Chapter 12
Arsenic Control for Hazard Risk Reduction

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Abstract Arsenic (As) is natural element, which can spread in the environment to a great extent due to human activities. Exposure to As in drinking water and soils has become a global and regional concern. Mining, metal smelting and processing, cultivation of plants, and disposing of wastes from different sources are the main anthropogenic sources of As in the environment. Consumption of arsenic-contaminated water and food are significant exposure pathways for As. Poisoning by As has proven both carcinogenic and noncarcinogenic impacts on human health. New technologies are being developed for As treatment in contaminated water and soil. At this chapter, we discuss geochemical behavior of As in the environment, human health risks of As, and As hazard controls and reduction methods. Several case studies including those performed by authors of this chapter are discussed.

Keywords Arsenic · Hazard · Risk reduction · Contamination · Geochemical behavior · Health risk

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1 Introduction

Exposure to arsenic is an issue of global and regional concern. Arsenic (As) is a ubiquitous potentially harmful metalloid found in rocks, soil, water, sediments, and air. Although constitutes less than 1% of rocks, coals, and soils (Alam et al., 2002), arsenic is known human carcinogen by both inhalation and ingestion exposures (Mahimairaja et al., 2005). Arsenic is the 47th most abundant natural element with average crustal abundance of 2.5 ppm. It is more enriched in the upper continental crust, in marine shales and mudstones, hydrothermal ore deposits, coal, and lignite deposits (Bowell et al., 2014).

Arsenic in the environment can have either geogenic or anthropogenic sources. Human activities can spread geogenic As to a great extent (Tarvainen et al., 2020). Weathering of rocks and hydrothermal and volcanic emissions are the geogenic sources of As in the environment (Mahimairaja et al., 2005). Anthropogenic sources of As are agriculture activities including uses of As-based pesticides, herbicides, and fertilizers and industrial activities such as wood and tannery treatment, painting, chemicals, and electroplating industries. Other anthropogenic sources of As are indiscriminate disposal of domestic (sewage) and industrial wastes, mining activities, and metals smelting and processing activities (Missimer et al., 2018; Li et al., 2020a, 2020b; Bowell & Craw, 2014; Mahimairaja et al., 2005). Infiltration of irrigation water and excessive groundwater exploitation are other anthropogenic resources of As contamination in groundwater (Li et al., 2020a, 2020b). Although the anthropogenic origin of As contamination is increasingly becoming important, extensive As contamination of groundwater in regions such as Bangladesh, West Bengal, and Taiwan is of geogenic sources (Mahimairaja et al., 2005; Ravenscroft et al., 2011; Chen et al., 2013). Geogenic As mobilization to groundwater systems is sometimes complicated multistep geochemical mechanisms which control pH-Eh changes (Russell et al., 2021).

Behavior of arsenic and its toxicity depends on several factors, e.g., its source, speciation, and biogeochemical processes (Bowell et al., 2014). Arsenic is found as major component of As-bearing minerals and minor component or adsorbed or disseminated species in structure of different minerals. Pyrite is one of principal As-bearing minerals (As up to 10 wt%) and is found as an authigenic mineral in sediments of different aquatic environments under strongly reducing condition (Nordstrom, 2000). Pyrite oxidizes in aerobic systems to hydrous iron oxides and release sulfate, acidity, and associated trace constituents such as As (Nordstrom & Archer, 2003). Arsenic can occur in the environment in several oxidation states although it mostly occurs either as the arsenite As(III) and arsenate As(V). Arsenite is more toxic, more mobile, and more bioavailable than arsenate (Amend et al., 2014). The pH-Eh conditions in aquatic environments control the prevalent As speciation and sorption or desorption of As (Sracek et al., 2004). Due to redox disequilibrium, both arsenite and arsenate may occur together in water (Bowell et al., 2014). Organic As species are rarely quantitatively determined in soil and surface and groundwater. Reduction-oxidation reactions during metabolism in body of living organisms are causing conversion of arsenite and arsenate and methylation of As(III) to yield methylated arsenic species (Rasheed et al., 2016). The most

Table 12.1 Inorganic and organic arsenic species

Arsenic type	Species	Abbreviation
Inorganic arsenic	Arsenate (arsenic acid)	As^{+5}
	Arsenite (arsenous acid)	As^{+3}
Organic arsenic	Monomethylarsonic acid or methylarsonic acid	MMA^{+5}
	Monomethylarsonous acid or methylarsonous acid	MMA^{+3}
	Dimethylarsinic acid	DMA^{+5}
	Dimethylarsinous acid	DMA^{+3}
	Arsenobetaine	AsB
	Arsenocholine	AsC
	Arsenosugars	–

Rasheed et al. (2016)

prevalent inorganic and organic arsenic compounds found in water, food, soil and hair, urine, and nail of humans as biomarkers are listed in Table 12.1 (Rasheed et al., 2016).

Both organic and inorganic species of As are strongly absorbed across the human gastrointestinal tract (Mahimairaja et al., 2005). There are several medical symptoms of arsenicosis including, e.g., melanosis, leucomelanosis, keratosis, hyperkeratosis, dorsum, non-petting edema, gangrene, and skin and internal cancer (Alam et al., 2002; Shankar et al., 2014; Smedley & Kinniburgh, 2002; Choong et al., 2007; Charlet & Polya, 2006). Arsenic poisoning is reported from more than 70 countries (Ravenscroft et al., 2011).

Potential routes of exposure to arsenic are ingestion of foods and drinking water, dermal exposure, and inhalation of aerosols containing arsenic (Missimer et al., 2018). Among various sources of As in the environment, drinking water probably poses the greatest threat to human health (Smedley & Kinniburgh, 2002). The impact of airborne arsenic may be difficult to detect in not highly contaminated regions due to very low concentrations observed in atmosphere compared to soil (Bowell et al., 2014). However, some studies show that anthropogenic airborne emission has the highest arsenic hazard quotient and cancer risk in the regions where geogenic arsenic exposure is limited because the use of contaminated groundwater is well controlled (Chen et al., 2013) or occur in industrial non-geogenic contaminated region (Tavakoli et al., 2020).

Gold mining and processing are identified as the sources of some of the highest recorded concentrations of As in water (Coudert et al., 2020). Exploitation of low-grade refractory As-bearing sulfide minerals (e.g., arsenopyrite, and arsenian pyrite) led to the production of As-rich effluents through the oxidation of gold ores, via hydrometallurgical routes, and/or release of As from mine tailings due to their exposure to air and water (Coudert et al., 2020; Wang et al., 2019). The weathering of As-rich sulfide residues of gold mining is another source of As contamination in environment.

In this chapter, the following subjects are discussed: (a) how human health risk of arsenic is evaluated in contaminated regions and (b) which methods are used for As remediation of polluted soils and water and for hazard risk reduction in the environment (Eslamian & Eslamian, 2021).

2   Assessment of Human Health Risk of Arsenic

Cancer is the most common cause of death in the world, and environmental pollutants are one of main responsible factors (Karakurt, 2019). Arsenic-contaminated water and food (agriculture products irrigated with polluted water or food prepared and cooked with polluted water) are significant exposure pathway for arsenic (Juhasz et al., 2006; Rasheed et al., 2016). The risk of arsenic exposure for an individual depends on many factors, such as water intake and body weight (Chen et al., 2018; Li et al., 2020a, 2020b; Yang et al., 2018). Contaminant bioavailability referred to as the fraction of administered dose of contaminant that reaches blood from the gastrointestinal tract is an important criterion used in human health risk assessment (Ruby et al., 1999). Arsenic speciation is the main control of arsenic bioavailability. Most of arsenite and arsenate species can be absorbed in the body and transported via the blood stream to the body tissues (De Capitani, 2011). Arsenic bioavailability in rice, the staple food of many people in the world, depends on arsenic speciation, which depends on rice crop, arsenic concentration, and its speciation in irrigation water and in cooking water. Juhasz et al. (2006) determined the speciation of arsenic in greenhouse-grown and supermarket rice and determined arsenic bioavailability in cooked rice using swine in vivo model. Results indicate that in supermarket rice, arsenic was found only in the inorganic form but was in dimethylarsinic form in greenhouse-grown rice. Due to low bioavailability of dimethylarsinic acid, only 33 ($\pm$3)% of the total rice-bound arsenic was bioavailable. In contrast, in supermarket-bought rice cooked in water with sodium arsenate, arsenic was found only in the inorganic form, and its bioavailability was much higher, reaching 89 ($\pm$9)%.

Assessing As health risk for humans only by comparing the concentration of total As (ignore speciation) with safe limits in drinking water (10 μg L^{-1}) or in ingested food is not reliable and comprehensive approach. Recent study by Ahmad et al. (2020e) showed that even the WHO guideline for drinking water (10 μg L^{-1} As) is not sufficiently protective and does not correspond to the excess lifetime cancer risk of 10^{-6} in the Netherlands (considered as virtually safe dose according to Dutch policy). Based on their calculations, exposure to each additional μg L^{-1} As through drinking water can increase the risk for lung cancer development in the Dutch population by 0.025%.

There is a need to know more about arsenic fate after input to soil through irrigation water in water-soil-plant system especially from the perspective of arsenic speciation and fractionation changes for evaluation of arsenic health risk in soil and crops for human. There are different approaches for bioavailability of soil As. Rahman et al. (2017) evaluated human bioavailability of As in old and recent arsenical pesticide-contaminated soils using an extraction test replicating gastric conditions, an operationally defined bioavailability extraction test with 1.0 M HCl and in vivo bioaccumulation test using earthworms (Fig. 12.1).
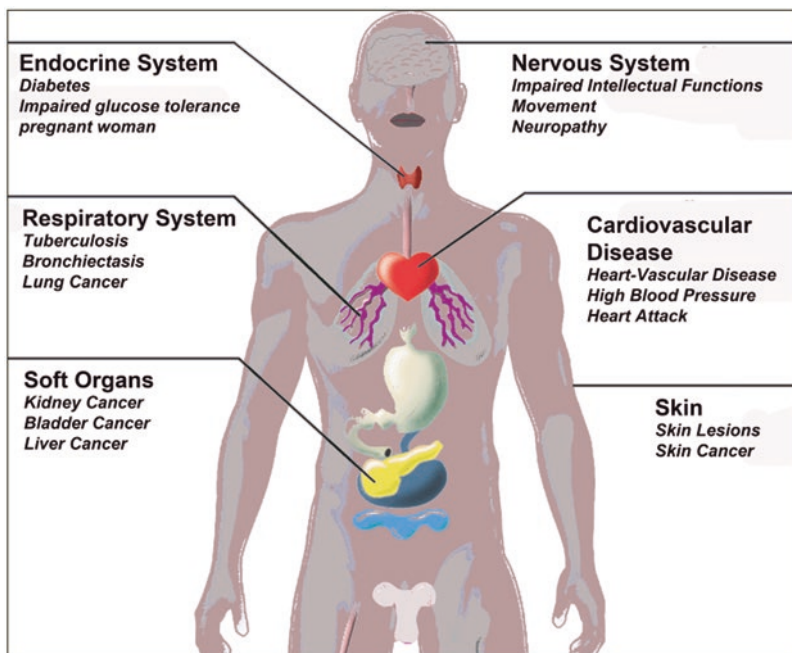
Fig. 12.1 Adverse health effects due to chronic As exposure through drinking water (Ahmad et al., 2020e)

3   Arsenic in Soil and Water Resources

Arsenic above the European Union (EU) recommended maximum acceptable limit for agricultural soil (20 mg/kg) has been associated with mining activities and the metal processing industry (Tarvainen et al., 2020) and contaminated groundwater used for irrigation (Bhattacharya et al., 2012). In addition, As can be enriched significantly in soils from past use of arsenical pesticides (Yokel & Delistraty, 2003). High loading rates of arsenical pesticides, coarse soil texture, low organic matter content, and use of irrigation promote deeper movement of As into soil and even contamination of underlying shallow groundwater (Peryea & Creger, 1994). However, atmospheric pollution and the phosphate fertilizers application are regarded as the main sources of anthropogenic arsenic in agricultural soils (EFSA Panel on Contaminants in the Food Chain (CONTAM), 2009).

Sequential extraction procedure often revealed As fractionation in soil which controls bio-accessibility and bioaccumulation of As. Soluble, exchangeable, and sorbed fraction of total As are considered as bioavailable and bioaccumulative As in soils. Bioavailability and bioaccumulativity of As decreased due to aging of arsenical pesticide-contaminated soils (Quazi et al., 2010; Rahman et al., 2017). Arsenic

release from mobile (bioavailable) fractions contributes to its content in immobile fractions such as crystalline Fe(oxide-hydroxide) and residuals. Organic-rich soils often have high As concentrations (Missimer et al., 2018; Li et al., 2020a, 2020b). In anaerobic soils, As is typically found in combination with sulfur. In aerobic soils arsenate is the predominant species, whereas in anaerobic soils, arsenite is the dominant species (Campbell & Nordstrom, 2014). However, in soil and sediments, in metastable As-bearing phases, and under the impact of anaerobic microbial activities, speciation of As does not frequently adjust to pH-Eh condition (Xu et al., 2011; Khosravi et al., 2019).

Data about As concentration, its fractionation, and speciation and physiochemical characteristics of soil such as pH-Eh condition, texture, organic matter contents, and age of contamination can be useful for successful remediation and management. The default Soil Cleanup Target Level (SCTL) that varies greatly by state and country depends on background concentrations and is determined by regulatory agencies according the site-specific risk assessments (Missimer et al., 2018).

While there are anthropogenic sources of arsenic, geological weathering is the primary cause of arsenic release into groundwater (Rasheed et al., 2016). For example, several million people are at risk from drinking As-contaminated water in West Bengal (India), Bangladesh, China, Vietnam, Taiwan, Chile, Argentina, and Mexico (Mahimairaja et al., 2005; Ravenscroft et al., 2011; Bretzler & Johnson 2015). People with hard manual work in tropical regions surpass the average daily water intake by a factor of 2–3, and for them the limit should still be decreased (Chakraborti et al., 2010). People drinking As-contaminated water over prolonged periods often show typical arsenical lesions, which are manifestations of As toxicity (Mahimairaja et al., 2005). In response to health concerns about arsenic in drinking water in the United States, the US Environmental Protect Agency (USEPA) reduced the drinking water standard for arsenic from 50 μg L^{-1} to 10 μg L^{-1} in 2001 which also matches the World Health Organization health-based recommendation (U.S. Environmental Protection Agency (USEPA), 2001).

4 Arsenic Removal from Contaminated Soil and Water

In recent years, there have been extensive efforts to find effective and economical methods for As removal from contaminated soils and water in all over the world. The methods can be divided according to their mechanism to seven major classes including: ion exchange, phytoremediation, adsorption, phytobial remediation, chemical precipitation, electrokinetic technics, and electrocoagulation (Alka et al., 2020b). Each method has its advantages and disadvantages (see Table 12.2). Ion exchange, chemical precipitation, electrocoagulation, and membrane technology have been used for As removal from water, wastewater, and synthetic water. On the other hand, adsorption, phytoremediation, nanophytoremediation, phytobial

Table 12.2 Advantages and disadvantages of methods used for As removal from the soil and water system

Method of removal	Advantages	Disadvantages
Adsorption	Easy operation handling; flexibility; low cost; sludge-free; high removal efficiency	Sorbents have to be replaced after adsorption bed gets and loses the capacity for removal; sorbents have low specific surface area when using metal oxides; suitable for water with low as concentrations
Ion exchange	Total removal; limited toxic Sludge production	Must be rejuvenated frequently to ensure complete removal; costly; each exchanger is specific for different as species; the resin is also responsive to natural anions
Phytoremediation	Eco-friendly and highly economic; occurs by using plants to assimilate arsenic from soil; land restoration	Time-consuming process; climate affects hyperaccumulating plants; microbes produce additional pollutants; pollutants may interact with metabolic processes of plants and hinder their growth
Nanophytoremediation (NP)	Improves phytoremediation efficiency and in situ remediation; transforms pollutants into less toxic forms; cost-effective	–
Phytobial remediation	Eco-friendly and cheap; Enhances phytoremediation Rate; increases plant defense responses to stress; assists in control of phytopathogens and plant growth	–
Chemical precipitation	Simple and effective; specific components removed	It forms residual products and processing cost is high
Electrokinetic processes	Cost-effective in removal of heavy metals	Small portion of soil at one time is treated
Electrocoagulation	A novel and promising arsenic removal strategy; efficient and relatively cheap Operates with local materials	Not effective for extracting trivalent arsenic; producing sludge; strongly affected by coagulant form and dose, solution pH and competing anions
Membrane technology	High efficiency; low energy Consumption; high filtration performance	Costly, large volumes of contaminated water

Adapted from Alka et al. (2020b)

Case Study 1: Arsenic hazard from Domestic Well Water

Arsenic concentration in domestic wells needs to be measured and controlled because they are the major sources of water in some regions of many countries. Any changes in pH-Eh condition in groundwater system can lead to geochemical disequilibrium in aquifers and release of contaminants such as As due to dissolution/precipitation of oxides (hydroxides) or sulfides, which are hosts of different trace elements. This can have negative implications on home water treatment such as reduced contaminant removal efficiency and iron fouling, which can lead to contaminant exposure from domestic well water (Erickson et al., 2021).

For example, according to the study of 250 new domestic wells in Minnesota, North Central United States, well installation changes geochemical conditions in aquifers for more than 12 months. In one well, which had extremely high initial arsenic about 1550 μg L^{-1}, contamination decreased after 15 months to 5.2 μg L^{-1}. Erickson et al. (2021) investigated reasons for increase of As concentration after well installation. They found that well installation procedures introduce oxic drilling fluids and hypochlorite, a strong oxidant used for disinfection, thus inducing geochemical disequilibrium. The oxidation of arsenic-containing sulfides (which lowers pH) combined with low pH dissolution of arsenic-bearing Fe (oxyhydr)oxides caused the very high observed arsenic concentration.

remediation, and electrokinetic processes have been used for both water and soil remediation.

The restrictions for soil and water treatment technologies, local and national authorities' requirements, a country's development stage, and local regulations for arsenic levels determine which method can be used for arsenic treatment (Alka et al., 2020b).

4.1 Ion Exchange

Ions retained electrostatically on the solid phase surface can be exchanged with ions of similar charge in solution. This effective method is used to remove contaminants such as arsenate from contaminated water (or wastewater, Alka et al., 2020). It should be noted that this method extracts only arsenate (not arsenite) from water due to its negative charge in water (unlike dissolved arsenite in water which often is neutral, Jadhav et al., 2015). In this method, different synthetic resins are typically used as exchanger (Lee et al., 2017). Resins are regularly regenerated to displace the exchanged ions after application period. The ion exchange process does not generate large amount of waste and is affordable (Lee et al., 2017). Increase of TDS in water leads to decreasing in efficiency of arsenic removal (Jadhav et al., 2015).

Case Study 2: Use of ArsenX^{np}, a Hybrid Anion Exchanger for arsenic Removal in Remote Villages in the Indian Subcontinent

Highly As-contaminated groundwater used as drinking water is the cause of widespread arsenic poisoning affecting nearly 100 million people living in Bangladesh and West Bengal. Arsenic concentration exceeds the maximum contaminant level of arsenic (50 μg L^{-1}) in drinking water in India. In thousands of villages in Indian subcontinent, arsenic-rich groundwater is the only viable source of drinking water.

Sarkar et al. (2007) investigated the performance of hybrid anion exchangers which are essentially spherical anion exchange resin beads containing dispersed nanoparticles of hydrated ferric oxide (commercially available as ArsenX^{np}) for arsenic removal over a long period. In addition, they investigated the regeneration of the media, elucidated arsenic removal mechanism, and also studied strategies of arsenic remediation in Indian subcontinent.

The sorption columns used in the field for removal of arsenic are either single columns or split columns, which allow entry of atmospheric oxygen to promote the oxidation of dissolved Fe(II) species in arsenic-contaminated well water to insoluble Fe(III) oxides or hydrated ferric oxide particulates (Fig. 12.2). Apart from the usual role played by the sorbents like ArsenX^{np} or
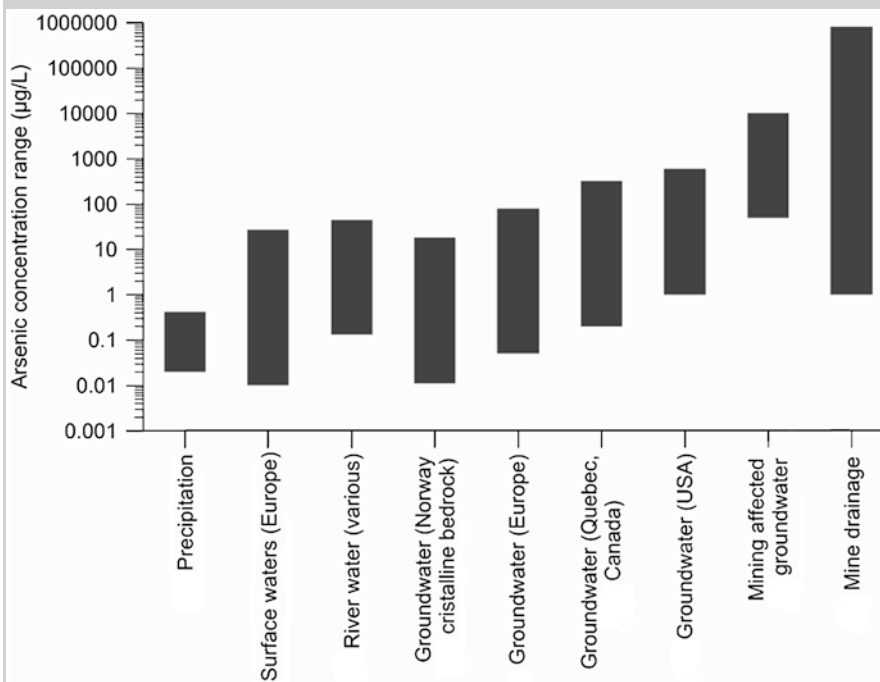

Fig. 12.2 Arsenic concentration ranges in water resources. (Adapted from Coudert et al., 2020)
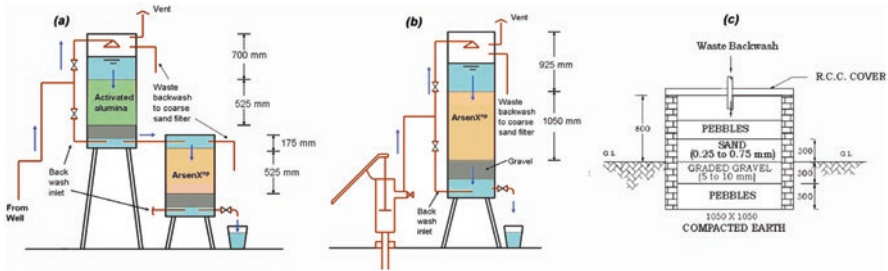
Case Study 2: *Continued*

Fig. 12.3 Schematic detail of construction and operation of a (**a**) split-column unit and (**b**) single unit used in the field, (**c**) details of the coarse sand filter for entrapment of waste back-wash containing hydrated ferric oxide particles. (Sarkar et al., 2007)

activated alumina, hydrated ferric oxide particulates also improve the treatment process. Each As removal system is attached to a hand pump-driven well and is capable of providing arsenic-safe water for approximately one thousand people in villages. No chemical additives, adjustment of pH, or electricity is required to run these systems. Every system was running for more than 20,000 bed volumes before a breakthrough of 50 μg L^{-1} of arsenic was reached. Upon exhaustion, the media was withdrawn and taken to a central regeneration facility where 2% NaCl and 2% NaOH solutions were used for regeneration. After regeneration, the spent solutions, containing high arsenic concentration, were transformed into solid residuals and were disposed safely to avoid any significant arsenic leaching. Laboratory investigations have confirmed that the regenerated ArsenX^{np} was amenable to reuse in multiple cycles without any significant loss in capacity (Fig. 12.3).

4.2   Phytoremediation

Phytoremediation uses hyperaccumulating plants with extensive root system, high tolerance, and rapid growth to remove pollutants, and the technique requires very little nutrient input and is easy to manage (Manoj et al., 2020). This method is further classified as phytoextraction, phytostabilization, phytofiltration, and phytovolatilization, which are based on the metal uptake and transport routes (DalCorso et al., 2019). However, a plant may use more than one strategy simultaneously (Favas et al., 2014) (Fig. 12.4).

The efficiency of As removal by phytoremediation method can be enhanced by using chelators, inoculation by microbes, (Irshad et al. 2020a, 2020b), soil amendments (Irem et al., 2019), and introduction of nanoparticles (Moameri & Khalaki, 2019). For example, inoculation with the strain of *Cupriavidus basilensis* could increase the abundance of aioA-like genes in the rhizosphere and As accumulation in *P. vittata* by up to 171% (Yang et al., 2020). Nanophytoremediation (NP) that combines phytoremediation with nanotechnology for contaminants remediation is
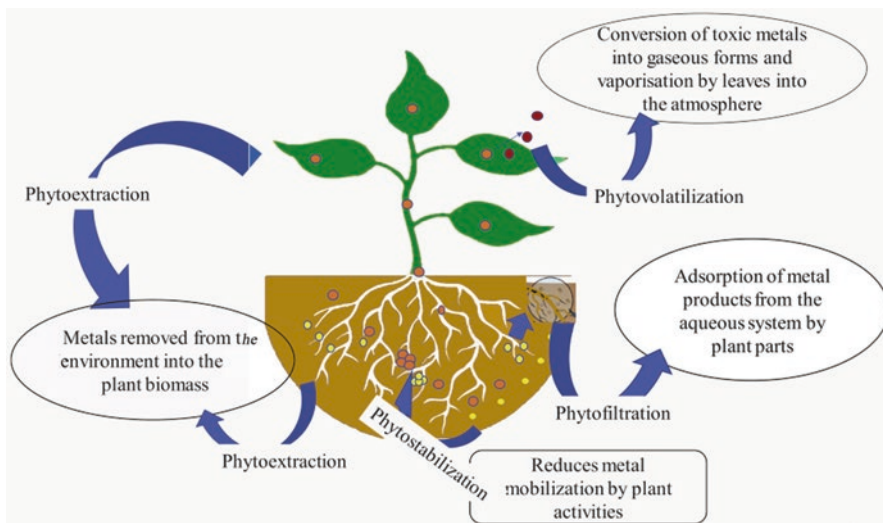
Fig. 12.4 Description of phytoremediation processes occurring in plants. (Modified from Alka et al. (2020))

known as a green and eco-friendly technology. Nanomaterials can eliminate the need for treatment, reduce the cleanup time, and remove and dispose toxic substances from soils and water (Chen et al., 2021). However, few studies exist on the removal of arsenic using nanophytoremediation and underlying mechanisms governing the synergistic removal bioremediation technologies (Alka et al., 2020b).

Case Study 3: As- and Cd-Contaminated Soil Remediation with Multi-Walled Carbon Nanotubes Combine with Hyperaccumulator Solanum Nigrum L.

Nanomaterials have been increasingly applied for the remediation of contaminated soils, but few researches have been reported on the complex interactions of nanomaterials with heavy metals in phytoremediation. Some studies give a strong evidence to promote the phytoremediation for As-contaminated soils by using nanomaterials.

Chen et al. (2021) have conducted a pot experiment to investigate the effects of different doses of multi-walled carbon nanotubes on the plant growth and accumulation of Cd and As in hyperaccumulator plant. Hyperaccumulator of Cd *Solanum nigrum L.* (*S. nigrum*) was cultivated in Cd- and As-contaminated soils amended with carbon nanotubes at 100, 500, and 1000 mg kg^{-1} for 60 days. Root and leaf growth was inhibited by dose of 1000 mg kg^{-1} carbon nanotubes. However, the application of carbon nanotubes in doses of 100 and 500 mg kg^{-1} has increased the shoot length and plant dry biomass by 5.56–25.13% and 5.23–27.97%, respectively. Meanwhile, multi-walled carbon nanotubes at 500 mg kg^{-1} significantly enhanced the accumulation of As

by about 32.47% in *S. nigrum* and alleviated Cd- and As-induced toxicity, by motivating plant growth, stimulating antioxidant enzymatic activities, and increasing micronutrient content ($p < 0.05$). The bioconcentration factor of As decreased (15.31–28.08%) under carbon nanotubes application, which played an important role in the alleviation of phytotoxicity. Besides, bioavailable Cd and As were reduced in rhizosphere soils and the most significant reduction 16.29% for Cd and 8.19% for As was found in 500 mg kg^{-1} carbon nanotubes treatment. Generally, these findings demonstrated that suitable concentration of carbon nanotubes can enhance remediation efficiency in As- and Cd-contaminated soils.

4.3   Adsorption

Adsorption is a suitable As removal technique that is highly efficient for arsenite and arsenate remediation (>95%, Alka et al., 2020). Since it has low cost and do not need so skilled personnel to run treatment procedure, it can be used for arsenic removal from drinking water in rural and peri-urban areas in developing countries (Kumar et al., 2019). Adsorbent type, initial concentration of As, speciation and interfering species, exposure period, pH, and temperature influence the efficiency of this technique of As removal (Litter et al., 2010; Sarkar & Paul, 2016). The oxidation process of arsenite by atmospheric oxygen is very slow, and at pre-treatment step of this method, arsenite is converted to arsenate using different oxidants such as ozone (O_3), hypochlorite (HClO), potassium permanganate ($KMnO_4$), and hydrogen peroxide (H_2O_2) (Ahmad et al., 2018; Ahmad et al., 2020a, 2020b, 2020c). Arsenic adsorbents have been successfully applied for arsenic remediation including granular adsorbents, metal oxides coated on sand, iron oxide-based sorbents engineered biochar, zero-valent iron, clinoptilolite-rich zeolitic tuff, iron/olivine composite, activated carbon, agriculture wastes, and activated alumina (Bundschuh & Chatterjee, 2013; Bretzler & Johnson 2015; Alka et al., 2020b). Different adsorbents materials including the composite materials and nanomaterials have been classified by Maity et al. (2020) according to their application in laboratory or in field, type of water (surface water, groundwater, drinking water, and waste water), pH, initial As concentration, and the percent of arsenite and arsenate removal. See this article for further details about different As adsorbents. They concluded that especially Fe nanoparticles and/or composite materials are quite effective for the As treatment process.

Case Study 4: Removal of Arsenic Using Natural Geological Materials at Chaco-Pampean Plain in Argentina (Bundschuh & Chatterjee, 2013)
There are high concentrations of As in groundwater around Santiago del Estero in Chaco-Pampean Plain in NW Argentina. Groundwater is of Na-HCO$_3$ type and frequently shows high pH values. Redox status is generally oxidizing and As is present as As(V). Local population is poor and there is a need for low- or zero-cost As removal material. Several natural materials have been tested including soils rich in clay from Santiago del Estero region and lateritic soils from Misiones Province in the north of Argentina. Adsorption experiments performed in batch mode showed much higher As removal for lateritic soils from Misiones (up to 99%) compared to soils from Santiago del Estero (40–53%). This was consistent with lower oxalate extractable Al and Fe soils. Potential limiting factor can be the cost of lateritic material transport from Misiones to Santiago del Estero.

Case Study 5: Field Removal from Natural Water by Zero-Valent Iron Assisted by Solar Radiation
The towns of Camarones, Esquiña, and Illapata are in the Atacama Desert in northern Chile. Local residents use water from the Camarones River (TDS ranges between 1 and 2 g L^{-1}) for both human consumption and agricultural activities. The concentration ranges between 1000 and 1300 µg L^{-1}, and is mainly in the form of As (V). High As concentration in river has chronically affected the rural populations living near the river and caused a variety of health problems. In addition, high concentrations have been reported in the soil, plants, and animals in the area. In situ removal method was applied to this highly contaminated water by Cornejo et al. (2008). High removal efficiency (above 99%) was necessary to obtain the World Health Organization (WHO) recommended level of 10 µg L^{-1}. Cornejo et al. (2008) approach was based on the use of steel wool, lemon juice, and solar radiation. They used surface method analysis to optimize the amount of zero-valent iron (steel wool) and the citrate concentration (lemon juice). The optimal conditions when using solar radiation were 1.3 g L^{-1} of steel wool and 0.04 mL (one drop) of lemon juice. Under these conditions, removal percentages were higher than 99.5%, and the final concentration was below 10 µg L^{-1}. The main characteristics of method were highly effective removal, easy to use, and inexpensive implementation.

4.4 Phytobial Remediation

The toxic metals cannot be degraded entirely from the environment, but they can be accumulated by microorganisms, and the uptake of metals by bacteria can take place (Harms et al., 2011). In phytobial method, plants and microbes including algae, bacteria, and fungi are combined to mitigate soil and groundwater As (Roy et al., 2015). It is cost-effective and is widely accepted by society (Sodhi et al., 2019). Under phytoremediation technology, heavy metal hyperaccumulator plants have been extensively employed to extract high concentrations of heavy metals, but slow growth, limited biomass, and stresses caused by heavy metals decreased the efficiency of hyperaccumulators (Asad et al., 2019). Recently, plant growth-promoting bacteria that assists phytoremediation have been applied for both improving plant metal tolerance and promoting plant growth while achieving the goal of large-scale removal of As (Alka et al., 2020a). Plant growth-promoting rhizobacteria produce several metabolites, including growth hormones, siderophores, and organic acids, which aid in solubilization and provide essential nutrients (e.g., Fe and Mg) to the plants (Asad et al., 2019). Although phytobial remediation of As was proved a very successful technology, there are various pathways of As tolerance mechanisms not fully understood (Irshad et al., 2021). Arsenite oxidizing bacteria such as *Alcaligenes faecalis* and arsenate reducing bacteria including of *Sulfurospirillum arsenophillum* and *Sulfurosprillum barnesii* are examples of bacteria involved in bioremediation of arsenic (Sodhi et al., 2019).

The impacts of symbiosis between microorganisms and plants on As remediation and detoxification were recently investigated by Irshad et al. (2020b), Roy et al. (2015), Zeraatkar et al. (2016), Guarino et al. (2020), and Wang et al. (2020).

Case Study 6: Alleviation of Arsenic-Induced Phytotoxicity of Rice Plant Using Groundwater Inhabited by Bacillus and Paenibacillus Strains
Arsenic contamination in agricultural soil is causing several hazardous health effects through percolation in food chain. Rice plants are being affected more than other agricultural crops due to the use of arsenic-contaminated groundwater for irrigation, their higher arsenic uptake, and mobilization tendency (Kalita et al., 2018).

A hydroponic experimental setup has been conducted by Banerjee et al. (2020) for evaluating the effects of two potent arsenic-tolerant bacterial strains including *Bacillus thuringiensis A01* and *Paenibacillus glucanolyticus B05* for possible mitigation of the arsenic-induced phytotoxicity and to maintain overall growth of the rice seedlings. Miniket cultivar-IR-50 widely cultivated in West Bengal was used in this study. Their results show that:

- *Bacillus thuringiensis A01* could reduce arsenic uptake up to 56% (roots) and 85% (shoots) and *Paenibacillus glucanolyticus B05* up to 31% (roots) and 65% (shoots) in a hydroponic environment.

- Germination percentage has been enhanced significantly.
- Expressions of oxidative stress defensive enzymes such as superoxide dismutase, peroxidase, and catalase have been augmented at seedling stages (21 days) toward detoxification of arsenic imposed excess ROS generation.
- There was increment of leaf thiobarbituric acid reactive substances.
- Phenolic and flavonoid mediated free radical scavenging ability of the test plants increased significantly.

This study results revealed that selected bacterial strains could perform efficient bioremediation of arsenic-contaminated rice cultivation.

4.5   Chemical Precipitation

Chemical precipitation is a technique that uses reagents such as ferric salts, sulfides, magnesium salts, and calcium salts for removal of heavy metals like arsenic (Alka et al., 2020b). These reagents help in removing arsenic by converting dissolved arsenic to low-solubility compounds. The most common reagents used in the removal of As especially in As-rich wastewater are ferric arsenate and calcium arsenate (Alka et al., 2020b). Treatment of arsenic-rich acid wastewater produced in the mining and smelting process with iron salt and lime is an economical and effective method (Wang et al., 2019).

Also, ferric-based coprecipitation low-pressure membrane filtration is a promising As removal method. Coprecipitation of arsenate with $Fe(III)$ (oxyhydr)oxides is a widely used As removal method. However, arsenite coprecipitation with $Fe(III)$ (oxyhydr)oxides is less effective in the pH range of most groundwaters because arsenite is uncharged and has a significantly lower affinity for adsorption to $Fe(III)$ (oxyhydr)oxide surfaces (Ahmad et al., 2020c). Laboratory investigations indicated that As coprecipitation with $Fe(III)$ (oxyhydr)oxides rapidly reached equilibrium before membrane filtration, within 1 min. Therefore, As removal efficiency was not improved by increasing water residence time (Ahmad et al., 2020c). The As removal rate increased with increasing the Fe/As ratio in water (Wang et al., 2019). Also, a higher $Fe(III)$ dose was required to reduce $As(V)$ to sub-µg/L levels for feedwater containing high concentration of oxyanions such as phosphate and silicate and low concentration of cations such as calcium (Ahmad et al., 2020c). Even stability of amorphous coprecipitates is usually poor if the Fe/As ratio is lower which can release As after disposing the coprecipitates (Wang et al., 2021).

4.6   Electrokinetic Technics

The electrokinetic technique (EK) is an innovative, in situ, and effective method that guides the movement and transport of free pollutants in soil within electrical field in processes such as electrophoresis, electromigration, water electrolysis, and electro-osmotic flow (Xu et al., 2019). The EK has limitations in removing As because dissolved As is difficult to treat, and using this method increases the risk of mobility of other heavy metals (Alka et al., 2020b). Coupling with other techniques may increase this method efficiency, making it more economically and environmentally friendly. The EK method can be coupled or combined with permeable reaction barrier (PRB, Yuan & Chiang, 2007; Yao et al., 2020; Ji et al., 2020), anaerobic bioleaching (Lee et al., 2009; Kim et al., 2012), application of chemical reagents (Yuan & Chiang, 2008), humus, and humic and fulvic acids (Li et al. 2020a, 2020b; Xu et al., 2021).
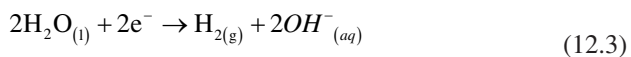
4.7   Electrocoagulation

Electrocoagulation (EC) process is a very efficient and economic tool for removing various water pollutants such as turbidity, phosphate, fluoride, and various heavy metals (Nidheesh & Singh, 2017). The EC process is also effective promising technology for removal of arsenic from water (Mohora et al., 2018; Demirbas et al., 2019) and other solutions (Alka et al., 2020b). Electrocoagulation is using various sacrificial metal anodes such as aluminum, iron, magnesium, zinc, etc. for in situ generation of metallic coagulants. Iron and aluminum are more common to use in EC method, and aluminum anodes are less efficient than iron electrodes (Alka et al., 2020b). The electrolytic oxidation of the anode occurs after the application of direct current and the metallic anode dissociates into di- or trivalent metallic ions (Nidheesh & Singh, 2017). The EC is using electrical energy destabilize the colloidal suspensions, leading to the dissolution of different heavy metals and metalloids including As and then their flotation and flocculation (Maitlo et al., 2019).
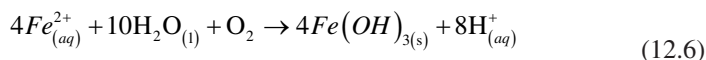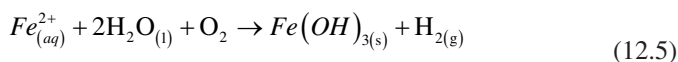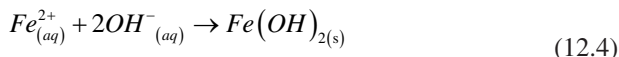
The EC process is started when a potential is applied through an external power source and the sacrificial electrode undergoes oxidation as given below for mild steel anode (Balasubramanian et al., 2009):

$$Fe_{(s)} \rightarrow Fe^{2+}_{(aq)} + 2e^-$$

$$(12.1)$$

and the cathodic reactions can be written as:

$$2H^+ + 2e^- = H_2$$

$$(12.2)$$

$$2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH^-_{(aq)}$$

$$(12.3)$$

Release of OH^- at the cathode continues due to the formation of cationic monomer species attributable to the dissolution of electrolyte that occurs at the sacrificial mild steel anode (Demirbas et al., 2019):

$$Fe^{2+}_{(aq)} + 2OH^-_{\,(aq)} \rightarrow Fe(OH)_{2(s)}$$

(12.4)

$$Fe^{2+}_{(aq)} + 2H_2O_{(l)} + O_2 \rightarrow Fe(OH)_{3(s)} + H_{2(g)}$$

(12.5)

$$4Fe^{2+}_{(aq)} + 10H_2O_{(l)} + O_2 \rightarrow 4Fe(OH)_{3(s)} + 8H^+_{(aq)}$$

(12.6)

Arsenate absorbed/precipitated by forming complex with Fe^{3+} as below:

$$4Fe(OH)_{3(s)} + AsO^{3-}_{4(aq)} \rightarrow \left[Fe(OH)_3 * AsO^{3-}_4 \right]_{(s)}$$

(12.7)
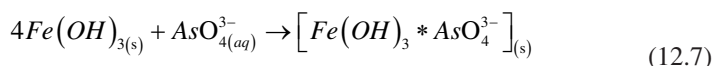
Performance of the EC method depends on (a) speciation of As, (b) materials used in cathode and anode, (c) anions and cations concentrations in water, and (d) pH (Nidheesh & Singh, 2017). The removal of arsenate is easier than that of arsenite, and arsenate can be completely removed (Nidheesh & Singh, 2017). Then oxidation of arsenite to arsenate occurs during the EC process (Kumar et al., 2004). Testing of As removal efficiency through the EC process with different anode-cathode pairs of iron and aluminum implies that Al-Fe and Fe-Fe systems are more efficient compared to Al-Al system (Gomes et al., 2007). Presence of iron in water has positive impact, but fluoride, phosphate, silicate, boron, and bicarbonate decrease the efficiency (You & Han, 2016; Wan et al., 2011; Silva et al., 2018; Goren & Kobya, 2021). Silicate and phosphate ions affect performance more than boron, bicarbonate, and fluoride (Goren & Kobya, 2021). Generally, sulfate presence in water didn't affect the As removal efficiency by the EC process at low concentration. However, when sulfate concentration increased to 100 mg L^{-1}, As removal efficiency decreased significantly (Wan et al., 2011; You & Han, 2016). Similar to sulfate, the impacts of magnesium and calcium cations in water on efficiency of As removal by EC process are dependent on their concentrations in water (You & Han, 2016; Hu et al., 2014). High pH and high initial arsenic concentration decrease the As removal (Wan et al., 2011). For example, Can et al. (2014) study shows that the highest removal efficiency was observed at initial pH = 4, and You and Han (2016) observed the fastest removal efficiency at pH = 7. Also, the external addition of air (O_2) enhanced the efficiency of electrocoagulation process and improved As removal (Syam Babu et al., 2021). The obtained experimental results showed that the efficiency of arsenic removal increased with increasing current density (Can et al., 2014).

4.8   Membrane Filtration

Membrane technology is addressed as a pressure-driven process, widely known as one of the most efficient technologies, and it has the potential of reducing arsenic concentration by 96% (Alka et al., 2020b). High pressures are required to force the water to pass across a membrane to change water from concentrated to diluted. Driving pressure increases as selectivity increases (Choong et al., 2007). Laboratory investigations by Nguyen et al. (2009a, 2009b) implied that the arsenic separation efficiency increased only by a few percent with increasing applied pressure (from 138 to 552 kPa). Membranes are commonly divided into four categories based on increasing selectivity: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and hyperfiltration or reverse osmosis (RO) (Choong et al., 2007). The MF can be used to remove bacteria and suspended solids (pore sizes of 0.1 to micron) and UF to remove colloids, viruses, and certain proteins (pore size of 0.0003–0.1 microns), and NF relies on physical rejection based on molecular size and charge (pore sizes are in the range of 0.001 to 0.003 microns). The RO uses pore size of about 0.0005 microns and could be used for desalination. Different types of membranes are used in removal of arsenic from water systems (Pramod et al., 2020), such as NF (Gonzalez et al., 2019), UF (Ahmad et al., 2020c), and RO (Schmidt et al., 2016). The NF and RO are the most promising technologies for selective removal of arsenic (Figoli et al., 2020). Cake-layer formation is the predominant membrane fouling mechanism (Ahmad et al., 2020c). The molecular weight cutoff (MWCO), electrokinetic charge, and individual salt rejection characteristics are the important membrane properties governing the separation of arsenic (Nguyen et al. 2009a, 2009b). Removal of arsenic by membrane filtration is highly dependent on As speciation and properties of membranes (Nguyen et al. 2009a, 2009b). Investigations imply that the removal efficiency of MF was low and only 37% of As(III) and 40% of As(V) were removed by MF (PVA membrane, Pure-Envitech, Korea) due to its large pore size (Nguyen et al. 2009a, 2009b). However, the removal efficiency of MF increased dramatically up to 90% for As(V) and 84% for As(III) when an amount of 0.1 g L^{-1} (nZVI) was added into arsenic solution (Nguyen et al. 2009a, 2009b).

The performance of the NF is better for removing of As(V) than As(III) (Nguyen et al. 2009a, 2009b) which results in the dominance of Donnan exclusion over steric exclusion in controlling the As removal capacity of the membrane (Nguyen et al. 2009a, 2009b).

Quality of raw water is important factor, which controls coagulation/MF (C/MF) hybrid method. Zhang et al. (2012) show that increase of dissolved organic carbon and HPO_4^{2-} and HCO_3^- concentrations would moderately decrease As(V) removal by (C/MF). However, ions such as Cl^-, NO^{3-}, F^-, SO_4^{2-}, K^+, Ca^{2+}, and Mg^{2+} have little effect on As(V) removal. However, in As removal by NF membrane, the rejection of monovalent As(V) increased by the presence of Cl^- and decreased by the presence of SO_4^{2-} due to mutual interactions between anions (C. M. Nguyen et al., 2009a, 2009b). Also, pH range is important in control of As(V) and As(III) removal

through NF (Nguyen et al., 2009a, 2009b). For example, C. M. Nguyen et al.'s (2009a, 2009b) results show that the removal of As(V) increased with increasing pH over the range from 4 to 10, while As(III) removal significantly increased over the pH range 8–10.

Boussouga et al. (2021) investigated the removal of As(V) by NF and concluded that rejection of As(V) is not affected by increasing salinity but increases with increasing pH. Also, their results show that the presence of humic acid enhances As(V) rejection by 10–20% with the loss of NF due to increase of membrane surface charge.

Assessment of NF and RO membranes for simultaneous removal of arsenic and boron from spent geothermal water implies that tested NF and RO membranes were successful in arsenic removal and RO membranes showed better performance in terms of both arsenic and boron removals (Jarma et al., 2021). Remediation of natural As(V)-contaminated groundwater (As concentration ranging from 59 to 118 ppb) in Calabria, Italy, was treated by NF using two types of membranes commercialized by GE Osmonics, named HL and DK, both made of polyamide thin film and with the same molecular weight cutoff (MWCO). This remediation led to As concentration lower than 10 pbb in groundwater, and the highest water flux was obtained with the HL membrane (Figoli et al., 2020). Arsenic removal from water by UF membrane can be enhanced extremely when coupled with different complementary methods such as photocatalysis (Molinari & Argurio, 2017), adsorption, and sand filtration (Ruiping et al., 2009). In addition, there are several different ways to increase the As removal performance of UF membranes such as micellar-enhanced UF (Iqbal et al., 2007; Yaqub & Lee, 2020) and electro-UF (Hsieh et al., 2008).

5   Switching to Save Water and Soil

Arsenic intake to the environment increased several times due to industries development in recent decades, which led to catastrophic changes in ecosystem. For example, total As concentration increases several times in sediments of freshwater lake in China (Chen et al., 2015). This resulted in >tenfold loss of crustacean zooplankton and > fivefold increase in highly metal-tolerant alga. It is possible to control As intake, but it is difficult due to complexity involved in dealing with contaminated sites, exacerbated by site characteristics, hydrogeological conditions, unknown source term, chemical form, and complex land use (Mahimairaja et al., 2005). Then prevention of As contamination of water resources and soil is always preferred approach compared to As mitigation.

Sometimes geological and geomorphological conditions trigger the release of arsenic to groundwater. Alluvial aquifers with reactive organic matter, low-lying topographic features, and slow groundwater flow caused by low hydraulic gradients provide favorable conditions for arsenic enrichment (Hasan et al., 2007; Bhowmick et al., 2011; Li et al., 2020a, 2020b; Ravenscroft et al., 2011). During the exploration for new groundwater resources of drinking and irrigation water, the geological,

geomorphological, geochemical, and hydrogeological conditions in aquifers should be considered to find the best sites for exploitation of fresh and As-free water. In most affected countries such as Bangladesh, shallow groundwater shows higher As concentration than deep groundwater (Ravenscroft et al., 2011). It is due to young reactive organic matter in sediments, excessive irrigation pumping, evaporation, and groundwater level fluctuation which changes the pH-Eh condition and lead to geo-chemical disequilibrium in aquifers (Ravenscroft et al., 2011; Zabala et al., 2016; Bandara et al., 2018; Das & Banerjee, 2020; Li et al. 2020a, 2020b). It can be rec-ommended to use deep groundwater with low As concentration at least for drinking and use shallow water only for irrigation, but pumping rate in deep wells has to be controlled to avoid the penetration of shallow As-rich groundwater to deep aquifers (von Brömssen et al., 2014).

Several recommendations have been given to prevent of As contamination of soils and groundwater due to cultivation and irrigation: (a) large-scale use of rain-water in irrigation systems in regions where water used for cultivation has high As concentration; (b) cultivation in controlled conditions such as hydroponic systems especially for hyperaccumulator plants such as rice; (c) prevention of using treated or nontreated wastewater from municipal, urban, industrial, and mining resources in cultivation; and (d) limiting application of fertilizers and pesticides. In addition, agricultural activities on As-contaminated soils and with high As concentration in irrigation water should be abandoned.

Important anthropogenic sources of As release to the environment are disposed solid wastes and wastewater. For example, the application of sewage sludge (Tarvainen et al., 2020, Li et al., 2020a, 2020b) and mine drainage (Coudert et al., 2020) are potential sources of arsenic. Generally, As treatment of As-contaminated water, wastewater, and soils should not only maximize As removal but also should allow for the production of residues that are geochemically stable over long term (Coudert et al., 2020). Successful treatment of As-contaminated water, wastewater, and soil treatment should take into consideration appropriate disposal methods for arsenic-bearing wastes (solid or liquid wastes generated by different treatment pro-cedures). Arsenic-bearing wastes should be disposed in environments that limit the potential risks of As release and contamination of water and food sources. Current disposal options for As-bearing wastes include landfilling, stabilization, cow dung mixing, passive aeration, pond disposal, and soil disposal (Clancy et al., 2013). Suitable environments for As-bearing wastes (solid or aquatic) are those with simi-lar conditions under which As was mobilized (Clancy et al., 2013), for example, a passive aeration system for oxidized As-bearing wastes or anaerobic system for reduced As-bearing wastes. Additionally, it is necessary to consider microbially mediated transformations of As and Fe, and wastes should be stored in environ-ments with limited microbial activity.

Case Study 7: The Application of Groundwater Flow Modeling for of Deep Low Aquifer at Matlab, Southeastern (von Brömssen et al., 2014)
The Matlab site in southeastern is heavily contaminated by As at shallow and intermediate depths with As concentrations up to 360 µg/L and As present mostly as As(III). On the other hand, deeper aquifers show much lower As concentrations or are As free. Switching of groundwater pumping to deeper aquifer as viable option was tested by groundwater flow modeling. Parameters for the modeling were obtained using hydraulic head monitoring, pumping tests, and ^{14}C dating. Based on the modeling results, local flow system reaches the depth about 30 m below ground surface. This is a consequence of strong anisotropy of hydraulic conductivity ($K_z/K_{x,y} < 1000$) obtained by calibration of monitoring data. Below this depth, regional flow system with much older groundwater with low As concentration is located. Currently, the risk of cross-contamination by irrigation pumping is low because irrigation pumping wells are installed in relatively shallow depth, but installation of deep irrigation wells could increase the risk in future. It is suggested to develop deeper wells for drinking water purposes, but installing deep irrigation wells is discouraged.

6   Conclusions

Arsenic is a naturally occurring element. However, anthropogenic activities such as mining metal processing and cultivation of crops change geochemical equilibrium and mobilize arsenic. Fast urbanization and industrialization also mobilize arsenic and increase its transfer to the ecosystems. High arsenic concentrations in air, soil, and in surface waters and groundwater have an impact on human health due to inhalation of As-contaminated air and ingestion of As-contaminated drinking water and food. Loss of some species living in ecosystems and decrease of cultivation producibility are the results of increasing of As concentration in aquatic environments and soil. Input of As from drinking water and food depends on many factors such as lifestyle, age, gender, race, and weight. It is necessary to consider and anticipate the changes in As mobility and bioavailability for human, animals, and plants before starting extraction of groundwater and mining, industrial, and agricultural activities or other processes changing pH-Eh conditions and geochemical equilibrium. Disposal of wastes from different sources should be done as safely as possible to reduce the possibility of As release from disposing sites to the environment. Principal methods of arsenic remediation from contaminated water and soil including adsorption, ion exchange, phytoremediation, phytobial remediation, chemical precipitation, electrokinetic processes, electrocoagulation, and membrane technology discussed in this text. We need to find more practical, economical, and eco-friendly remediation methods and technologies with a minimum waste production for As treatment. Global climate change can exacerbate water crisis and As-contaminated soil and water resources problems.

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