed H$_2$O$_2$ oxidation of As(III) to As(V) studied. Efficient oxidation and adsorption results were seen over a wide range of pHs.	[194]
Milli-Q water	CuFe$_2$O$_4$ activating peroxymonosulfate.	Oxidation and adsorption	A novel material was introduced for the efficient oxidation and adsorption of As/As(III).	[195]
Paddy rice water	Reactive intermediates (RIs) from paddy water.	Oxidation	RIs improved the oxidation rate of As(III) to As(V) by 1.8–4.1-fold in paddy water compared to surface water with oxygen during a dark reaction.	[196]
Deionized water	Fe-modified biochar (FeBC)	Oxidation and adsorption.	With the use of FeBC, the efficiency of [As] removal increased from 99.2% and 86.4%, respectively, for As(V) and As(III) in the absence of 0$_2$, to >99.9% in the presence of O$_2$.	[197]

Table 8. *Cont.*

Water Type	Materials	Method	Study Characteristics	Reference
Sediment water	O_2 nanobubbles	Bio oxidation and air oxidation	With the application of O_2 nanobubbles to sediment water sample with algal-induced hypoxia, the concentration of [As] in the sample rose to <10 µg/L from 23.2 µg/L as nanobubbles catalysed the oxidation of As(III) to As(V) (65–75%) and methylated [As] (10–15%).	[198]
Deionized water	molybdenum-disulfide/iron-oxide (MoS2/FeOx@BC)	Oxidation and adsorption	MoS2/FeOx@BC oxidize the As(III) to As(V) and then adsorb with greater efficiency as compared to MoS2/BC and FeOx@BC.	[198]
Ultrapure water	WTRs-chitosan beads (WCB) and Mn-WCB	Oxidation and adsorption	Results suggest that oxidation ability improved; hence, adsorption ability of WCB improved with MnO_2 added to WCB.	[199]
Ultrapure water	Sulfite	Oxidation	A novel sulfite activation method using ultrasound for achieving an improved oxidation rate of As(III) compared to oxidation supported by ultrasound (US) showed a 2.9% rise in As(III) oxidation rate at 7 pH.	[200]
Distilled water	Bentonite/chitosan/titania (BT/CS-TiO2).	Photooxidation and adsorption	BT/CS-TiO2 was prepared and then tested under UV, which produced 97% oxidation of As(III) to As(V).	[201]

6.2. Precipitation Method

The process of transmuting a dissolved material from a supersaturated solution into an insoluble solid is called precipitation. Under this category, co-precipitation of As with metal hydro-oxides occurs with conventional methods such as lime softening, coagulation/flocculation and iron/manganese and As mitigation by O^{2-} filtration [202].

Removal of As from water by softening with caustic soda or lime is one of the more conventional methods [203]. It is vital to control the pH of water between 8.5 and 9.5 to perform softening of normally occurring hard water; however, this pH range is not suitable for softening of water containing As, as a pH value above 10.5 is required to attain magnesium-dependent precipitation, which is desired as magnesium has a high tendency to adsorb [As] and is a cheaper option—the pH of the water medium must be normalized, which can be costly. Oxidation of As(III) (if present) to As(V) and the addition of a coagulant can increase the As removal efficiency [204].

Coagulation and flocculation are the most employed and documented techniques of As mitigation from water [205]. Positively charged coagulants (such as ferric chloride ($FeCl_3$) or aluminium sulfate ($Al_2(SO_4)_3$)) are introduced to As-containing water. The precipitation method of iron/manganese removal by aeration for the mitigation of [As] in water is similar to the addition of iron-based coagulants, with the difference being that precipitation of naturally present ferrous iron Fe(II) occurs. The coagulant reacts and forms larger particles with reduced As in water. When ionic flocculants are added to the system, they transform the already formed particles of effluents into larger sizes by chemically binding the smaller particles and resulting in flocs. This is followed by a process of filtration or sedimentation [206] (Figure 7).
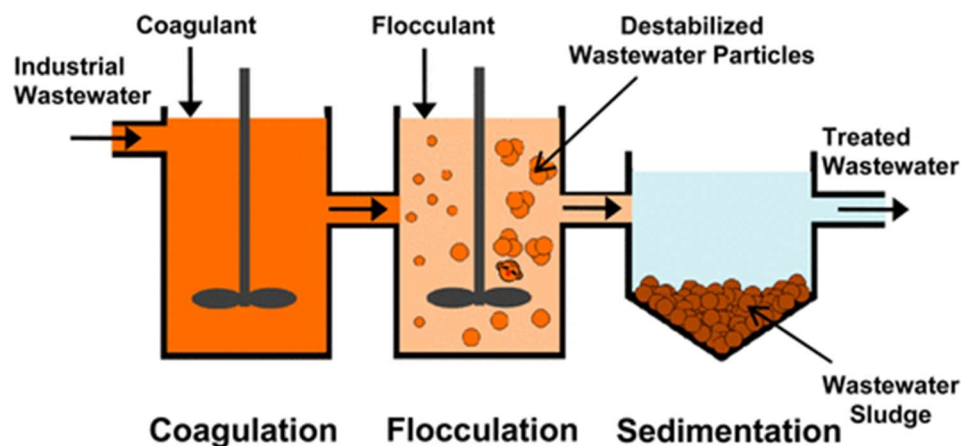
Figure 7. Illustration describing the coagulation, flocculation and sedimentation of As in water, adapted with permission from [206], publisher ACS Publications.

As removal from water with coagulation/flocculation is impacted by pH. At a pH ≤ 7.6, widely used coagulants (iron-based and aluminium-based) have the same efficiency; however, at a pH > 7.6, $Al_2(SO_4)_3$ is reported to have less operational efficiency as compared to $FeCl_3$. However, both coagulants decrease the concentration of As in saturated solution to a safe level that is less than 10 ppm (WHO) [207–209]. The by-product of these methods is challenging to handle, is mainly in the form of sludge and is costly to treat to avoid any secondary pollution [201]. Table 9 reviews conventional and contemporary coagulants, the operating pH noted by scientists, formulas, initial concentration of As and brief results.

Table 9. Various chemical coagulants used for [As] removal along with brief method result, pH value and initial concentration.

Coagulant	Chemical Formula	pH	Initial Conc. of As (μg/L)	Results	Reference
Ferric chloride	$FeCl_3$	8.0–8.4	400–600	The technology was deployed at a large scale in northern Chile for provision of drinking water. The result shows reduction in As concentration from 400 μg/L to 10 μg/L.	[207]
Alum	X $Al(SO_4)_2 \cdot 12H_2O$	7	2000	The results showed that at 125 mg/L and 100 mg/L doses of alum at pH 7, the removal efficiency of As was between 82 and 86%.	[210]
Zirconium (iv) Chloride	$ZrCl_4$	7.5	50	A total of 55% removal of [As(V)] was observed with 2 mg/L does of coagulant. The efficiency is found directly proportional at pH range 6.5–8.5, whereas removal of As(III) only found as 8% irrelevant to pH.	[211]
Titanium (iii) chloride	$TiCl_3$	7.5	50	With 2 mg/L dose of coagulant, 32% of [As(III)] and 75% of [As(V)] removal achieved. The process remained pH dependant.	[211]
Titanium (iv) chloride	$TiCl_4$	7.5	50	A total of 55% removal of As(V) was observed on distilled water sample with 2 mg/L dose of coagulant. The efficiency is directly proportional at the pH range 6.5–8.5, whereas removal of As(III) was only found as 26% irrelevant to pH. When tested on wastewater, the results showed that coagulant performs excellently in waters with high DOC and low alkalinity.	[211,212]

Coagulant	Chemical Formula	pH	Initial Conc. of As (µg/L)	Results	Reference
Titanium(IV) oxychloride	Cl_2OTi	7.5	50	With 2 mg/L of coagulant dose, 37% and 20% removal efficiency were achieved for [As(V)] and As(III). The methods remained pH dependent.	[211]
zirconium oxychloride	Cl_2H_2OZr	7.5	50	With a 2 mg/L dosage of coagulant, 59% and 8% removal efficiency was achieved for As(V) and As(III).	[211]
Ferric Sulphate	$Fe_2(SO_4)_3$	7	500 and 1000	Dosages of 32 mg/L (1000 µg/L initial concentration) and 28 mg/L (500 µg/L initial concentration) of coagulant give 100% results.	[213]
Titanium (IV) Sulphate	$TiOSO_4$	7	1000	Removal of 90% of As(III) was achieved with 25 mg/L dosage of coagulant.	[214]

Some latest developments on coagulation and flocculation for As removal in water are presented in Table 10. The precipitation method for mitigating As from water is used with multiple modifications. The methods of coagulation and flocculation are combined with adsorption to attain better results. The process of electro-coagulation has been improved with the introduction of novel materials electorates. The efficiency of precipitation of dissolved As in water has also been enhanced using pre-oxidation of [As] and biological materials. Combining coagulation-flocculation with sand filtration has also produced improved results with the potential of low cost. Developing small-scale and household devices for mitigating As in water and incorporating the technology of precipitation shows that the technology is cost-effective and in reach of the public.

Table 10. Case studies using coagulation and flocculation process for arsenic mitigation from water.

Water Type	Materials	Method	Study Characteristics	Reference
Groundwater	Aluminium sacrificial anodes	Electrocoagulation (EC) and flocculation	Hydrated silica and aluminium react in an EC system to form aluminosilicates, and As becomes adsorbed on newly formed compound. The result showed achievement of As concentration in water less than 10 ppm.	[215]
Tap water	Cellulose and chitosan-based natural biopolymer and ferric chloride (FeCl₃)	Coagulation, flocculation and adsorption	In the comparative study, initial concentration of 2 mg/L removed with efficiency of 69.25% using FeCl₃ as coagulant, whereas the use of cellulose and chitosan-based ferric chloride FeCl₃ has enhanced the removal efficiency to 84.62%.	[216]

Table 10. *Cont.*

Water Type	Materials	Method	Study Characteristics	Reference
Deionized water	Hydrophilic ligands and FeCl$_3$	Coagulation and flocculation	Hydrophilic ligands were introduced for the separation of As from water. It is noted that smaller doses of coagulant (Fecl$_3$) are required to achieve the higher As(III,V) removal, comparatively.	[217]
Groundwater	Mining drainage effluent and exchange resin with iron oxides.	Coagulation, flocculation, and adsorption	To achieve high removal rate and low cost, coagulation, flocculation, and adsorption were combined to test in comparison with other conventional systems, and the new system has produced better results.	[218]
Milli-Q purified water	Fe(II), H$_2$O$_2$ and [Fe(III)]	Coagulation	Introduction of H$_2$O$_2$ and Fe(II) to conventional iron-based Fe(III) coagulation was studied, and it was found that addition of H$_2$O$_2$ significantly enhanced the performance efficiency of removal of As from water.	[219]
Deionized water	Cactus mucilage and Ferric [Fe(III)] Salt.	Coagulation and flocculation	The addition of mucilage treatment (flocs) to the conventional system of coagulation enhanced As removal efficiency and removed 75–96% of As in 30 min of contact time.	[220]
Mine water	Ferric sulphate [(Fe$_2$(SO$_4$)$_3$]	Coagulation, lime precipitation, ballasted flocculation, and sedimentation	Fe$_2$(SO$_4$)$_3$ used for single-stage and two-stage coagulation, lime precipitation, ballasted flocculation, and sedimentation was compared, and found the two-stage system is more efficient.	[221]
Groundwaters	Coagulants: FeCl$_3$, Fe$_2$(SO$_4$)$_3$, FeSO$_4$, and oxidants: H$_2$O$_2$ and KMnO$_4$	Oxidation, coagulation and flocculation	Different oxidants and coagulants were used to treat [As] bearing groundwater, and 97% removal efficiency is achieved with the initial concentration of 204 µg/L.	[222]
Groundwater	Ferric(III) sulfate (FS) and Polyferric sulfate (PFS)	Coagulation and flocculation and sand filtration	FS and PFS coagulants were compared using microscopic techniques to analyse the surface complexes of Fe flocs and two-bucket systems, combining coagulation and sand filtration. PFS is found superior.	[223]
Lake water	Ferric chloride	Facile remediation strategy by coagulation process	The coagulant was directly sprayed on the lake affected with As without any pre-treatment, and a removal efficiency of 95.1–96.7% was achieved.	[224]

Table 10. *Cont.*

Water Type	Materials	Method	Study Characteristics	Reference
Millipore water	Iron Cathode and Aluminium anode	Electrocoagulation (EC)	EC method used with iron cathode and aluminium anode to check removal efficiency. At pH 5–8, 3 volts and 12 min processing time, initial concentration of 2–5 mgL^{-1} conc. of As(V) was brought down to <10 ppm.	[225]
Deionized water	Aluminium chloride (AlCl$_3$) and polyaluminium chloride (PACl)	Coagulation	Alum coagulants (AlCl$_3$ and PACl) were investigated for removal efficiencies of As(III) and As(V). Both were found not impressive for the removal of As(III), whereas they showed 100% removal efficiency for As(V).	[226]
Deionized water	Multiple-Review	Coagulation/flocculation and adsorption	A review of small-scale and household technologies using coagulation/flocculation and adsorption methods were deployable in rural/underdeveloped areas with the least cost.	[227]
Raw water	Sodium hypochlorite (NaClO) and FeCl$_3$	Oxidation and coagulation-flocculation	Enhanced coagulation in an integrated efficient-whirling clarifier was performed with FeCl$_3$ to achieve the result of 10 μg/L from the initial 100 μg/L concentration.	[181]

6.3. Membrane Technologies

Different membrane-based technologies, microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO), require a pressure difference between the feed side and outlet to ensure the system functions optimally. Under a broader classification, the membrane systems which require operating pressure ranging from 3 to 120 bars (RO and NF) are called high-pressure membranes, and UF and MF are termed low-pressure (0.1–5 bars) membranes [228,229]. Pressure-driven membranes are products of natural and synthetic materials (polyuria polyamide, polysulfone, straw, etc.) with pores through which fluid passes under pressure, leaving behind dissolved and suspended non-essential materials such as tiny ions, molecules, bacteria and larger particles up to 200 μm in diameter [230–232]. The As mitigation with membrane processes depends on membrane technologies' pore size range. Pore sizes are typically measured in nanometers (nm) and diameters range between 100 and 10,000 for MF, 2 and 100 for UF, 0.5 and 2 for NF and <0.5 in the case of RO, Figure 8 [233].

NF and RO are conventionally adopted for direct As remediation in water. However, the process of MF and UF also opted but with in-line coagulation to increase the size of the particles of compounds bearing As and the effect of pH cannot be neglected [232]. A more sophisticated method of UF, called micellar enhanced UF, may be used to remove As from water [231]. NF and RO have nearly similar characteristics deemed technically fit for As remediation from water; however, there is a distinction between both methods. Mono- and multivalent ions are completely removed by RO with very high retention, whereas NF has lower retention of monovalent ions with higher water fluxes [233]. Determining and understanding the physical and chemical properties of As speciation in water is vital

for optimising the separation of [As] through membrane technologies and operational efficiency. Due to the impact of pH on chemical properties such as hydration, charge and size of the As species, this can affect the retention mechanism. Change in pH may also affect membrane properties such as pore size and surface charge. Therefore, pH significantly controls the retention of As in NF/RO systems [234]. Other properties such as temperature, operational conditions, the concentration of total dissolved solids and other contaminants in water are also important [232,233,235]. Studies suggest that retention of As(III) is typically lower than retention of As(V) [235]; therefore, pre-oxidation may be required to control the suspended particulate and gain maximum operational efficiency.
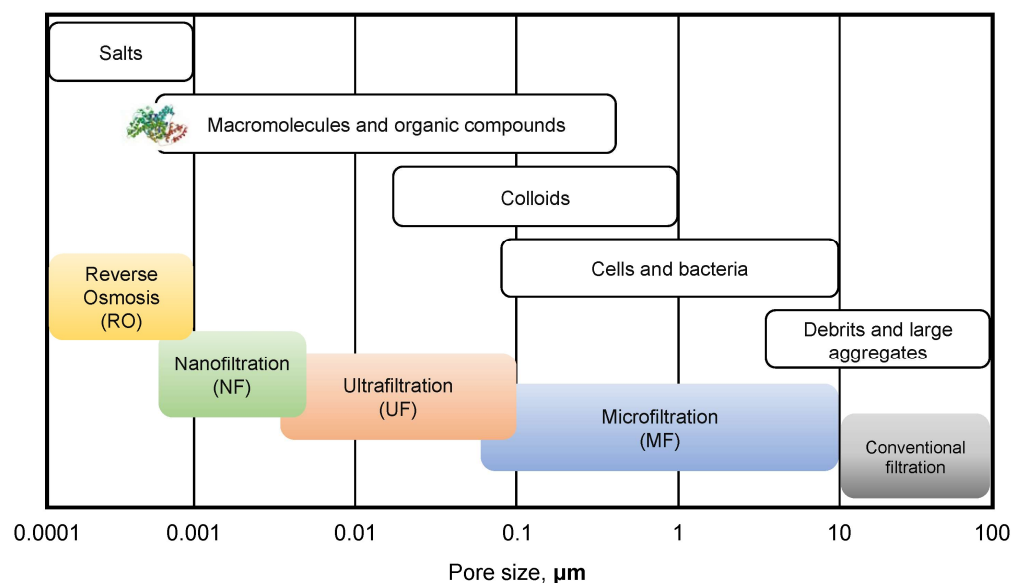

Figure 8. Pore size classification of different membrane technologies with characteristic dimensions of various foulants, adapted with permission from [233], publisher MDPI Publications.

Further improvements related to membrane-based As mitigation methods have been introduced with various processes and new technologies. Table 11 is a review of some latest developments in membrane-based technologies for As removal in water. New technological processes with various methods have been deployed to improve As removal by membrane technology science. Membrane filtration processes embedded with novel materials/cheap adsorbents/nanomaterials have been proven effective for separating As from water. Novel materials have been developed which produce better removal efficiency when used in the fabrication of membranes and avoid fouling of membranes to some extent. Membrane distillation in which hydrophobic membranes with micro-pores were found to be operationally more effective as non-condensable gases and vapours are only present in the membrane's pores, thus avoiding contact with water. Liquid membranes, especially HFSLM and emulsion liquid membranes, were also demonstrated to be efficient for these purposes [235,236].

Table 11. Case studies using membrane technologies for arsenic mitigation from water.

Water Type	Materials	Method	Study Characteristics	Reference
Deionized Water	Silk nanofibrils	Membrane filtration	Silk nanofibrils-based, multi-layered, novel material membranes were prepared. The system produced a flux of >890 L/m^2/h (high) and >89% of rejection of pollutants, including As. The membrane was also found to avoid fouling.	[236]

Table 11. *Cont.*

Water Type	Materials	Method	Study Characteristics	Reference
Deionized Water	Nano zerovalent iron-kaolin clay (nZVI-Kaol)	Membrane filtration	Membranes were prepared with nZVI-Kaol clay in different compositions and tested for As(III) removal from water. Greater than 50% removal efficiency for As_2O_3 was achieved.	[237]
Freshwater	Chelating polymer	Membrane distillation (MD), photocatalytic oxidation and polymer-enhanced ultrafiltration (PEUF)	MD deployed with an efficiency of 98.8% from an initial contraction of 0.059–5 mg/L. In addition, MD retentate was applied with the photocatalytic process for the oxidation of As(III) to As(V), which was subsequently removed by PEUF with 98.2% efficiency.	[238]
Deionized Water	Nano-aluminium oxide and polyphenylsulfone (PPSU)	Membrane filtration	Ultra-membrane containing nano-aluminium oxide with PPSU prepared. A membrane fabricated with 1.5% w/w e nano-aluminium oxide in PPSU removed 98.41% of As(V).	[239]
Deionized Water	Hydrous manganese oxide (HMO) nano clay and polyacrylonitrile (PAN) nanofibers	Electrospinning technique (ET) and membrane filtration	A membrane was fabricated with ET by impregnating HMO into PAN forming nanofiber membrane. The novel absorbent showed high efficiency for As(V) uptake and was found reusable after cycles.	[240]
Synthetic wastewater	Microporous polypropylene fibres.	Membrane filtration	A hollow fibre-supported liquid membrane (HFSLM) was used to mitigate As ions from water. The results demonstrated that mitigation can be conducted up to the discharge limit of wastewater standards of Thailand (250 ppb).	[241]
Deionized Water	Micro tetravalent manganese feroxyhyte (µTMF) and micro granular ferric hydroxide (µGFH).	Adsorption and membrane filtration	µTMF and µGFH, by-products of manufacturing granular absorbents, were tested with hybrid submerged microfiltration membrane adsorption and adsorption kinetic method. µTMF is found better in mitigating As(V) from modelled water with shorter residence time (~3 h) and greater efficiency (1.4-fold) compared to µGFH.	[242]
Tap water	Hydrophilic nickel-carbon nanotubes.	Membrane filtration	Cathode penitential using hydrophilic nickel–carbon nanotubes deployed to enhance the performance of UF membranes to mitigate As(III), which has lower removal efficiency when mitigated in water by NF and RO. A significant increase in As(III) rejection in water is monitored.	[243]
Surface water	NaOCl	Membrane filtration	Membrane fouling is a significant challenge in the wastewater industry. The RO membrane is recycled by oxidation and converted into a UF membrane. Application of pe-oxidation to feed was also made, and As was later removed by UF. Economic and environmental benefits are highlighted.	[244]

Water Type	Materials	Method	Study Characteristics	Reference
Waste-water	Polycaprolactone and iron- intercalated montmorillonite filler.	Membrane filtration and adsorption	In the batch scale study, a membrane fabricated with polycaprolactone matrix and iron-intercalated montmorillonite filler was studied for As removal and found that the membrane is a promising alternative for As removal from wastewater.	[245]
Surface water	Zeeweed UF membrane by Suez Water Technologies and Solutions.	Oxidation, coagulation, flocculation, sedimentation and membrane filtration	Dead-end UF incorporated with conventional water treatment systems comprising oxidation, coagulation, flocculation and sedimentation systems. The threshold value for As in water was achieved.	[246]
Deionized water	μTMF and μGFH	Adsorption and membrane filtration	MF membrane was modified by depositing iron oxide adsorptive materials on the membrane. The membrane was tested by application study and mathematical modelling on water, and this procedure may increase the life of the membrane filtration cycle for As(V) removal with other advantages.	[247]
Geothermal water	Commercial membranes; CK- NF, XLE BWRO, Osmonics and NF90 and spiral wound membranes; BW30, TR-BE-BW and TR-NE90-NF	Membrane filtration	The various membranes of NF and RO were tested for the removal of As from geothermal water on a mini-pilot scale (membranes; BW30, TR-BE-BW and TR-NE90-NF) and cross-flow flat-sheet membrane testing unit (commercial membranes; CK-NF, XLE BWRO, Osmonics and NF90). Maximum As removal efficiency of 99% with XLE BWRO commercial membrane was achieved. Removal efficiency of 90% was performed on all membranes tested on the mini-pilot scale.	[248]
Deionized water	Zinc-magnesium oxides and cellulose	Membrane filtration	Hollow membranes with polyphenylsulfone/cellulose acetate derivatives and zinc–magnesium oxide combinations were fabricated and tested. Membrane prepared with novel binary effect showed increase As retention efficiency.	[249]
Deionized water	Chitosan and Polypropylene	Membrane filtration	A total of 75% removal efficiency was achieved when tested for removal of As(V) after the fabrication of membrane from newly synthesised material (modified polypropylene with chitosan).	[250]

6.4. Adsorption

Adsorption methods are a popular process for As mitigation. In a simplified approach, water is passed from a bed packed with sorbent filled in a container [251–253]. Adsorption involves van der Waals forces and electrostatics. The method involves the separation of As in phase one and then the accumulation of [As] on the sorbent material surface in phase two, Figure 9 [246].
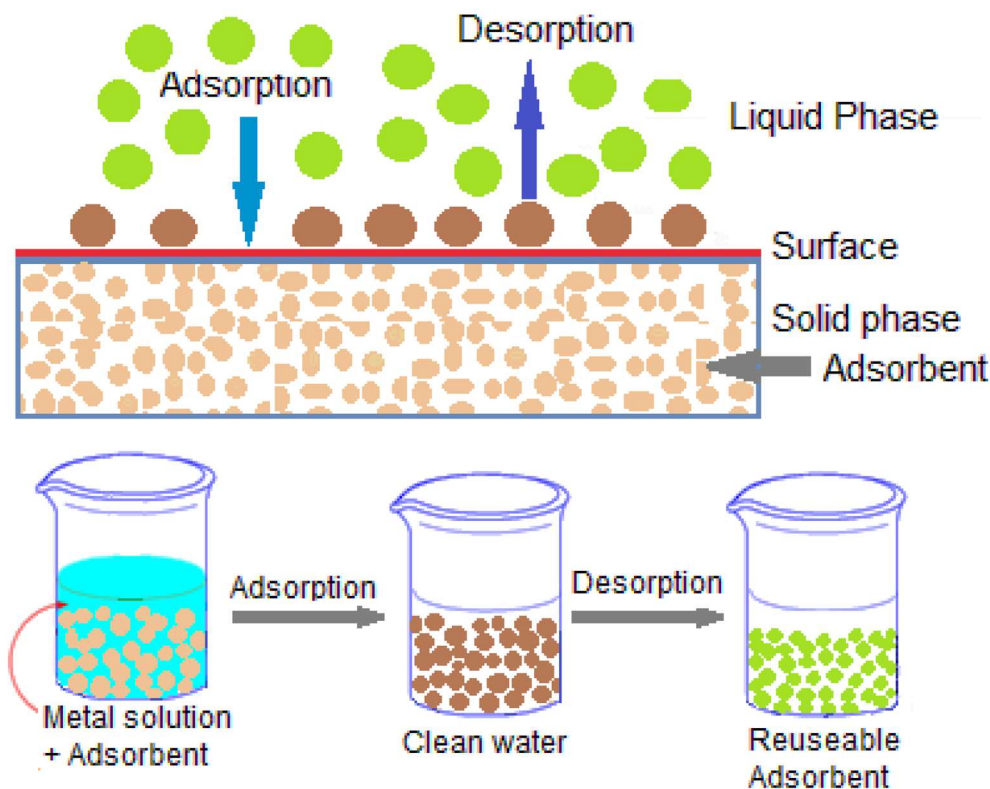
Figure 9. Schematic describing the adsorption process, adapted with permission from [253], publisher MDPI Publications.

One benefit of adsorption is the selective mitigation of As in water without disturbing other quantitative parameters. In addition, there is no sludge generation, and it is simple and easy to implement; moreover, it is suitable for point-of-use scale applications. As speciation, temperature, pH, and ionic strength are the properties of the solution that impact the process, whereas properties of adsorbents such as surface area, particle size, pore size/shape, zeta potential, etc., are important. The process of mitigating As by adsorption depends on these absorbent characteristics [169,232].

For the definition of adsorbent replacement frequency, column size for adsorbent containment and loading/dose rate, the main design parameter is termed 'empty bed contact time (EBCT)', which is 5–6 min in range for As mitigation in water by adsorption [232,254]. Adsorption of As(V, III) from water is mainly a function of pH and the concentration of the chemical system. Generally, when pH ranges between 1 and 4, As(V) is adsorbed by sorbent, whereas in the range of pH between 4 and 9, As(III) is adsorbed [168,255]. Phosphate and silicate, when in ionic form, compete with arsenate and arsenite for the adsorption material. The performance of the adsorption materials is mainly limited by factors such as difficulties in the regeneration of sorbent material, non-regularity in the pore structure of sorbent, chemical reactions of other metal ions with sorbents and small surface area [256–258].

There are various materials which are used as sorbents for [As] from water, such as aluminium-based sorbents, iron-based sorbents, nanomaterial sorbents, magnetic sorbents, bio-based sorbents, polymer-based sorbents, ash-based sorbents, hybrid sorbents and miscellaneous sorbents, and specific examples are montmorillonite [259], red mud [260], activated carbon [261], $Fe(OH)_2$ [262], TiO_2 [263], Al_2O_3 (activated) [264], ion-exchange polymers [265], chitosan [266], $Al_2Si_2O_5(OH)_4$ [267] and many more [268,269]. A recent review classified the sorbents in detail in a contemporary style, as shown in Figure 10.
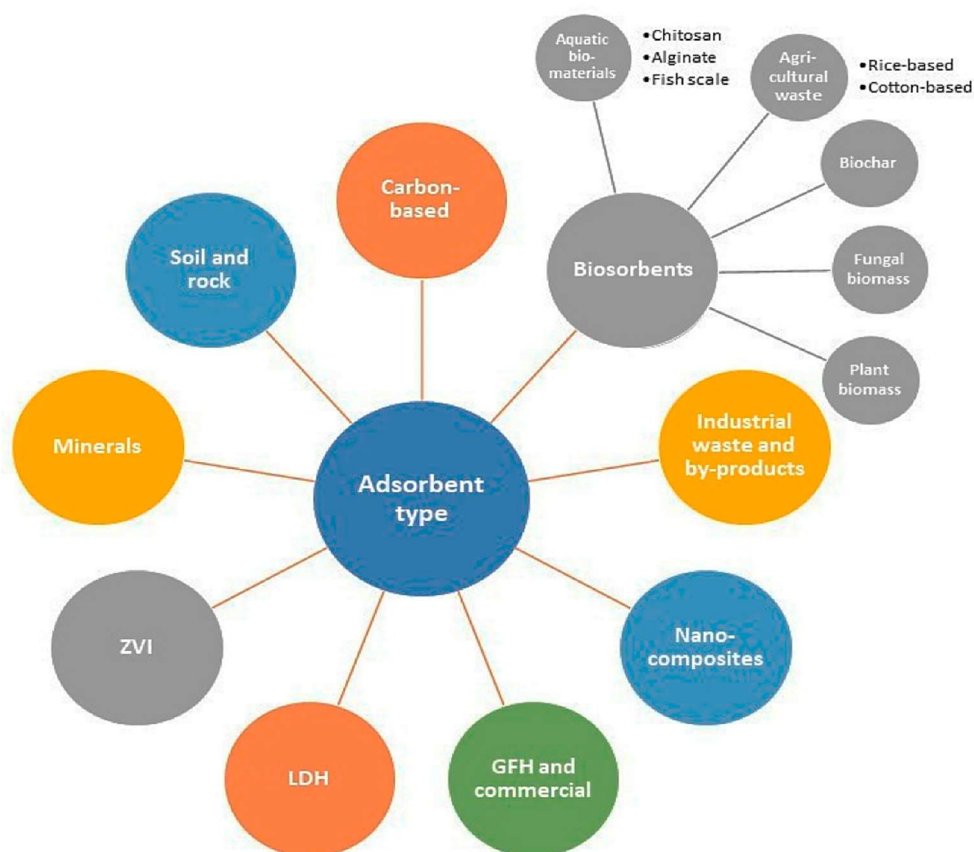
Figure 10. Classification of sorbents adapted with permission from [269], publisher MDPI. ZVI = zero valent ion; LDH = layered double hydroxides and GFH = granular ferric hydroxide.

Iron-based sorbents are popular due to their low cost and comparatively large surface area to attract inorganic species of As to form bonds with iron. In addition, iron can remove As from water by reducing As(III) to As(V) as an oxidizing agent, by containing and immobilizing the [As] species and by co-precipitation along with adsorption [270].

Various nanomaterials have been developed as novel adsorbents for contaminants, especially heavy metalloids, including [As]. They have attracted significant attention from the environmental community due to their large surface areas, high reactivity, and high specificity. The most widely used and researched nanoparticles for the treatment of arsenic-contaminated water are carbon nanotubes and nanocomposites, titanium and iron nanoparticles, and iron-based nanoparticles [168,271]. Nanocrystalline TiO_2 has a higher adsorption capacity, possibly due to a larger surface area than nonporous TiO_2 [168,272]. Nanomaterials based on iron, including zero-valent iron nanoparticles (nZVI) and iron oxide nanoparticles (Fe_3O_4 and Fe_2O_3), are highly relevant for treating arsenic-contaminated water. Iron-based nanoparticles' ability to remove contaminants is heavily influenced by the oxidation state of iron; however, they are cheap and effective in use [273,274]. Ceria nanoparticle adsorption is pH-dependent when mitigating As from water [275]. Zirconium oxide nanoparticles are chemically stable, non-toxic, and insoluble, making this variant an excellent option in the nanomaterial family for drinking water purification and As removal from water [276]. Biosorbents are proven promising media for adsorption with high adsorption capacity. They are prepared from natural material extracted from animals, plants, or waste biomass and are relatively low cost. The sorbents found to be effective for the uptake of heavy metals/metalloids are generally from the groups of biomasses such as biopolymers from aquatic, biomass from plants, biomass from microorganisms, biochar and biomass from agricultural waste and can be used as sorbents without modification or with pre-treatment [269,277–279]. One of the most popular is chitosan (CS) which originates

from arthropods' exoskeletons as a natural aminopolysaccharide. A free amino group that binds with heavy metal/metalloid ions makes it applicable for the uptake of [As] and other metals. Other advantages of CS sorbents are renewability, biodegradability, low cost and natural non-toxicity [280,281]. Another important biosorbent is alginate, a biopolymer that can be extracted from sea tangle and seaweed cell walls. The biopolymer is made up of β-D-mannuronate and α-L-guluronate. Divalent cations react with acids of alginate to form hydrophobic gels and are thermally irreversible [269,282]. Another material from nature carrying carbon as an adsorption tool is biochar, a black-coloured product developed from various biomasses. The characteristics of the material, such as its low cost, non-toxicity and large surface area, amorphous structure and functionalized surface with chemical groups to offer affinity to heavy metals/metalloids ions, make biochar an attractive option for further research and development. Biochar has been used in composites with other supporting elements [283,284]. Waste from agricultural activity mainly comprises hydrocarbons, water, hemicellulose, lignin, lipids, etc., which has also been evaluated as a potential source of sorbents for the uptake of (primarily) heavy metals/metalloids including As [285]. Other biosorbents such as codfish scale, rice polish, cotton-based adsorbent, fungal biomass and plant biomass have also been tested under various conditions for effectiveness and reasonable results are achieved and have paved the path for further improvement and research in the areas [286–290]. In Table 12, a review of some latest improvements claimed and tested regarding the adsorption of As from water is presented. The adsorption technique seems to be the method of choice, outperforming other methods such as membrane filtration and precipitation. Conventional adsorption processes can be enhanced by principally modifying the sorbents achieving removal efficacies of up to 100%. The modified sorbents can be regenerated, contributing to a circular economy and have shown tremendous potential with increased reuse cycles. Furthermore, the method of adsorption is sludge free, reducing the chances of secondary pollution from the treatment process. There is a drive to identify materials that are locally sourced and cost-effective for the preparation of sorbents and methods; therefore, the potential of point-of-use scale usage of the technique increases, which will be preferable in developing countries keeping in view the decentralised water supply and treatment systems.

Table 12. Case studies using adsorption process for arsenic mitigation from water.

Water Type	Materials	Method	Study Characteristics	Reference
Groundwater	Chitosan and titanium oxysulfate (TiOSO$_4$)	Adsorption	Using the sol-gel method, a novel granular adsorbent is fabricated with titanium and chitosan, which is reasonably amorphous for adsorption As(V). Small-scale column tests results reveal that at pH range 8.0–8.5, 165.6 µg/L of initial concentration was brought down to <10 µg/L at 126-bed volume with adsorption capacity of novel adsorbent around 1.22 mg/g at effluent equilibrium (1370 BV). The adsorbent is found to have good prospects compared to other adsorbents, with a capacity of 70% after four cycles.	[291]
Ultrapure water	Chitosan, graphene oxide and polyethyleneimine	Adsorption	Combining chitosan, graphene oxide and polyethyleneimine, a new magnetic novel absorbent formed abbreviated as MCS/GO-PEI. A Langmuir isotherm model showed that the new material was a single-layer absorbent. Adsorption equilibrium was achieved in 8 min for As at pH 7. The adsorbent has shown reasonable characteristics for reusability.	[292]

Water Type	Materials	Method	Study Characteristics	Reference
Deionized water.	Iron-manganese treatment sludge and chitosan	Adsorption	Iron-manganese treatment by-product (backwashing sludge) in powered form converted to granular with the combination of chitosan. Langmuir model gave an adsorption capacity of 14.95 mg/g for As(V), followed by a filtration test in the plexiglass column. With a bed depth of 15 cm, total sorption (10.755 mg) and unit sorption capacity (0.779 mg/g) were calculated. Compared with when bed depth was 60 cm, the total and unit adsorption was calculated as 20.405 mg and 0.370 mg/g, respectively. At a bed height of 30 cm, EBCT is given 9 min and a breakthrough time of 13 h. After regeneration of the material for adsorption, a removal efficiency of 96% for As(V) also showed potential for reuse.	[293]
Groundwater	Chitosan and N-vinylcaprolactam/N-N-dimethylacrylamide (net-CS)-g-NVCL/DMAAm hydrogels	Adsorption	A new novel material for adsorption of As from groundwater (pH range 7–8.3) formed with a grating of hydrogel on chitosan. Evaluation by isotherms and kinetics sorption tests was conducted. An adsorption capacity of 0.0022 mg/g was measured on the new sorbent after 50 h. Removal efficiency of 40% (isotherms) and 46% (kinetics sorption test) for As was achieved.	[294]
Ultra-pure water	Chitosan and 3-mercaptopropyl trimethoxysilane ($C_6H_{16}O_3SSi$)	Adsorption	With the reaction of $C_6H_{16}O_3SSi$ and chitosan, a new chitosan-based material was formed. Removal efficiency for As(III) of 99% was achieved from the initial concentration of 10 mg/L at an extensive pH range (3–10). A maximum adsorption capacity of 21.01 mg/g is monitored at pH 7. With oxalic acid, the material can be regenerated with 80% [As] removal efficiency in 2 cycles.	[295]
Deionized water.	Coal fly ash, $ZrOCl_2$ and $FeCl_3 \cdot 6H_2O$	Adsorption	Zeolite W (ZW) (fly ash-based) was modified using microwaves, with the resultant material having iron and zirconium at 6.9 and 5.04% *w/w*, respectively. The modified material tested for adsorption of As(V) and achieved 99.87% removal efficiency at pH from 2 to 10. Maximum adsorption of 42.31 mg/g was attained at pH 2.	[296]
Double distilled water	Calcium alginate, Iron Oxide Magnetic Nanoparticles and methionine	Adsorption	Reacting calcium alginate with magnetic iron oxide nanoparticles functionalized with methionine formed a new bead absorbent. Laboratory-based tests confirmed that novel material can remove As(III) up to 99.56% at an equilibrium time of 1.83 h and pH 7.0–7.5. Langmuir isotherm calculation provided an adsorption capacity (qm) of 6.6533 mg/g.	[297]
Deionized water.	Seeds skin extract of Sapindus, CeO_2-ZrO_2 and Iron alginate	Adsorption	Nanooxides of CeO_2-ZrO_2 (mixed) formed. As stabilizing agent to control the size of nano mixed oxides, Sapindus plant seeds' skin was extracted. These nano oxide particles are embedded in iron alginate to prepare beads. After the test of adsorption, the results gave an adsorption capacity of 140.5 mg/g and 153.25 mg/g for As(V) and As(III), respectively.	[298]
Wastewater	Waste red mud and calcium-alginate	Adsorption	Beads of novel material formed by doping red mud with calcium alginate. The material beads are tested to remove As(III) ions from wastewater and achieved results by reducing the concentration in effluent from 0.101 mg/L to 0.008 mg/L. A total of 1.807 mg/g is found to be peak adsorption capacity. The material has shown the potential to be used for the removal of As from water and wastewater.	[299]
Livestock farm wastewater	Yttrium (III) chloride hexahydrate, graphene oxide and alginate hydrogel	Adsorption	A new novel hydrogel was prepared by reacting graphene oxide, yttrium (III) chloride and alginate hydrogel and was tested by an adsorption experiment under room temperature. Maximum adsorption of As(V) was calculated as 273.39 mg/g, which is reported to be far higher than other absorbents available.	[300]

Water Type	Materials	Method	Study Characteristics	Reference
Distilled water	Pectin, Glutaraldehyde, *N,N*-methylene bisacrylamide, *N,N,N, N*-tetramethyl ethylenediamine and EGDMA	Adsorption	The pectin-based hydrogel was prepared, loaded with Fe(II) and tested to remove As(V) from water. Hydrogel was designed by crosslinking pectin in the presence of crosslinkers and 2-acrylamido-2-methylpropanesulphonic acid. Maximum uptake of As(V) was achieved at 35 °C temperature and 7.0–9.2 pH with 5% glutaraldehyde crosslinked hydrogels.	[301]
Distilled water.	Solid particulate waste collagen and APPN-g-pentapolymer (apple-based pectin) hydrogels	Adsorption	Collagenic fibres from tannery waste are incorporated into apple pomace pectin to form pectin-grafted penta-polymers created by one-pot facile polymerization of APPN and synthetic monomers. These polymers were tested for adsorption of As(V) and achieved 180.47 mg/g of As adsorption at three pH with a 25 mg dose of polymer.	[302]
Distilled water.	Waste orange juice residue	Adsorption	Pectin, cellulose and hemicellulose from waste orange juice were converted into acidic polysaccharides (pectic acid)/hydrogels by saponification and phosphorylation. Gels produced showed a strong affinity for iron ions and absorbed other metal ions (oxo-anions) such as As.	[303]
Deionized water	Steam-exploded straw and $CeCl_3$	Adsorption	Ce^{3+}-based biochar-loaded nanoparticles formed as a novel adsorbent for the adsorption of As. The material was tested using spectroscopy detection technologies and batch experiments. The Langmuir model showed a sorption capacity of 219.8 mg/g for As(V) at room temperature with 5 pH. Therefore, a promising new material from As(V) absorption was invented.	[304]
Water	Phosphorene-based materials—Theoretical study	Adsorption	Calculation based upon density functional theory was performed to evaluate the nanoflakes' phosphorene oxide (PO) ability to mitigate methylarsenicals from polluted water sources by using solid phase adsorption. The study was conducted to understand PO's adsorption characteristics for peril methylarsenicals, providing a valued framework for future development. The study suggests PO is suitable for the function as it has a large surface area, no pre-oxidation required for the process, high adsorption capacity and easy recovery.	[305]
Deionized water.	Dry sewage sludge and nano-zero-valent iron	Adsorption	Sewage sludge and nano-zero-valent iron passed through the co-pyrolysis process to prepare biochar. Biochar was tested for As(V) adsorption with batch experiments with initial pH and concentration as 2 and 20 mg/L, respectively. The biochar dose was 10 g/L, at a temperature of 298 K and contact time of 24 h, As(V) removal efficiency was given as 99%. With the Freundlich model, the adsorption capacity (max) is calculated as 60.61 mg/g for As(V).	[306]
Deionized water	SiO_2 and titanium chloride ($TiCl_3$)	Adsorption	Two nanomaterials, aggregated dendritic anatase-titanium dioxide and aggregated dendritic anatase-titanium dioxide, reacted to the surface of SiO_2 and were evaluated. As removal performance by both materials was tested under different conditions by laboratory batch experiments. The maximum removal efficiency of 95% was given by aggregated nanomaterial.	[307]
Deionized water	Attapulgite, sodium thiosulfate ($Na_2S_2O_3$) and Ferrous sulfate heptahydrate ($FeSO_4 \cdot 7H_2O$)	Adsorption	Nanoscale zero-valent iron supported by attapulgite and modified by sulfide to prepare a new nanomaterial for As(III) remediation in water. With support and modification, the specific surface area of nanomaterial increased to 46.04 m^2/g from 19.61 m^2/g. As a comparative result, the removal efficiency of As(III) increased to 65.1% from 51.4% at 20 min and 51.4% to 65.1% at 20 min. The increase in performance of nanomaterial is seen at a pH range of 3.0–6.0. A total of 193.8 mg/g is given as adsorption capacity (max).	[308]

Water Type	Materials	Method	Study Characteristics	Reference
Deionized water	Milli-Q water	Adsorption and photo-oxidation.	Composite oxide of nanostructure properties of iron, titanium, and manganese was prepared to treat the para-arsanilic acid. A total of 424.7 m^2/g is calculated as the specific area of the new material. Adsorption capacity (maximum) is given as 45.6 mg/g. In addition to adsorption, the photo-oxidation of the pollutant from As(III) to As(V) is also performed in the system to achieve better results.	[309]
Ultrapure water	Ferric chloride, Ferrous chloride and Copper chloride	Adsorption and photo oxidation.	Magnetised CuO-Fe_3O_4 material deployed for photo-oxidation of p-arsanilic acid with visible light irradiation and subsequent removal by adsorption. The As is seen completely oxidized from As(III) to As(V) in 36 min, and adsorption by nano-CuO-Fe_3O_4 is also observed with efficiency of 95% at a pH range of 4 to 7.	[310]
Ultrapure water	Iron sludge	Adsorption	A novel nanomaterial prepared by Fe_3O_4@C magnetic nanoparticles of amorphous iron oxides from iron sludge for the removal of As(V) from water. BET analysis calculated the surface area as 122.7 m^2/g. At a pH range of 3–9, the adsorption of As(V) is noted as high. The adsorption capacity (maximum) at room temperature is given as 13.47 mg/g, which was 3.29-fold higher than non-modified material (Fe_3O_4@C).	[311]
Deionized water	Fe $(NO_3)_3 \bullet 9H_2O$, $MnSO_4 \bullet H_2O$ and $CoSO_4 \bullet 7H_2O$	Adsorption	$CoFe_2O_4$ and $MnFe_2O_4$ nanoparticles are prepared with magnetic properties and tested to remove As. Freundlich model equation obeyed to show multilayer adsorption with adsorption capacity (maximum) as 230 mg/g for $MnFe_2O_4$ and 250 mg/g for $CoFe_2O_4$.	[312]
Milli-Q ultrapure water	Magnesium chloride hexahydrate, Aluminum chloride hexahydrate and Ferric chloride	Adsorption	Facile pyrolysis method was adopted to prepare four types of magnetic biochar, out of which one is prepared as magnetic biochar only, while the other three are anchored with Aluminium, manganese and both (Al and Mn) in addition. Upon testing, the optimal pH range was given as 4–6. Aluminium-based magnetic biochar outperformed all other comparisons for the co-up-take of As(V) and fluoride. From adsorption isotherm information, the adsorption capacity (maximum) is given as 34.45 mg/g for As(V) at a temperature of 10 °C and pH of 5.	[313]
Ultrapure water	Cerium oxide and polyhedrons	Adsorption	With the combination and application of calcination and impregnation, cerium oxide anchored on polyhedrons (from MIL-100(Fe)) to fabricate magnetic mesoporous nanocomposite was denoted as Fe_2O_3/CeO_2-t with a unique octahedral nanostructure. Fe_2O_3, a conventional absorbent, when modified on a surface with CeO_2 and tested as an absorbent of As(III), reduced the concentration to 10 μg/L from 180 μg/L in 20 min, which is 9-fold faster than the conventional comparison. By an isotherm method, the adsorption capacity (maximum) is 68.25 mg/g, which was higher than many similar absorbents.	[314]
Deionized water	Fly ash and ferrous sulfate heptahydrate	Adsorption	Zeolite 5A loaded on nanozero valent iron to form a new material. New material denoted as NZVI-5A and reported with a specific area of 238 m^2/g with a removal capacity of 72.09 mg/g from the isotherm model. pH range was reported as 4–12, and the method was not reported as pH sensitive. Removal efficiencies remain unaffected by the co-existence of other pollutants. NZVI-5A zeolite worked with 84% removal efficiency after 5 cycles.	[315]
Deionized water	Sodium alginate, Chitosan and banana peels	Adsorption	Two adsorbents were prepared by combining alginate and chitosan separately with banana peel ash and biopolymeric matrix ash. Both sorbents were characterised and tested for the removal of heavy metals. The material formed with alginate was proven to be an excellent absorbent for heavy metals with 100% efficiency.	[316]

Table 12. *Cont.*

Water Type	Materials	Method	Study Characteristics	Reference
Deionized water	Oyster shell powder and rice husk ash	Adsorption	Rice husk ash (TRHA) and oyster shell (OS) powder are used to prepare pallets for use as adsorbent for As(III) removal from water samples. A ratio of 0.3:0.7 = TRHA: OS has been reported as the best ratio for removal with 26.2 mg/g as adsorption capacity with breaking. Further analysis concluded that CaO reacted with As(III), which was 25% by weight of the pellet while the remaining 75% is CaSiO$_3$, which helps the pellet to keep in a stable position (avoiding cracking).	[317]
Bi-distilled water	Soluble starch, cellulose and iron chloride	Adsorption	Soluble starch and cellulose, used as carbon sources, and iron chloride, used as the source of iron, were deployed to make a few composite materials to test as sorbent for As(V) in water samples. The maximum adsorption capacity was measured as 280 μg As(V)/g of novel material.	[318]
Deionized water	Cobalt acetate, Aniline, Ferric chloride and Chitosan from shrimp	Adsorption	Magnetization and functionalization of biopolymer to prepare cobalt oxide doped magnetic-chitosan graphene oxide grafted with polyaniline. Take up of As(V) by new material tested by batch experiments and theoretical investigation. An adsorption capacity of 90.91 mg/g was given at 7 pH—the highest removal efficacy of 89% given at 50 min of interaction.	[319]
Deionized water	1-(2-Hydroxyethyl) piperazine, 3-cyanopropyltriethoxysilan, Sporopollenin and Ferric chloride	Adsorption	With 1-(2-hydroxyethyl) piperazine, the sporopollenin from Lycopodium clavatum was magnetized/functionalized. The novel bio-polymer-based sorbent was used to test in batch experiments for the adsorption of As(III) and Pb(II). Maximum adsorption of 69.85 mg/g was calculated for As(III).	[320]
Distilled water	Cetyltrimethylammonium bromide surfactant, tetraethylorthosilicate and β-cyclodextrin polymer	Adsorption	A combination of β-cyclodextrin polymer and mesoporous silica was performed to prepare a hybrid structure for the mitigation of As(V) and Hg (II). Maximum adsorption capacity calculated as 265.6 mg/g. As per data, Langmuir equilibrium is followed to attain a monolayered homogenous absorbent. Column test (fixed bed) achieved a removal efficiency of 72.8% for As (V).	[321]
Canal water	Arsenic, 2-hydroxyethyl methacrylate and 4-vinyl pyridine	Adsorption	As-imprinted polymer prepared by polymerization and precipitation using 2-hydroxyethyl methacrylate and 4-vinyl pyridine. The absorbent is tested for the adsorption of As(III) in aqueous environments. Adsorption capacity (maximum) calculated as 106.3 mg/g. A total of 99% removal efficiency achieved for removal of As(III) by the polymer as adsorbent.	[322]

6.5. Social Mitigation

Secondary preventions from As in water are paramount in preventing As exposure in water. Studies have suggested that technological measures taken to mitigate the vulnerability of As to human health from As-polluted water alone is not enough and does not reduce the risk to safe levels [323,324]. There are scientifically based suggestions that even after cessation of exposure to As, the risk of exposure may not be reduced for decades. Studies on the health impact (cancer) over long-term periods (50 years and 20 years) are presented [325–327]. Hence, based on evidence, it is critical to suggest that it may be important to consider other potential strategies in combination with technical mitigation of As exposure. Various methods that involve different fields and sectors will be required for long-lasting, cost-effective solutions. This might include changing water sources, adjusting how we cook and irrigate, setting up community water filtration plants, and making new policies to reduce the danger [324,328].

Information, testing and awareness are critical and paramount steps in mitigating As through secondary prevention measures. People may take water from polluted wells if the information is not provided, a test has not been conducted, or awareness of harmful effects is not clearly understood. Free testing services for any water source should be available

with field kits to gain the information required for choosing the right water source. A simple test costs around 2.30 USD, with the cost of supplies at 0.30 USD per test [325]. Awareness and information campaign designs are essential as studies have reported that with information and awareness, ½ to ¼ of the exposed population will stop using the affected well/source of water [329]. Introducing a policy to subsidise testing kits may help people understand the value of testing and may increase the demand for the kits [330]. Switching of water sources from contaminated to cleaner sources may only happen if the people know the harmful health consequences of As first. Policy intervention that joins the awareness and test campaigns has been noted with increased popularity [331].

Combined with testing, sourcing alternative sources of water such as surface water/rain harvesting, dug wells, tube wells (very shallow shrouded), infiltration galleries, tube wells (baritone) and tube wells (shallow shrouded) are an effective strategy [328]. The health risk substitution may properly be analysed before switching. Health risks are associated with all types of water supplies—substituting danger while reducing one may come up in greater magnitude. Pathogens and cyanobacteria-derived toxins in surface water and pollution from chemical containments are major health risk substitutes which may be faced and need to be comparatively assessed before the substitution of a water source [324]. Rice is the primary and cheap source of food/carbohydrate for the majority of the world's population and resultantly becomes the staple reason of contribution of As in comparison with other food sources. Growing rice requires irrigation by water more than many other crops. Irrigation by As-polluted groundwater poses a significant threat to the food chain. It is suggested that bioaccumulation of As in rice may be reduced by changing the agronomic techniques. A study suggests that changing the irrigation system of paddy fields from flood irrigation to sprinkler irrigation systems reduced the accumulation of As in rice. In addition, some changes in cooking practices may also reduce the risk of As exposure, as it is observed that washing the grains before cooking and then cooking in excess water can contribute to a decrease in exposure of As [328]. Centralised and community-based treatment systems are more attractive alternatives to household-small-scale mitigation of As strategies. Such systems may supply water free from As to homes ranging from 100 to 200 families. Currently, available systems may produce a water supply of 1 million liters before replacement of media/membrane/equipment. Monitoring water quality in community-based plants is easier compared to small-scale plants. Such systems also have the advantage of disposing of the by-products from the filtration plants, reducing the risk of secondary pollution. However, concerns may arise concerning social behaviours, cost and maintenance requirements which could impact sustainability and long-term viability. Hence, the policy intervention and government system mechanism may step in for sustainable utility service delivery [325]. As the scale of the policy implementation increases, such as from a town to the national level, the complexities also increase. For example, spillover effects of the policy interventions on communities may change the prices of equipment and services in the market, which may impact the policy intervention's effectiveness. Therefore, research on the effects in specific areas/countries may make the information available for policymakers on unseen costs and benefits, which cannot be noted when policy is implemented on a small scale [325–328].

7. Conclusions

In conclusion, this review has provided evidence that the pervasive issue of arsenic contamination in water is a significant global health challenge, with detrimental impacts ranging from organ failure to various cancers. Recognising the global scale of arsenic enrichment, numerous countries have taken stringent measures to lower permissible concentrations in drinking water. The complexity of arsenic's chemical behaviour, influenced by factors such as pH and Eh, necessitates ongoing research to understand its mobility and reactivity in natural settings. Current detection and quantification methods for arsenic, while advanced, underscore the need for accurate speciation to address the varied toxicity of different arsenic compounds. Innovations in analytical techniques have enhanced

our ability to monitor arsenic more precisely, contributing to more effective water quality management. Traditional treatment methods, including oxidation and coagulation, have evolved with advancements in material science, improving efficiency and efficacy. Membrane filtration technologies and novel materials have shown promise in achieving better arsenic removal while minimising environmental impact. Adsorption, as a preferred mitigation method, continues to be refined with the development of new sorbents that offer high removal efficiencies, cost-effectiveness, and low environmental footprints. The exploration of biosorbents and other innovative materials holds potential for further advancements in arsenic remediation technologies. To address the complex challenges of arsenic contamination, a multifaceted approach that includes trans-disciplinary and multi-sectoral strategies is essential. Emphasis on sustainable, zero-carbon emission technologies and materials will be critical in developing effective solutions. The goal is to achieve maximum removal efficiency while ensuring that methods are environmentally benign, energy-efficient, and economically viable. Future efforts are needed, especially in the direction of development of sustainable green material-based technologies, and due focus should be given to integrating these advanced technologies into practical applications, supported by robust policy frameworks and community engagement. By prioritising sustainable innovation and comprehensive arsenic management strategies, we can make significant strides towards safeguarding public health and ensuring access to safe drinking water globally.

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