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FOCUSED REVIEW

A review for recent advances on soil washing remediation technologies

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Abstract
Contaminated soils have caused serious harm to human health and the ecological environment due to the high toxicity of organic and inorganic pollutants, which has attracted extensive attention in recent years. Because of its low cost, simple operation and high efficiency, soil washing technology is widely used to permanently remove various pollutants in contaminated soils and is considered to be the most promising remediation technology. This review summarized the recent developments in the field of soil washing technology and discusses the application of conventional washing agents, advanced emerging washing agents, the recycling of washing effluents and the combination of soil washing and other remediation technologies. Overall, the findings provide a comprehensive understanding of soil washing technology and suggest some potential improvements from a scientific and practical point of view.

Keywords Soil remediation · Soil washing · Eluent · Heavy metal · Organic pollutant

Introduction

With the intensification of human activities such as mining, coal burning, livestock breeding and agricultural production, a large number of heavy metals and organic pollutants, including lead, cadmium, antibiotics, polycyclic aromatic hydrocarbons, phthalates and pesticides, have been discharged into various environments (Khan et al. 2021; Tao et al. 2020). Due to the strong retention capacity of organic matters and minerals, soils have maintained a large number of pollutants in the past few decades. In addition, sewage irrigation, surface runoff and atmospheric dry and wet deposition will also transfer many pollutants into the soil environment, resulting in serious pollution. Most of these pollutants are highly toxic and threaten the safety of ecosystems (Liu et al. 2021a; Trellu et al. 2021). To avoid potential harm to human beings, the remediation of contaminated soils is important and urgent and has also attracted the attention of many scientists all over the world.

Currently, there are many remediation technologies for contaminated soils, such as solidification, stabilization, soil washing, chemical redox, and electric remediation. Among them, the soil washing method, leaching pollutants from contaminated soil by desorption, chelation, dissolution and other chemical actions under the addition of specific solutions, has the advantages of flexible application, simple operation, short period, low cost and high removal efficiency (Fig. 1) (Chen et al. 2021; Fazle Bari et al. 2022; Tran et al. 2022). In particular, pollutants in soils are permanently

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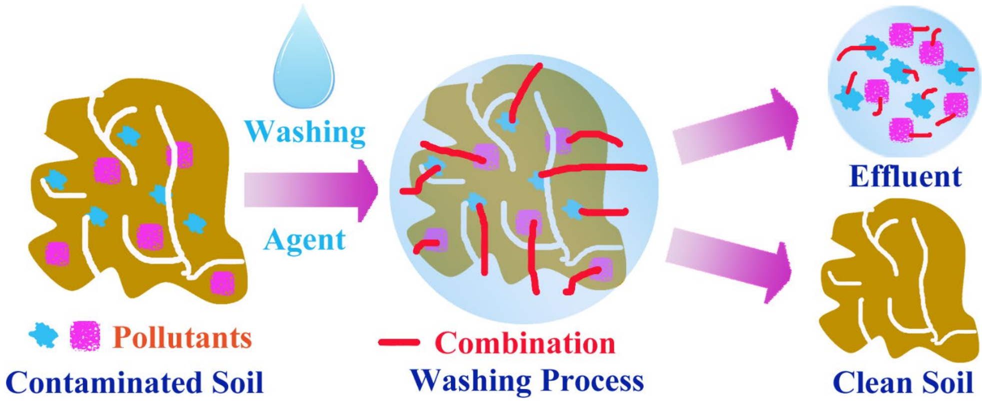

Fig. 1 Mechanism schematic of soil washing technology

removed by soil washing without subsequent continuous monitoring and remediation (Lee and Son 2021; Liu et al. 2021a). Therefore, soil washing remediation technology has good applicability and can be further popularized.

In recent years, many advanced scientific achievements have been reported but are rarely summarized. Our review aims to bridge this gap by describing the application of conventional washing agents, emerging washing agents, the recovery of washing effluents and the combination with other remediation technologies. The advantages and limitations of the latest studies on soil washing are introduced. The related findings will contribute to the development of soil washing remediation technology.

Conventional washing agents

Washing agents are crucial for the washing performance of contaminated soils. In the past few decades, many specific washing agents have been developed and applied, mainly including inorganic agents, organic chelating agents and surfactant agents (Table 1). Inorganic agents, such as ferric chloride, phosphoric acid, nitric acid and hydrochloric acid, have often been used to remediate heavy metal-contaminated soils (Liu et al. 2021a). Wang et al. compared the removal performance of six conventional inorganic washing agents, phosphoric acid, potassium carbonate, potassium acetate, potassium dihydrogen phosphate, nitric acid and potassium nitrate, on farmland soil contaminated by heavy metals (Wang et al. 2020d). The removal ratio of nitric acid is the highest due to mineral acidolysis, metal compound dissolution, soil desorption and ion exchange. However, the soil enzyme activities and soil microbial diversities decreased, and the residual state of heavy metals could be transformed into bioavailable heavy metals after the washing process because of the significant change in soil properties, such as low pH (Zhai et al. 2018). Using calcium hydroxide to neutralize nitric acid-washed soil is a feasible method to improve the content of soil available nutrients and reduce adverse effects. Compared with strong acid agents, mild washing agents have attracted more extensive attention. Ferric chloride leads to the production of H^+ and forms a competitive adsorption process with heavy metals. Cl^- can also react with heavy metals to improve solubility (Guo et al. 2016). The results show that the removal ratio of lead in farmland soil by ferric chloride is extremely high, similar to hydrochloric acid (Moon et al. 2021). In addition, the soluble and exchangeable parts of lead are significantly reduced after washing with ferric chloride. However, the removal of copper and zinc is quite limited.

Organic chelating agents are also conventional washing agents, including natural chelating agents such as citric acid, malic acid and oxalic acid, as well as artificial chelating agents such as ethylenediaminetetraacetic acid and aminotriacetic acid (Fazle Bari et al. 2022; Shukla et al. 2022). Mineral loss in soil was the important limiting factor for citric acid-like chelating agents. Recently, a mixed cleaning agent (destabilizing agent, oxalic acid, citric acid, tartaric acid; exchanger, potassium dihydrogen phosphate) has been proposed to effectively remove arsenic but with little loss of minerals. Iron minerals are destroyed by organic acids and release arsenic first; then, potassium dihydrogen phosphate is immobilized on the surface of iron minerals by a competitive adsorption process with arsenic, which reduces the loss of iron minerals (Wei et al. 2022). Natural organic chelating agents are generally biodegradable and environmentally friendly, which can prevent the occurrence of secondary pollution, but their complexation ability with pollutants is much lower than that of artificial chelating agents (Liu et al. 2021a). Therefore, to explore efficient and bio-degradable organic chelating agents is challenging. Begum et al. compared four artificial organic chelators and proved that 2-[bis(carboxymethyl) amino]pentanedioic acid and 2-(1,2-dicarboxyethylamino)-3-hydroxy-butanedioic acid had higher washing efficiency for radioactive strontium (Begum et al. 2020; Gluhar et al. 2020). Wang et al.

Table 1 Typical conventional and emerging soil washing agents

Washing Agent	Dosage (g/mL)	pH	Time (min)	Temperature (℃)	Leaching Compound and Removal Ratio	Removal Mechanism	Advantage	Disadvantage	Reference
$FeCl_3$ (HCl)	1:2	1.5 (< 0.5)	60	20	55% for lead (53%)	mineral acidolysis; dissolution; ion exchange; competitive adsorption	high removal capacity	low pH; nutrient loss; soil microbial diversity decreased	Moon et al. 2021
GLDA	1:5	5	120	—	52% for cadmium; 72% for lead; 34% for zinc	acid dissolution; ion exchange; surface complexation	biodegradable	high cost	Wang et al. 2020b
RL	1:2	—	1440	25	63% for petroleum hydrocarbon	solubilization; degradation	biodegradable	high cost	Lai et al. 2009
NC	1:20	6	1440	30	23% for phenanthrene	interruption of PHE/SOM-metal/mineral linkages	detoxification	low efficiency	Yin et al. 2021
CH-M	1:7	—	30	40	92% for lead	dissolution; complexation	rapid; low cost; few changes on soil surface	—	Huang et al. 2022

Note: $FeCl_3$, ferric chloride; HCl, hydrochloric acid; GLDA, glutamate–N,N–diacetic acid; RL, rhamnolipids; NC, nanocellulose; CH-M, choline chloride-malonate acid; —, not mentioned

proposed four biodegradable artificial organic chelators to remove cadmium, lead and zinc and confirmed the great application potential of iminodisuccinic acid and glutamate-N,N-diacetic acid due to strong and synergistic effects of acid dissolution, ion exchange dissolution and surface complexation dissolution (Wang et al. 2020b).

Surfactants, another conventional washing agent, are amphiphilic chemicals with both hydrophilicity and hydrophobicity in their molecular structures. The unique molecular structure of surfactants can improve the water solubility of pollutants, especially organic pollutants, to improve the washing effect (Liu et al. 2021b; Rahman et al. 2022). In recent years, biosurfactants have been proposed and have attracted extensive attention because of their biodegradability and low toxicity (Mishra et al. 2021). Rhamnolipid is the most common biosurfactant in soil washing technology and can form ionic bonds with heavy metals and rhamnolipid metal complexes. Lai et al. demonstrated that rhamnolipids can also be used to leach organic pollutants efficiently from soil, such as petroleum hydrocarbons, whose solubilization effect is much higher than that of the nonbiosurfactants Tween 80 and Triton X-100 (Lai et al. 2009). In addition, biosurfactants can stimulate microbial activity for further degradation of organic pollutants (Sun et al. 2021). Guo et al. reported a novel biosurfactant derived from swine wastewater by using *Pseudomonas frederiksbergensis* for the treatment of hydrophobic organic pollutants. In a wide range of pH values and salt concentrations, it can improve the water solubility of benzo(a)pyrene and promote the biodegradation process (up to 84.8%) (Guo and Wen 2021). In addition, the properties of contaminated soil were not destroyed, while the microbial activity, enzyme activity, and water holding capacity were increased. Christopher et al. proposed a surface-modified amino acid-enhanced biosurfactant, which is used to efficiently wash aromatic hydrocarbons in industrial soil with low phytotoxicit (Christopher et al. 2021). Although biosurfactants have higher washing efficiency, better biodegradability and lower toxicity, the greatest challenge is the high price due to the complex production process. To improve the usability and efficiency of conventional washing agents, the modification of washing agents, optimization of washing conditions and combination of multiple technologies will be the main topics in the future.

Emerging washing agents

In recent years, many research groups have also been committed to developing various novel soil washing agents to improve removal efficiency and environmental friendliness (Table 1) (Guo et al. 2022; Shukla et al. 2022). Yin et al. proposed an environmentally friendly nanocellulose washing agent for the green remediation of phenanthrene-contaminated soil (Yin et al. 2021). The hydroxyl groups of nanocellulose can react with Fe–O, Si–O, and Mn–O to destroy the phenanthrene/organic metal/mineral bond and release phenanthrene from soil. In addition, nanocellulose can be deemed a barrier between algae cells and toxic phenanthrene, which confirmed that nanocellulose is beneficial to reduce the harm of residual phenanthrene to living beings. For heavy metals, the extracellular polymer *Aspergillus tubingensis* F12 was proposed, and the maximum leaching capacity reached 3.7 mg/g (Tang et al. 2021a). The key mechanisms are ion exchange, biosorption and redox (extracellular polymeric substances as reduction agents) in the two-step ionization/re-immobilization. In addition, the ecological impact was evaluated, with little impact on soil characteristics and biological community structure. Wu et al. provided a sodium alginate-coated silicon sulfide nano zero valent iron nanocomposite to remove nickel (25.94%), cadmium (44.50%) and chromium (62.6%) in a variety of heavy metal-contaminated soils through adsorption, reduction and coprecipitation (Wu et al. 2020). The composite could be easily separated, and heavy metals could also be recycled. Interestingly, the composite could also be used repeatedly with high performance. To have high sustainability, the removal capacity of the washing agents should be also important. The deep eutectic solvent mixed with choline chloride and malonic acid or ethylene glycol has a good removal efficiency of lead in contaminated soil (92.12% or 95.79%) (Huang et al. 2021, 2022). In the washing process, lead nitrate will be dissolved and converted into complexes with carboxyl groups (Zhang et al. 2021). Additionally, there was no significant change in the soil mineral phase and surface functional groups, which was beneficial for the reuse of the washed soil. Dilution with water can reduce the viscosity and cost of the proposed deep eutectic solvent to further reduce the remediation cost(Tang et al. 2021b). For emerging washing agents, in addition to the removal performance and environmental impact, the potential of large-scale production and recovery should also be studied for practical application.

Recycling of washing effluents

After the washing process, the effluent is produced, contains a large number of dissolved metal ions or organic pollutants, and could result in secondary pollution or even more serious harm to the ecosystem. Therefore, the treatment of soil washing effluent is necessary and is also conducive to recycling and reducing the remediation cost greatly (Fig. 2) (Bianco et al. 2022; Trellu et al. 2021). Simple reactions were always feasible to remove pollutants from soil washing effluents. Recently, an inorganic coagulant composed of thenardite, calcium carbonate, and tychite crystals has been successfully applied to remove suspended soil, strontium, and caesium and make radioactive soil washing effluent recyclable (Lee et al. 2022). Additionally, Kim et al. proposed a reduction reaction to selectively recover ferrous oxalate and remove arsenic from soil washing effluents. Ferrous oxalate with low solubility was formed by the addition of dithionite and was collected as a resource. Then, the sulfide produced by the decomposition of dithionite can react with arsenic (Kim and Baek 2019). For organic pollutants, the degradation process was promising. Liu et al. reported a feasible method, an electrochemical process coupled with Fe^{2+}/persulfate (Fe^{2+}/PDS) oxidation, to remove the organic pollutant diesel from Tween 80-derived soil washing effluent (Liu et al. 2022). The removal ratio of diesel can reach 88.6%, and the recovery rate of Tween 80 can reach 70.0%. Soil respiration experiments confirmed that the electric/iron/persulfate process does not produce highly toxic products when degrading diesel in soil washing effluent. Additionally, a novel photoelectrochemical cell was proposed that was efficient for the remediation of soil washing effluent containing chlorinated hydrocarbons (Cotillas et al. 2020). Under the condition of low electric charge (<5 Ah dm^{-3}), organochlorine compounds could be completely mineralized. The removal ratio of total organic carbon reached 94.58%, and the toxicity of the soil washing effluent was significantly reduced. Ultraviolet radiation and its combination with peroxydisulfate are also effective in selectively degrading phenanthrene and recovering sodium dodecyl sulfate used as a soil washing agent (Wang et al. 2020c). $^1O_2$, •OH and $SO_4^{\bullet-}$ were the main oxidizing species, and the removal ratio was 98.7% with a time consumption of only 8 min. The recovered sodium dodecyl sulfate displays great washing performance for phenanthrene-contaminated soil. To enhance the treatment performance for the soil washing effluent, chemical and physical processes were combined. Wang et al. proposed a hierarchical porous CX-TiO_2 composite that can selectively adsorb 79.87% phenanthrene directly and then degrade 97.8% phenanthrene in TX-100-derived washing effluent (Wang et al. 2020a). Generally, soil composition, pH, ionic strength, dissolved organic matter, types and forms of pollutants are important influencing factors during the treatment of soil washing effluents, and related studies are extremely limited. There are still lots works needed to do for practical recycling of washing effluents.

Soil washing coupled with other remediation technologies

To maximize remediation efficiency and avoid potential risks, soil washing can be combined with other remediation technologies and show excellent performance (Fig. 3) (Harati et al. 2021; Muñoz-Morales et al. 2021). Compared with the traditional soil washing process, stirring and ultrasonic-assisted soil washing can improve the removal efficiency because particle movement, particle collision and scrubbing are conducive to the desorption of pollutants from the soil (Park and Son 2017). Choi et al. reported that ultrasonic processes can assist in extracting a large number of heavy metals and organic pollutants under less extreme

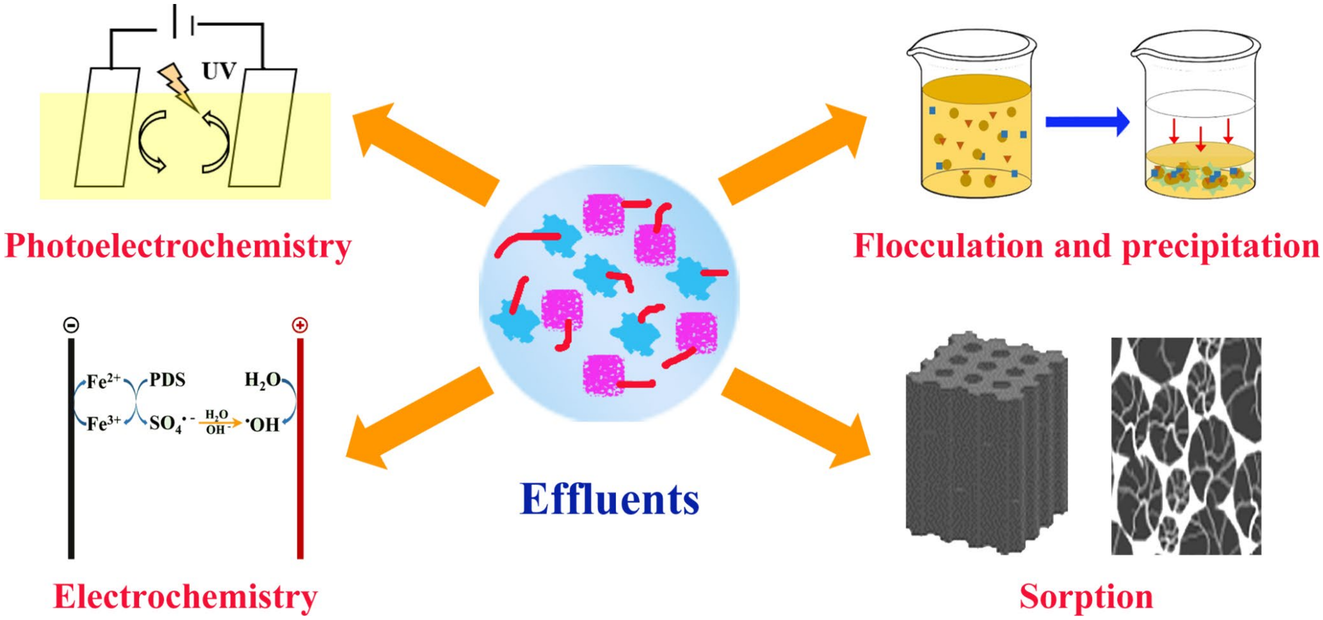

Fig. 2 Main methods for the recycling of washing effluents

washing conditions, such as lower washing agent concentrations, lower ratios of liquid to soil, and small soil particle sizes (Choi et al. 2021; Lee and Son 2021). Recently, a two-step strategy was proposed, soil washing first and then oxidation, which was beneficial to extract organic pollutants from soil and degrade extracted organic pollutants from washing effluents simultaneously. Liu et al. and Suanon et al. combined soil washing and electrochemical advanced oxidation and found that the removal efficiency of diesel and organochlorine pesticides increased significantly, even up to 88% (Liu et al. 2020; Suanon et al. 2020). A washing and subsequent photoelectrochemical method was also developed and showed an economical and effective method. Tao et al. used ethylenediaminetetraacetic acid and Tween 80 to extract copper (73.5%) and phenanthrene (68.1%) simultaneously (Tao et al. 2020). Then, the photoelectro-persulfate process was carried out, 83.6% of copper was reduced, 99.6% of phenanthrene was eliminated, and 36.8% of ethylenediaminetetraacetic acid and 94.0% of Tween 80 were recovered. In addition to advanced oxidation, electrokinetics can also assist soil washing with high performance. Ma et al. showed that the removal ratio of cadmium can be increased to 97.79% from soil by combining electrokinetics and soil washing (Ma et al. 2019). Immobilization was feasible to remediate heavy metal-contaminated soil, especially combined with soil washing. The bioavailability of heavy metals remaining in soil can be decreased significantly by immobilization materials, and the soil environment can be improved (Zhai et al. 2018). From the point of environmental friendliness and efficiency, combining soil washing and bioremediation is more promising (Srivastava et al. 2022). However, the relevant research is very limited to date. Fanaei et al. developed a method, biosurfactant washing combined with H_2O_2-stimulated biotreatment, for the green remediation of heavy oil-contaminated soil (Fanaei et al. 2020). 86% of petroleum hydrocarbons were released from the soil and biodegraded by peroxidase produced by biostimulation with H_2O_2. In addition, Xiao et al. proposed first soil washing and then phytoremediation for heavy metal-contaminated soils (Xiao et al. 2019). Compared to strong acids (hydrochloric acid) and chelators (ethylenediamine tetraacetic acid and nitrotriacetic acid), low molecular mass organic acids are more suitable for use in the phytoremediation of ryegrass. The removal efficiency is similar, but low molecular mass organic acids have little interference on soil structure and microbe activities. For multitechnology combinations, most of them are at the laboratory scale, lack confirmation and are optimal for engineering applications.

Conclusions

Soil washing remediation technology has broad application prospects for various contaminated soils. For developed washing agents, the best operating conditions for various soils and pollutants and potential risk should be further confirmed. Additionally, the specific washing mechanisms are of great significance to scientifically improve the removal performance, which should be given much attention. Faced with the complex pollution status, such as combined pollution, the study of mixed, graded or alternate washing should be strengthened for the simultaneous removal of various

Fig. 3 Main remediation technologies combined with soil washing

different types of pollutants. In addition to existing soil washing agents, developing advanced soil washing agents with the advantage of high efficiency and environmental friendliness was also the focus in soil washing technology. It is important to note that for emerging pollutants, such as resistance genes, nanoparticles and microplastics, specialized soil washing agents are limited. To reduce secondary pollution risk and remediation cost, developing more feasible methods to recycle soil washing effluents would play important roles. Combining soil washing with other remediation technologies, especially emerging technology, such as plasma technology, has great prospects, expands advantages and reduces disadvantages. Additionally, green remediation oriented to the combination of soil washing, phytoremediation and microbial remediation will become a future focus. Given the destruction of the soil environment by the washing process, seeking methods to restore soil quality, such as the addition of targeted soil conditioners, is urgent and necessary in the future.

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