

A review of green remediation strategies for heavy metal contaminated soil

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Abstract

Heavy metals and metalloids can accumulate in soil, with potentially toxic effects to human health and ecosystems, threatening the sustainable use and management of soil resources. Although a number of remediation technologies, such as Solidification/Stabilization (S/S), soil washing, electrokinetic remediation and chemical oxidation/reduction can be applied for the immobilization, removal or detoxification of heavy metals in soil, the environmental, social and economic impacts associated with these conventional approaches hinder their overall sustainability. More attempts have been made to maximize the 'net environmental benefit' in various ways, including recovering resources, embracing nature-based solutions (NBS), and saving energy with the emergence and development of the 'green and sustainable remediation' (GSR) movement. This review critically discusses these green remediation strategies, and the novel soil amendments being utilized in these sustainable approaches. Iron-based amendments are the most promising candidates in green remediation due to the highest stabilization performances for both oxyanions and metallic cations as well as relatively low disturbance to soil. In comparison, waste-derived materials suffer from risks of contaminant release in the long run, reducing the overall sustainability despite their low costs. It has been found that phytoremediation and green amendment-based S/S are typically the 'greenest' remediation strategies, but wise decisions should be made on the basis of case-specific sustainability assessment results. Finally, it is proposed that integration of several green remediation techniques may have a synergistic effect on remediation efficiency.

KEYWORDS

biochar, metal, nature-based solutions, phytoremediation, sustainable remediation

1 | INTRODUCTION

To promote human well-being and ensure healthy lives, the United Nations has set an ambitious sustainable development goal to reduce illnesses and deaths associated with soil contamination by 2030 (UN, 2015). Soil is a fragile, non-renewable resource that is fundamental to life on earth (FAO,

2015; Hou et al., 2020). However, anthropogenic activities have led to widespread soil degradation and contamination (FAO, 2018). In Europe, there are approximately 2.5 million sites that are potentially contaminated by heavy metals and organic contaminants (EEA, 2018). In USA, 235,000–355,000 sites need to be remediated (US EPA 2004). In China, it is estimated that 16.1% of the nation's land contains

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heavy metals and other pollutants that exceed soil quality standards (MEP, 2014). Toxic metals and metalloids in soil (hereinafter referred to as 'heavy metals') have aroused much concern due to their toxic effects on human beings and ecosystems (O'Connor et al., 2020; Wang et al., 2021). Therefore to achieve the goal of sustainable development, it is necessary to seek sustainable approaches for soil heavy metal remediation.

Heavy metals can be removed or stabilized using various approaches. Conventional remediation technologies, including Portland cement-based Solidification/Stabilization (Shen et al., 2019b), containment (Liu et al., 2018), soil washing (Zhang et al., 2010), electrokinetic remediation (Al-Hamdan & Reddy, 2008), thermal desorption (Park et al., 2015), and chemical oxidation/reduction (McCann et al., 2018), have proven effective for the immobilization, removal, or the transformation/detoxification (i.e. arsenic (As), chromium (Cr) and mercury (Hg)) of heavy metals. However, they have been criticized due to environmental concerns such as high energy consumption, greenhouse gas emission, long-term metal leaching risks and air pollution, economic concerns such as high capital cost, and social concerns such as low public acceptance. To overcome those criticisms attempts have been made to maximize environmental, social and economic benefits through the 'green and sustainable remediation' (GSR) movement to assure the sustainability of the remediation processes.

Over the last 20 years, research on GSR as well as GSR-associated topics such as nature-based solutions (NBS), green chemistry and sustainability assessment has boomed (Figure 1). Environmentally friendly soil amendments with less secondary impact on the environment, lower cost and

higher social acceptance have been gradually adopted, including waste-derived materials (e.g. biochar, compost and red mud), natural minerals (e.g. montmorillonite, palygorskite, zeolite and diatomite) and green-synthesized nanomaterials, to remediate heavy metal contaminated soils in a more sustainable way. With the aims of recovering valuable resources, minimizing the human impacts, and saving the energy, green remediation approaches have also emerged and are being adopted by remediation practitioners. However, there is a lack of literature summarizing these novel attempts critically and systematically.

This review focuses on the remediation mechanisms as well as the environmental, social and economic impacts of the green materials and remediation technologies. Green remediation technologies were divided into several categories, namely resource recovery strategies, nature-based solutions, and energy-efficient techniques. Roles of organisms, including plants and microorganisms, were examined, and the relationships between nature-based remediation technologies and the GSR movement were investigated. Several challenges and potential future directions have also been critically proposed.

2 | OVERVIEW OF GREEN AND SUSTAINABLE REMEDIATION

2.1 | An emerging movement

The remediation industry has been through three stages (Hou et al., 2020). Owing to unrealistic regulator demands and pressure from the public, remediation practitioners were once expected to 'remove the last bit of contaminants'

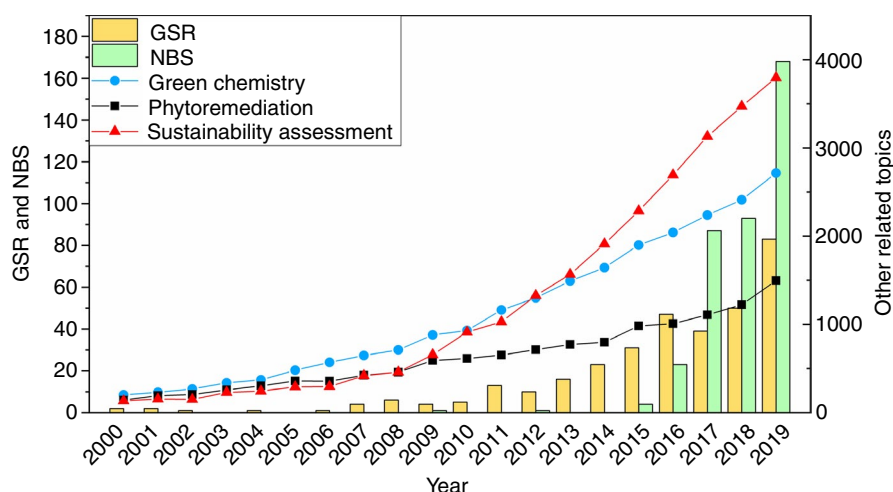


FIGURE 1 Growing numbers of publications on green and sustainable remediation (GSR)-related topics over the last 20 years. Literature search was carried out in Web of Science Core Collections on 25 October 2020. Keywords include 'green remediation', 'green and sustainable remediation', 'sustainable remediation', 'nature-based solution*', 'nature based solution*', 'green chemistry', 'phytoremediation', 'sustainability assessment', 'life cycle assessment', 'life-cycle assessment', 'multicriteria assessment' and 'multi-criteria assessment'

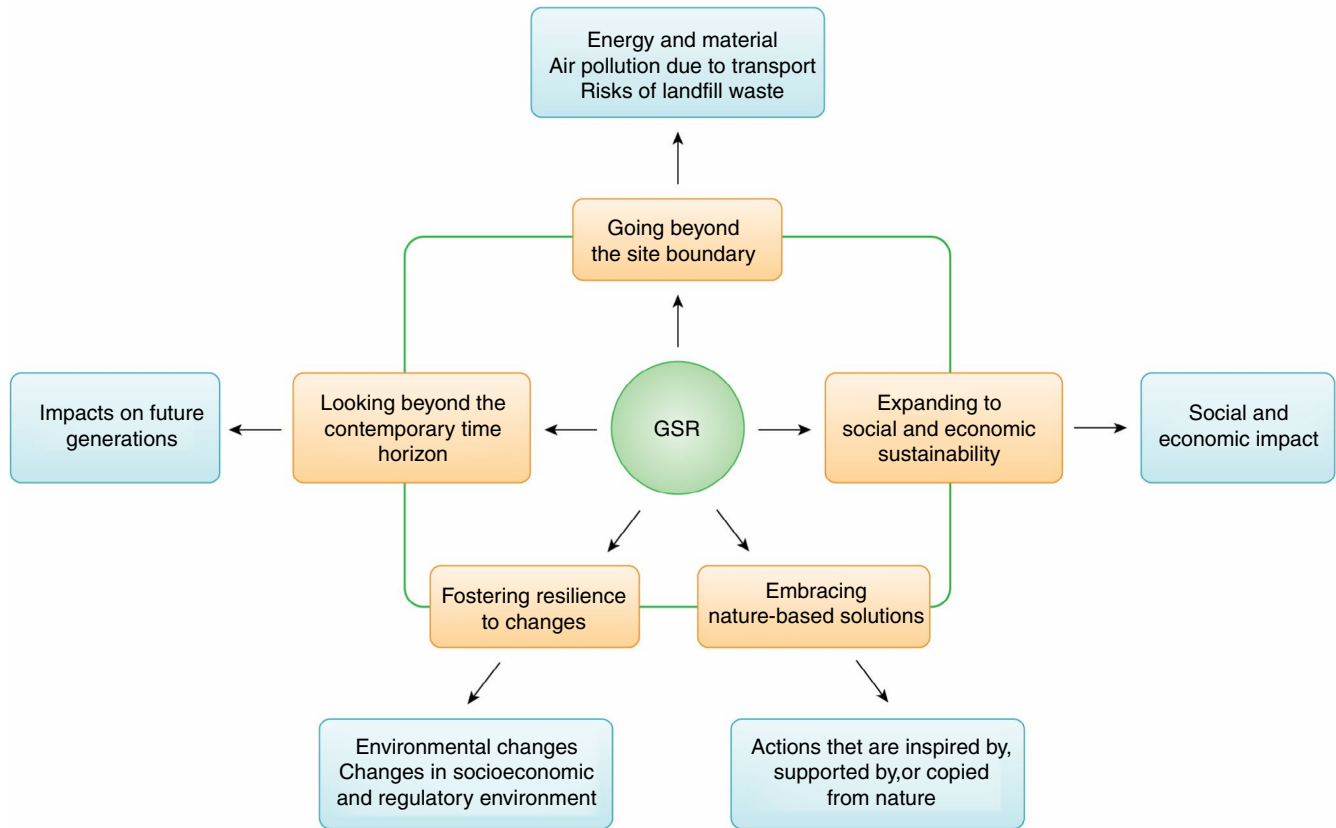


FIGURE 2 Five general principles of GSR. Information obtained from (Hou et al., 2020)

(stage 1). However, by the 1990 s, many nations realized that the cost of a ‘remove all’ strategy would significantly outweigh the perceived social benefits. Remediation practitioners have also recognized that significant biogeophysical constraints made it impossible to return contaminated soils to pristine conditions. A compromise solution was therefore put forward that remediation should aim to make land suitable for certain uses (German Bundestag, 1998; Hou, 2020). Risk-based remediation therefore emerged, allowing the contaminated site cleanup standards to be set at a more realistic and acceptable level (stage 2). However, based on the scientific findings that remediation operations themselves may bring about adverse impacts (e.g. emissions of greenhouse gas, air pollution, groundwater contamination and eutrophication) (Anon, 1990; Diamond et al., 1999; Tadesse et al., 1994), and the fact that more remediation stakeholders came to realize the importance of ‘sustainability’ in a modern society, demand for a sustainable remediation approach that minimizes environmental, social and economic impacts has been growing. Therefore, the concepts of ‘green remediation’ (in the USA) and ‘sustainable remediation’ (in Europe) have emerged, and the term ‘green and sustainable remediation (GSR)’ has been adopted to unite these two concepts (Hou & Al-Tab baa, 2014; Hou et al., 2012; Hou et al., 2015; NICOLE, 2008; US EPA, 2010). The GSR movement has

blossomed in the last 10 years. With careful design and implementation, GSR could help to optimize resource utilization, promote human well-being, improve environmental quality and help establish an energetic remediation market (Hou et al., 2020).

2.2 | Fundamental principles

GSR is a holistic remediation strategy where environmental, social and economic benefits are maximized. To achieve this goal, several general principles are proposed (Figure 2) (Hou et al., 2020):

- A GSR strategy should look beyond the site boundary. Secondary impacts, such as the environmental risks of landfill waste, noise and air pollution during transport and greenhouse gas emissions during the whole remediation life cycle should be quantitatively assessed;
- A GSR approach should consider the impacts on future generations. For instance, passive containment treatments appear superior to active remediation strategies due to low capital cost. However, the high maintenance cost may hinder its overall sustainability in the long term;
- A GSR approach should focus on social and economic sustainability. Although the environmental impact of the

TABLE 1 Mechanisms, advantages/disadvantages and sustainability concerns of green soil amendments

Amendment	Metal immobilization mechanisms	Advantages	Disadvantages	Sustainability concerns	References
Biochar	Surface complexation, electrostatic interactions, co-precipitation with minerals.	Utilization of biomass wastes, including crop residues, wood, animal wastes and sewage sludge. Easy to fabricate (i.e. pyrolysis). Tunable functionality, well-developed pores.	Contaminated biomass-derived biochars may release metals after being applied to soil. Biochar itself contains polycyclic aromatic hydrocarbons (PAHs) that can be released to the soil environment.	Environmental—Biochar stores carbon in ground, with its field application offsetting 12% of anthropogenic carbon dioxide (CO ₂) equivalent emissions. Biochar immobilizes heavy metals while simultaneously increasing the crop yield. Social—In situ biochar application to soil cause little occupational risks. The concept of sustainable biochar to restore degraded soils is widely acknowledged by the public. Economic—Biochar is a waste-derived material. The cost of biochar production is usually below \$500 per tonne. The cost may even be as low as \$50 per tonne. Bio-oil and syn-gas produced during biochar fabrication can be used as clean energy.	(Filiberto & Gaunt, 2013; IBI, 2015; Wang et al., 2017; Wang et al., 2020f; Wang et al., 2020g; Woolf et al., 2010)
Coal fly ash	Surface complexation and precipitation with metal oxides in coal fly ash, and the liming effect.	Large amounts of coal fly ash (i.e. 780 million tonnes) are produced annually. Application of coal fly ash to soil offers a way to handle it properly.	Coal fly ash itself may be contaminated by heavy metals and various organic contaminants.	Environmental—Coal fly ash application increases the risks of metal and organic contaminant release to the environment. Social—High occupational risks, as well as low social acceptance. Economic—Extremely low cost below \$40 per tonne, or even cost-free.	(Leelarungroj et al., 2018; Palansooriya et al., 2020a; Robl et al., 2017; Zha et al., 2019)
Red mud	Surface complexation and precipitation with metal oxides in red mud, and the liming effect.	Large amounts of red mud (i.e. 120 million tonnes) are produced annually. Application of red mud to soil offers a way to handle it properly.	Release of heavy metals after application of red mud to soil. Red mud usually has high pH (i.e. >12), not suitable for As-contaminated soils.	Environmental—Red mud application increases the risks of metal release to the environment. High Na content (i.e. up to 6,090 mg kg ⁻¹) and high pH of this soil amendment cause elevated soil salinity and unwanted soil pH rise to unacceptable levels (i.e. >9). Social—High occupational risks, as well as low social acceptance. Economic—Extremely low cost below \$40 per tonne, or even cost-free.	(Feigl et al., 2012; Hua et al., 2017; Lacatusu et al., 2014)

(Continues)

TABLE 1 (Continued)

Metal immobilization mechanisms		Advantages	Disadvantages	Sustainability concerns	References
Amendment					
Clay minerals	Surface complexation and precipitation with hydroxyl, ion exchange and the liming effect.	Clay minerals are abundant in nature.	Unmodified clay shows poor metal stabilization performances.	Environmental—Clay-based immobilization causes little disturbance to soil. However, evidence has shown that clay alone may not stabilize metals effectively. Social—Some types of clay minerals such as palygorskite are possibly carcinogenic to humans. Economic—Relatively low cost, ranging from \$12 to \$162 per tonne.	(USGS, 2021; Wang et al., 2020e; Xu et al., 2017)
Iron oxides	Surface complexation and precipitation.	Extremely useful for the simultaneously immobilization of oxyanions and metallic cations, especially for soils contaminated by As and heavy metals. Iron oxides, such as goethite and hematite, are abundant in nature.	Not reported.	Environmental—Iron oxides have proven to be the most effective soil amendment with long-term effectiveness. Social—Little occupational risk with high social acceptance. Economic—Relatively low cost, below \$100 per tonne.	(Komárek et al., 2013; Porter et al., 2004; USGS, 2015)
Magnesium oxide	Precipitation and Solidification.	The hydration of MgO promotes metal encapsulation and chemical precipitation.	MgO application results in elevated mechanical strength and soil pH, not suitable for the remediation of agricultural soils.	Environmental—The hydration product Mg(OH) ₂ stabilizes metals effectively in the long run. Social—Little occupational risk. Economic—Relatively high cost, usually above \$500 per tonne.	(Jin et al., 2016; Shen et al., 2019a)
Nanomaterials prepared via green synthesis	Oxidation/reduction, precipitation, surface complexation.	High reactivity towards heavy metals.	Ecological risks associated with their application must not be overlooked.	Environmental—Excellent remediation performances with high ecological risks. Social—Low social acceptance. Economic—Relatively high cost, hindering the practical applications.	(Bardos et al., 2018; Mueller et al., 2012; Wang et al., 2019c)

entire life cycle has been addressed by many studies, social and economic impacts are often overlooked. A proper selection of measurable socioeconomic indicators, as well as the combination of quantitative and qualitative methods in sustainability assessment is therefore needed;

- Sustainable remediation strategies should be resilient to environmental, social and economic changes. A GSR approach should be able to meet evolving environmental standards, adapt to various future site reuse purposes and resist the changing environment (e.g. sea level rise and groundwater depletion);
- Adoption of NBS ensures environmental, social and economic sustainability (section 5). However, nature-based remediation strategies are not equivalent to 'GSR'. For instance, although phytoremediation may extract, volatilize or stabilize soil metals effectively, the long duration of the process decreases the overall sustainability. To meet the goal of GSR, nature-based remediation strategies may need to be modified (e.g. phytomanagement).

3 | GREEN SOIL AMENDMENTS

3.1 | Biochar

As a carbon-rich solid material derived from the pyrolysis of hydrothermal carbonization of biomass feedstock, biochar has been widely used in soil remediation due to the well-developed porous structure, dynamic surface functionality, and the relatively low cost compared with activated carbon (Table 1) (El-Naggar et al., 2019a; IBI, 2015; Wang et al., 2020f). Biochar can be prepared *via* the thermal treatment of various biological wastes, including crop residues (e.g. corn stalk, rice straw, rice husk and rapeseed stem), grass, wood, sewage sludge, anaerobic digestate and animal waste (e.g. poultry litter, swine manure and chicken manure) (El-Naggar et al., 2019b; Palansooriya et al., 2019; Shaheen et al., 2019). Biochar interacts with heavy metals in various ways (Table 1, Figure S1). Outer-sphere complexation, inner-sphere complexation, electrostatic interactions, surface precipitation and ion exchange are potential metal immobilization mechanisms (Ahmad et al., 2014; Lebrun et al., 2020; Shaheen et al., 2019). To enhance the metal binding affinities of biochar sorbents, new trends have emerged in biochar pyrolysis and post-pyrolysis modification strategies (Wang et al., 2020g). For instance, to enhance inner-sphere complexation between biochar surface and heavy metals, novel pyrolysis strategies, such as NH_3 -ambiance pyrolysis and co-pyrolysis, or modification methods, such as alkaline activation, acid activation and oxidant activation can be applied to introduce more oxygen or nitrogen-containing functional groups on the biochar surface. In addition, steam activation,

microwave-assisted pyrolysis, modification with clay minerals and acid/alkaline modification can be applied to remove the tars or improve the pore structure of the resulting biochar, favouring the non-specific interactions such as physical adsorption and ion exchange.

Apart from immobilizing soil metals directly as a stabilization agent, biochar could also assist in phytoremediation processes. The activities of rhizosphere microorganisms will be enhanced, and nutrients in the biochar will be released in the longer term, thus promoting plant growth (Lu et al., 2015; Nejad et al., 2017; Vamerali et al., 2012; Wang et al., 2020d). Furthermore, biochar may act as a carbon source in the anode of microbial fuel cells (section 6) (Liu et al., 2020b; Md Khudzari et al., 2019). In general, biochar is a sustainable soil amendment, offering a variety of benefits, including waste management (O'Connor et al., 2018b), renewable energy generation (the by-products such as bio-oil and syn-gas can be used as energy sources) (Thangalazhy-Gopakumar et al., 2015), improving soil fertility (Xiao & Meng, 2020; Ye et al., 2020), preserving microbial communities (Wu et al., 2020), preventing soil acidification (Shi et al., 2020), and most importantly, carbon sequestration for climate change mitigation (Table 1) (El-Naggar et al., 2018; Hou et al., 2020; Woolf et al., 2016).

3.2 | Industrial waste-based materials

Industrial by-products have received much attention due to the large amounts produced each year (Table 1). Reusing them as soil amendments is a feasible approach for the sustainable utilization of these low-value by-products. Coal combustion fly ash is a typical coal industry by-product; it is estimated that 780 million tonnes are produced annually. Despite the fact that the chemical compositions of fly ash may vary due to different sources and compositions of the coal combusted, all types contain considerable amounts of SiO_2 , Al_2O_3 , Fe_2O_3 and CaO (Leelarunroj et al., 2018; Palansooriya et al., 2020; Robl et al., 2017; Zha et al., 2019), and have a similar metal immobilization mechanism with oxides (i.e. liming & precipitation, surface complexation) (section 3.4). Red mud is a by-product from the Bayer process of the alumina industry (Taneez & Hurel, 2019). Similar to fly ash, red mud is also a mixture of oxides, including Fe_2O_3 , Al_2O_3 , SiO_2 , Na_2O , CaO and TiO_2 (Zia-ur-Rehman et al., 2019). Therefore, red mud can also be adopted as an immobilizing agent for heavy metals since it has similar remediation mechanisms (Huang & Hao, 2012; Taneez & Hurel, 2019; Wang et al., 2018c). Other oxide-containing industrial wastes, such as steel slag (León-Romero et al., 2018; Ning et al., 2016) and coal gangue (Chu et al., 2020), have also been applied for the sustainable remediation of heavy metals. However,

industrial wastes may contain considerable amounts of toxic metals and organic contaminants (Table 1). Once applied to the soil, these contaminants may be released and migrate in the long run, posing risks to the environment (section 7).

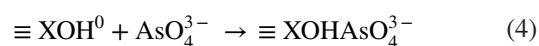
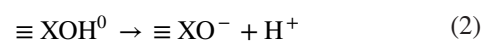
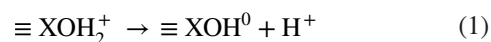
3.3 | Natural minerals

Minerals are promising candidates for GSR due to their relatively low costs and their environmentally benign features. Clay minerals have attracted much attention for metal stabilization due to their high specific area, liming (pH-increasing) effect, excellent ion exchange capacity and abundant surface hydroxyl groups (Table 1) (Chen et al., 2019; Doni et al., 2020; Xu et al., 2017). In clay-based S/S approaches, liming, surface complexation and precipitation are the dominant stabilization mechanisms (Wang et al., 2019a, 2020e; Xu et al., 2017; Yang et al., 2017a). However, unmodified clay minerals suffer from low adsorption capacity, low selectivity towards metal types and rapidly diminishing stabilization performance (Table 1) (Wu et al., 2011, 2016; Xu et al., 2017). To overcome these obstacles, several attempts have been made to modify natural clays to produce more 'powerful' immobilizing agents. For instance, Wang et al., (2020e) modified montmorillonite with humic acid to enhance Cd and Hg stabilization. The oxygen-containing hydroxyl, amino, carbonyl and carboxyl groups in the humic acid layer favoured surface complexation, resulting in a decrease in leachable metal concentrations (> 65% for both metals). Liang et al., (2019) used a novel mercapto-modified attapulgite for the immobilization of soil Cd. Modified attapulgite could reduce plant accumulation of Cd by 75%, due to enhanced sorption of bioavailable Cd onto the modified clay mineral. Interestingly, Wang et al., (2019a) found that sulphur-modified organoclay mobilized soil Hg (the water and acid-soluble fractions of soil Hg increased by 700%), thus promoting the phytoextraction efficiency. Apart from clay, several other types of natural minerals can also aid in heavy metal remediation. For instance, zeolite, an alkaline aluminosilicate mineral with a well-developed microporous structure, immobilizes toxic metals *via* non-specific adsorption and the liming effect (Lee et al., 2019; Shi et al., 2009; Tahervand & Jalali, 2017). Diatomite, a naturally occurring siliceous mineral, is able to stabilize metals through surface complexation and non-specific adsorption, while promoting plant growth simultaneously due to its high Si content (Radziemska et al., 2020; Ye et al., 2015). Compared with metal-rich industrial waste-derived amendments, minerals have few adverse impacts on soil (Table 1). Once the obstacle of long-term stability has been solved

through proper modification strategies, mineral-based materials have great potential for the sustainable remediation of soil metals.

3.4 | Metal oxides

Oxides (including hydroxides, oxyhydroxides and hydrous oxides) have been extensively investigated as immobilizing agents for heavy metals due to their excellent sorption affinities. Iron oxides (e.g. α -FeOOH, γ -FeOOH and α -Fe₂O₃), manganese oxides (e.g. γ -MnOOH and β -MnO₂), aluminum oxides (e.g. γ -AlOOH, α -AlOOH and γ -Al(OH)₃) and magnesium oxide (MgO) are four major types of metal oxides in heavy metal stabilization (Komárek et al., 2013; Shen et al., 2019a). Metal oxides are regarded as promising immobilization agents with long-term stability, which is due to inner-sphere complexation (Eq. 1–4) (Karamalidis, 2010; Komárek et al., 2013):



where $\equiv \text{XOH}^0$ refers to the surface of metal oxide (hydroxide) particles, M^{2+} refers to the divalent metal cation. Both cationic and anionic metal(loid)s can be stabilized through the formation of complexes, offering more stability in the long term than non-specific adsorption (such as Van der Waals force and electrostatic interactions) (Table 1) (Hou et al., 2020).

Precipitation may also play a vital role in oxide-based metal stabilization processes. For instance, due to the alkaline environment created during the hydration process of magnesia, MgO-based binders could achieve promising stabilization performances for Cd and Pb due to precipitation (Table 1) (Shen et al., 2018, 2019a; Wang et al., 2016b). However, the pH-increasing (liming) effect is highly dependent on the amendment type. When it comes to the application of ferrous sulphates, a reverse trend is observed where the oxidation of FeSO₄ releases protons, resulting in the acidification of soil (Eq. 5) (Di Palma et al., 2015; Komárek et al., 2013):



In this case, alkaline amendments (e.g. biochar and lime) should be added simultaneously to avoid a remobilization of cationic metals (e.g. Cd, Hg, Cu and Zn).

3.5 | Nanomaterials produced by green synthesis methods

Conventional manufacturing strategies for nano-sized remediation agents that involve the use of toxic reagents (e.g. NaBH_4) have been criticized (US EPA, 2017a; Zhang et al., 2020b). Green synthesis aims at minimizing the environmental impact and preventing pollution at the molecular level (US EPA, 2017a; Wang et al., 2019c). This can be achieved either by (a) using green materials, such as plant extracts and other nature-derived materials as precursors; or (b) running chemical reactions under mild conditions (ACS, 2020; US EPA, 2017a).

Green-synthesized nanomaterials can aid in green remediation in various ways. Iron-based nanoparticles can be used as immobilization agents directly. For instance, nano zero-valent iron (nZVI) derived from waste tea could be applied for Cr(VI) reduction in the soil (Chrysochoou et al., 2012). Iron oxide nanoparticles prepared using leaf extract could stabilize Cd and As in the soil through co-precipitation (Lin et al., 2019; Su et al., 2020). For more information regarding the applications of nZVI and iron oxide nanoparticles, we refer readers to Wang et al., (2019c). Green nanoparticles can also assist in soil remediation in an indirect way. Plant extract-mediated Ag nanoparticles could promote plant growth through increased soil pH, bioavailability of nutrients and water holding capacity (Das et al., 2018). Environmentally benign nano-sized mineral-based soil conditioners can be prepared with feldspar and lime using a mild hydrothermal method (Liu et al., 2017). However, nanomaterials are usually criticized due to their potential toxic effects on soil microorganisms, plants and human beings. In addition, the relatively high cost of nanomaterials has hindered their practical applications (Table 1).

4 | RESOURCE RECOVERY STRATEGIES

4.1 | Phytomining

Plants have different response patterns towards heavy metals in soils. Most plant species are sensitive to a high metal concentration, while others have developed tolerance and resistance, and may even accumulate them in their tissues (Jia et al., 2019; O'Connor et al., 2019; Wang et al., 2020d). The phenomenon of accumulating unusually high concentrations of toxic metals in plant tissues is termed hyperaccumulation. A hyperaccumulator is typically defined as a plant that can accumulate metals to a concentration that is over 100 times higher than ordinary plants growing in the same environment (Lopez et al., 2019; Rosenkranz et al., 2017; Tognacchini et al., 2020). Phytomining takes advantage of hyperaccumulation to recover valuable metals. Hyperaccumulators extract metals from the metal-rich soils (e.g. mine tailings, low grade ores and metalliferous soils) and translocate them in aboveground tissues. After harvesting and drying, the plant is reduced to ash, which is further subjected to sintering or smelting, allowing the metals in plant tissues to be recovered (Anjum et al., 2012; Novo et al., 2017; Zhang et al., 2014).

A high biomass yield and metal concentration in crop tissues assure the success of a phytomining process. In order to produce a 'crop' of metals, several issues must be considered. It is widely established that an increase in metal bioavailability results in high bioaccumulation in plant tissues; therefore, an increase in labile forms of metals favours phytomining. This can be achieved either through changes in soil pH (e.g. an increase in pH mobilizes the oxyanion As, while a decrease in pH mobilizes most metallic cations) (Huang & Chen, 2003; Zheng et al., 2019), or the addition of

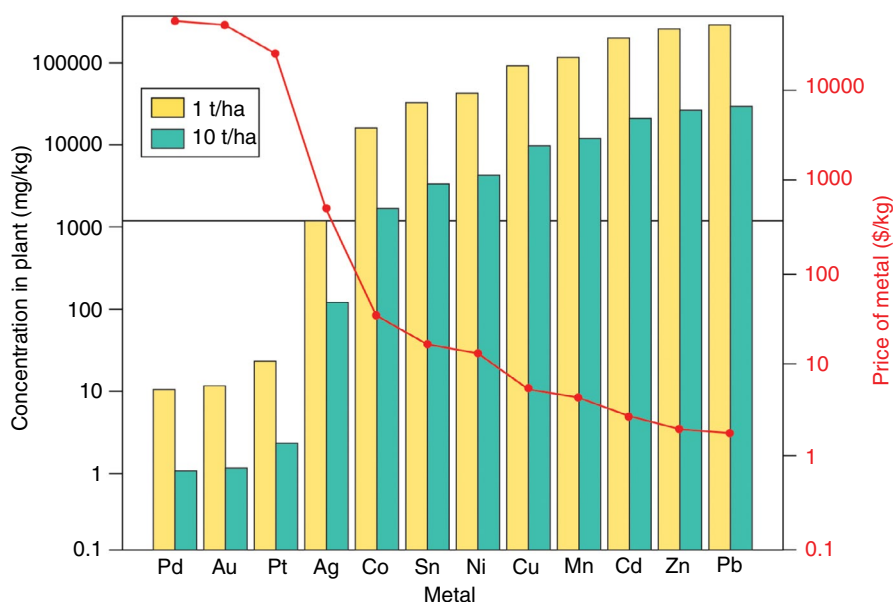


FIGURE 3 Concentrations of metals in plant tissues that would be need to achieve a gross value of $\$500 \text{ ha}^{-1}$. Prices of metal (in 2019) were obtained from London metal exchange (<https://www.lme.com/>) and the KITCO (<https://www.kitco.com/>). For hyperaccumulators, the threshold concentration is regarded as $1,000 \text{ mg/kg}$, except for Au (1 mg kg^{-1}), Cd (100 mg kg^{-1}) and Zn ($10,000 \text{ mg kg}^{-1}$)

chelating agents (such as EDTA) (Nedelkoska & Doran, 2000; Robinson et al., 1999). Besides, a higher biomass can be harvested through the addition of various amendments such as inorganic fertilizers and organic amendments (such as compost, biochar and animal waste). In addition, the selection of a proper planting density, as well as a suitable planting and harvesting strategy (such as double cropping and double harvesting), will also lead to higher biomass yields. The mechanisms involved in these promising strategies have been discussed by Wang et al., (2020d).

Phytomining is a 'green' method for metal mining, since its impact on the environment is minimal compared with the erosion caused by opencast mining activities (Table 3). Phytomining can also be regarded as a remediation strategy for highly contaminated soils. Natural revegetation of a mining-affected area may take hundreds of years, while phytomining offers a relatively fast approach for the restoration of degraded lands (i.e. cutting the restoration duration to several years) (Table 3) (Sheoran et al., 2009). The plant cover minimizes wind erosion and run-off, and the rhizosphere also reduces the migration of toxic elements to groundwater. Therefore, it is an environmentally benign, non-destructive and aesthetically pleasing strategy with high public acceptance. However, many studies have neglected the economic considerations of this 'mining' process. The most important factor determining the economics of the operation is the price of the metal being phytomined (Figure 3). In addition, metal concentrations and biomass yield also affect the gross value that can be achieved in a phytomining process. For hyperaccumulators, there exists a maximum concentration of heavy metals that can be accumulated in plant tissues (Alford et al., 2012; Burge & Barker, 2010; Clemens, 2017; Deinlein

et al., 2012; Sheoran et al., 2009; Tsadilas et al., 2018). As shown in Figure 3, extremely high concentrations of Ni, Cu, Mn, Cd, Zn and Pb (much higher than their threshold values) are required in order to achieve a gross value of \$500 ha⁻¹ (assuming that the biomass yield can reach 10 t ha⁻¹), which is impossible in practical applications. In comparison, the best candidates for phytomining approaches are precious metals such as Pb, Au, Pt and Ag. However, this does not mean that hyperaccumulators of other metals are not useful in environmental applications. Indeed, they have been widely used for remediation purposes (section 5.1).

4.2 | Soil washing

Soil washing can drastically reduce contaminant concentrations and return the soil to below the threshold of regulatory environmental standards. Typically, the soil has to be screened first to remove the less contaminated coarse particles. The remaining fine material needs to be mixed with a washing solution to extract the toxic metals (Hou, 2020; US EPA, 2017b). This process is metal-specific, and an ideal washing solution should be capable of elevating the mobility of metals while maintaining the soil physicochemical properties (for reuse purposes). Therefore, soil washing can be regarded as a resource recovery strategy (to reuse the soil).

The selection of a proper extractant is the key to the success of soil washing. Water, acids, bases, solvents, chelating agents and surfactants can be used in the washing process (Ferraro et al., 2016; Liu et al., 2020a; US EPA, 2017b). However, conventional washing solutions may be detrimental to the environment. For instance, the remaining EDTA in

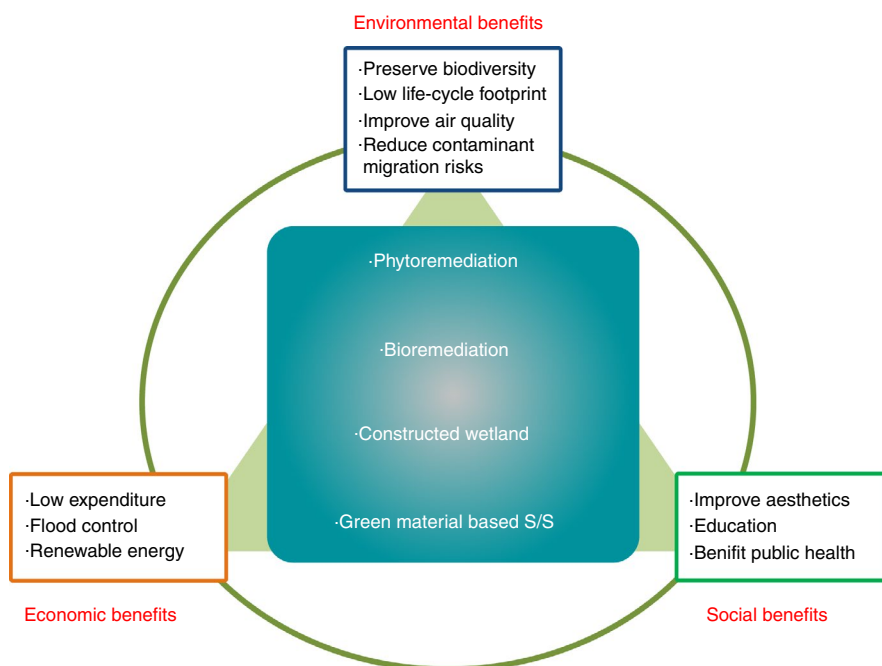


FIGURE 4 Environmental, economic and social benefits of nature-based remediation approaches

the soil after chelant-assisted washing is hard to decompose, which will be harmful to soil organisms (Duo et al., 2019; Epelde et al., 2008). Besides, EDTA in the remediated soil may migrate to groundwater or surface water, resulting in water pollution (this chemical is regarded as a major contaminant in water bodies) (EEA, 1996; WHO, 2011). Strong acids, such as HCl or H₂SO₄, result in the acidification of the soil, making the soil unsuitable for agricultural reuse purposes (Ash et al., 2016). Addition of surfactants can aid in the remediation of metal and organic pollutant co-contaminated soils, but may have adverse effects on soil microbes (Singh & Cameotra, 2013). To minimize the impact on soil properties, many attempts have been made to seek 'greener' washing solutions. Natural organic substances (e.g. humic substances) (Kulikowska et al., 2015; Meng et al., 2017), biodegradable chelants (e.g. iminodisuccinic acid, methylglycinediacetic acid, citric acid and poly- γ -glutamic acid) (Begum et al., 2012; Guo et al., 2018), plant-derived washing agents (Cao et al., 2017a, 2017b) and biosurfactants (Singh & Cameotra, 2013) have emerged as novel extractants in the soil washing process. To date, the applicability of these novel extractants has only been tested in the laboratory. Negative impacts of the washing solutions on the environment (soil, groundwater, plants and organisms) can be expected when applying in situ and should be minimized. For this purpose, more pilot-scale studies should be conducted, and sustainability assessment tools should be adopted to further assess the primary, secondary and tertiary impacts of these novel approaches (section 7).

5 | NATURE-BASED SOLUTIONS

Nature-based solutions (NBS) are defined as "solutions that are inspired and supported by nature, which are

cost-effective, simultaneously provide environmental, social and economic benefits and help build resilience" (European Commission, 2015). In the context of soil remediation, NBS are those techniques that aim at working with nature rather than against it, including phytoremediation (to work with plants), bioremediation (microorganisms) and constructed wetland (the whole wetland ecosystem). In addition, immobilization using natural or waste-derived green materials (e.g. natural minerals and biochar) can also provide environmental, social and economic benefits as compared with conventional Portland cement-based Solidification/Stabilization (O'Connor et al., 2018a; Shen et al., 2019b; Wang et al., 2020b). The environmental benefits of NBS (Figure 4) align well with the aims of metal remediation approaches. Apart from removing the toxic metals or mitigating the risks of metal migration (e.g. remove metals using phytoextraction or reduce the metal migration using phytostabilization), a nature-based remediation method should also preserve biodiversity and improve air quality (e.g. plants used for NBS remediates the land while simultaneously purifying the air) (Seddon et al., 2019; Song et al., 2019b). In addition, a nature-based remediation approach promotes human well-being through improving aesthetics and bringing in health benefits (Colléony & Shwartz, 2019; Dick et al., 2019). The aesthetic value of nature-based remediation approaches (Figure 5) will not only promote social acceptance, but also improve people's psychological states through providing spaces for public recreation. A relatively low cost, utilization of renewable energy (i.e. solar energy) and the ability to control non-point pollution (e.g. constructed wetland) also ensure economic viability (Faivre et al., 2017; Maes & Jacobs, 2017). The following subsections discuss four types of nature-based remediation approaches and explain how nature 'works' with us in soil remediation.

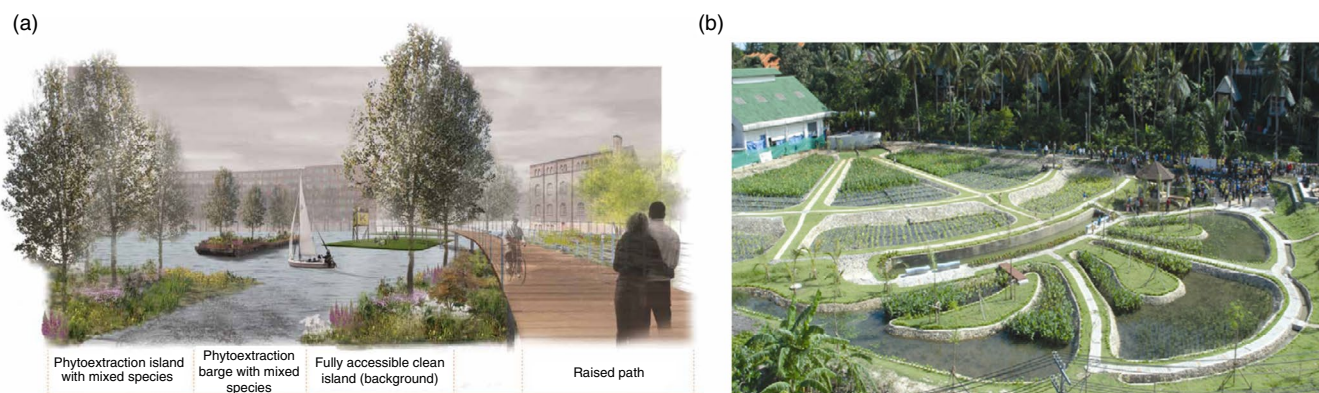


FIGURE 5 The aesthetic values of nature-based remediation approaches. (a) Combining phytoextraction processes with public recreation. A raised path limits the access to the phytoextraction area. Reproduced with permission from Wilschut et al., (2013). Copyright 2013 Elsevier. (b) A flower-and-butterfly shaped constructed wetland on the tourist island of Koh Phi Phi, Thailand. Reproduced with permission from Brix et al., (2011). Copyright 2011 Elsevier

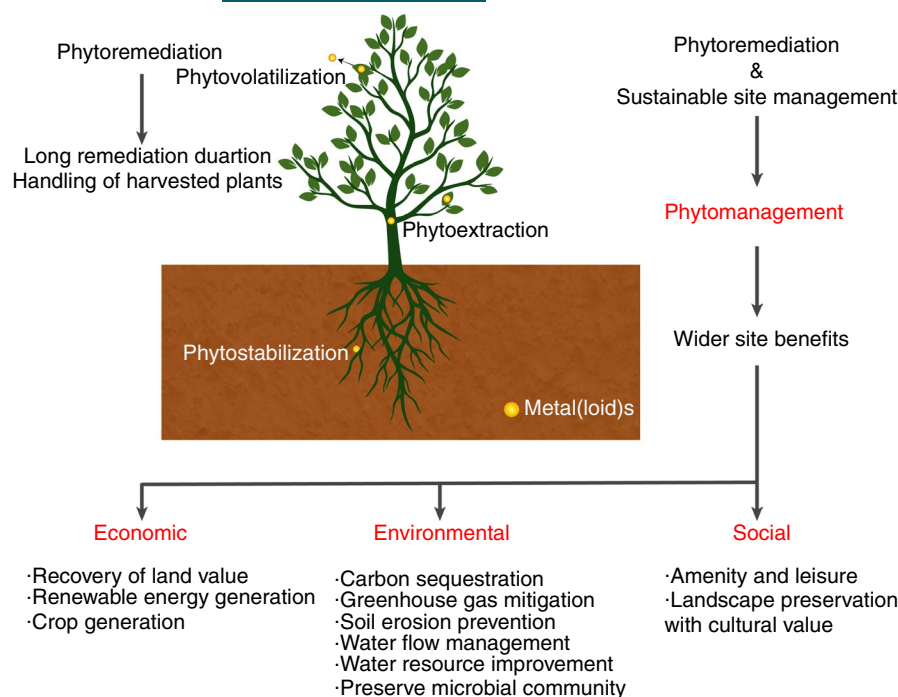


FIGURE 6 Phytoremediation and phytomanagement of heavy metals in soil. Modified with permission from Burges et al., (2018). Copyright 2018 Taylor & Francis. Additional information obtained from (Fässler et al., 2010; Mench et al., 2018; Song et al., 2019a; Wang et al., 2020d)

5.1 | Phytoremediation and phytomanagement

Phytoremediation uses plants to extract (phytoextraction), immobilize (phytostabilization), or volatilize (phytovolatilization) toxic metals (Hou et al., 2020; O'Connor et al., 2019; Wang et al., 2020d). Compared with phytomining which aims to recover valuable elements (i.e. the mining process using plants) (section 4.1), phytoremediation focuses more on the contaminant removal or immobilization performances of plants (i.e. the remediation process). Metal-tolerant plant species (also known as metallophytes) are considered to be ideal candidates for phytoremediation. Based on their physicochemical strategy as a response to high soil metal concentrations, metallophytes can be divided into three types, namely accumulators, indicators and excluders (Burges et al., 2018; Lago-Vila et al., 2019; Lam et al., 2018; Shi et al., 2016). Accumulators tend to uptake and translocate toxic metals in aboveground parts (for phytoextraction), indicators regulate the bioaccumulation of metals so that internal concentrations (in plant tissues) reflect external concentrations (in soil), while excluders restrict the root uptake and translocation of toxic metals, resulting in a phytostabilization effect.

Three main strategies have been applied for metal phytoremediation (Figure 6). Phytoextraction is the most widely adopted approach that uses the metal uptake ability of accumulators to transfer metals from the soil to plant tissues. Phytovolatilization of toxic elements from plant tissues to the atmosphere may occur if the metal accumulated is highly volatile (e.g. Hg) (Awa & Hadibarata, 2020; Nagata et al., 2010). Phytostabilization is also widely used to reduce

metal mobility within the rhizosphere. This can be achieved either through metal precipitation, complexation or physical adsorption induced by plant itself, or through the immobilization effects caused by stabilization agents (such as industrial waste, biochar and minerals). In the latter case, plants play a secondary role in assisting soil immobilization agents. For detailed discussions on the remediation mechanisms, plant selection strategies and influencing factors, we refer readers to Anjum et al., (2012), Ashraf et al., (2019) and Wang et al., (2020d).

Although phytoremediation has been extensively explored in pot, greenhouse and field studies, the commercialization of this nature-based remediation approach has not taken off yet. Phytoremediation has been applied only once amongst 188 US Superfund remediation projects between 2012 and 2014 (US EPA, 2017b). One issue is that highly contaminated sites typically require hundreds or thousands of years for plants to extract toxic metals to meet the regulatory levels (Burges et al., 2020; Cheng et al., 2015). After harvesting, the proper handling of contaminated biomass is also a tough challenge (Michelon et al., 2019; Vociante et al., 2019). For phytostabilized sites, long-term monitoring is needed to assess the risks associated with metal leaching, since toxic metals are not removed (Table 3) (Epelde et al., 2014; Houben et al., 2013; Labidi et al., 2017).

To reach the aim of providing economic, social and environmental benefits simultaneously, a novel concept of 'phytomanagement' or 'phytoattenuation' has emerged (Figure 6). Compared with conventional phytoremediation approaches, sustainable risk-based land use is regarded as the first objective in a phytomanagement process, while metal remediation

is a secondary aim (Burgess et al., 2018; Meers et al., 2010). Profitable crops, rather than hyperaccumulators, are grown in contaminated land, which can be used for various purposes such as bio-energy production, animal feeding and soil fertility improvement after harvesting (Burgess et al., 2018; Fässler et al., 2010; Zhu et al., 2016). Phytomanagement provides much wider site benefits, including the generation of renewable energy, greenhouse gas mitigation, water flow management and landscape reservation (Table 3, Figure 6). As a combination of gentle remediation options with profitable site uses, phytomanagement offers an opportunity to achieve risk mitigation, land restoration and economic gain simultaneously.

5.2 | Microbial-based bioremediation

Unlike organic contaminants, heavy metals cannot be degraded. Microbial-based bioremediation relies on the stabilization, bioaccumulation or detoxification of these toxic elements. Microbial-induced stabilization involves the generation of oxides or carbonates for surface complexation or co-precipitation. For instance, Mn(II) oxidizing bacteria (e.g. *Providencia* sp.) has the ability to generate biogenic manganese oxides (BioMnO_x), which are promising stabilizing agents with high binding affinity towards toxic metals (Li et al., 2020). Ureolytic bacteria could induce calcite precipitation, favouring metal co-precipitation with CaCO_3 (e.g. PbCO_3 , CdCO_3 and CuCO_3) (Kang et al., 2016). Bioaccumulation in organisms can be achieved through the direct uptake of toxic metals. Soil animals, such as earthworms, are ideal candidates for this process (Boughattas et al., 2019). However, the long-term stability of microbial immobilization processes should be further assessed. Considering the fact that the stabilizing agents are generated during microbial metabolism, environmental factors may greatly affect the stabilization performance, indicating poor resilience to changes (section 2.2). How to deal with the organisms with high metal concentrations remains a tough problem, because improper handling of contaminated biomass may increase the secondary impact (section 2.2).

Microbial-based detoxification seems to be a more feasible method for the green remediation of Hg, As and Cr. It involves the oxidation or reduction of toxic metal(loid)s, thus decreasing their toxicity. Microbial reduction of Hg(II) to Hg(0) can be achieved using bacteria with *mer* operon (Mahbub et al., 2016; Wang et al., 2020c). In brief, Hg(II) enters the cytoplasm with the help of transporter proteins encoded by *merP* and *merT* genes. After that, the expression product of *merA* gene, mercuric reductase enzyme, will reduce Hg(II) to less toxic, highly volatile Hg(0) (Naguib et al., 2018; Petrus et al., 2015; Wang et al., 2020c). Owing

to the high volatility of elemental Hg, it will diffuse passively out of the cell and volatilize from soil to the air. However, Hg in the atmosphere can be transported globally, and further returned to the soil via wet or dry deposition (UNEP, 2018; Wang et al., 2020c). Detoxification of As can be accomplished through the microbial oxidation of As(III) to less toxic As(V) (Yang et al., 2017b). In oxygen-depleted flooded paddy soils, this can also be achieved through anaerobic oxidation (using nitrate as electron acceptor) (Zhang et al., 2017). The relative abundances of genera *Acidovorax* and *Azoarcus* increased, and the abundance of As(III) oxidase genes was enhanced during this process (Zhang et al., 2017). Cr(VI) is a potent carcinogen, teratogen and mutagen with toxicity and mutagenicity over 100 times higher than that of Cr(III) (Saha et al., 2011; Shelnutt et al., 2007). A wide variety of Cr reducing bacteria, including *Bacillus*, *Aeromonas*, *Pseudomonas* and *Enterobacter* have the potential for Cr(VI) contaminated soil remediation (Dogan et al., 2011; Tan et al., 2020).

Although microbial-based bioremediation of organic contaminants provides a balance between cost and effectiveness while keeping the site function (Hou et al., 2020), this nature-based remediation technique may not be green when applied to heavy metals (Table 3). During this process, metals cannot be removed from the contaminated soil, indicating the need for long-term management including proper monitoring. Compared with green material-based S/S (section 5.4), microbial-induced stabilization is much more sensitive to changes. Moreover, microbial detoxification involves much uncertainty, since the reversible 'detoxification' reaction may lead to 'toxification' when the environment changes. For instance, an increase in Eh may lead to the suppression of Hg(II) reduction process (Beckers et al., 2019a, 2019b; Frohne et al., 2012), making the microbial volatilization of Hg impossible (Chen et al., 2018; Nascimento & Chart one-Souza, 2003). Low resilience to changes may be the greatest obstacle of this nature-based remediation approach (Table 3).

One plausible role of microorganisms, however, is to assist in the phytoremediation process. Instead of working alone, a combination of microorganisms and plants can attain promising remediation performances. Plant-growth promoting bacteria (PGPB) can either act as a biocontrol agent towards plant pathogens in an indirect way or improve plant growth directly by moderating phytohormone levels (e.g. cytokinins, auxins and gibberellins) and increasing the bio-availability of nutrients (e.g. solubilize phosphorous and nitrogen fixation) (Kong & Glick, 2017). The promoting mechanisms of PGPB in phytoremediation processes include the increase of biomass, improvement of metal tolerance, and alterations to metal accumulation in plants (to decrease or increase the bioaccumulation through changes in metal bio-availability) (Alka et al., 2020; Antoniadis et al., 2017; Kong & Glick, 2017; Wang et al., 2020d). PGPBs can therefore assist in both phytoextraction (Kong et al., 2019; Konkolewska

TABLE 2 Microbial fuel cells (MFCs) in sustainable remediation of heavy metals in soil

Metal (mg kg ⁻¹)	Dominant microbial community	Current (mA)	Power density (mW m ⁻²)	External resistance (Ω)	Remediation duration (days)	Remediation mechanism	Performance	Reference
Zn (11,600), Cd (840)	Not investigated	1.03	~37	300	78	Migration to the cathode and form Cu oxides	Removal rate 25% and 18% for Zn and Cd, respectively	(Chen et al., 2015)
Cr(VI) (50) ^a	Firmicutes, Bacteroidetes, Patescibacteria, Gammaproteobacteria, Chloroflexi	-	-	1,000	10 months	Migration to the anode	An increase in metal mobility resulted in higher bioconcentration factor	(Guan et al., 2019a)
Cr(VI) (500) ^a	Not investigated	0.429	-	1,000	96	Cr(VI) reduction and precipitation as Cr(OH) ₃	Removal rate 75.4%	(Guan et al., 2019b)
Cd (3.4), Cu (78.6), Ni (67.7)	Proteobacteria, Actinobacteria, Acidobacteria, Nitrospirae	1.20	22.2	500	110	Precipitation with reduced sulphur; migration to the cathode and form oxides	Reduce Cd, Cu and Ni accumulation in rice grains by 35.1%, 32.8%, and 21.3%	(Gustave et al., 2020)
Cd (98) ^a , Pb (910) ^a	Not investigated	-	7.5 for Cd, 3.6 for Pb	1,000	143 for Cd, 108 for Pb	Migration to the cathode	Removal rate 31.0% and 44.1% for Cd and Pb, respectively	(Habibul et al., 2016a)
Pb (291.1), Zn (814.2%)	Not investigated	0.355	25.7	1,000	100	Migration to the cathode	Removal rate 37.2% and 15.1% for Pb and Zn, respectively, with 3% straw addition in the anode	(Song et al., 2018a)
Cu (600) ^a	Not investigated	-	65.8	1,000	56	Migration to the cathode	199.7 mg kg ⁻¹ Cu migrated to the cathode	(Wang et al., 2016c)
Cr (1,000) ^a	Not investigated	-	-	100	16	Cr(VI) reduction	Removal rate 99.1%	(Wang et al., 2016a)
Cu (670) ^a	Not investigated	-	-	20	9	Migration to the cathode	Reduce Cu concentration by 99.9% in 9 days	(Wang et al., 2020a)
Cd (30) ^a	Proteobacteria, Bacteroidetes, Euryarchaeota, Actinobacteria	0.39	-	1,000	35	Not mentioned	Translocation factor < 0.1 for <i>Typha latifolia</i> L.	(Yang & Shen, 2019)
Cr(VI) (100) ^a	Not investigated	0.141	-	-	14	Migration to the anode and adsorption by leaves	Removal rate 40%	(Zhang et al., 2019)
Cu (500) ^a	Not investigated	~0.5	60 (with 1 mol L ⁻¹ HCl)	1,000	74	Migration to the cathode	Removal rate 41.6% with 1 mol L ⁻¹ HCl as auxiliary reagent	(Zhang et al., 2020a)

^aArtificially spiked soil.

et al., 2020) and phytostabilization (Fatnassi et al., 2016; Honeker et al., 2019; Saran et al., 2020). With microbial assistance, metal extraction or stabilization efficiencies can be enhanced substantially. For instance, the PGPB *Burkholderia phytofirmans* inoculation enhanced the *Brassica juncea* phytoextraction efficiency of Zn, Cd and Pb by 37%, 21% and 10%, respectively (Konkolewska et al., 2020). Sunflower inoculated with *Bacillus proteolyticus* accumulated 40% less Cd and 20% less Pb in aboveground tissues, suggesting enhanced phytostabilization induced by PGPBs (Radziemska et al., 2020; Saran et al., 2020).

5.3 | Constructed wetland

Although a constructed wetland is typically designed for wastewater treatment, it is suggested that this artificial system may also be applied for land reclamation purposes. Several mechanisms are involved in metal retention/removal in a constructed wetland, including sorption, bioaccumulation, precipitation, biotransformation and sedimentation (Marchand et al., 2010). Sorption is a major mechanism dominating the environmental behaviours of metals in a constructed wetland. Both short-term retention and long-term immobilization can be achieved through various types of sorption mechanisms, including physisorption and chemisorption (Hu et al., 2020; Tran et al., 2017; Walaszek et al., 2018b). Macrophytes (i.e. aquatic plants) play a vital role in metal bioaccumulation, but only a few of them are hyperaccumulators (e.g. Ni hyperaccumulator *Limnobium laevigatum* and Cd hyperaccumulator *Potamogeton pectinatus*) (Arán et al., 2017; Lu et al., 2018), indicating that the uptake of metals may be secondary to other metal retention/removal mechanisms. In wetland ecosystems, both iron oxides and carbonates could co-precipitate with other metals (i.e. Cu, Zn, Ni, As, Cd and Pb) (Du Laing et al., 2009; Marchand et al., 2010; Sochacki et al., 2014). Mechanisms involved in biotransformation and detoxification of metals in wetlands are similar to those of microbial-based bioremediation. Microorganism-mediated oxidation or reduction will greatly affect the biogeochemical processes of heavy metals in constructed wetlands. Sedimentation occurs when macrophytes decrease water flow rates. In this sense, a studied wetland was like a stagnant pond in which high concentrations of suspended matter favour the formation of flocs, reducing metal mobility through coagulation and sedimentation (Tao & Haynes, 2016; Walaszek et al., 2018a). A constructed wetland provides a number of environmental, social and economic benefits, including flood control, biodiversity preservation, low life-cycle carbon footprint and improvement of aesthetics. However, as a water treatment technology, constructed wetland is rarely used for soil remediation with no practical applications reported to date. Besides, metal removal in the constructed wetland may take several years

(Marchand et al., 2010; Song et al., 2019a), and the uncertainties such as the death of macrophytes may hinder the metal remediation efficiency (García-Mercadoa et al., 2017).

5.4 | Stabilization using green materials

Originating in the late 1950 s, Solidification/Stabilization (S/S) is an old remediation technology with new vitality (Shen et al., 2019b). In China, the remediation market has reached 2.9 billion US dollars in 2018, with S/S being adopted by nearly half of the remediation projects (48.5%) (Shen et al., 2019b). A shift from labile forms (e.g. exchangeable and acid soluble) to stable forms (e.g. bound to organic matter, residual) of the metal geochemical fractions is the major mechanism involved in S/S; this can be achieved through surface complexation, precipitation, physical adsorption, ion exchange or electrostatic interactions (Hou et al., 2020) (Figure S1). Portland cement (PC) is the most widely applied S/S agent in metal immobilization. However, its production is associated with a high carbon footprint, accounting for 8% of the global CO₂ emission (Andrew, 2019). With the increasing awareness of global climate change, developing novel S/S agents with low environmental impact is necessary. Many attempts have been made to seek for low-carbon supplementary cementitious materials, including pulverized fly ash (Wang et al., 2019b), silica fumes (Goodarzi & Zandi, 2016), ground granulated blast-furnace slag (Wang et al., 2018a), incinerated sludge ash (Su & Shih, 2017), calcium carbide residue (Darikandeh, 2018) and reactive magnesium oxide cement (Wang et al., 2018b). However, a cement-like stabilization agent hinders the sustainable remediation of agricultural soils, since the increased mechanical strength after S/S treatment may be harmful to plant growth and yields. In addition, the decrease in soil biodiversity might be detrimental to the soil health and the agroecosystem (US EPA, 2015).

In the context of NBS, biological waste-derived stabilizing agents may be a better choice having a higher 'net environmental benefit' (Table 3). Apart from diminishing the above-mentioned environmental impacts of cement-like agents, addition of biological waste-derived materials (e.g. biochar and compost) could also promote soil fertility and plant growth. However, simply using green immobilization agents is not equivalent to being 'green and sustainable'. According to the general principles of GSR (Figure 2), several sustainability concerns must be addressed (Figure S2). For instance, utilization of some agents, such as metal-rich sewage sludge and compost, may increase the environmental risks of metal leaching. In addition, a sustainable remediation approach should look beyond the contemporary time horizon (section 2.2). For nature-based metal immobilization processes, long-term stability should be assessed. Various environmental

TABLE 3 A critical assessment of how “green” the “remediation strategies are”

Strategy	Target metal	Environmental considerations	Environmental greenness	Economic considerations
Resource recovery approaches				
Phytomining	Ni, Ti, Co, Ag, Au.	Compared with opencast mining operations, phytomining offers a metal recovery method with low impact to the environment. The plant cover prevents metal cross-media migration (i.e. through wind or percolation to groundwater)	***	Highly sensitive to the metal price—only metals with high values, such as Pb, Au, Pt and Ag, are suitable for phytomining.
Soil washing	All types of heavy metals.	Remove heavy metals thoroughly and rapidly from the soil. Extractants, such as strong acids and chelating agents, may cause contamination.	**	Moderate cost—around \$187 per tonne.
Nature-based solutions				
Phytoremediation	Metals that can be extracted, volatilized (e.g. Hg), or stabilized by plants.	Improve soil health and air quality via the function of plants.	***	Reported cost of phytoremediation ranges from \$15 to \$2,322 per cubic metre, highly site-specific.
Phytomanagement	Metals that can be extracted or stabilized by plants.	Remove heavy metals slowly while preventing their cross-media migration.	***	Profitable crops can be used for animal feeding, energy production or other purposes to realize sustainable use and management of contaminated soils. The cost ranges from \$0.4 to \$26 per cubic metre.
Microbial-based bioremediation	Hg, As and Cr.	Low disturbance to soil. Cause air pollution.	**	Hardly applicable for heavy metals. Only a few proof-of-concept investigations have been made, with no practical applications.
Constructed wetland	All types of heavy metals.	Improve soil health and air quality via the function of plants. Decrease the flooding risk.	***	Rarely used for soil remediation. The construction cost reaches around \$10,000 per acre.
Green S/S	All types of heavy metals.	Improve soil health with low disturbance.	***	Highly dependent on the soil amendment used. Natural minerals and biochar application render low cost.
Energy-efficient strategies				
Low-temperature thermal desorption	Hg	The soils treated at a relatively low temperature can be reused for agricultural purposes. Reduce energy input by ~ 35% compared to traditional high-temperature thermal desorption.	**	Reduce marginal cost from \$57 to \$38 following citric acid-facilitated thermal desorption.
Bio-electrokinetic remediation	All types of heavy metals.	Low environmental impact as metals can be removed from the soil but properly sequestered.	***	Commercialization has not taken off.

Note: The greenness scores indicate that certain remediation strategies ***well, **moderately or *poorly align with the principles of GSR.

Economic greenness	Social considerations	Social greenness	Limitations	References
*	High social acceptance. Improve aesthetics while creating place for leisure.	***	The handling of metal-rich biomass may cause secondary pollution.	(Brooks et al., 1998; Novo et al., 2017; Sheoran et al., 2009)
**	High social acceptance as a traditional remediation technology.	**	High disturbance to the environment. A high humic content in soil adds difficulty to soil washing. Not suitable for fine particles with a size distribution < 0.063 mm.	(FRTR, 2020b; US EPA 1996)
**	High social acceptance. Improve aesthetics while creating place for leisure.	***	The handling of contaminate biomass may result in secondary contamination. Phytovolatilization results in air pollution. Phytostabilization reveals poor long-term effectiveness. Long remediation duration hinders the practical applications.	(FRTR, 2020a; GWRTAC, 1997; US EPA, 2001; Wan et al., 2016)
**	Easy to be acknowledged due to the restoration and reuse of degraded barren lands. A relatively new concept as compared with phytoremediation.	**	Long duration of phytomanagement has hindered its practical application. Remediation practitioners tend to select more aggressive remediation technologies rather than gentle remediation options. Low resilience to environmental changes.	(Burgess et al., 2018; Meers et al., 2010; Zine et al., 2020)
*	Low social acceptance due to secondary contamination.	*	Low resilience to environmental changes.	(Dogan et al., 2011; Naguib et al., 2018; Zhang et al., 2017)
*	High social acceptance. Improve aesthetics while creating place for leisure.	**	Long remediation duration, low remediation efficiency.	(Tyndall & Bowman, 2016; US EPA, 1995; Vymazal, 2011)
***	High social acceptance for biochar, mineral and iron-based S/S.	***	The long-term effectiveness should be monitored.	(Komárek et al., 2013; Wang et al., 2020g; Xu et al., 2017)
**	Raise public concerns of possible Hg air pollution due to the release of this metal from soil.	*	Concerns of air Hg pollution.	(Hou et al., 2016; Ma et al., 2015)
*	A new technology that needs time to be accepted by the public.	*	This system is sensitive to changes, since microorganism plays vital roles in electric field generation.	(Chen et al., 2015; Habibul et al., 2016b; Yang & Shen, 2019; Zhang et al., 2019)

stresses, including freeze-thaw cycles, wetting-drying cycles, microbial metabolism, UV irradiation and plant growth may affect the stabilization performances in the long term (Figure S2, Figure S3) (Shen et al., 2019a; Wang et al., 2020e, 2020f). A diminished immobilization reliability results in the release and migration of metals. More long-term field studies are definitely needed to better examine the effectiveness of 'green' material-based S/S approaches.

6 | ENERGY-EFFICIENT STRATEGIES

6.1 | Low-temperature thermal desorption

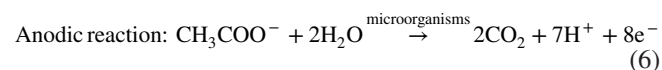
Considering its high volatility, thermal desorption has proven to be effective for Hg contaminated soil remediation. However, the high temperature (i.e. $>600^{\circ}\text{C}$) required in this process indicate relatively high costs (i.e. 480 USD t^{-1}) (He et al., 2015; Wang et al., 2020c) and a high carbon footprint (Hou et al., 2016). Furthermore, the substantial changes in physicochemical properties of the treated soil render it impossible for agricultural reuse purposes (Hou et al., 2016). Therefore, low-temperature thermal desorption approaches are required to minimize both primary and secondary impacts, and maximize the 'net environmental benefit' (Hou & Al-Tab baa, 2014). The addition of chemical agents such as citric acid (Ma et al., 2015) and FeCl_3 (Ma et al., 2014) could successfully reduce the heating temperature to 400°C while maintaining Hg removal efficiency. The enhancement in Hg removal at low temperatures may be either due to the acidic environment provided by citric acid, or the formation of volatile Hg species (i.e. HgCl_2 and Hg_2Cl_2). Microwave-induced thermal desorption is another effective means for Hg removal. Cao et al., (2018) observed that a high Hg removal efficiency (i.e. 87%) could be reached after microwave irradiation (400 W for 40 min) with granule activated carbon (GAC) as the receptor of microwave (the soil was heated to 350°C). It was observed that the exterior temperature was much lower than the interior temperature for a certain soil particle. In this way, off gas could migrate from the soil much more easily (compared to conventional desorption approaches) due to diminished heat transfer resistance.

However, a much longer time is required in low-temperature thermal desorption processes to reach the same removal rate of Hg. Utilization of chemical agents or GAC may also threaten the sustainability over the whole life cycle of the remediation process. Therefore, it is necessary to investigate the overall impact of low-temperature thermal desorption processes. Hou et al., (2016) conducted a life-cycle assessment of a thermal desorption treatment of an agricultural soil and reported that citric acid-facilitated low-temperature thermal desorption could effectively reduce the

life-cycle greenhouse gas emissions from 357 kg $\text{CO}_2\text{-eq}$ to 264 kg $\text{CO}_2\text{-eq}$ due to the reduction in electricity usage. In addition, the soil subjected to low-temperature thermal desorption could be immediately reused on-site for agricultural purposes. However, the feasibility of this method has only been confirmed at the bench scale. More studies should be undertaken at the field scale to get deeper and better insights and to further assess the feasibility of this energy-efficient strategy.

6.2 | Bio-electrokinetic remediation

Electrokinetic remediation is an effective means to mobilize contaminants using an external electric field, thus removing toxic metals from soil (López Vizcaíno et al., 2018; US EPA, 2017b). However, the relatively high energy consumption has hindered its practical applications. In order to overcome this obstacle, renewable energy should be used to generate the electric field. In recent years, microbial fuel cells (MFCs) have emerged as novel bio-electrochemical systems for sustainable wastewater treatment and soil remediation (Aarthi et al., 2019; Fang & Achal, 2019). In an MFC, electricity is generated by the microbial metabolism of organic matter under anaerobic conditions. This results in the release of electrons in the anode, which will flow to the cathode through an external circuit. The protons and electrons will produce water with electron acceptor (i.e. oxygen) in the cathode. A typical reaction mechanism is provided below (Eq. 6–7):



The key to electricity generation in this system is the microbial oxidation process in the anode. Several studies have examined the anode microbial community structure (Table 2). Compared with phytoremediation and S/S with a relatively long remediation or monitoring duration, this technique offers a quicker way to remove metals thoroughly from soil (i.e. the remediation duration ranges from 9 days to 10 months) (Table 2). A wide variety of phyla, such as Proteobacteria, Actinobacteria and Bacteroidetes, can contribute to this process. To enhance the microbial activity in the anode, inorganic nutrients (e.g. Ca^{2+} , Mg^{2+} , K^+ , PO_4^{3-} and NH_4^+) can be added in the anode chamber (Chen et al., 2015; Habibul et al., 2016a; Wang et al., 2016a). Sometimes, external organic carbon sources can also be added in various forms, such as straw (Song et al., 2018a) and leaves (Zhang et al., 2019).

Although MFC is still in its infancy, this kind of bio-electrochemical system has derived into distinct forms (Figure

S4). A typical three-chamber MFC can be applied either horizontally (Figure S4a) or vertically (Figure S4c). In this system, positively charged metals (e.g. Cu, Zn, Cd and Pb) will migrate to the cathode, while negatively charged metal(loid)s (e.g. Cr(VI) and As(III)) will migrate towards the anode. In a plant, MFC (P-MFC, Figure S4b), rhizodeposits, root exudates and other organic matter generated through photosynthesis can act as carbon sources for the anodic reactions. In this sense, plants can enhance the generation of electricity, but do not dominate this process. Another example is the adsorption-MFC system (A-MFC, Figure S4d). In this system, after the migration of contaminants towards the cathode or anode, the adsorbent in the vicinity of electrodes will adsorb metal cations or anions effectively. For instance, Zhang et al., (2019) designed an A-MFC for soil Cr(VI) removal. Fallen leaves were adopted as the natural biosorbent and the anode organic carbon source. Sorption tests confirmed that this sorbent has high affinity towards Cr(VI) (adsorption capacity 14 mg g^{-1}), and the removal efficiency of Cr(VI) in soil reached 40%.

The migration of heavy metals is highly dependent on their geochemical fractions. Compared with less-labile forms (i.e. residual, bound to organic matter), labile forms (i.e. exchangeable and acid soluble) migrate more easily under the external electric field (Chen et al., 2015; Wang et al., 2020a). Thus, a high portion of labile fractions may result in a promising remediation efficiency. In order to achieve this goal, auxiliary reagents, such as citric acid, acetic acid or hydrochloric acid can be adopted to mobilize the metals through soil pH reduction (Zhang et al., 2020a). Utilization of MFCs in soil remediation is a novel technology. However, application in the field seems less successful so far, most probably due the soil heterogeneity and the complex interactions of various environmental factors. Therefore, more efforts are needed to examine the feasibility of this technique in field studies. Bio-electrokinetic remediation is a new technology that requires time to be accepted by the public. Current studies are mainly proof-of-concept investigations of this technology (Tables 2, 3). Feasibility of practical applications should be assessed via field demonstrations.

7 | SUSTAINABILITY CONCERNS, CHALLENGES AND FUTURE DIRECTIONS

A critical summary of how 'green' these aforementioned remediation strategies have been provided in Table 3. It is suggested that overall phytoremediation and green S/S are the 'greenest', while microbial degradation, bio-electrokinetic remediation and soil washing may have more significant environmental, economic and social limitations diminishing the overall sustainability. However, remediation practitioners should bear in mind that adoption of green strategies is not

equivalent to 'green and sustainable remediation', since 'sustainability' is very subjective, and highly site-specific. To assess whether a remediation strategy can achieve the maximum environmental, social and economic benefits, sustainability assessment (SA) should be conducted. Sustainability assessment of a remediation approach depends on the selection of a wide range of environmental, social and economic considerations amongst various stakeholders (Bardos et al., 2016). Various SA tools can be adopted to examine the sustainability of a heavy metal remediation approach. Life-cycle assessment (LCA), a quantitative method standardized by the International Organization for Standardization (ISO, 2006), is generally considered to be the most integrated sustainability assessment tool. LCA will provide fresh insights into the green remediation processes and assist in wise decision-making through combination with other useful tools, such as health risk assessment (Hou et al., 2017) and multi-criteria analysis (Song et al., 2018b). To aid in the selection of green remediation strategies for certain contaminated sites, more attempts should be made to further evaluate the overall sustainability of 'green' technologies in the future.

Green remediation approaches, including the utilization of green materials, resource recovery strategies, nature-based solutions and energy-efficient strategies, have received much attention with view to the remediation of soils contaminated with heavy metals. However, several challenges exist in this field, which may hinder the overall sustainability:

Firstly, in plant-based remediation technologies, the metal-rich plant tissues collected after harvesting may become a contaminant source if not properly disposed. Phytostabilization is only effective within the rhizosphere, making it impossible for the remediation of deep soils. In addition, phytovolatilization of Hg result in air pollution, and the volatilized Hg (mostly in the form of elemental mercury) in the atmosphere may return to the ecosystems through both dry and wet deposition.

Secondly, green remediation materials, such as biochar synthesized from the contaminated biomass (e.g. plants used in phytoextraction) and metal-containing industrial waste-derived materials, may result in metal solubilization and mobilization. Although green remediation materials have proven effective for soil metal immobilization by a number of studies, their long-term stability has not been fully investigated. Due to various natural forces such as freeze-thaw erosion, wetting-drying cycles, UV irradiation, plant growth and groundwater flow, the stabilized metals may be remobilized, causing severe risks on the longer term.

Moreover, with view to microbial-based remediation approaches, the low resilience of microbial-based strategies hinders the sustainability. Microorganisms are sensitive to changes. For instance, in bioremediation or bio-electrokinetic remediation approaches, slight changes in temperature, redox potential, moisture, nutrients and organic matter may induce

a significant shift in the metabolism pathways of microorganisms, leading to the failure of microbial-based detoxification and electrokinetic removal processes.

In addition, the long remediation duration hinders the adoption of green remediation technologies due to decreased overall sustainability. Although plant-based and microbial-based remediation technologies appear to increase the environmental sustainability, remediation strategies with shorter time frames are often preferred, in order to allow a rapid return on the investment to assure economic sustainability. Therefore, from an applied point of view, a balance should be made between the environmental and economic benefits.

Considering that a single type of green remediation strategy may have significant limitations, it is suggested that future studies should integrate 'green' approaches to produce more effective remediation strategies. For instance, green stabilization materials, such as biochar and minerals, or a mixture of both, can act as both immobilizing agents and soil fertilizers, and assist in phytostabilization through enhancing metal immobilization and promoting root growth. Novel plant-based chelating agents can not only be adopted in soil washing, but also aid in phytomining, phytoextraction, low-temperature thermal desorption and bio-electrokinetic remediation by increasing the metal mobility. Carbon-rich remediation materials (i.e. biochar) may even act as a carbon source and enhance the activities of microorganisms in microbial-based remediation and bio-electrokinetic remediation approaches. The combination of green strategies will reveal a 'synergistic effect', thus promoting the metal remediation efficiencies.

8 | CONCLUSIONS

Soil contamination by heavy metals has raised much concern owing to the potentially toxic effects on human beings and ecosystems. With the emergence and development of the GSR movement, green remediation strategies have been adopted for metal removal, stabilization or detoxification, including resource recovery strategies such as phytomining and soil washing, nature-based remediation methods such as phytoremediation, microbial oxidation/reduction and constructed wetland, and energy-efficient strategies including low-temperature thermal desorption and bio-electrokinetic removal. In those technologies, plants and microorganisms play vital roles as does the adoption of green amendments, such as biological and industrial waste-derived materials, natural minerals, oxides and green-synthesized nanomaterials, which increases the environmental, social and economic benefits. Various green remediation strategies, such as phytoremediation and soil washing, have been already widely adopted in commercial applications, while others such as biochar-based stabilization, citric acid-facilitated low-temperature thermal

desorption and bio-electrokinetic remediation have only been investigated on a laboratory scale. However, in both cases, future attempts should be made to further test the practical applicability of these green remediation strategies.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section.

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