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REVIEW ARTICLE

Review on remediation technologies for arsenic-contaminated soil

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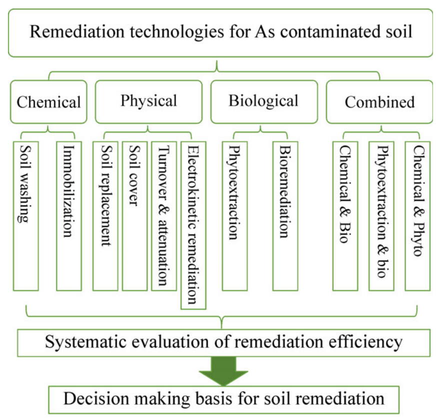
HIGHLIGHTS

- Recent progress of As-contaminated soil remediation technologies is presented.
- Phytoextraction and chemical immobilization are the most widely used methods.
- Novel remediation technologies for As-contaminated soil are still urgently needed.
- Methods for evaluating soil remediation efficiency are lacking.
- Future research directions for As-contaminated soil remediation are proposed.

GRAPHIC ABSTRACT



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ABSTRACT

Arsenic (As) is a top human carcinogen widely distributed in the environment. As-contaminated soil exists worldwide and poses a threat on human health through water/food consumption, inhalation, or skin contact. More than 200 million people are exposed to excessive As concentration through direct or indirect exposure to contaminated soil. Therefore, affordable and efficient technologies that control risks caused by excess As in soil must be developed. The presently available methods can be classified as chemical, physical, and biological. Combined utilization of multiple technologies is also common to improve remediation efficiency. This review presents the research progress on different remediation technologies for As-contaminated soil. For chemical methods, common soil washing or immobilization agents were summarized. Physical technologies were mainly discussed from the field scale. Phytoextraction, the most widely used technology for As-contaminated soil in China, was the main focus for bioremediation. Method development for evaluating soil remediation efficiency was also summarized. Further research directions were proposed based on literature analysis.

1 Introduction

Arsenic (As) has the properties of metals and metalloids but is often considered a heavy metal in environmental science owing to its high toxicity and inability for natural degradation. This element exists in the environment because of mining, mineral processing, smelting, and processing of As-bearing ores and secondary pollution in industrial or agricultural production and applications. As content in soils may vary from 1 mg/kg to 40 mg/kg, but high values have been reported owing to waste disposal, sewage irrigation, and pesticide and fertilizer application (Lazo et al., 2007).

Arsenic exists mainly as inorganic arsenate (AsV) or arsenite (AsIII), which are both highly toxic. AsV is an analog of phosphate and can be toxic when interfered with essential phosphate-required processes, such as ATP synthesis. By contrast, the toxicity of AsIII originated

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from its tendency to bind with thiols.

China accounts for 70% of total global As reserves. In this country, the total amount of As entering smelters exceeds 80,000 tons annually, and the annual As discharge is over 40,000 tons, accounting for approximately half of the total As emissions. According to a national survey on the environmental quality of soil in China released in 2014, 2.7% of the investigated soil samples were As-contaminated. A study revealed As distribution in the agricultural soils in China and As accumulation in topsoil. Such accumulation of As in topsoil became more evident in 2016 compared with that 10 years ago (Zhou et al., 2018).

Compared with water or air pollution, soil pollution is less visible and have been recognized by the public only since the beginning of this century. An excess amount of As in soil can lead to health risks from multiple exposure pathways. Dietary exposure is one of the main exposure ways for As to pose health risks to humans. Excess As in soil can be transported upward into the edible parts of crops, the consumption of which allows As to enter the human body. The oral pathway contributes >90% and 60% of the total noncarcinogenic risks of As for adults and children, respectively (Zhou et al., 2018). A study reported that 55% of investigated babies were exposed to unacceptable health risks owing to As contamination; hence, special concern has been given to the health risks of babies through breast milk (Samiee et al., 2019). Health risks from accidental direct ingestion of soil (Doyle et al., 2012) and inhalation of As-enriched soil particles (Gosselin and Zagury, 2020) have also been reported, although they occupy only a small portion of the population.

Given the great public awareness of the health risks posed by excess As in soil, the number of studies that identify remediation technologies to As-contaminated soil has increased. These technologies fall into two main categories, first to remove As from the contaminated medium, and second is to reduce its biotoxicity. Both can decrease the related health risks to humans to a certain extent. Remediation technologies can be classified as biological oxidation, electrokinetics, phytoremediation, coagulation–flocculation, and solidification/stabilization. Given that these methods are at their different stages of development, they have been applied to As-contaminated soil with varying levels of success.

This review presents the general overview of the remediation technologies for As-contaminated soil to provide information for landowners or managers to design an appropriate soil remediation technology. Attention was provided to laboratory- and field-scale soil remediation experiments based on their own research and development levels. Not all reported remediation technologies were listed, and those that have been applied or have the potential to be applied in the field were included. Evaluation can be regarded as the most important step during the remediation of a specific parcel of land. Hence, methods for evaluating the remediation efficiency of As-contaminated soil were summarized.

2 Chemical technologies

Chemical remediation technologies for As-contaminated soil mainly include soil washing and soil immobilization, with the former aiming to remove As from the soil, and the latter aiming to stabilize As content in the soil.

2.1 Soil washing

Soil washing or soil leaching refers to the technology that injects chemical reagents that can promote the dissolution or transport of soil pollutants and then collect the leachate enriched with pollutants, thereby removing pollutants from soil. Soil washing has been used in soils with varying levels of As contamination. The washing reagents include inorganic acids and bases, organic ligands, chelant, and recent biosurfactants (Jho et al., 2015; Rasmussen et al., 2015; Wei et al., 2016; Beiyuan et al., 2017; Wang et al., 2017).

Among these eluents, acids and bases are two of the earliest eluent categories that continue to show acceptable results under specific washing parameters by solubilizing Fe minerals and increasing the pH value, respectively (Im et al., 2015). With an increase in eluent concentration, the As removal rate also increased and led to a high destruction of soil properties and a high cost for the disposal of the leachate. Phosphoric acid is the most effective acid because it solubilizes As-absorbed Fe minerals and replaces AsV due to the similarity between AsV and PO_4^{3-}. The highest removal rate reached 90%. The possibility of using various phosphates, which showed high extraction capacity, to wash As-contaminated soil has also been studied (Zhao et al., 2016; Mukhopadhyay et al., 2017). Phosphate can compete with AsV during PO_4^{3-} or AsO_4^{3-} sorption to iron or manganese oxides, thereby replacing AsV from the absorbed fraction. The advantage of phosphate over phosphoric acid is the former’s low impact on soil properties, including soil pH and fertility.

Chelants alone have low extraction efficiency for As (Qiu et al., 2010; Wen and Marshall, 2011) because only a minor portion of As can be chelated by the functional groups. Chelants such as EDTA may result in secondary contamination because they are difficult to degrade and negatively affects the environment in the long run. Thus, biosurfactants and organic substances from natural materials with relatively low impacts on soil properties (Gusiatin, 2014) are recently widely used (Lin et al., 2017). The application of extracted dissolved organic carbon (DOC) can achieve a removal rate of 88% (Lin et al., 2017), which may result from the high pH of DOC and the binding capacity of the organic functional group.

The combined or sequential use of multiple eluents to maximize the advantages of several eluents with different characteristics can achieve an extraction efficiency of As from soil as high as 98% (Jang et al., 2007; Wei et al., 2016). The combination of biosurfactants and phosphate seems to be an efficient and non-destructive method. In addition to the category and dosage of eluents, the contact time, pH, and order of sequential washing are also important when determining the extraction efficiency.

Soil washing can be applied in two ways: in situ or ex situ. In situ washing is easy to apply, but the secondary contamination caused by the diffusion of As-enriched leachate needs to be strictly controlled. Ex situ washing is less likely to cause the diffusion of leachate, but the transportation of contaminated soil may result in secondary pollution to the environment along the transportation route.

Soil washing can efficiently transfer As pollution from soil to water, thereby decreasing the treatment difficulty. The main problems of this technology are the destruction of soil property (Table 1) and its high cost. Setting a soil washing plan requires the following: 1) establishing a washing agent with low environmental burden and low cost and high efficiency; 2) efficient treatment system for the leachate; 3) application of soil fertilizers; and 4) suitable soil properties, i.e., the fraction of clay is lower than 25%.

2.2 Soil immobilization

Soil immobilization refers to the application of chemical reagents that can immobilize soil pollutants and thus decrease the potential risks caused by excess pollutants in soil. Increasing attention has been paid to soil immobilization due to its low cost and convenience of operation. Table 2 provides several representative immobilization reagents applied to As-contaminated soil. Metallic compounds (especially Fe oxides), solid waste, and biochar are commonly used as immobilization agents for As (Doherty et al., 2017).

The positively charged surface of Fe oxides can form complexes with negatively charged AsV, thereby decreasing the mobility of As in soil. As absorption to the surface of Fe oxides, oxidation of AsIII to AsV, and precipitation of Fe with AsV may contribute to such immobilization.

Table 1 Reagents used in As-contaminated soil washing

Eluent		Eluent concentration	Original As concentration (mg/kg)	Removal rate (%)	Soil property change*	Reference
Acid	HCl	1 M	59	40	H	Im et al. (2015)
	H_3PO_4	2 M	165	90	H	Wang et al. (2017)
Base	NaOH	0.5 M	101	42	H	Beiyuan et al. (2017)
	NaOH	2 M	165	98	H	Wang et al. (2017)
Salt	Na_2CO_3	0.5 M	55	35	H	Beiyuan et al. (2017)
	$NH_4H_2PO_4$	0.5 M	35	25	H	Jho et al. (2015)
	$(NH_4)_2HPO_4$	0.5 M	59	35	H	Im et al. (2015)
	KH_2PO_4	0.1 M	150	63	M	Zhao et al., 2016
Chelant	Na_2EDTA	0.01 M	70	2	M	Qiu et al., 2010
	[S,S]-EDDS	2 mM	355	11	M	Wen and Marshall (2011)
Organic	Dissolved organic carbon (DOC)	3000 mg/L	390	88	L	Lin et al. (2017)
	Humic substances	0.05 M	990	18	L	Rasmussen et al. (2015)
	Citrate	0.05 M	990	64	M	Rasmussen et al. (2015)
	Nitrilotriacetic acid	0.05 M	990	60	M	Rasmussen et al. (2015)
	$H_2C_2O_4$	0.05 M	153	22	M	Wei et al. (2016)
	Oxalate	0.1 M:0.1 M	70	60	M	Qiu et al., (2010)
Surfactants	Tannic acid	3% weight	3574	38	M	Gusiatin (2014)
	Saponin	1.5% weight	85	75	M	Mukhopadhyay et al. (2017)
Combined/ Sequential	$Na_2S_2O_4$ in HCl	2%: 0.01 M	59	42	H	Im et al. (2015)
	$Na_2S_2O_4$ in EDTA	0.1 M: 0.1 M	165	95	H	Wang et al. (2017)
	$Na_2S_2O_4$-$C_6H_8O_7$-$NaHCO_3$	0.4 g:1 g:10 mL	101	81	H	Beiyuan et al. (2017)
	Saponin + KH_2PO_4	1.5% (w:v):150 mM	85	92	M	Mukhopadhyay et al. (2017)
	H_3PO_4-$C_2H_2O_4$ -Na_2EDTA	0.05 M:0.075 M:0.075 M	153	42	M	Wei et al. (2016)

Note: *H, M, and L indicate high, medium, and low impact on soil property, respectively.

Table 2 Reagents used in As-contaminated soil immobilization

Immobilization agent	Soil utilization	Total As concentration (mg/kg)	Soil pH	Reagent	Concentration	Immobilization efficiency (%)	Period	Reference
Laboratory-scale								
(hydro) oxides	Agricultural field	3500	7.4	Schwertmannite	5% weight	63	35 days	Yang et al. (2017)
	Mining site	2548	6.3	Maghemite nanoparticles	5% weight	99	10 days	Arenas-Lago et al. (2019)
	Mining site	479	4.6	Aluminum oxide	5% weight	31	2 days	Doherty et al. (2017)
	Mining site	479	4.6	Manganese (IV) oxide	3% weight	40	2 days	Doherty et al. (2017)
	Mining site	479	4.6	Kaolinite	10% weight	26	2 days	Doherty et al. (2017)
	Mining site	479	4.6	Ferric chloride + lime	1%:1% weight	71	2 days	Doherty et al. (2017)
	Mining site	479	4.6	Zero valent iron powder	1% weight	90	2 days	Doherty et al. (2017)
	Mining site	479	4.6	Ferrihydrite (synthesized)	3% weight	84	2 days	Doherty et al. (2017)
	Mining-metallurgy site	70200	6.4	Zero-valent iron nanoparticles	10% weight	92	72 hours	Gil–Díaz et al. (2017)
	Mining-metallurgy site	25900	6.4	Zero-valent iron nanoparticles	10% weight	91-95	72 hours	Gil–Díaz et al. (2017)
	Agricultural field	2047	4.2	$Al_2O_3 \cdot 2SiO_2 \cdot CaO$	6% weight	96.2	28 day	Wang et al. (2019)
biochar	Mining site	15,076.80	3.7	Biochar	2% weight	22	2 hours	Chen et al. (2018b)
	Landfill site	1202	4.6	Biochar	10% weight	25	7 days	Alozie et al. (2018)
	Paddy field	47	6.9	Biochar	1% weight	16	35 day	Zhu et al. (2019)
Solid waste	Paddy field	131.5	6.0	Acid mine drainage sludge	3% weight	93	25 days	Ko et al. (2015)
	Farmland	118	6.8	Coal mine drainage sludge	7% weight	98	28 days	Cui et al. (2018)
	Farmland	118	6.8	Waste cow bones	3% weight	74	28 days	Cui et al. (2018)
	Farmland	118	6.8	Steel making slag	7% weight	98	28 days	Cui et al. (2018)
	Agricultural areas	54	7.4	Municipal solid waste compost	3% weight	45	2 months	Abou Jaoude et al. (2019)
Combined	Paddy field	47	6.9	Bismuth-impregnated biochar	2% weight	69	35 day	Zhu et al. (2019)
	Paddy field	59	4.7	Fe:biochar	5%:1% weight	41	120 day	Qiao et al. (2018)
Bio-materials	Paddy soil	140	5.9	Soil microbial fuel cells bioanode	N/A	47	50 days	Gustave et al. (2018)
Field-scale								
(hydro) oxides	Brownfield	43300	7.0	Nanoscale zero-valent iron	2.5% weight	57	32 month	Gil-Díaz et al. (2019)
	Brownfield	7280	7.2	Nanoscale zero-valent iron	2.5% weight	74	32 month	Gil-Díaz et al. (2019)
	Agricultural field	600	8	Zero-valent iron	1% weight	95	15 years	Tiberg et al. (2016)
	industrial site	8280	7.4	Fe-Mn binary oxide	10% weight	7.8	10 months	Tiberg et al. (2016)
	Abandoned smelter	142	4.6	Fe-based sorbent (46.1% Fe_2O_3, 15.4% MgO, 14.3% CaO, 12.9% SO_3, 8.3% SiO_2, and 1.7% Al_2O_3)	1% weight	18.8	1 week	An et al. (2019)
Solid waste	Paddy field	131.5	6.0	Acid mine drainage sludge	3% weight	45	2 years	Ko et al. (2015)

Owing to the large surface area of nanoscale particles, Fe-based nanoparticles have enticed considerable interest in the remediation of As-contaminated soil (Gil-Díaz et al., 2017). These nanoparticles considerably decrease the mobility of As in mine soils (Arenas-Lago et al., 2019). However, without a stabilizer, nanoparticles easily aggregate, thereby decreasing their immobilizing efficiency. A study found that after adding a starch stabilizer, the As removal efficiency of magnetite nanoparticles was improved by 2.2 times (Liang and Zhao, 2014). Zero-valent iron nanoparticles remained active after 6–15 years, thereby substantially decreasing As mobility in soil (Tiberg et al., 2016).

The use of waste materials containing organic and inorganic components can achieve a high immobilization rate, and at the same time it can realize the disposal of solid waste; thus, it has received attention recently. Organic-based municipal solid waste compost, Fe-based coal mine drainage sludge, and steel-making slag (Ko et al., 2015) were found to effectively immobilize As in agricultural soil (Cui et al., 2018). Before the application of waste materials, the concentration and bioavailability of heavy metals in the waste materials need to be carefully considered.

Biochar alone can hardly achieve a high As immobilization rate because this ingredient is generally alkaline and negatively charged. With the appropriate combination of biochar with other materials, the extraction efficiency can be improved (Alozie et al., 2018). Combination with Fe is one of the most common measures to improve As immobilization efficiency (Zhu et al., 2019). The advantage of the combination of biochar and Fe is that it cannot only immobilize As but also immobilize other cations, such as cadmium (Qiao et al., 2018). Similar to the waste material, careful consideration of potential hazardous materials in biochar is necessary before its application.

Soil immobilization is used in field-scale experiments owing to its low cost and easy application. Fe-containing materials are the most commonly used. A field experiment found that nanoscale zero-valent iron can effectively immobilize As in a brownfield during the 32-month experimental period (Gil-Díaz et al., 2019). A lysimeter study indicated that Fe–Mn binary oxide waste substantially reduces the bioaccessibility of As by the coupled oxidation–sorption reaction (Tiberg et al., 2016).

In addition, the combined utilization of multiple agents has become a new trend in immobilizing As in soil. Mn oxide-modified biochar composite and concrete/maghemite can decrease the mobility of As in soil (Yu et al., 2015) and often immobilize multiple pollutants at the same time. The problem of this technology is the requirement of long-term monitoring of immobilization efficiency and the possibility of stabilized As returning to an active state.

Iron can help determine the behavior of As and has been used to establish an appropriate As mobilization or immobilization strategy. During remediation, the relationships between Fe and As, especially their speciation transformation and its effect on the mobility of As, require further investigation. Soil environmental conditions, such as acid and alkali, redox conditions, and changes in coexisting ions and organic matter, also affect the fate of As. Considering these impacting factors is important when establishing a soil remediation plan.

3 Physical technologies

The physical remediation technologies for As-contaminated soil mainly include soil replacement, soil cover, turnover and attenuation, and electrokinetic remediation.

3.1 Soil replacement and soil cover

Soil replacement and soil cover are similar; both need clean soil from other places. When the original soil level is lower than the surrounding soil, soil cover is used. If the original soil level is the same as the surrounding soil, soil replacement is used.

In soil replacement, the contaminated soil is replaced by clean soil to decrease the As concentrations in the original soil. This method is often applied to heavily contaminated soils owing to its high cost and energy requirement. In China, soil replacement is generally conducted when reclaiming mine land. Difficulty in obtaining sufficient clean soil is often the limiting factor for this technology. The prevention of secondary pollution during the transport of contaminated soil is another issue that must be considered. Soil cover (or soil dressing) is an efficient way to achieve the reduction of heavy metals in agricultural products. This method involves covering the contaminated soil with uncontaminated soil and setting a hardpan (mostly clay) layer between the contaminated and uncontaminated soils.

Based on the Japanese government report released in 2006, 87.2% of the total polluted land (7327 ha) in Japan has been remediated by applying soil replacement or soil cover (Ministry of Environment, Government of Japan, 2006). In China, soil exchange and soil cover are mainly applied to discarded mine zones. Although a layer usually separates contaminated and uncontaminated soil, the mix of clean soil with contaminated soil remains possible when the uncontaminated soil layer is not thick enough or the land is cultivated deeply. The thickness of soil covered on top is generally at least 20 cm and vary between 20 and 40 cm depending on the soil property, dressing method, and environment.

3.2 Turnover and attenuation

Turnover and attenuation involve mixing the contaminated top soil with clean deep soil to decrease the total concentrations of contaminants in soil. It is a widely used method in Japan, but it is not common in China because it is expensive and may negatively affect the growth of crops (Chen and Chiou, 2008). A field study of our research group (data not published yet) found that turnover and attenuation can efficiently lower the concentrations of pollutants in soil contaminated by sewage irrigation. The As concentration in top soil (0–20 cm) can be lowered by ~50%. However, the yield of wheat considerably decreases after turnover and attenuation because the fertile topsoil is buried underneath. Therefore, fertilizer application is necessary. In addition, the local soil conditions have to satisfy the following prerequirements: 1) the soil must be only slightly contaminated, 2) the contaminants are mainly distributed in topsoil, and 3) clean deep soil exists. Furthermore, the requirement of turnover machines implies that this remediation area needs to be suitable for machine farming. The key parameter is the turnover depth, which is usually set at 40, 60, or 80 cm. The turnover depth mainly depends on the distribution of contaminants on the soil profile and the maximum allowed cost.

These physical technologies can efficiently and quickly reduce As concentration in soil. Another advantage of physical technologies is that they cannot only decrease As concentration but also the concentration of all possible contaminants in soil. However, they also decrease the concentration of nutrients in soil, which may be a shortcoming for farmlands. Another shortcoming is that physical technologies often require high initial and maintenance investments and expert labor. In addition to the high cost, another factor that limits the application of this technology is the difficulty in finding sufficient clean soil sources (or clean deep soil for turnover and attenuation); this difficulty is increasing. The satisfaction of the respective prerequirements should be evaluated beforehand.

3.3 Electrokinetic remediation

Electrokinetic remediation (EKR) refers to the formation of a direct current electric field by inserting electrodes into contaminated soil solutions. Pollutants migrate from the treatment area to the electrode area along with the electric field; then, they can be removed by electrodeposition or ion exchange extraction.

EKR is a fast and efficient method to address As-contaminated soil. The As removal efficiency can be as high as 44.8% (Yuan and Chiang, 2008). Given that As removal by EKR technology has limited relationship with soil pore size, this technology can achieve high removal efficiency for soils with various properties, even for fine-grained soils (Ryu et al., 2017). EKR has been applied in the field, achieving varying results. EKR showed high removal rate for top soil (removal rate of 59% at the depth of 0–0.5 m) but comparatively lower removal rates in the deeper layers (Kim et al., 2014).

However, EKR only removes mobile fractions of As; therefore, other reactions that can improve the mobility of As, such as desorption, dissolution, and reduction, have been coupled with this process to achieve a high As removal efficiency (Amrate et al., 2006; Ryu et al., 2017). The addition of EDTA can improve the removal efficiency of EKR through chelation by 31% (Mao et al., 2016). Compared with EKR alone, addition of humic and fulvic acid improved the removal efficiency through competitive adsorption, reductive dissolution, and complexation by 1.5 and 1.8 times, respectively (Li et al., 2020). Phosphate is a good enhancing agent that can better improve the removal rate by 60% compared with oxalate-assisted EKR (Isosaari and Sillanpää, 2012). Similar to soil washing, the effect of an enhancing agent on soil property needs to be considered before its application.

Solar cells and microbial fuel have been applied to the EKR of As contaminated soil to reduce the cost due to energy consumption (Jeon et al., 2015; Habibul et al., 2016). Compared with traditional EKR, solar cell system-driven EKR reduced energy cost by 50% (Jeon et al., 2015). But the applications of these new energies are still mostly in laboratory experiments, which need to be further confirmed in the field.

The electrolysis of water is an important factor that affects the remediation of contaminated soil by EKR. The electrolyzed H^+ around the anode area can dissolve or desorb metals from soil particles, whereas OH^- may result in the precipitation of metals around the cathode areas. During remediation, water electrolysis changes the pH value near the electrode and affects the removal efficiency of As. How to control the pH value near the electrodes is an important way to improve the removal efficiency of EKR.

Despite the high remediation efficiency of these several physical technologies, they are often regarded as too expensive to be implemented in large-scale projects. Thus, field experiences of physical technologies, especially for a large-scale field remediation project, are few compared with chemical and biological ones.

4 Biological technologies

The biological remediation technologies for As-contaminated soil mainly include phytoremediation, microbial remediation, and animal bioremediation, with the former two having been studied more extensively.

4.1 Phytoremediation

Phytoremediation here mainly refers to phytoextraction, which involves growing a special plant that can accumulate high concentrations of heavy metals in the shoots from the contaminated soil, enabling the removal of contaminants through harvesting the aboveground parts. The As hyperaccumulator *Pteris vittata* can achieve an aboveground As concentration of ~1% (dry weight) (Ma et al., 2001). The advantages of this hyperaccumulator also include its high biomass, perennial property, and adaptation to variable environmental conditions. In situ phytoextraction projects using *P. vittata* have been conducted worldwide, with the highest As removal efficiency of 18% (weight) per year achieved in China (Chen et al., 2018a). Scientists from the US obtained an even higher As removal ratio. Two years after the transplanting of *P. vittata* to the contaminated soil, the total concentration of As in soil decreased from 190 mg/kg to 150 mg/kg, achieving a removal rate of 26.3% (Kertulis-Tartar et al., 2006). Differences in As removal efficiency may result from the hyperaccumulator species or populations, bioavailability of As in soil, and the climatic conditions.

The hyperaccumulation mechanism of this special fern has been widely studied. Several systematic and comprehensive reviews of *P. vittata* and its As hyperaccumulation mechanisms have been published (Xie et al., 2009; Han et al., 2017; Souri et al., 2017; Li et al., 2018). In the current review, the main hyperaccumulation mechanisms of As are summarized briefly.

The As hyperaccumulation ability of *P. vittata* is a constitutive characteristic, although it exhibits huge differences among its ecotypes/populations. The As concentration of the habitat soil where spores of *P. vittata* are collected is the main determining factor for the populational difference: the higher is the As concentration in the habitat soil, the lower is the As accumulating ability. Physiologically, the As accumulation mechanisms include: 1) root exudates that can dissolve As in the soil; 2) utilization of efficient phosphate transporter to transport As; 3) reduction of AsV to AsIII, which indicates higher upward transportation rate; and 4) detoxification in the shoots by compartmentation, precipitation, and efflux. From the molecular–biological view, a full-length *P. vittata* transcriptomic–tonoplast proteomic database suggested six families related to As hyperaccumulation, including ACR3 responsible for As efflux; the ABC superfamily responsible for As-thiol chelation; P-type ATPase that may transport AsIII, the major facilitator superfamily mediating AsV uptake; MIP mediating As transport; and nitrate transporter 3.1 that may be a novel AsV transporter (Yan et al., 2019).

Owing to the advantages of As phytoextraction technology as cost effective, easy to operate, and environmentally friendly, this technology has been utilized in ~20 field-scale soil remediation projects. During the implementation of these phytoextraction projects, several issues have been encountered: 1) the reproduction process of ferns is extremely slow, thereby implying a long nursery cycle and consequently a long remediation period; 2) the removal efficiency decreases with time owing to the depletion of bioavailable As in soil; and 3) the pollution control during harvest and disposal of As-enriched biomass. To deal with these issues, efforts have been exerted to establish appropriate sporeling nursery techniques and equipment, efficient soil amendments to increase As uptake efficiency during remediation, facilitation of As removal using microorganisms and plant growth regulators, and a series of technologies to dispose or even recycle the As-enriched biomass safely (Eze and Harvey, 2018; da Silva et al., 2019; Franchi et al., 2019).

Table 3 provides several representative phytoextraction projects and the utilized facilitating measures. Adding phosphate as a soil amendment is one of the most commonly and efficiently used measures to improve phytoextraction efficiency (Yang et al., 2018). Another simple but efficient strategy is to select ecotypes that can greatly adapt to the local environment and can accumulate As efficiently. *P. vittata* is an ancient fern with a long evolutionary history; it has evolved into different ecotypes with varied specialties that may be useful for humans. The selection of appropriate *P. vittata* ecotypes with stronger As tolerance and accumulation ability or with multi-element co-accumulating ability can improve the As removal rate by 10 times (Wan and Lei, 2018).

In addition to these macrolevel environmental adjust-

Table 3 Several representative phytoextraction projects of As-contaminated soil in China

Place	Facilitating measures	Area (ha)	Annual As removal rate (%)	Reference
Baoding, Hebei Province	Warming facilities for plants in winter	0.5	8	unpublished data
Chenzhou, Hunan Province	Phosphate amendment	1	6.0	Chen et al. (2018a)
Fangshan, Beijing City	Phosphate amendment and warming facilities for plants in winter	0.2	17	Chen et al. (2018a)
Gejiu, Yunnan Province	Harvest scenario optimization	5	18	Chen et al. (2018a)
Huanjiang, Guangxi Zhuang Autonomous Region	Phosphate amendment, ecotype selection, harvest scenario optimization	10	14	Wan et al. (2016)
Huize, Yunnan Province	Phosphate amendment and harvest scenarios optimization	0.5	12	Chen et al. (2018a)
Jiyuan, Henan Province	Warming facilities for plants in winter	1	14	Zhang et al. (2017)
Shimen, Hunan Province	Phosphate amendment and water adjustment	10	13.0	Yang et al. (2018)

ment measures, microlevel measures, such as genetic engineering of plants, are suggested to clean the environment. Transporter families responsible for AsV and AsIII, exclusion pathways for AsIII, and some regulatory systems are responsible for the unique As hyperaccumulation in *P. vittata*. However, these microlevel mechanisms are mostly experiments conducted in the laboratory (Ramírez-Rodríguez et al., 2019), with few studies of their application to the phytoextraction practice.

In practice, considering the requirement from farm owners or farm workers, intercropping emerges as an alternative, which can remove As and, at the same time, provide additional income to the farm owners. A slightly contaminated farmland can produce safe products without increasing environmental risks by intercropping the hyperaccumulator *P. vittata* with *Morus alba* L., a cash crop that is used to produce textiles for clothe production (Wan and Lei, 2018). The species of the cash crop and the intercropping models are key factors that determine the intercropping efficiency. However, this technology has a disadvantage, which is that the remediation period can be very long, implying increased monitoring cost and extensive labor requirement. Furthermore, the harvesting can be a problem because the hyperaccumulator and cash crops may need to be harvested at different times.

P. cretica and *Pityrogramma calomelanos* are two other As hyperaccumulating ferns that have been used in As-contaminated soil remediation (Anh et al., 2018; Eze and Harvey, 2018). They have a similarly strong As accumulating ability but smaller biomass than *P. vittata* and thus are less promoted than *P. vittata*. Another fern, *P. melanocaulon*, has Cu and As accumulating ability (Claveria et al., 2019). Willow (*Salix* sp.) is not an As hyperaccumulator but rather a plant widely used in the phytoextraction of As-contaminated soil. Compared with *P. vittata*, willow has huge biomass, extensive root systems, rapid growth rate, and potential to produce energy (Navazas et al., 2019). The number of potential plants that can extract As from soil is increasing, which should originate from field experience more than from laboratory experiments.

Phytoextraction is an environmentally friendly technology but is not as cheap as expected. The safe disposal of As-enriched biomass accounted for about one-fourth of the total cost. Therefore, developing methods for disposing hyperaccumulator biomass economically or for recycling anything useful from the biomass should be the focus of further research.

4.2 Microbial remediation

Microbes are essential in controlling the speciation of As, affecting the bioavailability of As in soil. The most common pathway of microorganisms in affecting As bioavailability is changing As speciation in soil.

In general, microbial remediation of As-contaminated soil can be classified as immobilization and As mobilization. The mobility of As can be lowered by oxidizing AsIII to AsV and thus decrease its bioavailability, especially in paddy soil (Mallick and Mukherjee, 2015). In addition to reduction/oxidation, other immobilization mechanisms include chelation, pH change, biosorption, bioaccumulation (Marwa et al., 2019), coprecipitation (Achal et al., 2012), codissolution (Lee et al., 2012), biomethylation (Liu et al., 2011), or changing the organic metallic complex to radionuclides (Pratush et al., 2018). Microbes used in As immobilization process can directly immobilize As. A rhizospheric fungi, *Aspergillus flavus*, can biotransform mobile As to immobilized As particles, which shows lower bioavailability to microbes and plants (Mohd et al., 2019). The microbial immobilization can also be indirect. It may utilize an ureolytic bacteria, secreting urease to precipitate calcite, and then immobilizes As by the adsorption of As to calcite (Achal et al., 2012). Iron-oxidizing microbes can oxidize Fe and then adsorb an increased amount of As on the solid particles, thereby decreasing the mobile fraction of As in soil (Tong et al., 2019). Sulfate-reducing bacteria decreased the extractable phase of As by 75% in a field experiment (Ko et al., 2017).

By contrast, As mobility can be increased by reducing AsV to AsIII or dissolve minerals, thereby facilitating its removal (Yamamura et al., 2005). Several As-resistant bacteria isolated from the *P. vittata* rhizosphere were able to solubilize As from $FeAsO_4$ and $AlAsO_4$ minerals (Ghosh et al., 2011). However, because separating microbes from contaminated soil is difficult, these As mobilization microbes are usually utilized together with other remediation technologies, such as phytoextraction. The combination of multiple technologies will be discussed in another section. As volatilization through methylation is the only way that microbes alone can remove As from soil. *Penicillium* sp. was able to produce 57.8% of volatile As species (Guimaraes et al., 2019). Similarly, several fungal strains were able to volatilize As from 0.23 to 6.4 mg/kg, as detected by silver nitrate-impregnated filter paper (Singh et al., 2015a). However, due to the potential health risks caused by As in the aerosols (Tanda et al., 2019), we need to be careful when utilizing As volatilization.

Table 4 lists some examples of microorganisms used in the bioremediation of As-contaminated soil. Reduction, bioleaching, and biovolatilization are commonly used to remove As from soil. If these mechanisms can be used together, then the As removal efficiency can be as high as 79% (Petkova et al., 2013). Oxidation and coprecipitation can decrease the mobile fraction of As by ~99% within 10 days (Achal et al., 2012).

Despite the high As remediation efficiency achieved in laboratory experiments, the field application of bioremediation technology alone on As-contaminated soil remains limited. An important reason for this limitation may be the environmental adaptability of these special microorgan-

Table 4 Examples of microorganisms used in bioremediation of As-contaminated soil

Microorganism	Category	Main function	Efficiency	Reference
Bacillus sp. SF-1	Dissimilatory As reducing prokaryotes	Mobilization by reduction	Increase the concentration of dissolved As by 56% within 70 h	Yamamura et al. (2005)
Geobacter metallireducens GS-15	Metal reducing bacterium	Mobilization by reductive dissolution of Fe and co-dissolution of As	Increase the concentration of dissolved As from 10 uM to 230 uM	Lee et al. (2012)
Rhodopseudomonas palustris	Genetically engineered bacterium	Mobilization by biovolatilization through methylation	Remove 2.2%–4.5% of As by biovolatilization during 30 days	Liu et al. (2011)
Acinetobacter junii	Plant growth-promoting rhizobacteria	Mobilization by biovolatilization through methylation	Volatize 14% of As within 72 h	Marwa et al. (2019)
Aspergillus niger	Microscopic fungi	Mobilization by bioleaching, bioaccumulation, and biovolatization	17% of total As was bioleached, 13% of total As was bioaccumulated, and 49% of total As was volatilized	Petkova et al. (2013)
Aspergillus flavus	Microscopic fungi	Mobilization by bioleaching, bioaccumulation, and biovolatization	Immobilization rate reached 84%	Mohd et al. (2019)
Bacillus flexus	Plant growth-promoting rhizobacteria	Bioaccumulation in the cell system	Accumulated 12% of As in the biomass	Marwa et al. (2019)
Brevibacillus sp KUMAs1	As-resistant bacterium	Immobilization by oxidation	Adsorption rate of 55% within 96 h	Mallick and Mukherjee (2015)
Sporosarcina ginsengisoli CR5	As-tolerant bacterium	Immobilization by microbially induced calcite precipitation	Exchangeable fraction of As decreased by 99% within 10 d	Achal et al. (2012)

isms (Mallick et al., 2015). Biological processes are often highly specific, and they need some highly specific environmental conditions, which are difficult to satisfy in the field. The common strategy is to apply bioremediation together with phytoextraction or chemical immobilization/ mobilization as a supplementary strategy, which will be discussed in the section for combined technologies.

Compared with chemical or physical technologies, biological remediation depends on organisms, which can be greener and more environmentally friendly but also less stable. The adaptability of organisms to the local environment is an important factor to consider when applying bioremediation technology. Moreover, the long-term stability of bioremediation technology is the most difficult target to achieve.

5 Combined technologies

A single technology can hardly satisfy the remediation requirement owing to the complexity of the soil environment. The combination of several soil remediation technologies by applying them at the same time or sequentially can achieve a better remediation result. Numerous ways for remediating As contaminated soil can be combined. In the current review, only the most commonly used combinations of remediation technologies were summarized. The combined application of technologies to As-contaminated soil often uses phytoextraction or chemical immobilization as the main technology, with other technologies applied together as facilitating measures.

The addition of microbes can act as a facilitating measure for chemical immobilization mainly by transforming the speciation of As. Adding leonardite and the microorganism *B. pumilus* at the same time can decrease As in wheat grains by ~96% and double wheat yield (Dolphen and Thiravetyan, 2019) relative to the combined action of a chemical adsorption process and a microbial immobilization process. The combination of microbial AsIII-oxidizing bacterium and schwertmannite can remove 99.3% of water-soluble AsIII and 82.6% of $NaHCO_3$-extractable AsIII (Yang et al., 2017). Similar to micro-remediation alone, the instability of such a combination mainly results from the unstable adaptability of microbes to the local environment.

The addition of microorganisms or chemical immobilizers can act as facilitating measures for phytoextraction mainly by increasing the bioavailability or mobility of As in soil. Poplar (*Populus deltoides* LH05-17) combined with the plant growth-promoting rhizobacterium D14 removed 54% of As in soil within five months; this value was 18% higher than using phytoextraction alone, mainly by promoting plant growth and enhancing As translocation (Wang et al., 2011). The As hyperaccumulator *P. vittata* in combination with several *Bacillus* strains can remove more As than can *P. vittata* alone mainly by the bioaccumulation of As by microorganisms themselves and the mobilization role of microbes (Singh et al., 2015b). The combined use of drinking water treatment residuals (WTRs) and a metal hyperaccumulating grass *Chrysopogon zizanioides* L. can efficiently control As pollution diffusion. WTRs indicated high metal binding and acid-neutralizing capacity, whereas *C. zizanioides* has an extensive root system. Both processes worked together, demonstrating the potential to treat As-contaminated soil effectively (RoyChowdhury et al., 2019). The combined application of soil amendments, microorganisms, and phytoextraction achieved higher efficiency than a single technology. The use of K_2HPO_4 improved the phytoextraction efficiency of *Brassica juncea* by up to 80% by enhancing the bioavailability of As. Furthermore, the addition of plant growth that promotes bacteria further increased the total uptake of As by *B. juncea* (Franchi et al., 2019).

Phytoextraction can act as a facilitating measure for soil washing and vice versa. The combined treatment of phytoextraction and soil washing removed 54% of As from soil compared with 47% in the soil washing treatment. The hyperaccumulator can improve the percent of the labile As fraction, thereby increasing the soil washing efficiency compared with soil washing alone. The soil washing increased the bioavailable As, thereby enabling additional As to be taken up by *P. vittata*. Therefore, the combined application of phytoextraction and soil washing can act as an efficient way to remove As from soil. However, the use of some eluents may have a negative effect on the growth of hyperaccumulators, which needs to be considered before its application.

Chemical technologies can be combined with physical technologies to achieve a higher removal rate. EKR efficiency can be improved by different reducing agents, which enabled the As removal efficiency of EKR to improve from 1% to 25% (Ryu et al., 2017). However, the highest removal rate was only 25%, which was suggested to be related to acidification during the process. Therefore, precise control of the pH values using chemical amendments may further help improve EKR efficiency. By the addition of EDTA, the removal efficiency of As(V) was improved from 35.4% to 44.8% in an electric kinetics remediation system. The addition of other chemicals, such as surfactants and small molecular organics, can also improve EKR efficiency to different extents (Yuan and Chiang, 2008). Similarly, microbes that can affect the speciation of As can also be applied to remediation together with EKR.

The most commonly used remediation technology for As contaminated soil is phytoextraction and chemical immobilization, coupled with biological or chemical amendments. The appropriate selection of plant species and amendments is a key factor for the effectiveness of remediation technology.

6 Evaluation methods for soil remediation efficiency

To date, no clear definition of soil remediation efficiency exists. The earlier evaluation procedure of soil remediation efficiency is comparatively simple. Only the reduction of risks caused by excess heavy metals in soil is considered. Therefore, for technologies that remove As from soil, the decrease in the total soil As concentration is used to evaluate the soil remediation efficiency. For technologies that immobilize As, the appropriate evaluation of remediation efficiency is more complex (Yoon et al., 2016). The commonly used methods include chemicals from the sequential extraction procedure, toxicity characteristic leaching procedure, indicator plant method, in vitro digestion, and modern physics method. Recently, microorganism whole-cell reporter was reported to monitor the bioavailability of As in soil, and it focused on the importance of considering the ecological influence of soil pollution and soil remediation (Yoon et al., 2016). In vitro digestion by monitoring digestion in the human body has become a new and acceptable method for evaluating the bioaccessibility of heavy metals in soil or food (Morais et al., 2019). The use of one method alone or of multiple methods together can be found in the literature. Among these methods evaluating the bioavailability or bioaccessibility of As, indicating which one is better is difficult. The selection of an evaluation method should be made based on the evaluation target.

In addition to the risk reduction effect of soil remediation, other effects, such as environmental influences (positive or negative), cost, and social acceptability, are also important factors to consider when choosing an appropriate soil remediation technology.

During the application of a soil remediation technology, it not only changes the concentration and bioavailability of heavy metal in soil but also affects the environment from other aspects, such as alleviation of greenhouse effect by planting hyperaccumulators on wasteland, increased energy use when transporting contaminated soils, space occupation during remediation, and the final disposal of solid waste. These positive or negative influences on the environment should also be considered when evaluating remediation efficiency. Environmental influences include many different categories; therefore, including all of them is difficult, especially when secondary or even tertiary effects are involved. The following difficulty is to assign a weight to each affecting category when putting all these influences together to obtain an integrated environmental influence score.

Cost is another important factor. An increasing number of studies have calculated the cost of a specific soil remediation technology. The cost often includes fixed and variable costs. The former is the one-time investment at the beginning of the remediation technology, such as the nursery equipment for the cultivation of hyperacumulator sporelings or seedlings or the incineration equipment for the harvested hyperaccumulator biomass. The latter is the other costs involved in the remediation process, such as the labor required to weed and harvest, the cost for fertilizers or irrigation when applying phytoextraction, and the money needed to buy various kinds of materials. With the extension of remediation time, the percentage of the fixed cost decreases. Notably, the economic conditions are different among countries and always changing. Caution must be observed when comparing the costs of different remediation technologies applied to different areas.

Owing to the consideration of not only the removal of contaminants but also the environmental merits and the cost requirements, Life Cycle Assessment (LCA) has been used to assess soil remediation efficiency but mostly on industrial lands (Rebitzer et al., 2004). Minimal information is available on farmland remediation assessment. According to the LCA principle, some technologies and software that calculate a value for risk reduction, environmental effect, social impact, and financial effect have been developed to determine the whole effects of soil remediation technologies.

The risk reduction, environmental merits, and cost model is one of the models used to evaluate soil remediation efficiency (Nijboer et al., 1998). This model was developed in Europe and recently adopted in China for the screening of remediation technologies for contaminated sites. Despite the general advantages of this model, many default parameter values may not be fit for a specific piece of land. The weights of different indexes, which reflect the importance of different factors, need to be carefully determined. Additional field experiences are necessary to provide accurate parameters. Further research on a systematic and standardized procedure for the evaluation of soil remediation efficiency is necessary. In this method, social benefit is a less focused topic than the other aspects considered during soil remediation. Social benefits of a remediation project usually include safety consideration for remediation workers, public acceptance for the remediation technology, and impact to the local community. The level of education, popularization of science, and the general development degree of a specific area all affect the social benefits. Similar to the environmental influence, quantifying social benefits and selecting a method for assigning specific weight for each category are difficult and sometimes subjective.

7 Conclusions

The research on As-contaminated soil remediation technologies has made huge progress over the past 20 years. However, the number of laboratory experiments is still substantially higher than that of field trials. Only phytoremediation and chemical immobilization have been tested on the field scale. Although field experiments are considerably more costly and have a higher possibility to fail, further experiences from the field are necessary. The combination of multiple technologies is used to increase remediation efficiency. The widely used technologies have their own advantages and disadvantages, implying that novel soil remediation technologies are still needed. Further research on As-contaminated soil remediation technology should focus on green, environmentally friendly, biological remediation, and the combination of potential facilitating measures. Another research gap is the evaluation or comparison of different soil remediation technologies, which is important for decision-making. Additional field experiences are needed to establish appropriate models that evaluate different aspects of remediation technology.

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