

Review

The Removal of Heavy Metals by Low Molecular Organic Acids (LMWOAs) from Contaminated Soils and Key Influencing Factors: A Literature Review

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Abstract

With the widespread consensus on the sustainable development of economy and society, the environmental issues of heavy metal pollution in soils have received considerable attention. Confronted with these issues, soil washing has been widely studied, due to its advantages of simple operation, wide application, high efficiency and low costs. However, reducing the secondary contamination of the washing agents and selecting the best washing agents have been a barrier to the development of washing technology. Based on this background, this study comprehensively investigated the concentration levels of heavy metals in different types of contaminated sites, analyzed the current research status in this field, and pointed out the urgency and challenge of soil remediation for heavy metal pollution. Meanwhile, the advantages and disadvantages of different remediation techniques and their application case studies for heavy metal contaminated soils were summarized. In addition, the washing effects of different types of low molecular weight organic acids on heavy metal pollutants in soils and their key influencing factors were also fully discussed. Finally, the limitation in this field area and future research directions were pointed out. This review would be expected to provide practical engineering application and development for soil washing.

Keywords: heavy metals, soil remediation, soil washing, low molecular organic acids, influencing factors

Introduction

As an important part of the ecosystem, soil are the source and sink for the migration and transformation of pollutants. Over the past 200 years, the continued industrialization worldwide has led to severe soil contamination [1]. According to incomplete statistics, 15% of agricultural land in China has been polluted by anthropogenic activities during the past 10 years. Soil heavy metal pollution is one of the most serious environmental problems worldwide. Several pollution sources contribute to heavy metal pollution in soils, including weathering of soil parent materials, mining, smelting, traffic, pesticide, and fertilizer use. Heavy metal pollutants in soils can transfer through soil-food chain-human body and other exposure routes. Because of their bioaccumulation and non-degradation, their significant enrichment will damage the functions of human small intestine, stomach, kidney, lung and cardiovascular tissues, and even may lead to cancer, malformation and mutation [2]. According to the Institute for Health Metrics and Evaluation (IHME), lead (Pb) exposure amounted to 540,000 deaths worldwide in 2016, and the annual cost of cognitive defects of children attributed to Pb contamination was 1154 billion US dollars [2]. Consequently, heavy metal pollution in soils has received widespread attention and is considered as a key indicator of pollution.

From the perspective of sustainable utilization and development of lands, it is urgent to develop economically feasible, environmentally friendly and efficient remediation technologies. It has witnessed an increase in papers about soil remediation during 1985-2022. In particular, the publication number of papers on soil remediation and soil remediation with heavy metal pollution has increased dramatically since 2010. It is clearly illustrated that the studies on the remediation technologies of contaminated soil, especially heavy metal contaminated soils, have attracted worldwide attention (Fig. 1). Soil remediation can reduce pollutants or prevent pollutants from entering the human bodies. Furthermore, it facilitates the redevelopment and use of abandoned contaminated lands, aiding economic and social development. In terms of the remediation principle, soil remediation refers to the removal of heavy metal pollutants from soils and the reduction of their mobility in soils. The remediation techniques can prevent heavy metals from posing the potential threats to human beings, animals and plants, and restoring the ecological function of soils [4]. For example, washing reagents such as surfactants, chelating agents and desorbents can remove heavy metals from contaminated soils, while chemical stabilization materials can significantly reduce their mobility and bioavailability. When it comes to the types of soil remediation technologies, physical remediation (soil replacement, electrokinetic and thermal desorption), chemical remediation (chemical washing, chemical stabilization, solidification/stabilization, chemical

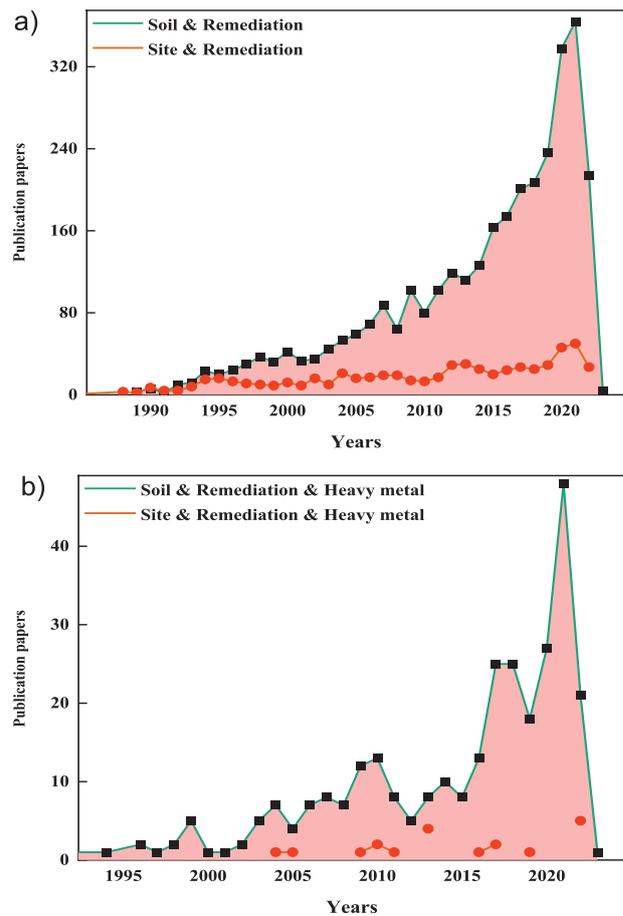


Fig. 1. a) Published literature statistics related to soil remediation; b) Published literature statistics related to remediation of heavy metals pollution in soil (source: web of Science; Key words: Remediation, Soil, Site and Heavy metal; Retrieved: September 4, 2022).

oxidation/reduction), bioremediation (phytoremediation, microbial remediation, plant-assisted remediation and microorganism-assisted remediation) are included [5-6]. It is reported that the application rates of physical, chemical and biological remediation technologies used for the remediation of heavy metal contaminated sites from Guangxi, Fujian, Liaoning and Yunnan provinces in China have reached up to 27.1%, 18.9% and 10.7%, respectively [7]. The case studies of the application of different remediation techniques are summarized in Table 1. In fact, these techniques have their own advantages and disadvantages, in terms of applicability, efficiency and costs. Physical remediation technology can directly remove or separate heavy metals from contaminated soils, but the workload and remediation costs are high, and it is also easy to destroy soil structure. Bioremediation by the use of plants and microorganisms can reduce the concentrations and toxicity of heavy metals in contaminated soils without secondary pollution, but the remediation time is long and the subsequent treatment faces significant challenges [8]. The serious defect of chemical stabilization remediation is that long-term sampling is needed to

Table 1. Application cases of different remediation technologies.

Categories	Technologies	Soil types	Heavy metals	Results	Reference
Physical	Soil replacement	Farmland near mining area	As, Pb, Cu, Cd and Zn	It occurred decrease in the concentrations of As (49.7%), Pb (48.6%), Cu (45.1%), Cd (28.8%), and Zn (32.1%) after treatments	[61]
	Electrokinetic remediation	Farmland near an electronic dismantling plant	Cd	The average removal efficiency is 88.3%	[62]
	Vitrification	Mine soil	As, Mn, Hg, Cu, Pb, Ni and Zn	The immobilization of Fe, Mn, Ni, Cu and Zn in samples treated at the highest temperatures (1250 to 1350°C)	[63]
	Thermal desorption	Mine soil	Hg	The removal efficiency reached up to 99.1%.	[64]
Chemical	Stabilization/immobilization	Shooting range	Pb, Cd and Zn	The addition of P compounds resulted in the immobilization of Cd, Pb and Zn by 1.56~76.2%, 3.21~83.6%, and 2.31~74.6%, respectively.	[65]
	Washing	Battery metal-contaminated site	Cu, Ni, Zn, Cd and Pb	The Cd concentrations in the solution decreases from 166 to 0.04 mg/kg.	[66]
	Oxidative/reductive	Dewatered tannery sludge	Cr, Pb, Cd, Ni, Cu and Hg	The H ₂ O ₂ treatment enabled to extract 70% of chromium within less than 4 h.	[67]
Biological	Phytoremediation	Heavy metal polluted lateritic soil	Zn, Cd and Pb	The ability to uptake Zn and Cd was higher, approximately 13 times on average than its ability to uptake Pb.	[68]
	Animal remediation	Industrial and mining towns	Cd, Cu, Zn and Pb	Epigeic earthworm <i>Metaphire californica</i> bioaccumulated more Cd (0.27~0.60 mmol/kg), while endogeic earthworm <i>Amyntas hupeiensis</i> and anecic earthworm <i>Amyntas asacceus</i> bioaccumulated more Cu (0.55~1.62 mmol/kg) and Zn (2.86~6.46 mmol/kg) from soils, respectively.	[69]
	Microbial remediation	Rhizosphere soil	Cr(VI)	Maximun Cr(VI) dissipation of 40% was registered by <i>Streptomyces</i> sp. Z38.	[70]

monitor the release of heavy metal pollutants in the post remediation stage, and thus the remediation cost is too high [9-10]. Soil washing refers to the leaching of heavy metal pollutants from contaminated soils through chemical mechanism such as desorption, chelation and dissolution with the addition of specific solutions. Compared with other remediation technologies, soil washing has the advantages of flexible application, simple operation, short time, low cost and high removal efficiency [11]. Especially, the removal of heavy metal pollutants from soils can be accomplished with no need for subsequent long-term monitoring and remediation. As a result, soil washing has recently been studied extensively.

Based on the present scientific backgrounds, the main aims of this review were to (i) analyze the heavy metal pollution in contaminated sites, and then point out the urgency and challenge of soil remediation; (ii) introduce the principle and its research status of soil washing; (iii) present the application cases of low molecular organic acids as soil washing agent and highlight the key factors affecting their washing effects, and (iv) present the research perspective on the development trends of soil washing.

Results and Discussion

Pollution Characteristics of Heavy Metals in Contaminated Sites

The contaminated site is an area where soil contamination has been identified as posing the potential risks to humans, ecosystems or other receptors. Over the past few decades, the number of contaminated sites has increased dramatically at global scale [12]. There are over 10 million contaminated sites worldwide [12]. Australia has about 80,000 contaminated sites [13]. According to EU statistics, there are about 2.5 million potentially contaminated sites in European countries, most of which need to be urgently remediated [14]. At present, there are about 38000 potentially contaminated sites in China [15]. According to relevant reports, there are more than 200,000 contaminated sites in China, of which 320 are ranked in seriously polluted, covering 5.48 million hectares [16]. Based on the search results of Web of Science, large quantities of studies on contaminated sites have been conducted globally. The investigation results of about 2500 sites in 75 counties from China indicated that there are more

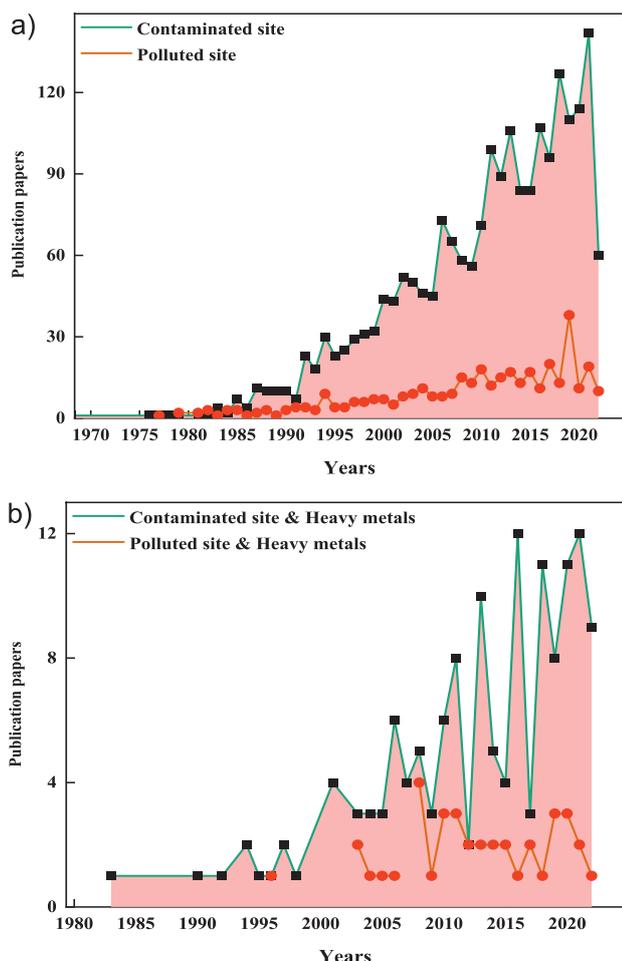


Fig. 2. a) Published literature statistics related to contaminated/polluted remediation; b) Published literature statistics related to heavy metals contaminated/polluted remediation (source: web of Science; Key words: Contaminated site, Polluted site and Heavy metal; Retrieved: September 4, 2022).

than 500 industries, which have produced or used 20644 kinds of chemicals and 1012 kinds of pollutants [17]. As shown in Fig. 2, 2483 research and review papers were published during 1968-2022. Among them, the number of research papers were 181 about heavy metal contaminated sites, indicating that heavy metal contaminated soils are the most common research hotspots in environmental science. Heavy metals generally refer to metals and metalloids with a density greater than 5 g/cm³, such as As, Hg, Zn, Cu, and Ni. Heavy metals are the most important inorganic pollutants in contaminated sites, accounting for about 34.8% of the soil pollution [18]. More than 5 million sites worldwide are reported to have heavy metal pollution in soils [19].

Although the studies on heavy metal pollution in contaminated sites have increased rapidly in the past decades, there is still a lack of the pollution investigation and assessment on the heavy metal concentrations in contaminated sites. Non-ferrous metal mining and smelting, electronic dismantling, electroplating,

tanning, as well as other industrial production activities are the main sources of metal pollution. For example, about 9.03 million tons of Zn were released into the environment, due to mining and smelting activities in China in 2018 [20]. Approximately 1.29×10^5 tons of Cr are released into the environment globally each year, most of which accumulate in soils, resulting in serious soil chromium contamination [21]. Hence, Table 2 summarized the contamination level of heavy metals in soils from different types of contaminated sites. The results showed that heavy metals such as Pb, Cd, As, Zn, Cr, Mn, Co, Hg and Ni were significantly enriched in soils, which were higher than their corresponding risk screening values of soil environmental quality standard in China (GB 15618-2018). To date, due to strict laws and regulations, some factories, companies or enterprises with pollutant emission have been recently closed or relocated, and thus resulted in many abandoned contaminated sites, most of which are difficult to be redeveloped and reused. It is worth noting that industrial contaminated sites not only cause the serious pollution of soil, groundwater and surface water, but also directly endanger public health. In recent years, there have been lots of safety incidents caused by contaminated sites with heavy metals [22-23]. For example, a serious pollution incident occurred in a high school campus in Changzhou, China in September 2015. The school is located opposite a chemical plant. About 500 students were found to suffer from dermatitis, abnormal blood indicators and other diseases that drew widespread concern in society [24-25]. Therefore, soil remediation with heavy metals has been a hot topic at the global environmental engineering community. In the United States, more than 1,000 mercury-contaminated sites have been taken account into the national priority remediation lists [26], and more than 1,300 contaminated sites have been added to the national priorities list by superfund. According to the action plan for the prevention and control of soil pollution of China, the safe utilization rates of contaminated land in China will reach more than 90% by 2020 and 95% by 2030, respectively [27]. In addition, local governments have a strong demand for the remediation and redevelopment of contaminated sites in order to sell land to increase fiscal revenue. Therefore, China is one of the fastest growing markets for soil remediation of contaminated sites worldwide. However, site remediation is very expensive, amounting for several percent of the country's GDP per year. In recent years, it has invested lots of money into soil remediation to promote social and economic development. For example, in May 2015 alone, Chinese government provided 2.8 billion yuan for the prevention and control of soil heavy metal pollution in 30 key areas [28]. In addition, the costs used for management and remediation of contaminated sites were EUR 6.53 billion per year in European countries [29].

Table 2. The pollution levels of heavy metals in contaminated soils.

Smelters	Location	Heavy metal concentrations	Ref.
The mineral smelters distributed on the edge of the main urban areas of Daye	Hubei province, China	Cd (1.14 mg/kg), Co (14.7 mg/kg), Cr (35.5 mg/kg), Cu (176 mg/kg), Mn (672 mg/kg), Ni (28.1 mg/kg), Pb (103 mg/kg), Zn (257 mg/kg)	[71]
Daye Cu smelter	Hubei province, China	Cu (664 mg/kg), Ni (90.6 mg/kg), Pb (4501 mg/kg), Cd (186 mg/kg), As (480 mg/kg)	[72]
Zhuzhou Zn smelter	Hunan province, China	Cu (945 mg/kg), Ni (186 mg/kg), Pb (2523 mg/kg), Cd (557 mg/kg), As (240 mg/kg)	[72]
Smelting-contaminated soils	Hunan province, China	Pb (2052 mg/kg), Mn (652 mg/kg)	[73]
Smelting-contaminated soils	Yunnan province, China	Pb (1118 mg/kg), Mn (687 mg/kg)	[73]
Três Marias smelting plant	Minas Gerais State, Brazil	Cd (612 mg/kg), Cu (1054 mg/kg), Pb (4729 mg/kg), Zn (32827 mg/kg)	[74]
Pb smelter	England, The UK	Pb (1450~106000 mg/kg), Zn (38~4080 mg/kg), Cd (9200~44700 mg/kg)	[75]
Pb smelter	Australia	Pb (1506~7796 mg/kg)	[76]
Lead contamination of the smelting district	Mitrovica, Kosovo	Pb (1.25×10^3 mg/kg)	[77]
a former non-ferrous metal refinery plant	Chungchungnamdo, Korea	As (61.2 mg/kg)	[78]
Mining areas	Location	Heavy metal concentrations	Ref.
Non-ferrous metals mining in Baiyin district	Gansu province, China	As (1180~2070 mg/kg), Cd (55.1~74.7 mg/kg), Cu (4920~7090 mg/kg), Pb (3340~4820 mg/kg), Zn (2450~3480 mg/kg)	[79]
Leiyang mining area	Hunan province, China	Cd (15.0 mg/kg), Cu (97.0 mg/kg)	[80]
A typical Pb/Zn mine	Guizhou Province, China	Pb (1.70×10^3 mg/kg), Zn (913 mg/kg), Mn (2.51×10^3 mg/kg), Cu (343 mg/kg), Cd (2.45 mg/kg), As (127 mg/kg)	[81]
Tongling Shizishan Copper mining area	Anhui Province, China	Cu (219 mg/kg), Zn (673 mg/kg), Pb (324 mg/kg), Cd (6.67 mg/kg), Ni (39.6 mg/kg)	[82]
Non-ferrous metal mining areas	Inner Mongolia, China	Cr (37.2 mg/kg), Ni (29.3 mg/kg), Cu (37.2 mg/kg), Zn (565 mg/kg), As (84.2 mg/kg), Cd (1.30 mg/kg), Pb (56.1 mg/kg)	[83]
Lanping and Huize Pb/Zn mining district	Yunnan Province, China	Pb (1.51×10^3 mg/kg), Zn (1.32×10^3 mg/kg), Cd (19.5 mg/kg)	[84]
Duobaoshan copper mine	Heilongjiang Province, China	As (12.1 mg/kg), Cd (0.110 mg/kg), Cr (73.7 mg/kg), Cu (53.3 mg/kg), Hg (0.0450 mg/kg), Pb (22.2 mg/kg), Zn (74.6 mg/kg), Mo (1.59 mg/kg), Mn (962 mg/kg)	[85]
Tongshan copper mine	Heilongjiang Province, China	As (12.8 mg/kg), Cd (0.130 mg/kg), Cr (62.5 mg/kg), Cu (29.1 mg/kg), Hg (0.0470 mg/kg), Pb (26.9 mg/kg), Zn (90.4 mg/kg), Se (0.169 mg/kg), Mo (1.27 mg/kg), Mn (1.24×10^3 mg/kg)	[85]
A typical Pb/Zn mining area	Jiangxi Province, China	Pb (2028 mg/kg), Zn (3794 mg/kg), Cd (14.8 mg/kg)	[86]
Derbyshire Pb mining area	England, The UK	Pb (4640~61720 mg/kg), Zn (317~3310 mg/kg), Cd (6.4~48.1 mg/kg)	[75]
The abandoned Lasail copper mining area	Oman	Zn (69~896 mg/kg), Cu (1746~8196 mg/kg), As (16~22 mg/kg)	[87]
E-waste contaminated soils	Location	Heavy metal concentrations	Ref.
Qingyuan e-waste recycling areas	Guangdong Province, China	Cr (27.9 mg/kg), Ni (13.1 mg/kg), Cu (33.8 mg/kg), Zn (141 mg/kg), As (20.6 mg/kg), Cd (0.500 mg/kg), Pb (34.4 mg/kg)	[88]

Table 2. Continued.

Guiyu e-waste recycling areas	Guangdong Province, China	Cr (21.0 mg/kg), Ni (8.34 mg/kg), Cu (17.3 mg/kg), Zn (128 mg/kg), As (6.65 mg/kg), Cd (0.130 mg/kg), Pb (34.5 mg/kg)	[88]
Qingyuan electronic waste dismantling site	Guangdong Province, China	As (3.32~54.6 mg/kg), Cd (0.140~115 mg/kg), Cr (20.5~712 mg/kg), Cu (34.3~29730 mg/kg), Hg (0.01~8 mg/kg), Mn (63~2613 mg/kg), Ni (3.24~1169 mg/kg), Pb (34.8~71655 mg/kg), Sb (3.81~2298 mg/kg)	[89]
Wenling electronic waste dismantling and scrap plant	Zhejiang Province, China	Cd (3.8~8.8 mg/kg), Cu (406~1450 mg/kg), Ni (42.4~155 mg/kg), Pb (47.4~252 mg/kg), Zn (68.9~1230 mg/kg)	[90]
Ziya electronic waste plant	Tianjin, China	Cd (0.62 mg/kg), Hg (0.29 mg/kg), As (0.68 mg/kg), Cu (135 mg/kg), Pb (48.5 mg/kg), Cr (112 mg/kg), Zn (222 mg/kg), Ni (50 mg/kg)	[91]
An e-waste dumpsite	Lagos State, Nigeria	Cu (8650 mg/kg), Pb (6800 mg/kg), Zn (1960 mg/kg)	[92]
Informal e-waste recycling hubs	Delhi, India	Pb (1570~2628 mg/kg), Al (81246~9864 mg/kg), Cu (3847~4109 mg/kg), Zn (354~676 mg/kg), Cd (2.41~3.56 mg/kg)	[93]
Informal e-waste recycling sites	Douala, Cameroon	Cr (2.42 mg/kg), Co (4.41 mg/kg), Cd (5.09 mg/kg), As (6.25 mg/kg), Ni (9.45 mg/kg), Hg (11.3 mg/kg), Zn (46.3 mg/kg), Pb (246 mg/kg), Cu (570 mg/kg)	[94]
An e-waste separating household area	Buriram province, Thailand	As (<0.012~1.38 mg/kg), Pb (0.110~15.3 mg/kg), Cd (<0.014~0.284 mg/kg), Cu (1.18~380 mg/kg)	[95]
Bui village e-waste recycling	Vietnam	Pb (460 mg/kg), Cd (8.22 mg/kg), Cr (57.5 mg/kg), Ni (93.6 mg/kg), As (8.70 mg/kg)	[96]
Electroplating contaminated soils	Location	Heavy metal concentrations	Ref.
An electroplating plant	Jiangsu Province, China	As (9.88 mg/kg), Hg (0.590 mg/kg), Cd (0.20 mg/kg), Pb (155 mg/kg), Cr (1324 mg/kg), Cu (615 mg/kg), Ni (324 mg/kg), Zn (436 mg/kg)	[97]
A contaminated site in an electroplating city	Guangdong province, China	Cr (4.7~2580 mg/kg), Cd (0.01~5.11 mg/kg), Pb (7.3~154 mg/kg), Cu (8.4~7240 mg/kg), Zn (8.5~30100 mg/kg), Ni (2.9~2790 mg/kg)	[98]
An electroplating plant	Guangdong province, China	Cu (78 mg/kg), Cr (VI) (61 mg/kg), Ni (146 mg/kg), Zn (38.6 mg/kg), Pb (58.1 mg/kg), Cd (0.14 mg/kg), As (3.57 mg/kg), Hg (0.064 mg/kg)	[99]
An electroplating plant	Beijing, China	Cu (77.6 mg/kg), Cr (15160.8 mg/kg), Cr (VI) (9441.6 mg/kg), Ni (51.6 mg/kg), Zn (1420 mg/kg), Pb (23.3 mg/kg)	[100]
A chrome plating site	Hebei province, China	Cr (948 mg/kg), Cr (VI) (139 mg/kg)	[101]
An abandoned plating site	Jiangsu Province, China	Zn (3849 mg/kg), Cr (9538 mg/kg), Cr (VI) (2175 mg/kg), Pb (68.7 mg/kg)	[102]
Relocation site of a typical electroplating plant	Guangdong province, China	Pb (117 mg/kg), Cd (0.7 mg/kg), Cr (178 mg/kg), Ni (278 mg/kg), As (21.3 mg/kg), Cu (984 mg/kg), Zn (136 mg/kg), Hg (0.1 mg/kg)	[103]
Tannery contaminated soils	Location	Heavy metal concentrations	Ref.
The former tannery campus of Hazaribagh	Zhejiang Province, China	Ni (69.1 mg/kg), Co (41.6 mg/kg), Cr (27200 mg/kg), Cr (VI) (51.5 mg/kg), Ti (3885 mg/kg), Mn (830 mg/kg), Ba (572 mg/kg)	[104]
The former tannery area	Dhaka, Bangladesh	Pb (54 mg/kg), Cr (4321 mg/kg), Zn (287 mg/kg), Cu (131 mg/kg), Ni (9 mg/kg)	[105]
Illegal disposal of tannery waste site	Campania, Italy	Cr (151~1705 mg/kg), Zn (166~609 mg/kg)	[106]
A tannery soil	India	Cr (28400 mg/kg), Pb (49.6 mg/kg), Cd (24.3 mg/kg), Ni (44.6 mg/kg)	[107]
The largest inhabiting tanneries area	Punjab Province, Pakistan	Cr (779 mg/kg)	[108]
Kombolcha tannery surrounding soil	Ethiopia	Cr (22.6 mg/kg), Cr (VI) (0.017 mg/kg)	[109]

Table 2. Continued.

Tannery Wastewater Treatment Plant	Modjo City, Ethiopia	Cr (III) (1811 mg/kg), Water-soluble Cr (VI) (0.183 mg/kg), Cr (VI) (0.621 mg/kg)	[110]
Tannery Joint Stock Company (ETSC) effluent discharge downstream contaminated area	Ethiopia	Cr (16.2-1582 mg/kg), Cr (VI) (1.17-2.23 mg/kg)	[111]

Soil Washing/Flushing

The publication number of papers on soil washing/flushing is shown in Fig. 3. From the Web of Science database, it has been found that the publication number on soil washing/flushing increased rapidly from 1975 to 2022, especially from 2010 to 2022, with a cumulative total of 526 publications. Moreover, China has the highest publications number on soil washing/flushing with 357 papers, which plays an important role in this domain, followed by the United States, Korea and Italy. As shown in Fig. 3, soil washing involves soil excavation, screening of oversized soil particles, and then mixing with the washing solution to desorb heavy metal contaminants from soil particles. In some cases, the eluent forms soluble complexes with the contaminants, causing the minerals containing the contaminants to dissolve or facilitating the desorption of the contaminants through competitive adsorption. Redox reactions may also occur during the washing process, causing changes in the valence states of metal ions, thereby affecting their bioavailability and toxicity [30]. In addition, multi-stage washing is adopted until the concentration of heavy metal contaminants is within the acceptable concentration range. In the final step, the waste solution needs to be reasonably treated and then recovered for their reuse in soil washing.

In the past years, many chemical agents, including inorganic salts, dilute acids and bases, organic acids, surfactants, and chelating agents, have been used to remove heavy metals from contaminated soils. Soil flushing generally is used in connection with in situ remediation. Typical soil flushing solutions include water and other solutions. By surface flooding or subface injection at contaminated sites, the contaminants are displaced or transferred to the flushing solution [31]. The remediation effects of soil washing/flushing are mainly influenced by soil properties (hydraulic permeability, particle size distribution and organic matter content, etc.), the occurrence states of heavy metal pollutants, the types of washing agent, as well as washing conditions. Among these factors, different washing agents have their distinct characteristics and remediation mechanisms. Therefore, how to choose an appropriate washing agent is a the most key factor influencing the performance of soil washing.

Common washing agents include surfactants, inorganic agents (acids, bases, salts), chelating agents (artificial chelating agents, natural chelating agents), etc. Compared with the washing agents that do damage to soil structure, and are also toxic to plants and animals, low molecular weight organic acids (LMWOAs) are classified as biodegradable chelating agents, such as citric acid, tartaric acid, oxalic acid,

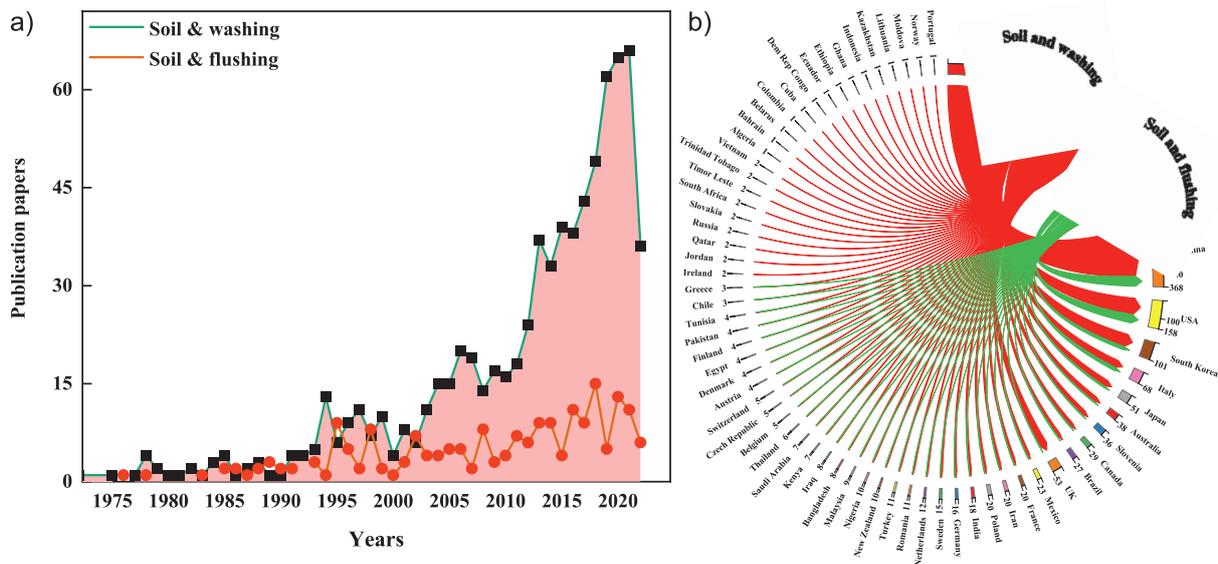


Fig. 3. a) Published literature statistics related to soil washing/flushing research; b) Published literature statistics related to soil washing/flushing research in different countries worldwide (source: web of Science; Key words: Soil, Washing and Flushing; Retrieved: September 4, 2022).

malic acid, acetic acid, propanedioic acid, fulvic acid, humic acid, hamaleic acid, lactic acid, methanesulfonic acid, salicylic acid, aspartic acid, ascorbic acid, etc. LMWOAs are able to be extracted from industrial and agricultural solid waste with additional economy benefits [32]. As a green and efficient soil remediation agent, LMWOAs play a vital role in reducing the absorption of heavy metals by plants, and can also mitigate the pollution risks from heavy metals entering the biological chain. Therefore, LMWOAs have been widely utilized in soil washing [33].

Different Types of Low Molecular Organic Acids and Their Application Effects

LMWOAs can be classified into the types of carboxyl, ammonium, sulfhydryl, according to the functional groups. It is proved that the LMWOAs with carboxylic groups can not only promote metal ion desorption by providing protons and increasing the net positive charge of soil mineral surface, but also enhance the mobility of heavy metal ions through solubilization, complexation, redox and competition for adsorption sites [34]. Besides that, LMWOAs with ammonium groups are able to form complexes with almost all metal ions [35]. Hoffmann et al. [36] demonstrates that sulfhydryl group is an important thiol intermediate in organic sulfur metabolism in soils, and that the functional group enables heavy metals with high

valence to be reduced, controlling the migration and transformation of heavy metals in soils. For example, the influence of the different functional groups on the release of As followed the order of sulfhydryl > ammonium > carboxyl, according to a previous study by Xu et al. [37]. This may be related to the fact that sulfhydryl groups are more reductive for As in the forms of oxides/iron oxides in soils. Furthermore, the number of functional groups also has a profound effect on soil washing. Generally, the removal efficiency of heavy metals from soils follows the decreasing orders of ternary carboxylic acid > dicarboxylic acid > monocarboxylic acid. The more functional groups and the larger molecular weight of the LMWOAs have, the more charge they may carry, and the stronger competitive adsorption and complexation they also show [38]. It was reported that the desorption process is a heterogeneous diffusion–reaction associated with the size of reaction interface [39]. Therefore, the number of functional groups from LMWOAs may also influence the release kinetics of metal desorption. The recent case studies of LMWOAs on heavy metal removal from soils are shown in Table 3.

The Main Factors Affecting the Washing Effects of LMWOAs

The concentrations of washing agents: When using the same types of LMWOAs, the removal rates of

Table 3. Different types of low molecular weight organic acids used for heavy metal removal.

LMWOAs	Soil types	Heavy metals	Washing condition	Results	Ref.
Citric acid	Contaminated soil from Pb/Zn mine	Cd, Pb and Zn	pH = 4.0, solid-liquid ratio of 1:10, and a washing time of 120 min, citric acid (0.2 M)	The removal efficiency of heavy metals in two soils were about 30–40%	[112]
	Suzhou National Hi-Tech District (SND)	Zn and Cu	pH = 6.58, solid-liquid ratio of 1:8, reaction less than 2 hours, citric acid (0.8 M)	The removal rate of Zn (II) and Cu (II) were more than 90%	[113]
	Soil on impact berm at military firing range	Pb	pH 5.65, at 3% soil slurry density, in a single batch for 4 h, citric acid (0.5 M)	The removal efficiency of Pb achieved 90.3%	[114]
	Agricultural soil near an old recycling lead smelter in Vietnam	Pb, Cu and Zn	pH = 3.5, solid-liquid ratio of 1:20, reaction 24 hours, citric acid (0.2 M)	The removal efficiencies of Pb and Zn were about 60%	[115]
	Agricultural soil in Dabaoshan District, Shaoguan City, Guangdong Province	Cu and Pb	pH = 4.51, solid-liquid ratio of 1:20, and contact time 360 min, 4:1 volume ratio (PFA: CIT = 4:1) of potassium fulvate (PFA, 3.2%) and citric acid (CIT, 0.16 M)	The removal efficiencies of Cu and Pb were 42.3% and 50.5%, respectively	[116]
Methanesulfonic acid	Devon Great Consols (DGC) Cu/As tailings	As, Cu	pH = 3.33, solid-liquid ratio of 1:10, shaking for 24 hours, Methanesulfonic acid (1 M)	The removal rates of As and Cu were 56% and 29%, respectively	[117]
Oxalic acid	Qingdao Hongxing Chemical Plant Chrome-containing Slag Dump	Cr	pH >7.7, solid-liquid ratio of 1:4, Oxalic acid (0.5M)	The most cumulative leaching amounts of Cr was 2304 mg/kg	[118]

Table 3. Continued.

LMWOAs	Soil types	Heavy metals	Washing condition	Results	Ref.
	Abandoned sewage treatment station soil from a relocated electroplating plant	Cu, Ni and Zn	pH = 3.0, soil-liquid ratio of 1:10, and shaking at room temperature for 6 h, oxalic acid (0.2 M)	Ni showed the highest removal of 70.4%, while Cu displayed the lowest removal of 51.1%	[119]
Tartaric acid	Topsoil from Zn mine	Zn	pH = 4.46, a soil-solution ratio of 1:10 and the drenching time of 120 min, tartaric acid (0.2 M)	The removal efficiency of Zn reached 89.4 %	[120]
Acetic acid	Farmland near a metal smelter	Cu, Zn	pH = 2.3, a soil-solution ratio of 1:50 and shaken for 24 h, acetic acid (0.1 M)	The dissolved fraction of Cd, Co, and Zn ranged from 60 to 90%, while Cu was only dissolved between 30 and 60%.	[121]
Lactic acid	Illegal disposal of soil from tannery sludge site	Cr (III)	pH = 7, solid to liquid ratio 1:10 and a washing time >30min, lactic acid (1.0 M)	About 70% of Cr(III) was removed	[122]
Aspartic Acid	Soil from a tailings dumping area	As	pH = 6.13, a solid-liquid ratio of 1:10 for 4 h, aspartic Acid (0.01 M)	94.0 mg/kg of As was leached from the soils	[123]
3-Mercaptopropionic acid	Soil from a tailings dumping area	As	pH = 11, a solid-liquid ratio of 1:10 for 48 h, 3-Mercaptopropionic acid (0.01 M)	247.3 mg/kg of As was leached from the soils	[123]
Ascorbic acid	Agricultural soil near an old recycling Pb smelter	Pb, Cu and Zn	pH = 3.5, a solid-liquid ratio 1:20 and reaction for 24 h, Ascorbic acid (0.2 M)	The removal efficiency of Pb and Zn were 14.2% and 21.8%, respectively, while the that of Cu was very low	[115]
Malic acid	Soil from an abandoned chromium salt chemical plant	Cr (VI)	pH = 4, a solid-liquid ratio of 1:10, and the washing time of 6 h	The removal rate of Cr (VI) was 79.3%	[124]

heavy metals increase at the initial stage followed by a slow decrease. This phenomenon may be related to the adsorption and desorption equilibrium between washing agents and heavy metals. It was demonstrated that the Cr released by citric acid increased from 107 to 147 mg/kg, while Cr released by malic acid ranged from 101 to 126 mg/kg, when the organic acid concentrations in the desorption solution increased from 5 to 20 mmol/L [38]. Xiao et al. observed a significant increase in the release of metal ion with the addition of LMWOAs in the range of 10 to 60 mmol/L [40]. It is proposed that the amount of washing agents contacting with per unit mass of contaminated soils increased in the initial stage, which provides more binding sites and promotes the formation of metal complexes [40]. In the later stage, although the concentration of washing agents is higher, heavy metals are predominantly adsorbed specifically in soils and less mobile, and thus their desorption rates are slower and less likely to be desorbed. For example, it was shown that the washing efficiency of As decreased, when oxalic acid concentrations were greater than 80 mg/L, which may be due to the formation of precipitates by oxalic acid at the higher concentration [41].

The pH of washing agents: The solubility of heavy metal contaminants is influenced by pH (Fig. 5). Significant high and low pH of washing solution tend to promote the mobility of heavy metals in soils. The lower solution pH is, more protons (H^+) it will contain. Low solution pH also means more competitive adsorption with heavy metal ions, which promotes the desorption of metal ions from soil particles. Moreover, low washing solution pH also enhances the solubility of metal hosting minerals in soils, resulting in the release of heavy metals from mineral lattices and increasing the mobility of heavy metal pollutants [42]. Higher solution pH can lead to metal sulfides being easily oxidized in the presence of dissolved oxygen, and thus heavy metals are more easily released from soils. When solution pH increases, the functional groups of LMWOAs carry more negative charges and obtain the stronger ability to chelate heavy metals, thus promoting the migration of metal ions from soils [43]. However, it should be considered that with the increase in solution pH, the amount of OH^- increases, which can promote precipitating with metal ions. In addition, metal ions are more easily adsorbed on clay minerals at high solution pH, which reduces their mobility in soils [44].

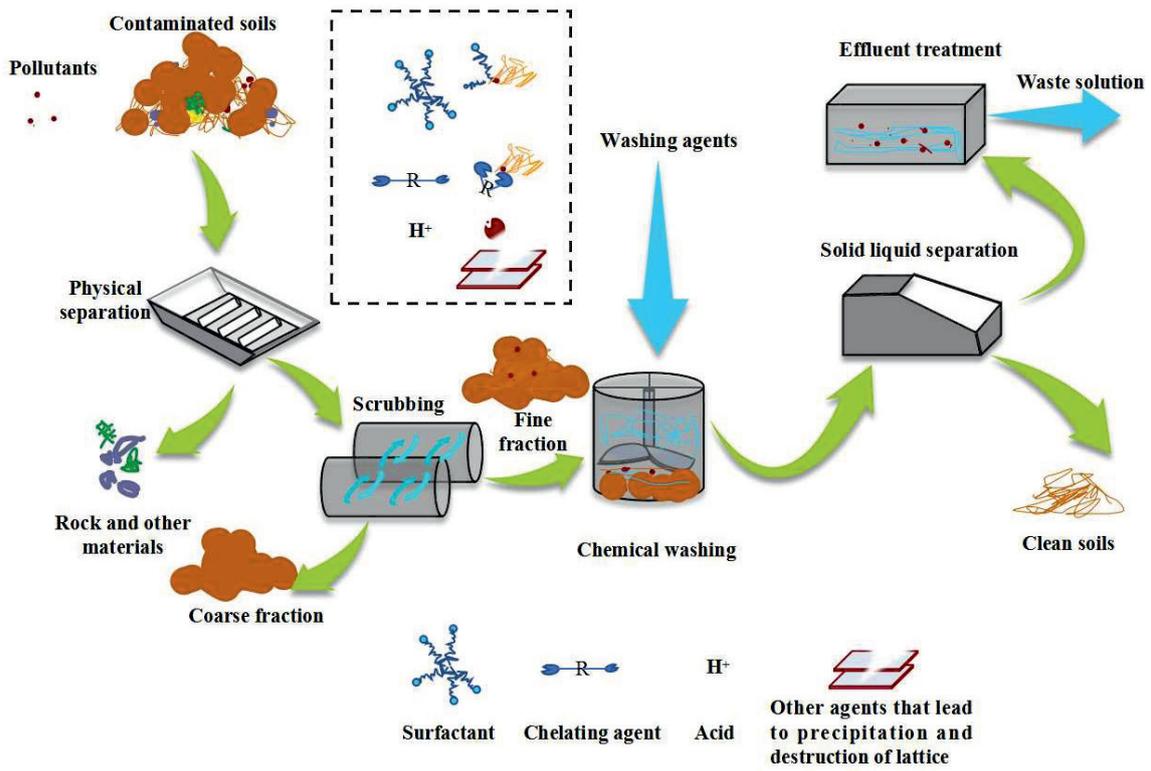


Fig. 4. Schematic diagram of soil washing process and principal mechanism.

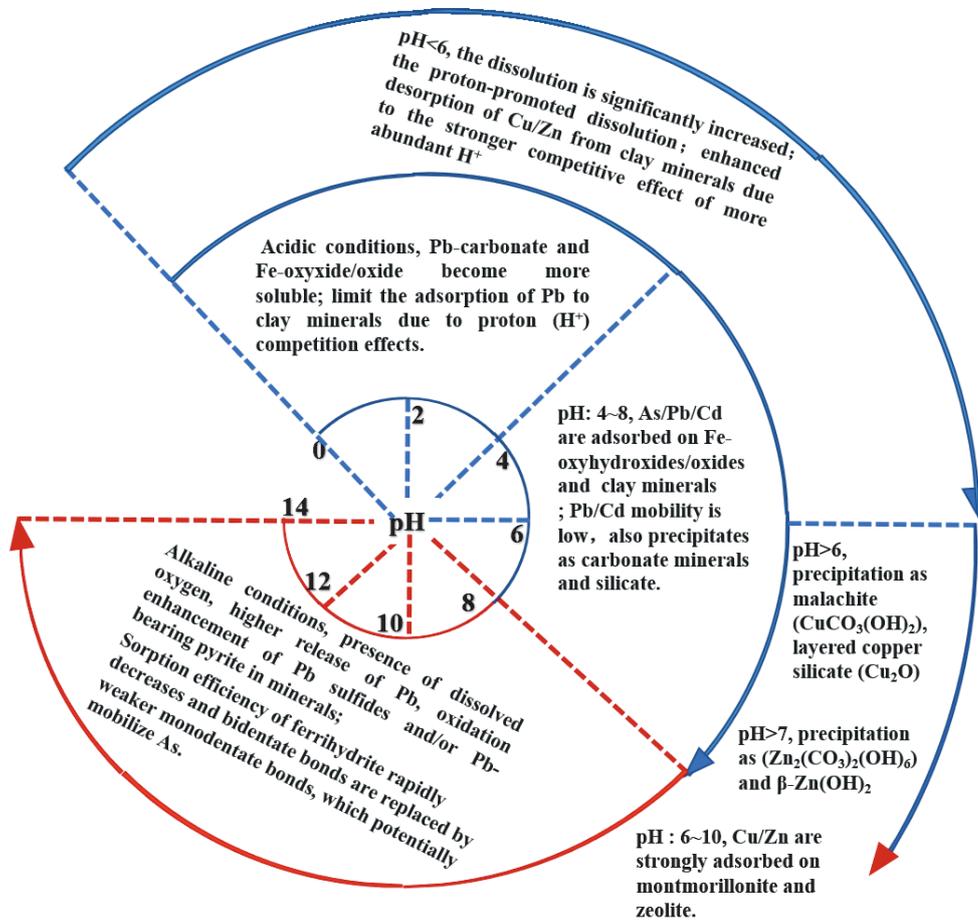


Fig. 5. Effect of pH on leaching, formation and migration of heavy metals (Compiled from literature [44]).

For example, it was found that when the pH value of EDTA–tartaric acid system increased from 3.0 to 7.0, the removal efficiency of Zn decreased from 82.1% to 69.5%, since appropriate pH value was conducive to the dissolution and further complexation removal of heavy metals, especially at low pH values [38]. Similarly, it was shown that the extraction of As, Pb, Fe and Mn increased with decreasing solution pH, reaching 70% at pH = 0.63 and only 1.6% at pH = 9.8 [45]. Therefore, it is necessary to master the leaching characteristics of different heavy metals under different pH conditions to determine the optimal pH of washing solution.

Solid-liquid ratio: The solid-liquid ratio has a significant effect on the contact areas and mass transfer process between the washing agents and heavy metals. The smaller the solid/liquid ratio is, the shorter the mass transfer time is and meanwhile the larger the desorption amounts of metal ions will be. The general solid–liquid ratio is between 1:3 and 1:20. When the fine soil fractions are at high percentage, it is suggested that lower solid–liquid ratio could promote the mixing of soils and washing agents and increase washing efficiency. The higher solid–liquid ratio is not conducive to agitation, while the lower ratio increases the consumption of washing agents, and the treatment costs of wastewater [46]. For example, Cheng et al. [47] found that the removal efficiency of Ni increased from 66.1% to 82.1%, when the solid-liquid ratio of citric acid-EDTA agent ranged from 1:5 to 1:10. However, when the solid-liquid ratio decreased from 1:10 to 1:20, the removal efficiency of Ni did not change significantly [47]. Similarly, Tan et al. [48] reported that the optimal solid–liquid ratio was 1:10, and the removal rate of Cr(VI) by citric acid was 73.5%, taking the treatment cost and washing effect into account [48]. Generally, the optimal solid-liquid ratio is set according to the results of feasibility experiments and pilot scale tests to achieve the balance between cost and efficiency.

Washing time: The effects of washing time is also related to the release kinetics of metal ion from soils. At the initial stage, the weakly bound metals in soils desorbed quickly, the acid solubilization of H⁺ increases the release of non-targeted elements such as Ca, Fe, Mg, K and Na with time, which consumes some of the reactive sites of the chelator and decrease the chances of them contacting with heavy metals. Meanwhile, chelating agents will be gradually adsorbed on soil particles, thus reducing their removal efficiency [49]. For example, Qin et al [50] found that when acetic, malic or citric acid was used for soil washing, the desorption amount of Cu, Cd and Pb from soils for 10 weeks were only 51.7%-87.6% of that in the first two days. Similarly, it was shown that the removal rates of Cu, Ni and Zn by EDTA-tartaric acid increased, as reaction time increased from 0 to 360 min, and their removal rates then tended to be stable, due to the slow release of strongly bound metal ions in soil particle [47]. The washing time of inorganic reagents is shorter than that of organic reagents. The washing time usually

ranges from 20 to 120 min. It has been shown that the extension in the washing time is beneficial to the removal of heavy metal pollutants, but will increase the treatment cost. Consequently, the pilot and field scales experiments need to be conducted determine the optimal washing time.

The occurrence forms of metal contaminants: Chemical extraction methods and instrumental characterization are often used for the studies on the geochemical behavior of metal contaminants. For instance, heavy metals in soils can be divided into four chemical fractions, according to the optimized BCR extraction method: acid extractable, exchangeable fraction, easily reducible, oxidizable and residual fractions. Among them, acid extractable and reducible fraction are considered to be unstable, because of their higher bioavailability and detriment to the environment. Previous studies have indicated that different chemical extraction methods differ in the classification of heavy metal fractions. Moreover, there are some problems in determining the chemical partitioning of heavy metals in soils. A typical example is that BCR methods can to a certain extent show errors in the exchangeable metal fraction. Meanwhile, BCR methods are not suitable to determine the fractionation classification of heavy metals in calcareous soils [51]. Thus, the uncertainty of chemical extraction methods will interfere with the determination of the actual chemical metal fractions. Fortunately, with the development of spectroscopic techniques, it has been possible to physically analyze the occurrence forms of heavy metals in soils from more microscopic perspective. For example, X-ray diffraction (XRD) can identify the main mineralogical composition in soil samples. However, XRD is not applicable for the identification mineral phase with poor crystallization in soils. As a new generation technique, X-ray absorption fine structure (XAFS) and X-ray absorption spectroscopy (XAS) can be used to study both crystalline and non-crystalline mineral phase [52]. Moreover, X-ray photoelectron spectroscopy (XPS) can also be used to characterize the changes in the valences and proportions of metal species in soils [53]. Additionally, Fourier transform infrared spectroscopy (FTIR) method is useful to compare soil mineralogy in specific areas [54]. However, there have been overlapped peaks in conventional FTIR. Two-dimensional correlation spectroscopy can resolve the overlapped peak by extending spectra along the second dimension and provide information about the relative directions and sequential orders of structural variations. Therefore, two-dimensional FTIR technique can be applied for further investigation of the interactions between heavy metals and the main components of dissolved organic materials [55].

In general, the unstable metal fractions are higher in smelter contaminated sites than that in the mine sites. In addition, there are significant differences in the geochemical fractions of different heavy metals in soils from different types of contaminated sites, which

are mainly related to their hosting mineral phases in soils. For example, fayalite, lead oxide, apatite, galena and wollastonite were identified as the dominant As, Cd, Pb and Zn bearing minerals in a typical Pb/Zn smelter contaminated soils [56]. Additionally, As is closely related to Fe (hydro) oxides and Fe bearing oxides, such as scorodite ($\text{Fe}(\text{AsO}_4)_2\text{H}_2\text{O}$, containing 34.6% Fe oxides and 49.8% arsenic pentoxide), and the non-specific/specific adsorbed As were present in goethite [57].

The desorption capacity of heavy metals by washing agents is closely related to their occurrence states in soils. A previous study by Yang et al. [58] showed that when 87% of Cd in soils was mainly present in the exchangeable and reducible fractions, citric acid reduced the exchangeable and reducible fractions of Cd by 78% and 55%, respectively. Similarly, Adamo et al. [59] found that the removal efficiency of Zn was lower than that of Cd, which was mainly due to the fact that Cd was mainly presented in the extractable fraction, while Zn mainly existed in the residual fraction. In addition, Xu et al. [60] showed that the existence of LMWOAs can transform residual As and Fe oxides bound As into adsorbable As in soils, i.e., the conversion of heavy metals from the stable fractions to the unstable fractions leads to their higher mobility.

Conclusions

Various anthropogenic activities not only have caused lots of abandoned contaminated sites, but also caused serious heavy metal pollution in soils, which poses the potential threat to the residents nearby. Among the remediation techniques, soil washing has been proved to be one of the best remediation techniques for heavy metals in contaminated soils. The selection of green, cheap and efficient chemical agents is an important factor to be considered in their application in soil washing. There may be some possible limitations in this study, and it is recommended to focus on the following aspects:

(i) Due to the complexity of the site characteristics and pollutant properties, as well as the treatment efficiency of single soil remediation technology, it is time to make full use of their respective advantages to combine different technologies and maximize the remediation efficiency. For example, more attention has been paid to the development of soil washing enhancement technologies, such as ultrasonic, microwave, photocatalytic and other physical enhancement, chemical redox enhancement and electrochemical enhancement.

(ii) From the aspect of managerial insights, the modern analytical instrumentation with high precision should be used to determine the hosting mineral phases of the major heavy metal pollutants in soils. Meanwhile, it is useful for soil washing to identify their occurrence forms by a suitable chemical sequential extraction

method. The effects of washing agents on the structure of different types of minerals need to be further studies.

(iii) The appropriate chemical agents and conditions of soil washing are reasonably chosen to reduce the amounts of waste solution. Meanwhile, studies on the washing process that can recycle the washing agents need to be carried out to reduce the remediation costs.

(iv) LMWOAs monomers generally show the lower remediation effects than artificial chelating agents (e. g. EDTA), which limit their wide application in practical remediation scenarios. It is suggested that LMWOAs polymers contain more functional groups that can react with heavy metal ions, which have wide application prospects. For example, polyacrylic acid, itaconic acid–acrylic acid copolymer, caboxyethylthiosuccinic acid, maleic acid–acrylic acid copolymer and epoxy polysuccinic acid have been proved to be able to significantly enhance the removal efficiency of heavy metals from soils.

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Conflict of Interest

The authors declare no conflict of interest.

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