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Arsenic in aquatic and sedimentary environments: Speciation, mobility, analytical advances, and implications for risk-based monitoring

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ABSTRACT

Arsenic contamination remains a major environmental and public-health concern due to its widespread occurrence, persistence, toxicity, and complex behavior in aquatic and sedimentary environments. This review synthesizes current knowledge on the occurrence, sources, speciation, biogeochemical transformation, mobility, toxicity, analytical determination, and risk-based monitoring of arsenic in water, sediments, porewater, and related environmental matrices. Particular emphasis is placed on the environmental significance of arsenic speciation, highlighting the contrasting behavior of arsenite [As(III)] and arsenate [As(V)] under varying redox and pH conditions. The review demonstrates that arsenic mobility and bioavailability are strongly regulated by sediment–water interactions involving iron and manganese oxyhydroxides, sulfide minerals, organic matter, microbial processes, and porewater dynamics. These interactions control arsenic retention, transformation, and secondary release, allowing sediments to function as both long-term sinks and potential contamination sources. Advances in analytical chemistry, particularly ICP-MS and HPLC-ICP-MS, have significantly improved the determination of total arsenic and species-specific characterization; however, challenges associated with species preservation, extraction efficiency, matrix interference, and quality assurance remain critical. The review further emphasizes that environmental risk cannot be adequately assessed using total arsenic concentrations alone. Instead, effective monitoring should integrate arsenic speciation, geochemical conditions, sediment–water processes, exposure pathways, and analytical reliability. Such an integrated, species-specific, and risk-based framework provides a stronger basis for understanding arsenic behavior, predicting environmental impacts, and supporting sustainable management of contaminated aquatic systems.

Keywords: arsenic speciation, aquatic environment, sediment-water interface, redox transformation, analytical determination, risk-based monitoring.

INTRODUCTION

Arsenic contamination remains one of the most persistent and complex environmental problems affecting aquatic and sedimentary systems worldwide. Its importance is not only related to its toxicity, but also to its widespread occurrence, diverse sources, long-term persistence, and strong dependence on geochemical conditions (Wang et al., 2022; Yadav et al., 2023). Arsenic can enter the environment through natural processes such as mineral weathering (Garelick et al., 2009), geothermal activity (Webster and Nordstrom, 2003), volcanic emissions (Kar, 2022), and reductive dissolution of arsenic-bearing minerals (Masuda, 2018), as well as through anthropogenic activities including mining, smelting, coal combustion, pesticide use, wood preservation, and industrial

discharge (Hanh et al., 2010; Patel et al., 2023). In aquatic environments, arsenic contamination is of particular concern because groundwater and surface water can act as direct exposure pathways for humans and ecosystems (Sharma and Sohn, 2009; Thakur et al., 2023). The World Health Organization recommends a provisional guideline value of 10 μg L^{-1} for arsenic in drinking water (Frisbie and Mitchell, 2022), reflecting the significant health risks associated with chronic exposure and the practical difficulty of reducing arsenic to very low levels in contaminated water supplies. However, environmental risk cannot be adequately evaluated using total arsenic concentration alone, because arsenic toxicity, mobility, bioavailability, and treatability are strongly controlled by its chemical form and environmental transformation behavior (Akter et al., 2005; Sharma and Sohn, 2009)

Recent studies and review articles have increasingly emphasized that arsenic speciation is central to understanding its environmental fate, mobility, bioavailability, toxicity, and associated environmental risks, highlighting the limitations of relying solely on total arsenic concentrations for environmental assessment (Ardini et al., 2020; Mukherjee et al., 2024; Patel et al., 2023). In natural waters, arsenic commonly occurs as inorganic arsenite As(III) and arsenate As(V), while methylated species such as monomethylarsonic acid and dimethylarsinic acid may occur in biologically active systems (Cullen and Reimer, 1989; Sharma and Sohn, 2009). As(III) is generally more mobile and toxic than As(V), particularly under reducing conditions where neutral arsenious acid species can dominate and interact weakly with mineral surfaces (Sharma and Sohn, 2009; Yang et al., 2015). In contrast, under oxic conditions, As(V) predominantly exists as negatively charged oxyanions ($H_2AsO_4^-$ and $HAsO_4^{2-}$) and is generally less mobile because it can be strongly adsorbed onto iron and aluminum (hydr)oxides, clay minerals, and other reactive mineral surfaces through inner-sphere surface complexation (Bowell et al., 2014; Dixit and Hering, 2003; Smedley and Kinniburgh, 2002). In sedimentary environments, arsenic mobility is further governed by redox gradients, pH variation, organic matter decomposition, microbial activity, porewater diffusion, sulfide formation, and the reductive dissolution or reprecipitation of iron and manganese oxyhydroxides (Barral-Fraga et al., 2020; Lloyd and Oremland, 2006; Nicholas et al., 2003). These coupled processes make sediments both sinks and secondary sources of arsenic, depending on the prevailing geochemical conditions (Acquavita et al., 2021; Liu et al., 2023). At the same time, analytical developments, particularly hyphenated techniques such as HPLC-ICP-MS and LC-ICP-MS, have enabled more accurate species-specific determination of arsenic in environmental and biological matrices (Acquavita et al., 2021; Viana et al., 2023). Emerging approaches, including field-based separation, microfluidic platforms, and rapid inorganic arsenic speciation methods, further indicate that arsenic monitoring is moving from total concentration measurement toward species-resolved and risk-oriented assessment.

Despite substantial progress, several critical gaps remain in the current understanding and monitoring of arsenic in aquatic and sedimentary environments. Many previous reviews have treated arsenic occurrence, toxicity, sediment behavior, and analytical determination as separate topics, whereas these aspects are strongly interconnected in real environmental systems (Mukherjee et al., 2024; Sharma and Sohn, 2009; Smedley and Kinniburgh, 2002). Total arsenic measurements are still widely used in environmental assessment, even though they provide limited information on toxicity, mobility, bioavailability, and treatment feasibility (Acquavita et al., 2021; Sharma and Sohn, 2009). In addition, arsenic species are chemically unstable and can transform during sampling, storage, preservation, and analysis, leading to uncertainty in speciation data if field handling and quality assurance are not rigorously controlled (Acquavita et al., 2021). The role of sediment–water interactions is also frequently underestimated, although arsenic release from sediments can sustain contamination even after external inputs are reduced (Acquavita et al., 2021; Nicholas et al., 2003). Furthermore, the integration of geochemical data, speciation analysis, exposure pathways, and risk-based monitoring remains limited, particularly in regions affected by mining, geothermal activity, groundwater contamination, and industrial discharge (Thakur et al., 2023; Wang et al., 2022). Therefore, a more integrated review is needed to connect arsenic biogeochemistry, environmental mobility, analytical reliability, and risk interpretation within a single framework.

This review aims to critically examine arsenic in aquatic and sedimentary environments by integrating its sources, occurrence, speciation, mobility, toxicity, analytical determination, and implications for risk-based monitoring. Particular

attention is given to the transformation of As(III) and As(V) under variable pH and redox conditions, the role of iron and manganese oxyhydroxides, sulfide minerals, organic matter, and porewater processes in controlling arsenic retention and release, and the analytical challenges associated with total arsenic and species-specific determination. The review also evaluates current analytical advances and identifies key limitations in sampling, preservation, matrix interference, and field-based speciation. By linking arsenic speciation, sediment–water interactions, analytical reliability, and exposure relevance, this review provides a conceptual basis for improving arsenic monitoring strategies and supporting more accurate environmental risk assessment in contaminated aquatic systems.

ENVIRONMENTAL OCCURRENCE AND SOURCES OF ARSENIC

Arsenic is widely distributed in the environment and occurs in terrestrial, aquatic, atmospheric, and biological compartments. In natural systems, arsenic rarely occurs as a free element, but is commonly associated with sulfur, oxygen, iron, and other metals (Bissen and Frimmel, 2003; Wang et al., 2022). Its environmental occurrence is strongly linked to the weathering and transformation of arsenic-bearing minerals, particularly sulfide minerals and their secondary alteration products (Yadav et al., 2023). In the Earth's crust, arsenic occurs in a wide range of mineral phases, including arsenates, sulfides, sulfosalts, arsenides, arsenites, oxides, silicates, and elemental arsenic (Herath et al., 2016; Santini and Ward, 2018). Among these, arsenopyrite (FeAsS) is considered one of the most important arsenic-bearing minerals and commonly occurs in association with sulfide ore deposits containing metals such as copper, nickel, lead, cobalt, gold, and iron (Basu et al., 2014; Gao, 2024; Tomkins et al., 2006). The presence of arsenic in these mineral systems is environmentally significant because oxidation, dissolution, and redox transformation of sulfide minerals can release arsenic into surrounding soils, sediments, surface water, and groundwater (Hechavarría-Hernández et al., 2023; Wang et al., 2022)

In aquatic environments, arsenic is present mainly as oxyanion-forming species rather than as simple cations. This characteristic distinguishes arsenic from many trace metals and strongly affects its mobility, adsorption behavior, and removal efficiency (Bissen and Frimmel, 2003). The dominant inorganic forms in natural waters are arsenite As(III) and arsenate As(V), with their distribution controlled primarily by pH, redox potential, dissolved oxygen, microbial activity, and the presence of reactive mineral surfaces (Bissen and Frimmel, 2003; Wang et al., 2022). Under oxidizing conditions, As(V) generally predominates and occurs as arsenate oxyanions, which can be strongly adsorbed onto iron and aluminum oxides, clay minerals, and other positively charged surfaces (Campbell and Nordstrom, 2014; Hooda, 2010; Nidheesh et al., 2022). Under reducing conditions, As(III) becomes more stable and may occur as neutral arsenious acid species, particularly at circumneutral pH, making it more mobile and less readily removed by conventional adsorption or coprecipitation processes (Kumari et al., 2017; Wang et al., 2022). Therefore, the environmental risk of arsenic in water is not determined only by its total concentration, but also by the chemical species present and the geochemical conditions controlling their transformation (Bissen and Frimmel, 2003; Wang et al., 2022). Several arsenic-bearing minerals are environmentally important because they can act as primary sources or secondary sinks of arsenic depending on redox condition, pH, weathering intensity, and mineral stability (Henke, 2009; Kar, 2022). The most relevant arsenic-bearing minerals and their environmental significance are summarized in Table 1.

Sediments play a central role in regulating arsenic behavior in aquatic systems (Ramadan et al., 2021; Wibowo et al., 2026). They can function as long-term sinks when arsenic is adsorbed onto iron and manganese oxyhydroxides, incorporated into mineral phases, or precipitated with sulfides under reducing conditions (Xing et al., 2024). However, sediments can also act as secondary sources of arsenic when environmental conditions change. For example, reductive dissolution of iron oxyhydroxides can release previously adsorbed arsenic into porewater, while diffusion and resuspension processes may transport arsenic from sediments back into the overlying water (Bhattacharya et al., 2016; Zeng et al., 2023). In estuarine and marine systems, salinity gradients, organic matter decomposition, sulfate reduction, and sediment diagenesis further influence arsenic retention and release (Aftabtalab et al., 2022; Baeyens et al., 2019). The dual role of

Table 1. Environmentally relevant arsenic-bearing minerals

Mineral	Formula	Mineral group	Environmental relevance
Arsenopyrite	FeAsS	Sulfide	Major arsenic-bearing sulfide; important source under oxidation
Realgar	AsS	Sulfide	Occurs in hydrothermal and mineralized settings
Orpiment	As_2S_3	Sulfide	Can form or dissolve under sulfidic conditions
Arsenolite	As_2O_3	Oxide	Secondary oxidation product of arsenic minerals
Scorodite	$FeAsO_4 \cdot 2H_2O$	Arsenate	Important arsenic-bearing Fe mineral under oxidizing acidic conditions
Pharmacosiderite	$Fe_3(AsO_4)_2(OH)_3 \cdot 5H_2O$	Arsenate	Secondary product associated with oxidation of arsenic minerals

Note: Adapted and synthesized from several studies (Bowell et al., 2014; Jia and Demopoulos, 2005; Krause et al., 2019; Smedley and Kinniburgh, 2002; Vaughan, 2006)

sediments as both sinks and sources highlights the need to consider sediment–water interactions in arsenic monitoring and risk assessment.

Natural sources of arsenic include mineral weathering, volcanic activity, geothermal inputs, hydrothermal processes, microbial transformation, and reductive dissolution of arsenic-bearing iron minerals (Wang et al., 2022; Yadav et al., 2023). Groundwater contamination is often associated with natural geogenic processes, particularly in aquifer systems where arsenic-bearing sediments undergo redox-driven mobilization (Li et al., 2026; Tripathi et al., 2025). Two major mechanisms frequently proposed for groundwater arsenic enrichment are oxidation of arsenic-bearing sulfide minerals and reductive dissolution of iron oxyhydroxides (Herath et al., 2016; Thakur et al., 2023). The first mechanism involves the exposure and oxidation of arsenopyrite or arsenic-rich pyrite, resulting in arsenic release into groundwater. The second mechanism involves microbial reduction of iron oxides under anoxic conditions, leading to the release of both dissolved iron and associated arsenic (Herath et al., 2016; Jiang et al., 2014). In some systems, competitive desorption by phosphate and other anions may also contribute to arsenic mobilization, although its importance depends strongly on local geochemical conditions (Aftabtalab et al., 2022; Bissen and Frimmel, 2003).

Anthropogenic activities have substantially increased arsenic inputs into the environment. Mining, ore processing, copper smelting, coal combustion, pesticide application, wood preservation, glass production, semiconductor manufacturing, and disposal of arsenic-containing wastes are among the major human-derived sources (Baeyens et al., 2019; Hanh et al., 2010). In metallurgical industries, arsenic may be released during the roasting and smelting of sulfide ores, often as particulate-bound arsenic or volatile arsenic oxides (Hanh et al., 2010; Zhao et al., 2021). Coal combustion also contributes to atmospheric arsenic emissions, especially where coal contains elevated arsenic concentrations or emission-control systems are insufficient (Hanh et al., 2010; Senior et al., 2020). Agricultural activities historically contributed arsenic through the use of arsenic-based pesticides, herbicides, insecticides, and animal feed additives (Baeyens et al., 2019; Hanh et al., 2010). Wood preservatives such as chromated copper arsenate have also introduced arsenic into soils and surface environments through leaching, disposal, and weathering of treated materials (Morais et al., 2021). Arsenic inputs to aquatic and sedimentary environments originate from both natural and anthropogenic sources. These sources differ in their release mechanisms, dominant transport pathways, and affected environmental compartments, as summarized in Table 2.

The relative contribution of natural and anthropogenic sources varies across environmental settings. In mineralized and mining-affected regions, arsenic contamination is often associated with sulfide oxidation, acid drainage, tailings, waste rock, and metal-processing residues (Jiao et al., 2023). In deltaic and alluvial aquifers, arsenic enrichment is more commonly controlled by natural sedimentary processes, organic matter-driven reduction, and iron oxide dissolution (Kar et al., 2011; Quicksall et al., 2008). In industrial and urban areas, atmospheric deposition, fossil fuel combustion, and contaminated waste streams may dominate arsenic inputs (Fuge, 2013; Liu, 2025). Therefore, source identification requires

Table 2. Major natural and anthropogenic sources of arsenic in the environment

Source category	Examples	Main release pathway	Relevant environmental compartment
Natural geogenic sources	Arsenopyrite, realgar, orpiment, geothermal fluids	Weathering, dissolution, redox transformation	Groundwater, sediment, soil
Mining and ore processing	Sulfide mining, tailings, waste rock	Oxidation, acid drainage, particle transport	Surface water, sediment, groundwater
Smelting and metallurgy	Copper, lead, zinc smelting	Atmospheric emission, dust, slag, wastewater	Air, soil, sediment
Coal combustion	Power generation, industrial combustion	Fly ash, atmospheric deposition	Air, soil, water
Agriculture	Historical arsenical pesticides, herbicides	Runoff, soil accumulation, leaching	Soil, surface water, crop systems
Wood preservation	Chromated copper arsenate-treated wood	Leaching, disposal, weathering	Soil, runoff, sediment

Note: Compiled and synthesized from several related studies (Bowell et al., 2014; Mandal and Suzuki, 2002; Mukherjee et al., 2024; Nordstrom, 2002; Sharma and Sohn, 2009; Smedley and Kinniburgh, 2002).

integrated assessment of geological setting, land use, hydrology, sediment chemistry, and arsenic speciation. Such source-based understanding is essential because the same total arsenic concentration may represent different levels of mobility, bioavailability, and exposure risk depending on whether arsenic is present in oxidized, reduced, adsorbed, precipitated, or organically bound forms (Bissen and Frimmel, 2003; Wang et al., 2022).

Thus, arsenic occurrence in the environment reflects the interaction between mineralogy, redox chemistry, hydrological transport, sediment dynamics, and human activities. The persistence of arsenic contamination is largely due to its ability to shift among dissolved, adsorbed, precipitated, and biologically transformed forms. Consequently, effective arsenic assessment cannot rely solely on measuring total arsenic in water. Instead, it requires an integrated understanding of source characteristics, environmental compartments, geochemical controls, and species-specific behavior. This perspective provides the basis for discussing arsenic speciation and biogeochemical transformation in the following section.

ARSENIC SPECIATION AND BIOGEOCHEMICAL TRANSFORMATION

Arsenic speciation is a fundamental factor controlling the environmental behavior, toxicity, mobility, and treatability of arsenic in aquatic and sedimentary systems. Unlike many trace metals that commonly occur as dissolved cations, arsenic generally occurs as oxyanions or neutral oxyacid species in natural waters (Fendorf and Kocar, 2009; Wang et al., 2022). This chemical behavior makes arsenic highly sensitive to pH, redox potential, dissolved oxygen, mineral surfaces, organic matter, sulfide availability, and microbial activity (Cui and Jing, 2019; Fendorf and Kocar, 2009). The environmental significance of arsenic is therefore not adequately represented by total arsenic concentration alone. Instead, risk assessment and remediation planning require information on the specific arsenic species present and the geochemical conditions that regulate their transformation.

Arsenic can occur in several oxidation states, including −3, 0, +3, and +5. However, in most aquatic and sedimentary environments, the trivalent and pentavalent forms dominate (Wang et al., 2015). Trivalent arsenic, commonly referred to as arsenite As(III), and pentavalent arsenic, commonly referred to as arsenate As(V), are the principal inorganic species controlling arsenic behavior in water and sediment systems. These two forms differ markedly in charge, adsorption affinity, redox stability, toxicity, and removal efficiency (Fendorf and Kocar, 2009; Wang et al., 2022). In general, As(III) is more toxic and mobile than As(V), whereas As(V) is more readily retained by mineral surfaces, particularly iron and aluminum oxides, under many oxic and circumneutral conditions (Fan et al., 2015; Wang et al., 2022). This contrast explains why arsenic speciation is often more environmentally meaningful than total arsenic concentration. The distribution of arsenic species in water is strongly governed by pH and redox potential. Because arsenic speciation is strongly governed by pH and redox potential, a conceptual illustration of the predominance of

As(V) and As(III) species under contrasting environmental conditions is presented in Figure 1. Under oxidizing conditions, As(V) is thermodynamically favored and occurs mainly as arsenate species. Depending on pH, arsenate may exist as H_3AsO_4, $H_2AsO_4^-$, $HAsO_4^{2-}$, or AsO_4^{3-}.

In most natural waters, which typically have pH values between mildly acidic and mildly alkaline conditions, $H_2AsO_4^-$ and $HAsO_4^{2-}$ are the dominant arsenate species. These negatively charged species can interact strongly with positively charged mineral surfaces, particularly iron oxyhydroxides, through ligand exchange and inner-sphere complexation (Fendorf and Kocar, 2009; Cui and Jing, 2019). Consequently, As(V) is often less mobile than As(III) in oxic environments, although its mobility may increase when adsorption sites are saturated or when competing anions such as phosphate, silicate, bicarbonate, or natural organic ligands are abundant (Cui and Jing, 2019). The pH-dependent dissociation behavior of arsenic species is a key factor controlling their charge, adsorption affinity, mobility, and removal efficiency in natural waters. The dominant inorganic and methylated arsenic species and their environmental implications are summarized in Table 3.

Under reducing conditions, As(III) becomes more stable and is often present as arsenious acid species. At circumneutral pH, As(III) commonly exists as neutral H_3AsO_3, which has weaker electrostatic interaction with mineral surfaces than arsenate oxyanions (Wang et al., 2015; Fendorf and Kocar, 2009). This neutral form is one of the main reasons why As(III) is more difficult to remove by conventional adsorption and coprecipitation processes. As(III) can become particularly mobile in reducing groundwater, organic-rich sediments, wetlands, flooded soils, and anoxic porewater, where iron oxyhydroxides are reductively dissolved and previously adsorbed arsenic is released into solution (Nicholas et al., 2003; Fendorf and Kocar, 2009). Therefore, the conversion of As(V) to As(III) represents a critical pathway for arsenic mobilization in many natural and contaminated systems.

In addition to inorganic arsenic species, methylated arsenic species such as monomethylarsonic acid and dimethylarsinic acid may occur in biologically active environments. These species are produced through microbial and biological methylation processes and are commonly found in soils, sediments, aquatic organisms, and some biological matrices (Lee and Wen, 2019; Wang et al., 2022). Historically, arsenic methylation was often interpreted as a detoxification process because several methylated arsenic compounds are less acutely toxic than inorganic arsenic (Chen and Rosen, 2020; Vahter and Concha, 2001). However, this interpretation is not universal because some methylated intermediates may still exhibit biological reactivity and toxicological significance. Therefore, methylated arsenic species should not be ignored in environmental assessment, especially where

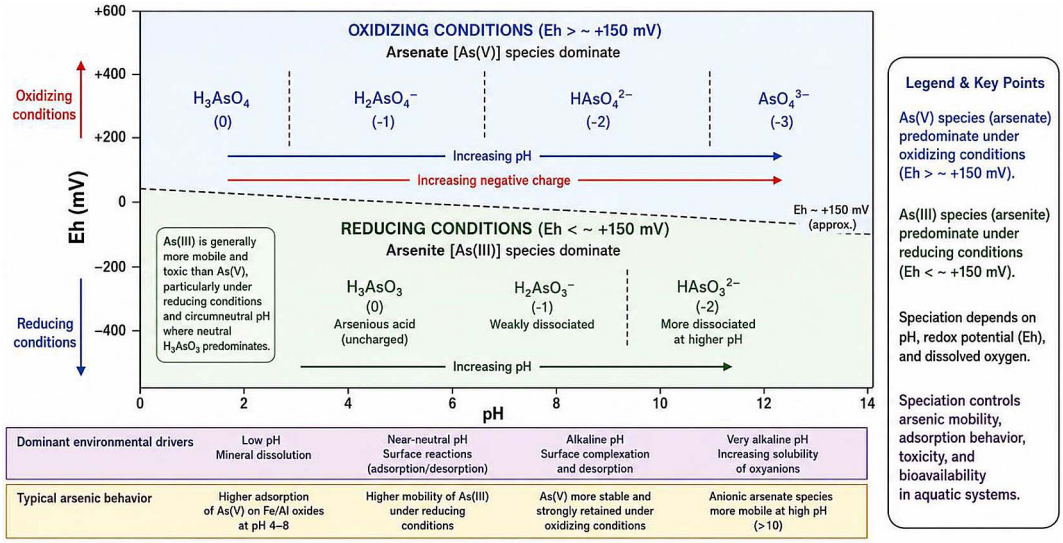

Figure 1. Conceptual diagram illustrating arsenic speciation as controlled by pH and redox conditions in aquatic environments. Under oxidizing conditions, arsenate species [As(V)] predominate, whereas under reducing conditions, arsenite species [As(III)] become more stable

Table 3. Dominant arsenic species and acid dissociation behavior in natural waters

Arsenic species	Main dissociation reactions	Approximate pKa	Environmental implication
Arsenic acid, As(V)	$H_3AsO_4 \rightleftharpoons H_2AsO_4^- \rightleftharpoons HAsO_4^{2-} \rightleftharpoons AsO_4^{3-}$	$pKa_1 \approx 2.2$; $pKa_2 \approx 7.0$; $pKa_3 \approx 11.5$	Dominant under oxic conditions; adsorbs strongly onto Fe/Al oxides
Arsenous acid, As(III)	$H_3AsO_3 \rightleftharpoons H_2AsO_3^- \rightleftharpoons HAsO_3^{2-} \rightleftharpoons AsO_3^{3-}$	$pKa_1 \approx 9.2$	Neutral at circumneutral pH; more mobile and harder to remove
Monomethylarsonic acid	MMA species	$pKa_1 \approx 4.2$; $pKa_2 \approx 8.8$	Product of biological methylation
Dimethylarsinic acid	DMA species	$pKa_1 \approx 6.3$	Common methylated arsenic species in biological systems

Note: Compiled from several previous studies (Cullen and Reimer, 1989; Langmuir, 1997; Sharma and Sohn, 2009; Smedley and Kinniburgh, 2002).

biological transformation, organic matter decomposition, and food-chain transfer are relevant (Lee and Wen, 2019; Zeng et al., 2023).

Biogeochemical transformation of arsenic is closely linked to redox cycling. Oxidation of As(III) to As(V) can reduce arsenic mobility by increasing its affinity for mineral adsorption sites, while reduction of As(V) to As(III) can enhance mobility and bioavailability (Dixit and Hering, 2003; Manning and Goldberg, 1997). These transformations may occur through abiotic reactions with manganese oxides, iron minerals, sulfide species, or dissolved oxygen, but they are often strongly influenced by microbial processes (Fazi et al., 2016). Microorganisms can mediate arsenic oxidation, arsenate reduction, methylation, and demethylation, thereby modifying arsenic speciation and redistribution across water, sediment, and biological compartments (Fazi et al., 2016). In sedimentary systems, such microbial transformations are particularly important because redox gradients develop naturally with depth as oxygen is consumed and alternative electron acceptors such as nitrate, manganese oxides, iron oxides, sulfate, and carbon dioxide are sequentially utilized (Nicholas et al., 2003). Recent evidence further indicates that nitrate-dependent processes can promote anaerobic oxidation of As(III) and alter arsenic redistribution in estuarine sediments (Ding et al., 2024).

The interaction between arsenic speciation and sediment diagenesis is central to arsenic mobility. In oxic surface sediments, As(V) may be retained by freshly precipitated iron and manganese oxyhydroxides (Fendorf and Kocar, 2009). As sediments are buried and reducing conditions develop, these oxyhydroxides may dissolve, releasing arsenic into porewater (Nicholas et al., 2003). The released arsenic may then diffuse upward toward oxic layers, where it can be reoxidized and readsorbed, or downward into more reducing zones, where it may interact with sulfide and form arsenic-bearing sulfide phases (Dissanayake and Chandrajith, 2009; Du et al., 2026). This dynamic cycling explains why sediments can function simultaneously as sinks, transformation zones, and secondary sources of arsenic. It also demonstrates why dissolved arsenic concentrations in overlying water may not fully represent the long-term arsenic reservoir stored in sediments.

Thermodynamic Eh-pH diagrams are useful tools for predicting the dominant arsenic species under specific redox and pH conditions. These diagrams show that As(V) generally dominates under oxic conditions, whereas As(III) becomes more stable under mildly to strongly reducing conditions (Smedley and Kinniburgh, 2002). In sulfidic environments, arsenic may also form arsenic sulfide phases or become associated with iron sulfides (O'Day et al., 2004; Omoregie et al., 2013). However, thermodynamic predictions should be interpreted carefully because natural systems are rarely at equilibrium. Kinetic limitations, microbial mediation, mineral surface reactions, organic ligands, colloidal transport, and hydrological changes can cause actual arsenic behavior to deviate from equilibrium predictions. Thus, Eh-pH diagrams are best used as conceptual guides rather than definitive predictors of arsenic fate.

Overall, arsenic speciation and transformation are controlled by a complex interaction of pH, redox potential, mineralogy, organic matter, microbial activity, and sediment–water exchange (Cui and Jing, 2019). The dominance of As(III), As(V), methylated species, or sulfide-associated forms determines whether arsenic is retained, mobilized, transformed, or transferred through environmental compartments. For this reason, species-specific analysis is essential for understanding arsenic contamination and for designing

effective monitoring, remediation, and risk assessment strategies (Fendorf and Kocar, 2009; Smedley and Kinniburgh, 2002). The following section further discusses how sediment–water interactions, iron and manganese oxyhydroxides, sulfide phases, organic matter, and porewater processes regulate arsenic retention and release in aquatic environments.

ARSENIC MOBILITY IN WATER-SEDIMENT SYSTEMS

The mobility of arsenic in aquatic environments is strongly controlled by sediment–water interactions. Sediments are not passive repositories of contaminants, but dynamic biogeochemical systems in which arsenic can be retained, transformed, released, or redistributed (Khalid et al., 2017; Martínez López et al., 2023). In riverine, lacustrine, estuarine, and marine systems, arsenic may enter sediments through the deposition of suspended particles, adsorption onto mineral surfaces, coprecipitation with iron and manganese oxyhydroxides, association with organic matter, or incorporation into sulfide phases (Benjamin and Honeyman, 1992; Cukrov et al., 2024; Santini and Ward, 2018). Once deposited, arsenic is exposed to changing geochemical conditions caused by sediment burial, organic matter degradation, microbial activity, redox zonation, porewater diffusion, and physical disturbance (Maqsood and Łobos-Moysa, 2025; Xing et al., 2024). These processes determine whether sediments act as long-term sinks or secondary sources of arsenic to overlying water. The major processes controlling arsenic exchange across the sediment-water interface, including adsorption, desorption, redox transformation, porewater diffusion, resuspension, and sulfide-associated immobilization, are conceptually illustrated in Figure 2.

Because arsenic mobility is controlled by multiple interacting geochemical processes, it is useful to identify the major sedimentary and aqueous factors that regulate arsenic retention and release. The key controls on arsenic mobility in water–sediment systems are summarized in Table 4.

In oxic surface sediments, arsenic is commonly retained by adsorption or coprecipitation with iron and manganese oxyhydroxides. These mineral phases have high surface reactivity and can bind arsenic through surface complexation, ligand exchange, and occlusion during mineral

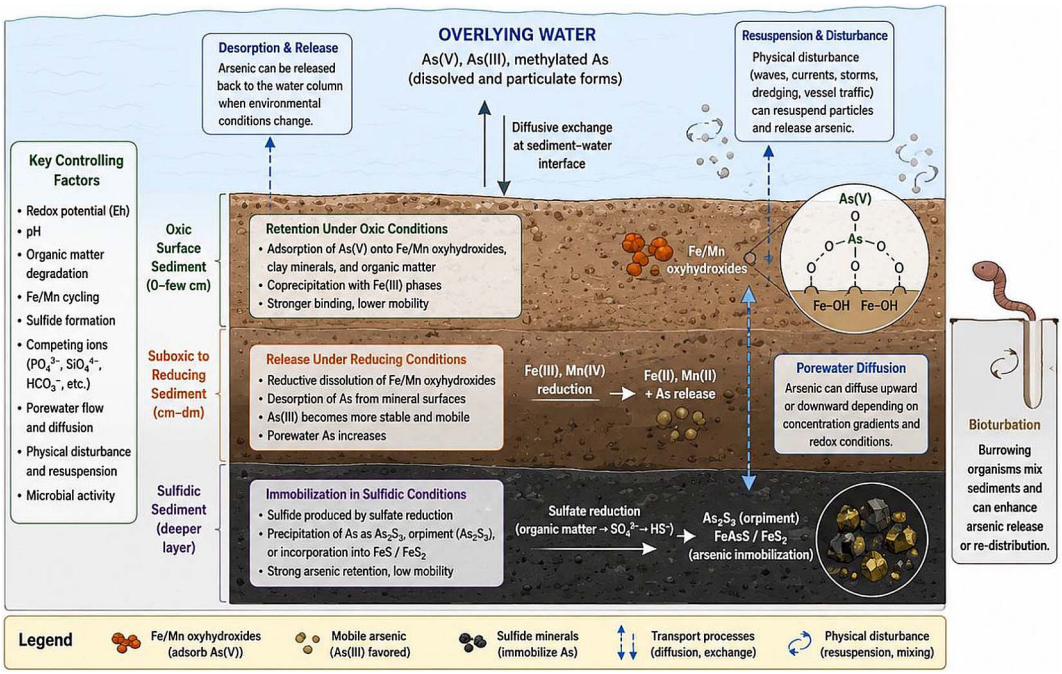

Figure 2. Conceptual framework of arsenic mobility and exchange across the sediment-water interface
The figure illustrates how arsenic can be retained in oxic surface sediments through adsorption and coprecipitation with Fe/Mn oxyhydroxides, released under reducing conditions through reductive dissolution, transported through porewater diffusion, immobilized in deeper sulfidic layers through sulfide-associated phases, or remobilized by desorption, resuspension, bioturbation, and hydrodynamic disturbance

precipitation (Aftabtalab et al., 2022; Foster, 2003; Miretzky and Cirelli, 2010). Iron oxyhydroxides are particularly important because they can strongly adsorb arsenate and, under certain conditions, arsenite. Freshly formed or poorly crystalline iron oxyhydroxides generally provide more reactive binding sites than well-crystallized mineral phases (Aftabtalab et al., 2022). Manganese oxides may also influence arsenic mobility, not only as adsorption surfaces but also as oxidants capable of transforming As(III) into As(V) (Ying et al., 2012). This oxidation process can reduce arsenic mobility by producing arsenate species that interact more strongly with mineral surfaces. Therefore, the presence, reactivity, and stability of Fe/Mn oxyhydroxides are central controls on arsenic retention in oxic and suboxic sediment layers.

However, the retention capacity of Fe/Mn oxyhydroxides is highly sensitive to redox changes. As sediments are buried and oxygen becomes depleted, microbial decomposition of organic matter progressively consumes available electron acceptors. Under reducing conditions, manganese oxides and iron oxyhydroxides may undergo reductive dissolution, releasing Mn(II), Fe(II), and associated trace elements into porewater (Farnsworth and Hering, 2011; Klinkhammer, 1980). Arsenic that was previously adsorbed or coprecipitated with these mineral phases can therefore be mobilized. This mechanism is widely recognized as one of the main pathways for arsenic enrichment in anoxic groundwater and sediment porewater (Bennett et al., 2012). In such systems, the release of arsenic is often linked to elevated dissolved iron, indicating that arsenic mobility is coupled to iron redox cycling.

Organic matter plays a dual role in arsenic mobility. On one hand, organic matter can contribute to arsenic retention through complexation, adsorption, and association with particulate phases (Aftabtalab et al., 2022). Functional groups such as carboxyl, phenolic, and sulfhydryl groups may interact with arsenic directly or indirectly by binding metals that form arsenic-bearing complexes. On the other hand, organic matter is also a major driver of reducing conditions in sediments. Its microbial decomposition consumes dissolved oxygen and promotes the sequential reduction of nitrate, manganese oxides, iron oxyhydroxides, sulfate, and carbon dioxide (Curtis, 1987; Pandey et al., 2024). As reducing conditions intensify, arsenic may be released from Fe/Mn oxyhydroxides into porewater. Thus, organic matter can either enhance arsenic immobilization or promote arsenic mobilization depending on its abundance, reactivity, and influence on sediment redox conditions.

Sulfide formation represents another major control on arsenic mobility in anoxic sediments. Under sulfate-reducing conditions, microbial sulfate reduction produces dissolved sulfide, which can react with iron to form iron sulfide minerals such as amorphous FeS, mackinawite, greigite, and pyrite (Fortin and Beveridge, 1997; Mansor et al., 2025). These sulfide phases can immobilize arsenic by adsorption, coprecipitation, structural incorporation, or precipitation of discrete arsenic sulfides. In strongly reducing and sulfidic environments, arsenic may therefore become less mobile as it is transferred from porewater into sulfide-associated solid phases. However, this immobilization is not necessarily permanent. If sulfidic sediments are later exposed to oxygen through

Table 4. Key controls on arsenic mobility in water-sediment systems

Control factor	Main process	Effect on arsenic mobility
Fe oxyhydroxides	Adsorption, coprecipitation, reductive dissolution	Retains As under oxic conditions; releases As under reducing conditions
Mn oxides	Adsorption and oxidation of As(III)	Can oxidize As(III) to As(V), reducing mobility
Organic matter	Complexation and microbial redox control	May bind As, but can also promote reducing conditions and As release
Sulfide phases	Precipitation, adsorption, incorporation into FeS/FeS_2	Immobilizes As under sulfidic conditions
Porewater gradient	Diffusion and redistribution	Controls vertical transport and sediment-water exchange
pH and competing ions	Surface charge and ligand competition	High pH, phosphate, and silicate may enhance desorption

Note: Compliated from several studies (Bowell et al., 2014; Dixit and Hering, 2003; Fendorf and Kocar, 2009; Oremland and Stolz, 2003).

resuspension, drainage, erosion, bioturbation, or water-level fluctuation, sulfide oxidation can release arsenic back into the aqueous phase. This reversible behavior makes arsenic mobility highly sensitive to environmental disturbance.

Porewater is the immediate medium through which arsenic is transported within sediments and exchanged with overlying water. The concentration of arsenic in porewater reflects the balance between release from solid phases, removal through precipitation or adsorption, transformation among arsenic species, and diffusive transport (Kocar et al., 2014; Smedley and Kinniburgh, 2013). When arsenic is released into porewater at depth, concentration gradients can drive upward diffusion toward the sediment–water interface or downward migration into deeper reducing zones. Upward-diffusing arsenic may be reoxidized and readsorbed in oxic surface layers, creating a recycling mechanism that limits its escape to overlying water. Alternatively, if the oxic layer is thin, disturbed, or chemically saturated, arsenic may diffuse into the water column and contribute to secondary contamination (Bowell et al., 2014; Smedley and Kinniburgh, 2013). Downward-diffusing arsenic may encounter sulfide-rich zones and become immobilized as arsenic-bearing sulfides or associated with pyrite.

The sediment–water interface is therefore a critical transition zone for arsenic cycling. It separates relatively oxic overlying water from progressively reducing sediment layers and acts as a zone of intense chemical and microbial transformation (Duan et al., 2024; Xing et al., 2024). Small changes in oxygen penetration depth, organic matter supply, pH, microbial activity, hydrodynamic mixing, or bioturbation can modify arsenic flux across this interface. In shallow lakes, wetlands, estuaries, reservoirs, and mining-affected water bodies, seasonal changes in temperature, water level, salinity, and organic matter input may further alter sediment redox conditions and arsenic release (Barral-Fraga et al., 2020). Consequently, arsenic concentrations in overlying water may vary temporally even when external arsenic inputs remain constant. The redox evolution of sediments strongly influences arsenic mobility because different electron-accepting processes control the stability of Fe/Mn oxyhydroxides, sulfide phases, and porewater chemistry. The main redox stages relevant to arsenic release and immobilization are summarized in Table 5.

Adsorption–desorption reactions also play a major role in regulating arsenic mobility. Arsenate generally adsorbs strongly onto iron and aluminum oxides through inner-sphere complexation, particularly at acidic to near-neutral pH (Aftabtalab et al., 2022; Miretzky and Cirelli, 2010). However, arsenate adsorption can decrease at higher pH because mineral surfaces become more negatively charged and competition with hydroxide increases. Arsenite adsorption is more complex because As(III) is often present as a neutral species under circumneutral pH, reducing its electrostatic attraction to mineral surfaces (Martin et al., 2014; Stachowicz et al., 2006). Competing anions such as phosphate, silicate, carbonate, bicarbonate, and dissolved organic ligands can further suppress arsenic adsorption by occupying reactive surface sites or altering surface charge (Bang and Meng, 2004; Roberts et al., 2004). Therefore, the mobility of arsenic is not controlled by arsenic concentration alone, but by the broader chemical composition of the water–sediment system.

Physical processes also influence arsenic transport. Sediment resuspension can transfer particle-bound arsenic into the water column,

Table 5. Sequence of redox reactions relevant to arsenic release in sediments

Redox stage	Dominant electron acceptor	Environmental relevance to arsenic
Oxic respiration	O_2	Promotes Fe/Mn oxide stability and arsenic retention
Denitrification	NO_3^-	Early reducing condition; limited direct As release
Mn reduction	Mn(IV) oxides	May release associated metals and affect As oxidation
Fe reduction	Fe(III) oxyhydroxides	Major pathway for As release from sediments
Sulfate reduction	SO_4^{2-}	Produces sulfide; may immobilize As as sulfide-associated phases
Methanogenesis	CO_2/organic matter	Strongly reducing condition; may alter As mobility and microbial transformation

Note: Compilated from several studies (Bowell et al., 2014; Fendorf and Kocar, 2009; Froelich et al., 1979; Oremland and Stolz, 2003; Smedley and Kinniburgh, 2002).

where changes in oxygen and pH may alter arsenic speciation and release. Erosion and deposition can redistribute arsenic-contaminated particles across river channels, floodplains, estuaries, and coastal zones. In estuarine systems, salinity gradients can promote flocculation, desorption, or competitive exchange reactions, changing arsenic partitioning between dissolved and particulate phases (Schindler et al., 2021). Flooding and drying cycles can also shift redox conditions, causing repeated dissolution and precipitation of arsenic-bearing phases (Fakhreddine et al., 2021). These processes highlight the need to evaluate arsenic at the system scale rather than as a static contaminant in a single environmental compartment.

Overall, arsenic mobility in water–sediment systems is governed by coupled geochemical, microbiological, and physical processes. Fe/Mn oxyhydroxides, organic matter, sulfide phases, porewater gradients, adsorption–desorption reactions, and sediment disturbance collectively determine whether arsenic remains immobilized or becomes available for transport and exposure. This complexity explains why total arsenic concentrations in water, sediment, or porewater may provide incomplete information when interpreted separately. A more reliable assessment requires simultaneous consideration of arsenic speciation, solid-phase associations, redox status, porewater chemistry, and sediment–water flux. These factors are essential for predicting arsenic persistence, secondary release, and long-term environmental risk.

TOXICITY, EXPOSURE PATHWAYS, AND RISK IMPLICATIONS

The toxicity of arsenic is strongly dependent on its chemical species, oxidation state, solubility, exposure route, dose, and duration of exposure (Ganie et al., 2024; Sharma and Sohn, 2009). This species-dependent toxicity is one of the main reasons why total arsenic concentration alone is insufficient for environmental and health risk assessment (Diacomanolis et al., 2016). Inorganic arsenic species, particularly arsenite As(III) and arsenate As(V), are generally of greatest concern because they are more toxic and environmentally relevant than most organic arsenic compounds (Druwe and Vaillancourt, 2010; Ganie et al., 2024). Among these, As(III) is commonly considered more toxic and more mobile than As(V), largely because of its strong affinity for sulfhydryl groups in proteins and enzymes, its ability to interfere with cellular metabolism, and its tendency to occur as a neutral species under circumneutral reducing conditions (Druwe and Vaillancourt, 2010; Sharma and Sohn, 2009). In contrast, As(V) can behave as a phosphate analogue and disrupt phosphate-dependent biochemical processes, including ATP production and cellular energy transfer (Druwe and Vaillancourt, 2010).

Human exposure to arsenic occurs mainly through contaminated drinking water, food, inhalation of arsenic-bearing particles, occupational contact, and, in some settings, ingestion of contaminated soil or dust (De Voogt, 2017; Kumari et al., 2017). Drinking water remains one of the most important exposure pathways because dissolved inorganic arsenic can be directly consumed over long periods. Groundwater contamination is particularly critical in geogenic arsenic regions, where arsenic can be released from aquifer sediments through reductive dissolution of iron oxyhydroxides or oxidation of arsenic-bearing sulfide minerals (Mahimairaja et al., 2005; Sharma and Sohn, 2009). In mining-affected and industrial areas, exposure may also occur through surface water, sediments, tailings, atmospheric deposition, and contaminated agricultural soils (Hoang et al., 2021). Therefore, arsenic risk depends not only on concentration in water, but also on source type, environmental compartment, exposure frequency, and the chemical form of arsenic present.

Food-chain exposure is another important pathway, especially in areas where arsenic-contaminated water is used for irrigation or where aquatic organisms accumulate arsenic from contaminated sediments and water (Rahman et al., 2012; Arslan et al., 2017). Rice is a major concern because it can accumulate inorganic arsenic under flooded soil conditions, where reducing environments favor arsenic mobilization and uptake by plant roots (Arslan et al., 2017; Pullyottum Kavil et al., 2020). Aquatic organisms may also accumulate arsenic, but the toxicological significance depends strongly on speciation. Many marine organisms contain arsenobetaine and other organoarsenic species that are generally considered less toxic than inorganic arsenic (Azizur Rahman et al., 2012). However, the presence of inorganic arsenic or reactive methylated species in food matrices remains relevant for health risk assessment. Therefore, food-chain risk evaluation requires species-specific determination rather than total arsenic measurement alone.

At the cellular level, As(III) can bind strongly to thiol-containing proteins, inhibit enzyme activity, induce oxidative stress, alter signal transduction, disrupt DNA repair, and interfere with cellular respiration (Druwe and Vaillancourt, 2010; Ganie et al., 2024). As(V), because of its chemical similarity to phosphate, can replace phosphate in biochemical reactions and impair energy metabolism. Chronic exposure to inorganic arsenic has been associated with skin lesions, pigmentation changes, hyperkeratosis, cardiovascular effects, neurological effects, diabetes-related outcomes, and cancers of the skin, lung, bladder, and other organs (Ganie et al., 2024; Malik et al., 2022). The severity and manifestation of these effects depend on exposure level, exposure duration, nutritional status, genetic susceptibility, arsenic metabolism, and co-exposure to other contaminants. These health outcomes highlight the need for monitoring approaches that connect environmental arsenic data with exposure relevance and toxicological significance.

Arsenic metabolism in humans involves reduction, oxidation, and methylation reactions. Ingested inorganic arsenic can be reduced and methylated to monomethylarsonic acid and dimethylarsinic acid, which are excreted primarily through urine (Sharma and Sohn, 2009; Ganie et al., 2024). For many years, methylation was interpreted as a detoxification pathway because methylated arsenic species are often more readily eliminated from the body. However, this interpretation is now recognized as oversimplified because some methylated trivalent intermediates may be highly reactive and biologically active (Druwe and Vaillancourt, 2010). Therefore, urinary arsenic speciation can provide useful information on recent exposure and metabolic profile, but interpretation requires careful differentiation between inorganic arsenic, methylated metabolites, and relatively non-toxic dietary organoarsenic species derived from seafood (Sharma and Sohn, 2009).

Different biological matrices provide different information on arsenic exposure. Urine is commonly used as an indicator of recent arsenic exposure because arsenic and its metabolites are excreted mainly through the urinary pathway. Hair and nails can reflect longer-term exposure because arsenic has an affinity for keratin-rich tissues (Chan and Huff, 1997). Blood arsenic is less useful for retrospective exposure assessment because arsenic is rapidly cleared from circulation. However, biomonitoring data must be interpreted cautiously because external contamination, dietary sources, species transformation, and analytical method limitations can influence measured concentrations. For this reason, biomonitoring should be combined with environmental measurements, exposure history, and arsenic speciation to support more reliable risk interpretation.

Ecological toxicity also depends on arsenic speciation and environmental conditions. In aquatic organisms, arsenic bioavailability is influenced by dissolved species, sediment binding, trophic transfer, salinity, organic matter, and redox conditions (Ghosh et al., 2022; Neff, 1997; Wang et al., 2022). Benthic organisms may be exposed to arsenic through direct contact with contaminated sediments, ingestion of particles, and porewater exchange. Plants can take up arsenic from soils and sediments, with uptake affected by phosphate availability, iron plaque formation on roots, soil redox conditions, and arsenic species (Khalid et al., 2017). Because arsenate behaves similarly to phosphate, it can enter plant uptake pathways and interfere with phosphorus metabolism. Arsenite, meanwhile, can interact with thiol-rich biomolecules and cause cellular toxicity (Druwe and Vaillancourt, 2010). These processes show that ecological risk cannot be inferred from total arsenic concentration alone without considering speciation, bioavailability, and exposure route.

The environmental risk of arsenic is therefore best understood through an integrated framework combining source identification, speciation, mobility, exposure pathway, and receptor sensitivity (Prasad et al., 2026). A water sample with low total arsenic may still be of concern if the dominant species is As(III), while sediment with high total arsenic may pose limited immediate risk if arsenic is strongly immobilized in stable mineral phases (Kanel et al., 2023). Conversely, arsenic-rich sediments may become important secondary sources when redox conditions shift, sulfide minerals oxidize, or Fe/Mn oxyhydroxides dissolve. Thus, risk assessment must consider not only how much arsenic is present, but also where it is located, in what chemical form it occurs, how stable that form is, and whether environmental conditions favor release, transformation, or biological uptake.

Overall, arsenic toxicity and risk are controlled by the interaction between chemical speciation, environmental mobility, exposure pathway, and biological response (Sharma and Sohn, 2009; Ganie et al., 2024). This reinforces the need for risk-based monitoring that moves beyond total

arsenic measurement toward species-specific and matrix-specific assessment. Such an approach is particularly important in groundwater systems, sediment-impacted water bodies, mining regions, agricultural areas, and ecosystems where arsenic can be transferred through water, sediment, soil, food, and biological tissues. The following section discusses analytical methods for arsenic determination and speciation, with emphasis on how analytical reliability influences environmental interpretation and risk assessment.

ANALYTICAL METHODS FOR ARSENIC DETERMINATION AND SPECIATION

Reliable determination of arsenic in environmental samples is essential for understanding its occurrence, mobility, toxicity, bioavailability, and associated environmental risks. However, arsenic analysis is analytically challenging because environmental samples often contain arsenic at trace or ultra-trace concentrations, while its chemical species differ substantially in toxicity, mobility, and environmental behavior (Ardini et al., 2020; Nearing et al., 2014). Total arsenic concentration provides important information regarding contamination status but does not distinguish among arsenic species that govern bioavailability, toxicity, and ecological significance. Consequently, modern arsenic monitoring increasingly requires both total arsenic determination and species-specific analysis in water, sediments, porewater, soils, biological tissues, and food matrices (Ardini et al., 2020; Bosch et al., 2010).

Total arsenic analysis is commonly performed using atomic spectrometric and mass spectrometric techniques. Inductively coupled plasma mass spectrometry (ICP-MS) is widely regarded as the benchmark method because of its exceptional sensitivity, low detection limits, broad dynamic range, rapid analysis, and multi-element capability. Detection limits frequently reach sub-μg L^{-1} concentrations, allowing accurate quantification of arsenic in both pristine and contaminated environments. Inductively coupled plasma optical emission spectrometry (ICP-OES) is also widely used but generally exhibits lower sensitivity than ICP-MS for trace-level arsenic determination. Atomic absorption spectrometry (AAS), particularly hydride generation atomic absorption spectrometry (HG-AAS), has long been applied because arsenic can be converted into volatile hydride species prior to detection. Hydride generation atomic fluorescence spectrometry (HG-AFS) offers additional improvements in sensitivity and selectivity and remains valuable for routine environmental monitoring where access to ICP-MS is limited (Mishra and Sankararamakrishnan, 2018; Yu et al., 2020)

Sample preparation strongly influences the reliability of total arsenic determination. Water samples are typically filtered and acidified immediately after collection to preserve dissolved arsenic fractions, whereas sediments, soils, plants, and biological tissues require digestion prior to instrumental analysis. Acid digestion using nitric acid, hydrochloric acid, hydrogen peroxide, or microwave-assisted digestion is frequently employed to release arsenic from solid matrices (Chen et al., 2006). However, digestion procedures must be selected carefully because incomplete digestion can underestimate total arsenic concentrations, whereas overly aggressive digestion or inappropriate reagent combinations may cause analyte loss, contamination, or species alteration. For solid environmental samples, quality control measures such as certified reference materials, procedural blanks, spike recovery tests, duplicate analyses, and matrix-matched calibration are essential to ensure analytical reliability (Fırat et al., 2017).

Although total arsenic determination is useful for contamination screening and regulatory comparison, it is insufficient for risk-based interpretation because arsenic toxicity and mobility are strongly species-dependent. Speciation analysis aims to separate and quantify individual arsenic species, including As(III), As(V), monomethylarsonic acid (MMA), dimethylarsinic acid (DMA), arsenobetaine, arsenocholine, arsenosugars, and other organoarsenic compounds (Bosch et al., 2010; Chen et al., 2014). The most powerful analytical approach combines chromatographic separation with element-specific detection. High-performance liquid chromatography coupled with ICP-MS (HPLC-ICP-MS) is widely considered the gold standard for arsenic speciation because it enables efficient separation of arsenic species followed by highly sensitive and selective elemental detection (Ardini et al., 2020; B'Hymer & Caruso, 2004). Ion-exchange chromatography is commonly applied to separate inorganic and methylated arsenic species, whereas reversed-phase and ion-pair chromatographic approaches may be employed for more complex organoarsenic compounds (Bosch et al., 2010).

The accuracy of arsenic speciation analysis depends not only on instrumental capability but also on species preservation during sampling, storage, extraction, and analysis. Arsenic species can undergo oxidation, reduction, methylation, demethylation, adsorption, precipitation, or microbially mediated transformation following sample collection (Wolf et al., 2011). For example, As(III) may oxidize to As(V) during storage, whereas As(V) may be reduced under anoxic or biologically active conditions. Consequently, field filtration, refrigeration, temperature control, exclusion of light, use of appropriate preservatives, minimization of storage duration, and validation of preservation protocols are critical for obtaining reliable speciation data. In porewater and sediment samples, maintaining in situ redox conditions is particularly important because exposure to oxygen may rapidly alter arsenic speciation as well as associated Fe/Mn oxyhydroxide and sulfide phases (Wolf et al., 2011).

Extraction remains one of the greatest challenges in arsenic speciation analysis of solid matrices. Unlike dissolved arsenic in water, arsenic in sediments, soils, plants, and biological tissues may be associated with minerals, organic matter, sulfides, or biological macromolecules. Therefore, extraction procedures must release arsenic species while preserving their original chemical forms. Mild extractants may preserve species integrity but yield incomplete recovery, whereas stronger extractants may improve extraction efficiency at the expense of species stability. Various extraction approaches, including phosphate-buffer extraction, water–methanol extraction, alkaline extraction, enzymatic extraction, microwave-assisted extraction, and acid extraction, have been applied depending on sample type and analytical objectives (M.-L. Chen et al., 2014; Kresa et al., 2026). Because no universal extraction procedure is suitable for all environmental matrices, method validation should evaluate extraction efficiency, species recovery, species stability, and mass balance between extracted species and independently measured total arsenic concentrations.

Matrix interference represents another important analytical challenge. In ICP-MS analysis, arsenic determination at m/z 75 may be affected by polyatomic interferences, particularly $^{40}Ar^{35}Cl^{+}$ ions generated in chloride-rich matrices (Ardini et al., 2020; Balaram, 2021; Wolf et al., 2011). Advanced instrumental configurations employing collision or reaction cell technologies have been developed to minimize such interferences and improve analytical accuracy. Additional matrix effects may arise from high dissolved solids, organic matter, iron, sulfur, chloride, and other coexisting constituents that influence nebulization efficiency, plasma stability, chromatographic separation, or detector response. Consequently, arsenic methods should be optimized according to matrix characteristics rather than applied as generic procedures. This consideration is particularly important for seawater, estuarine waters, acid mine drainage, industrial wastewater, sediment extracts, and biological samples.

Electrochemical methods have also emerged as promising alternatives for arsenic determination, particularly in field-oriented applications. Techniques such as stripping voltammetry and modified electrode sensors offer advantages including low cost, portability, rapid response, and relatively simple instrumentation (Borrill et al., 2019; Devnani and Sharma, 2023). These approaches are particularly attractive for decentralized monitoring programs and resource-limited regions where access to advanced laboratory facilities may be restricted. However, electrochemical methods are susceptible to interference from coexisting ions, electrode fouling, matrix effects, and oxidation-state dependence. Many systems exhibit higher sensitivity toward As(III) than As(V), necessitating additional oxidation or reduction steps when determining total inorganic arsenic concentrations. Therefore, rigorous validation remains necessary before electrochemical methods can be widely adopted for regulatory monitoring.

Field-based and rapid analytical approaches are becoming increasingly important for arsenic surveillance. Portable test kits, colorimetric assays, paper-based devices, microfluidic platforms, passive samplers, and sensor-based systems can provide rapid screening of arsenic contamination (Chillè et al., 2024). These tools are valuable for preliminary assessment, emergency response, community-scale monitoring, and spatial mapping of contaminated groundwater sources. Nevertheless, many rapid methods remain limited in detection capability, selectivity, species differentiation, and matrix tolerance. Consequently, field screening results should ideally be verified using laboratory-based methods when concentrations approach regulatory thresholds or when management decisions involve public health protection.

Quality assurance and quality control (QA/QC) are essential components of arsenic

determination and speciation analysis. Reliable monitoring requires appropriate calibration procedures, procedural blanks, duplicate analyses, spike recovery tests, certified reference materials, detection limit estimation, method validation, and uncertainty assessment (Fırat et al., 2017). Speciation analysis requires additional QA/QC measures, including species-specific standards, chromatographic resolution assessment, retention-time verification, species recovery evaluation, and comparison between summed species concentrations and independently determined total arsenic. Without these controls, analytical results may be difficult to interpret and may introduce uncertainty into environmental risk assessment, remediation planning, or regulatory decision-making. Because different analytical techniques offer different levels of sensitivity, selectivity, matrix tolerance, and speciation capability, analytical method selection should be guided by both monitoring objectives and sample characteristics. The principal methods used for total arsenic determination and arsenic speciation are summarized in Table 6.

Overall, arsenic analytical strategies should be selected according to the monitoring objective. Total arsenic determination is appropriate for initial screening, contamination assessment, and regulatory comparison, whereas species-specific analysis is necessary to understand mobility, bioavailability, toxicity, treatment behavior, and exposure risk. In complex environmental systems, the most informative approach integrates total arsenic measurements, arsenic speciation, matrix characterization, and supporting geochemical parameters such as pH, redox potential, dissolved oxygen, iron, manganese, sulfide, phosphate, salinity, and organic matter content. Such an integrated analytical framework provides a stronger scientific basis for interpreting arsenic behavior and for supporting risk-based monitoring and environmental management.

CURRENT CHALLENGES AND FUTURE PERSPECTIVES FOR RISK-BASED MONITORING

Despite substantial progress in arsenic research, several challenges remain in translating environmental measurements into reliable risk assessment and management strategies. A major limitation is the continued reliance on total arsenic concentration as the primary indicator of contamination. Although total arsenic is useful for screening and regulatory comparison, it provides limited information on toxicity, mobility, bioavailability, and environmental behavior. Two samples with similar total arsenic concentrations may represent very different levels of risk if they contain different proportions of As(III), As(V), methylated species, particulate-bound arsenic, or mineral-associated arsenic. Therefore, the interpretation of arsenic contamination requires a transition from concentration-based assessment toward species-resolved, process-based, and exposure-relevant monitoring.

One of the most important challenges is the instability of arsenic species during sampling, storage, extraction, and analysis. Arsenic speciation in natural waters, porewater, sediments, soils, and biological samples can change rapidly in response to oxygen exposure, microbial activity, pH shifts, redox alteration, light exposure, and interactions with mineral phases. As(III) may be oxidized to As(V), As(V) may be reduced under anoxic conditions, and dissolved arsenic may adsorb onto particles or precipitate during storage. In sediment and porewater samples, disturbance of in situ redox conditions during collection can significantly alter the original species distribution. Consequently, reliable speciation analysis requires carefully validated sampling protocols, field filtration, preservation procedures, low-temperature storage, short holding times, and documentation of geochemical conditions at the time of collection.

Another key challenge is the limited integration between arsenic measurements and sediment-water processes. Many monitoring programs focus primarily on dissolved arsenic in water, while sediments are treated as separate or secondary compartments. However, sediments can store large arsenic inventories and release arsenic when redox conditions shift, Fe/Mn oxyhydroxides dissolve, sulfide phases oxidize, or organic matter degradation intensifies. This means that water-column arsenic concentrations may not fully represent the long-term contamination potential of an aquatic system. Risk-based monitoring should therefore include not only water samples but also sediments, porewater, sediment-associated arsenic fractions, and, where feasible, measurements of arsenic flux across the sediment-water interface. Such integration is particularly important in lakes, reservoirs, wetlands,

Table 6. Analytical methods for total arsenic and arsenic speciation

Method	Target	Advantages	Limitations
ICP-MS	Total As and coupled speciation	Very sensitive, multi-element capability	Matrix interference, high cost
ICP-OES	Total As	Suitable for higher concentrations	Less sensitive than ICP-MS
]HG-AAS/HG-AFS	Total inorganic As	Sensitive and relatively established	Requires hydride generation conditions
HPLC-ICP-MS	Arsenic speciation	Species-specific, high sensitivity	Expensive, requires careful preservation
Voltammetry	Mainly As(III), sometimes total inorganic As	Portable, low cost	Matrix interference and electrode fouling
Colorimetric/test kits	Screening	Rapid and field-friendly	Limited accuracy and speciation capability

estuaries, mining-affected rivers, and groundwater-connected sedimentary systems.

Analytical reliability remains another critical issue. Advanced instruments such as ICP-MS and HPLC-ICP-MS provide highly sensitive arsenic determination and speciation, but their availability remains limited in many regions affected by arsenic contamination. In addition, complex environmental matrices such as seawater, acid mine drainage, industrial wastewater, sediment extracts, and biological tissues can introduce spectral interference, matrix suppression, extraction bias, and species transformation. For speciation analysis, the summed concentration of individual species does not always match independently measured total arsenic, indicating incomplete extraction, species degradation, or analytical loss. Therefore, future arsenic monitoring should place greater emphasis on method validation, certified reference materials, inter-laboratory comparison, uncertainty reporting, species recovery assessment, and matrix-specific quality assurance protocols.

Field-based arsenic monitoring also requires further development. Portable kits, colorimetric tests, paper-based sensors, electrochemical devices, and microfluidic platforms have considerable potential for rapid screening and decentralized monitoring. These tools are particularly valuable in remote areas, mining regions, and communities where laboratory access is limited. However, many field methods still face limitations related to detection limits, selectivity, speciation capability, matrix interference, user-dependent error, and quality control. Future research should focus on improving the sensitivity, robustness, and species discrimination capability of field-based technologies while ensuring that screening results can be validated against laboratory reference methods. Rather than replacing laboratory analysis, field technologies should be integrated into tiered monitoring frameworks in which rapid screening guides more comprehensive confirmatory investigations.

A further limitation is the weak connection between arsenic speciation data and exposure assessment. Environmental monitoring often reports arsenic concentrations in water, sediment, soil, or biota without sufficiently linking these measurements to actual exposure pathways and receptor vulnerability. For example, arsenic in groundwater may directly affect drinking-water exposure, whereas arsenic in sediment may pose risks through porewater diffusion, benthic uptake, resuspension, or food-web transfer. Arsenic in rice, fish, or other food matrices requires species-specific interpretation because inorganic and organic arsenic species differ substantially in toxicological relevance. Therefore, risk assessment should integrate environmental compartment, arsenic species, exposure route, receptor type, and bioavailability. Such integration would make monitoring outcomes more directly relevant to public health and ecological protection.

Future research should also strengthen the mechanistic understanding of arsenic mobility under changing environmental conditions. Climate variability, flooding, drought, sea-level rise, salinity intrusion, organic matter loading, land-use change, and mining disturbance can modify redox conditions and arsenic release. For example, flooding may enhance reducing conditions and mobilize arsenic from Fe oxyhydroxides, whereas drying and oxidation may destabilize sulfide-bound arsenic. Seasonal hydrological fluctuations can additionally alter groundwater-surface water interactions, sediment-water exchange, and arsenic transport pathways. Therefore, long-term and process-based monitoring programs are needed

to capture temporal variability rather than relying solely on single-time-point measurements.

Another important future direction is the development of integrated geochemical and predictive models. Models that combine arsenic speciation, pH, redox potential, Fe/Mn cycling, sulfide formation, organic matter degradation, adsorption-desorption processes, hydrological transport, and exposure pathways can improve predictions of arsenic mobility under different environmental scenarios. However, model reliability depends on high-quality analytical data and field validation. Thermodynamic Eh-pH diagrams are useful for conceptual interpretation, but they cannot fully represent kinetic limitations, microbial mediation, colloidal transport, heterogeneous sediment conditions, and temporal variability. Therefore, future modeling efforts should increasingly integrate equilibrium chemistry, reaction kinetics, microbial processes, and long-term field observations.

Risk-based arsenic monitoring should ultimately move toward an integrated framework. Such a framework should include source identification; total arsenic determination; species-specific analysis; characterization of pH, redox potential, dissolved oxygen, iron, manganese, sulfide, phosphate, organic matter, and salinity; assessment of sediment and porewater arsenic pools; evaluation of sediment-water exchange; identification of exposure pathways; and interpretation of toxicity based on speciation and bioavailability. This approach can better distinguish between systems where arsenic is present but relatively immobile and systems where arsenic is actively mobilized and capable of affecting human or ecological receptors.

Overall, the future of arsenic monitoring lies in moving beyond isolated measurements toward integrated, species-specific, process-based, and exposure-oriented assessment. Total arsenic concentration remains an important indicator, but it should be interpreted together with arsenic speciation, geochemical context, sediment-water interactions, analytical uncertainty, bioavailability, and exposure relevance. Such a strategy would improve the ability to diagnose contamination sources, predict secondary release, evaluate remediation requirements, and protect both human and ecosystem health. This risk-based perspective provides the foundation for a more reliable, defensible, and scientifically robust approach to arsenic assessment in aquatic and sedimentary environments.

CONCLUSIONS

Arsenic contamination in aquatic and sedimentary environments remains a major environmental and public-health concern because its occurrence, mobility, toxicity, and persistence are governed by complex interactions among geological sources, anthropogenic inputs, biogeochemical transformations, sediment-water exchange processes, and environmental conditions. This review highlights that the environmental significance of arsenic cannot be adequately interpreted from total arsenic concentrations alone. Instead, arsenic speciation is a critical determinant of mobility, bioavailability, toxicity, and exposure risk. The predominance of As(III), As(V), methylated species, or mineral-associated forms strongly influences arsenic transport, biological uptake, and environmental behavior under varying redox and pH conditions.

Sediment-water interactions represent a fundamental control on arsenic cycling in aquatic systems. Iron and manganese oxyhydroxides, organic matter, sulfide minerals, and porewater processes regulate arsenic retention and release, allowing sediments to function simultaneously as sinks, transformation zones, and secondary contamination sources. Consequently, dissolved arsenic concentrations in water may not fully reflect long-term contamination potential or future mobilization risks. Understanding the coupling among redox dynamics, microbial activity, mineral transformations, and sediment diagenesis is therefore essential for predicting arsenic behavior across environmental compartments.

Advances in analytical chemistry have significantly improved the ability to determine both total arsenic and individual arsenic species. Techniques such as ICP-MS and HPLC-ICP-MS provide powerful tools for environmental assessment, although challenges associated with species preservation, extraction efficiency, matrix interference, and quality assurance remain important considerations. Reliable interpretation of arsenic data requires analytical approaches that preserve species integrity and account for uncertainty throughout the monitoring process.

Overall, effective arsenic assessment should move beyond concentration-based evaluation toward an integrated, species-specific, and risk-based framework. Combining source characterization, arsenic speciation, sediment-water

interactions, geochemical indicators, exposure pathways, bioavailability assessment, and analytical reliability provides a more robust basis for predicting arsenic mobility, identifying secondary release mechanisms, supporting remediation decisions, and protecting both ecosystem and human health in contaminated aquatic environments.

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