

Soil Washing Methods for Effective Removal of Heavy Metal Contaminants

Jian Chong Chiu, Paran Gani*

Department of Civil and Construction Engineering, Faculty of Engineering and Science, Curtin University Malaysia

*Correspondence[: paran.gani@curtin.edu.my](mailto:paran.gani@curtin.edu.my)

SUBMITTED: 1 May 2024; REVISED: 14 June 2024; ACCEPTED: 16 June 2024

ABSTRACT: Soil pollution caused by heavy metals from anthropogenic activities poses a significant environmental and health threat globally. Traditional remediation methods like solidification/stabilization have limitations, prompting the need for alternative techniques. Soil washing emerges as a promising approach, employing physical and chemical methods to effectively remove contaminants. This paper explores soil washing methods, focusing on sites contaminated with heavy metals such as zinc, lead, nickel, mercury, arsenic, copper, chromium, and cadmium, particularly influenced by military and industrial activities. Several techniques, including physical separation and chemical extraction, are discussed, which consider a few factors such as magnetism, density, size, and hydrophobicity to concentrate metal contaminants and solubilize soils. Physical separation targets particulate contaminants, while chemical extraction addresses non-detrital metals or soils with adsorbed ionic forms. The study also analyses field applications of soil washing systems and the implementation of remediation techniques. It emphasizes the need for innovative soil remediation strategies to mitigate the adverse effects of heavy metal contamination on soil quality and human health.

KEYWORDS: Soil washing; heavy metals; remediation; physical separation; chemical extraction

1. Introduction

In most countries, pollution of heavy metals in soil has been a serious concern. Over the years, anthropogenic activities such as mining, smelting, and wastewater drainage have caused contamination of soils [1]. Heavy metals like zinc (Zn), Lead (Pb), and cadmium (Cd) have low degradability and are persistent in soil [2]. Furthermore, the presence of cadmium and lead can significantly aggravate the quality of the soil or environment as well as pose threats to human health whereas extravagant zinc dosages are toxic to plants [3]. Hence, remediation techniques are needed to control soil contamination by heavy metals.

Traditional remediation techniques for heavy metal contaminated soils involve the removal of polluted soils, accompanied by immobilizing the contaminants with solidification/stabilization (S/S) technology before disposing of treated materials on-site or authorized landfill [4]. However, S/S remedial technology is no longer deemed a viable response to persistent environmental solutions due to several reasons. Heavy metals are not effectively separated from polluted media and require subsequent on-site heavy metals monitoring. The durability of the solidified/stabilized materials is uncertain. Lastly, long-term

landfill management of S/S materials requires soil caps to suppress soil erosion issues. Therefore, research for soil remediation technologies to treat heavy metal contaminated soils is much needed. Soil washing is one of the long-term remediation techniques to remove heavy metals from contaminated soils using either physical or chemical methods.

In this paper, soil washing methods will be discussed based on sites contaminated with zinc, lead, nickel, mercury, arsenic, copper, chromium, and cadmium. The topic mainly focuses on soil matric remediation that is heavily influenced by military and industrial activities such as the shooting range site and recycling site. There is no discussion of soil washing methods relating to radioactive metals or organic contaminants. Methods such as physical separation by mineral procedure technology, dissolving or leaching chemical extraction, and a combination of chemical extraction and physical separation are discussed. Analysis of metal-contaminated soil treatment involving the field applications of soil washing systems will be discussed as well as the implementation of soil washing remediation techniques to the metal-contaminated soil treatments.

Soil washing is a technique used to remove metal contaminants from soil using physical and/or chemical processes knowingly physical separation and chemical extraction. By exploiting variations in specific physical characteristics between metal-bearing particles and soil particles, physical separation concentrates metal contaminants into smaller soil volumes. Magnetism, density, size, and hydrophobic surface properties are considered in the specific physical characteristics during the physical separation process. Chemical extraction is a technique attempting to solubilize soils with metal contaminants using an aqueous extraction fluid containing chemical reagents such as chelating agents or acids [5].

Currently, physical separation is viewed mainly as a process of particle separation, whereas chemical extraction can be regarded as a process of metal desorption/solubilization. Physical separation generally applies especially as the metal contaminants are in the form of particulates whereas chemical extraction is suitable mainly for non-detrital metals or soil with adsorbed ionic forms.

2. Physical Separation Methods

For physical separation, the general approach to remove the desired metal-bearing particles is to use techniques commonly used in mineral and mining processing industries from mineral ores. Mineral processing methods have a simple implementation, low-cost operation, and are well established with the process and equipment required [6]. Mineral extraction techniques are investigated in general to separate metal pollutants from soils [7]. Major technology classes are summarized according to the standards of separation used. Operation units such as froth flotation, magnetic separation, and others are discussed in the physical separation methods with three aspects to focus on. The first aspect is the applicability of engineering for physical separation by sources of metal contamination and soil matrix characteristics. Followed by the technological class reviewing and physical separation integrated processes.

2.1. Physical separation applicability and limitations.

Techniques of physical separation apply primarily to metal-bearing particles or discrete particles in the forms of particulate for metals. Physical separation is typically not suitable for the treatment of sobbed metal forms, although attrition scrubbing can greatly improve metal desorption during chemical leaching. To speculate the applicability of physical particle

separation methods, understanding the degree of liberation of the mineralogical phase involving heavy metals is crucial [8]. The degree of liberation is dependent on the mineralogical characteristics of particles of metal contaminant such as the morphology, shape, and mineralogical association. Degree of liberation refers to the releasing of the metal phase relative to the different associations with the soil particles or carrying phase. The metal phase refers to the mineral shape of the metal whereas the carrying phase refers to other mineral phases which can be associated with the metal phase. Table 1 below illustrates the variety of potential metal phase states with the inclusion of volume, association, and surface that is weakly bounded and liberated or free.

	Metal Phase Description		
Metal phase included in the volume	Carrying phase	Metal phase ٠	Low degree of liberation Density is influenced by the carrying phase minerals Surface properties are consistent but rely on the carrying phase Physical separation is extremely challenging or unfeasible Crushing is necessary
Metal phase associated	Carrying phase	Metal phase	Medium degree of liberation Density is influenced by the carrying phase and minerals of the metal phase Surface properties are not consistent Physical separation can be feasible
Metal phase weekly bounded on surface	Carrying phase	Metal phase	Medium degree of liberation Physical separation can be feasible if metal phase particles are liberated
Metal phase liberated or free		Metal phase	High degree of liberation Density is influenced by the minerals of the metal phase Surface properties are consistent Physical separation is feasible

Table 1. Applicability of physical separation of particulate forms according to the liberation degree of the metal phase [5,9].

The degree of liberation and applicability of particle separation is discussed briefly for each metal stage by froth flotation and gravity concentration. Microscopy and spectroscopy techniques like scanning microscopic electrons coupling with analysis of energy-dispersive Xray can examine the metal-bearing particle mineralogical aspects and solid phase speciation. Physical separation efficiency is dependent on a few characteristics of soil such as the shape of the particulate, particle size distribution, content of clay and humic, magnetic properties, density differences between metal contaminants and soil matrix, soil matrix heterogeneity, and particle surface hydrophobic properties [10]. In these following cases, the treatment is found to be inconceivable. Soil particles that are vigorously bound with metal contaminants. No

significant difference is found in surface properties or density between metal-bearing and soil matrix. High variety of metal chemical forms. Presence of metals in all contaminated soil particle size fractions. Silt or clay content that is 30 to 50 percent excess in soil. Soil that contains high viscosity organic compounds and high humic content. Particles of feed content are one of the most critical variables influencing the applicability of physical separation technologies, as polluted soils usually contain a broad range of particle sizes, and technological performances are often limited to a narrow range of particle sizes. Most gravity concentrators and hydroclassifiers usually have good sand fraction applicability. Gravity concentrators such as spirals, shaking tables, and jigs are typically not appropriate for fine particles [5]. Based on the equipment, there is a limiting factor in the amount of fine particles. The content of the soil with sand in 50 to 70 percent excess is more cost effective and primarily appropriate with physical separation [11]. A method that incorporates attrition scrubbing and wet screening or hydrocyclones can be used to remediate sediments in fine-grained matrices [12]. Froth flotation can also be effective in the treatment of relatively fine particles. The preferences of technologies for physical separation strongly depend on the types of soil and site to be treated. These methods are mainly applicable in industrial or urban areas to "anthropogenic" soils. Human activities such as disposal, landfills, and industrial artifacts heavily influence the soils and usually consist of toxic waste mixtures and anthropogenic or natural landfills. Physical separation methods, on the other hand, are not adequate for the treatment of agricultural or natural soils affected by diffuse contamination due to several reasons. Comparatively low concentration level of metals and mostly occur in sorbed forms. Typically, these soils are high in clay or silt and organic matter. Because most of the metals present in soils are in sorbed forms as opposed to discrete particles, physical separation is frequently correlated with chemical procedures to improve removal efficiency of metal.

2.2. Hydrodynamic classification.

Hydrodynamic classification requires particle separation based on the velocity of particles falling through the water flow with fluidisation, elutriation, and sedimentation or by centrifugal separation or separation into the water flow by hydroclone [7]. In the soil washing process, hydrocyclones were vastly used to separate the fine soil from bigger sand particles. The centrifugal force is stronger than the force due to gravity, thus significantly reducing the operating time for separation [13]. Compared to other classification machinery, hydrocyclones have low capital and operating costs. In the context of soil remediation, screw classifiers and elutriation-based hydroclassifiers can also be introduced [11]. In addition, more effective technologies have recently been developed for the mineral processing industry, such as the CrossFlow classifier and the HydroFloat separator [14].

2.3. Gravity concentration.

Gravity concentration method exploits the difference in particle gravity in the slurry to separate the metal-bearing particles from the soil matrix. Settling is caused by the particle size weight, shape, and density, but the key factor of settling is due to its density. Gravity separation is ineffective when it is used for treating particles that have a wide range of size distribution or a small distribution of density [10]. Gosselin [13] has stated that for sufficient separation, the difference in density must be larger than the value of $1g/cm³$. Taggart "concentration criterion" can be used to estimate the effectiveness of density separation [7]. During the separation process, gravity concentrators like spirals, shaking tables, and jigs can be affected by the effect of particle size. For this purpose, before performing gravity concentration, it is important to identify the classification of the soil size. If the difference in density between the particles of contaminant and soil is consequential, the technologies of gravity concentration should be distinctly appropriate for the separation of particles. For dense media separation, the spiral, shaking table, mineral jig, and MGS-Mozley the gravity concentrator volume capacities correspond to 25, 4, 100-500, and 5 t/h [13]. Conditions of the particle size range are stated in Figure 1 below.

Figure 1. Range of feed particle size range to apply physical separation methods [5].

Commonly used gravity concentrators for large-scale soil treatment are spirals, shaking tables, and jigs. Mineral jigs are usually used for treating coarse sand fractions between 800 to 2000 m or gravel fractions from 2000 to 6000 m whereas spirals and shaking tables are better suited to treat fine coarse or medium sand from 63 to 2000 µm. Fine sand fractions between 63 to 125 m and silt or clay that is below 63 m are to be processed with MGS-Mozley. Following long-term test results are reported by Bergeron [15] in Montreal, Canada for the brownfield soil remediation project. Two jigs in series were able to remove copper by 75 percent within the 1700 to 6400 µm fraction. Fraction of 106 to 1700 µm containing 1025 µg/g of copper was able to achieve efficiency of removal up to 54 percent using the spiral method. MGS-Mozley has a removal efficiency of 47 percent removing the copper in the initial soil of 924 μ m/g below the soil fraction of 106 µm. Due to the dense property of lead, gravity concentration can be used to remedy soils contaminated with lead in particulate forms. Using jigs for density

separation is a prominent technique for removing lead from the firing ranges of small arms where lead is substantially present in spent bullets [5].

2.4. Froth flotation.

Froth flotation is a physicochemical method that takes advantage of the difference in hydrophobic properties of the soil matrix metal-bearing particles [5]. The principle of separation is based on the affinity of the hydrophobic surfaces of a particle for the injection of air bubbles into the soil flurry. Three steps are involved in this separation process. The desired metal-bearing particles are attached to the air bubbles. Gathering of bubbles in foam portion and removal of floats up slurry in the foam fraction. By using a surfactant, the surface of the metal-bearing particles is frequently rendered as hydrophobic. In the mineral industry, froth flotation is commonly used and metal sulphides can be isolated more easily than oxides and carbonates [16]. There are various types of flotation structures, including the flotation column as well as the flotation cells. Metals such as cadmium, copper, lead, and zinc present in soils and sediments have been successfully removed by using the froth flotation method [17]. Using froth flotation as a remediation technique, however, is still less used than other techniques for soil washing. Froth flotation seems to be a pertinent technique for the treatment of matrices with fine grain, in particular, the anoxic dredged sediments with heavy metals mainly in the form of sulfides [18].

2.5. Magnetic and Electrostatic separation.

Particles in the soil are magnetically susceptible, ranging from negative, intermediate to larger positive ferromagnetic minerals [19]. A low intensity magnetic field can attract ferromagnetic material, while a high intensity magnetic field is required to separate paramagnetic material [8]. Debris from spent munitions at military sites and metallic or ferrous waste products with high concentrations of heavy metals in brownfields are recovered by using low intensity magnetism separation [20]. Heavy metals of soil matrix by magnetic separation are based on the interaction between the ferromagnetic materials and metal contaminants. Rikers [19] demonstrated that high-intensity wet magnetism separation is ideal for the extraction of chromium, copper, nickel, lead, and zinc from multiple soils. When there is no correlation of heavy metals between the ferromagnetic phase, the separation process will not be effective.

The heterogeneity of soil matrices in brownfields can significantly impact the efficiency of magnetic separation. Brownfield soils often contain a mix of particle sizes and types, ranging from fine clays to coarse sands and gravels, which affects separation efficiency [21]. Fine particles, such as clays and silts, may reduce effectiveness due to their lower magnetic susceptibility and tendency to form aggregates, shielding magnetic minerals from the magnetic field [22]. The presence of significant amounts of non-magnetic materials like organic matter, glass, and plastics can also hinder the process by occupying space in the separation equipment, thereby limiting the interaction between the magnetic field and the magnetic particles. Furthermore, different soil components have varying magnetic susceptibilities, where iron-rich particles are highly magnetic and easily separable, but non-magnetic particles like quartz and feldspar pose a challenge [23]. Moisture content plays a role as well; high moisture can cause clumping of particles, reducing the separation efficiency. Chemical contaminants can alter the magnetic properties of soil particles, whereas metallic contaminants can sometimes enhance magnetic properties, making separation easier, whereas non-metallic contaminants might indirectly affect separation by changing the physical behaviour of the soil.

Heterogeneous soil matrix of brownfields will result in the insignificant efficiency of removing metal contaminants in soil [8]. However, magnetic separation is particularly effective in soils with high iron content, low organic matter, and minimal moisture, which facilitate better exposure of magnetic particles to the magnetic field. It is less effective in soils with high clay or organic content and those with predominantly non-magnetic minerals, like peaty or siliceous soils. Therefore, the success of magnetic separation in brownfield remediation depends on a careful assessment of soil properties and the selection of appropriate conditions for the process. The application of electrostatic separation seems to be limited and hardly practiced in the field of soil remediation. A full-scale application for electrostatic separation is the lead-based paint chip separation and recovery technology of the PMET [24]. After gravity separation steps, ferromagnetic separation, and classification of size, this method uses the final step in the electrostatic separation technique. An overview of magnetic separation, electrostatic separation, and froth flotation is shown in Table 2.

Method	Principle	Applications	Advantages	Limitations	References
Froth Flotation	Utilizes differences in hydrophobic properties. Metal- bearing particles attach to air bubbles, forming froth for separation.	Commonly used in mineral processing; effective for metals like cadmium, copper, lead, and zinc. Used for fine- grained sediments and soils. especially those with metal sulfides.	Effective for fine particles and specific metals (e.g., sulfides). Allows selective recovery of minerals.	Less effective for coarse particles. Limited use in soil remediation compared to other methods.	$[5,16-18]$
Magnetic Separation	Exploits differences in magnetic properties. Ferromagnetic materials are attracted to magnetic fields, enabling separation.	Ideal for recovering ferrous materials from soil, particularly from brownfields and military sites. Effective for metals like chromium, copper, nickel, lead, and zinc.	Non-destructive. Can handle large volumes of material. Effective for both ferromagnetic and paramagnetic materials.	Ineffective for non-magnetic contaminants. Performance affected by soil heterogeneity.	[7,19,20]
Electrostatic Separation	Based on differences in electrical conductivity. Particles are charged and separated using an electric field.	Primarily used for the separation of lead-based paint chips and recovery of metals after preliminary treatments.	Can achieve high- purity separations. Effective as a final step in multi-stage separation processes.	Limited application in soil remediation. Requires pre- processing steps for effective separation.	$\lceil 24 \rceil$

Table 2. Overview of magnetic separation, electrostatic separation, and froth flotation methods for treating metal contaminated soil remediation.

2.6. Attrition scrubbing.

Attrition scrubbers typically operate with powerful counter-flows generated by dual impellers submerged in a soil slurry with a high solid content, approximately 70 to 80 percent [10]. This mechanical particle-to-particle scrubbing is designed to produce two primary effects: scouring and breaking. The scouring effect refers to the process of removing the film or coating from individual grains, which exposes fresh and clean surfaces. While the breaking effect includes the separation or dispersion of clay and silt bound to the particles of sand and gravel, and soil agglomerates disintegration. Soil scrubbing is carried out primarily by attrition of particles to particles, but also through the contact between the particles and paddles. This process is crucial as it improves the subsequent treatment processes by increasing the exposure and accessibility of contaminants.

In froth flotation, cleaner surfaces enhance the attachment of hydrophobic contaminants to air bubbles, thereby improving separation efficiency. Marino et al. [25] demonstrated that the mechanical attrition on the Wilfley table improves the metal removal efficiency through the gravity concentration. The scrubbing effect creates clean and fresh particle surfaces by scraping off oxidized coating and can improve the following process of froth flotation [16]. Attrition scrubbers are also used to enhance chemical extraction by improving the removal of contaminants superficially bound to solid particles. For chemical extraction, the removal of coatings and disaggregation of particles increases the contact area for solvents, enhancing contaminant removal. Attrition scrubbing is particularly beneficial for soils contaminated with heavy metals or hydrophobic organic compounds. Its beneficial condition when these contaminants are bound to fine particles or coated surfaces, such as in clayey or silty soils, where contaminants are tightly bound and direct remediation is less effective. According to Williford et al. [10], preconditioning by attrition scrubbing increases the efficiency of hydroclassification. Overall, by creating more favourable conditions for the removal of contaminants, attrition scrubbing reduces the need for chemical reagents and lowers overall remediation costs, making it a critical step in soil remediation efforts.

2.7. Integrated process train of physical separation.

In application of physical separation, typical physical separation treatment train is comprised of the following process. Isolation of oversized material by using mechanical screening as preliminary step of size classification. Preceded and accompanied of hydroclassification step by attrition scrubbing to give a sufficient particle size array for future treatment. Sand fraction treatment by froth flotation or gravity concentration. Treating the fine fraction and managing the residuals generated. Most physical separation procedure are established on basic separation of particles due to the fine fraction is usually observed as contaminated and coarse fraction as uncontaminated. Metal contamination, however, are able to spread across different particle size fractions and concentrations in sand fractions can be appeared as high, particularly in industrial or urban soils that are contaminated by disposal of heterogenous waste [26]. If contamination of metal is particulate in nature and is plentiful in all fractions of particle size, separation base on size alone cannot achieve the adequate metal contaminants separation. By this, the density and floatability of the separation shall be examined. Water treatment, desliming, crushing and dewatering are the essential to the physical separation treatment train. Physical separation can also be used as an independent method of volume reduction or as pre-treatment before recovery of metal.

2.8. Physical separation technologies advantages and disadvantages.

The advantages of physical separation technologies are that in the same treatment system, both organic and metal contaminants can be treated. Soil volume can be further recycled or disposed off-site can be significantly reduced. Treated soil can be put back at low cost to the site and some metal recovery can be used for recycling in some cases. Train treatment systems are simply scalable and certain mobile unknit systems for on-site remediation are available at full scale. Lastly, the technologies in the mineral processing industry are well developed and operating costs are generally low. For large-scale implementations, the physical separation of soil treatment has some drawbacks. Treatment system for physical separation technologies usually requires huge space for soil and big sized equipment for treatment of soil. In order to be cost-effective, large volume of soils must be required during treatment. Lastly, wash-water treatment and the disposal of solid residuals off-site might be necessary, thereby consequently increasing costs [7].

3. Chemical Extraction Methods

Chemical extraction method employs chemical reagent extracting fluid to carry the metals in soil to aqueous solution. In context of soil remediation, solubility can be enhanced by leaching solutions in dissolving metal contaminants or by transforming the compounds into more soluble forms. In this section, five leaching solution types will be discussed, including acids, chelating agents, surfactants, and oxidizing or reducing agents. The summary of these types is presented in Table 3.

3.1. Chemical extraction applicability and limitations.

The performance of metal removal through the chemical extraction methods depends on the geochemistry of the soil, characteristics of metal contamination, dosage chemistry of the extraction agent and the storage conditions. Fractionation and speciation of metals in soils are essential parameters for identifying the efficiency of metal removal through chemical treatments. The analysis of metal speciation can be complicated and the metal fractionation is often applied in accordance with soil substrates. The division of metals by contact with the soil substrates is generally determined by the sequential extraction process [27]. Typically, five fractions are used as the analytical protocols known as the exchangeable, carbonate or soluble bounded acid, Fe-Mn or reducible bound, sulfide and organic or oxidizable bound and residual [28]. Factors that might restrict the chemical extraction applicability and efficacy include the high content of clay or silt, humic, iron, and calcium elements, high calcite or buffering capacity, contamination of cationic and anionic heavy metals simultaneously, elevated soil heterogeneity, and Metals associated with fractions of residual rock, found in mineral lattices or isolated particles. Fine grained soils need greater contact times and might decrease the efficiency of chemical extraction. The high content of humic can impede metal extraction due to the high affinity of COOH groups on humic substances for heavy metals [29]. The respective cations of iron and calcium can disrupt the chelating process. The high content of calcite or high buffering capability may reduce the efficiency of acid leaching [30]. High soil heterogeneity may affect fluid extraction formulations and might need multiple steps in the process.

Method	Applicability and Limitations	Citation
Chemical Extraction	• Performance depends on soil geochemistry, metal contamination characteristics, dosage chemistry, and storage conditions • Fractionation and speciation of metals crucial for treatment efficiency • Factors restricting applicability include high clay or silt content, humic substances, iron and calcium presence, high calcite or buffering capacity, cationic and anionic heavy metal contamination, soil heterogeneity, and metals associated with residual rock • Fine-grained soils require greater contact times, while high humic content impedes extraction • Iron and calcium cations disrupt chelation, and calcite or high buffering capacity reduces acid leaching efficiency • Soil heterogeneity may necessitate multiple extraction steps	$[27 - 30]$
Acid Extraction	• Validated technology for metal-contaminated soils, sediments, and sludges • pH value of washing fluid crucial for metal extraction • Metal cation desorption via ion exchange and dissolution of soil mineral components and metal compounds • Low pH enhances metal cation desorption by binding protons to soil surface sites	[29, 31, 32]
Chelant Extraction	• Chelating agents form stable metal complexes, facilitating metal extraction • Selection criteria include stability over a wide pH range, non-adsorbable complexes on soil surfaces, low biodegradability for reagent recycling, and cost efficiency • EDTA offers less soil structure damage compared to strong acids but poses ecological threats and higher costs	[33, 34]
Surfactant Solubilization	• Surfactants assist in desorption or dispersion of contaminants from soil, enhancing metal mobilization • More effective when metals are closely attached to organic contaminants • Selection based on soil and metal substrates and acidity • Caustic surfactants suggested for organically-related metals • Acidic surfactants may extract carbonate and oxide-bounded metals • Anionic biosurfactants can extract cationic metal forms but not yet implemented on a large scale	$[35 - 37]$
Integrated Process Train	• Heap or pile leaching cost-effective for industrial soil treatment but slower and requires higher metal concentrations Agitation leaching enables rapid and efficient metal pollutant removal After dissolution, leaching solution treated with specific aqueous processes like complexation, sedimentation, or electrochemical techniques • Solvent extraction selectively transfers ionic metal species to an organic phase • Washed soil rinsed with water to remove residual extracting agents before disposal on-site or off-site depending on soil characteristics.	[38, 39]

Table 3. Overview of chemical extraction methods for metal contaminated soil remediation.

3.2. Acid extraction.

Acid extraction is a validated technology for the treatment of metal-contaminated soils, sediments, sludges, and commercial units [31]. Washing fluid pH value plays a major role in extracting heavy metals from soils. Metal cation desorption through ion exchange and dissolution of soil mineral components and metal compounds contribute to the mechanism for extraction of metals with the use of acid solution [32]. When the pH is low, the added protons can bind to soil surface sites and increase metal cation desorption, which is transferred to the washing liquid [29].

3.3. Chelant extraction.

Because chelating agents are capable of forming stable metal complexes, their use provides a favourable approach to metal extraction from contaminated soils. Five major factors are considered in the selection of chelating agents for the extraction of metals from soils. Highly stable complexes over a wide range of pH should be formed by chelating agents. Metal complexes built on the surfaces of the soil should be non-adsorbable. The biodegradability of chelating agents should be low if the reagents are being recycled for reuse. Metal recovery and

reagents used should be cost-efficient [33]. The primary benefit of using chelating agents such as EDTA compared to using strong acids is that chelating agents cause less soil structure damage. Nonetheless, EDTA has two major drawbacks in contrast with HCl. Using chemical products requires higher cost and EDTA might pose a major threat to ecology if it is not treated properly after the washing process due to its low biodegradability [34].

3.4. Surfactant-enhanced solubilization.

While the use of surfactants is more adequate for the treatment of organic contaminants, it is also a fascinating chemical technique to examine the extraction of metals from the soil by surfactants. The use of surfactants has recently been studied to boost the EDTA leaching cycle for the mobilization of metals from soil [35]. The addition of surfactant in the washing solution is intended to assist in the desorption or dispersion of contaminants from soil. Washing with surfactants can be more efficient when the metals are closely attached to organic contaminants. The combination of soil and metal substrates and acidic or simple conditions are critical criteria for effective soil washing with surfactants. Caustic surfactant was suggested by Mulligan et al. [36] to be used for removing organically-related metals while acidic surfactant may be used to extract carbon and oxide bounded metals. Several laboratory studies have shown that anionic biosurfactant solutions can extract cationic forms of metals from contaminated soils and sediments [37]. Nonetheless, for large-scale remediation ventures, metal extraction by biosurfactants has not yet been carried out.

3.5. Integrated process train of chemical extraction.

Leaching methods are classified into two main classes in large-scale operations known as leaching percolation such as leaching of heap or pile or vat leaching and agitated batch-based leaching and continuous process [38]. Throughout the process of heap leaching, the soil is stacked throughout a heap and the leach solution is sprayed over the top of the heap and allowed to percolate down through the heap. Under turbulent flow conditions, the agitated leaching method is carried out. Heap or pile leaching tends to be more cost-effective for industrial soil treatment [32]. Nevertheless, the processes of extraction can be sluggish and metal concentrations must be comparatively higher. Agitation leaching is an important approach for soil treatment because it can enable rapid and efficient removal of metal pollutants from soil [39]. After the metal compounds have been dissolved, the leaching solution must be isolated from the residual solid soil phase. The resulting metal-containing solution can be treated with specific aqueous processes such as complexation, sedimentation, precipitation, electrochemical techniques, exchange of liquid ions, exchange of resin ions, and metal recovery membrane technologies. Solvent extraction techniques enable certain ionic metal species in an aqueous washing solution to be selectively transferred to an organic liquid phase if the metal compounds extracted are preferably soluble in the organic phase [38]. Usually, the washed soil is rinsed with water to remove the residual soil extracting agents. The resulting clean soil fraction may be returned to the site or disposed of off-site depending on the soil characteristics and reagents used.

3.6. Chemical extraction technologies advantages and disadvantages.

Similar to physical separation, the main advantages of chemical procedures are generally treatable sorbed metal forms and dissolvable certain metal compounds. In some cases, finegrained soils may be treated and metals extracted can easily be recovered using a wide range of methods. If the chemical reagents are recycled, detoxified or not hazardous, chemically enhanced soil washing can become attractive. There are many disadvantages to the large-scale implementation of chemical extraction processes. The use of chemical agents can greatly increase the costs of processing. The treated soil may be inappropriate for revegetation and onsite disposal because it has compromised physicochemical and microbiological properties. There may be problems with the presence of toxic chemical agents in the final soil or residual sludge for disposal. Particular chemical agents in the washing fluid can make recycling and treatment of water more difficult, thus increasing the overall cost of the process. Metal-rich sludge treatment may also be difficult. Chemical agents may also cause environmental issues such as EDTA leaching may pollute and persist in the soils and treatment with wastewater can produce large quantities of toxic sludges that must be managed carefully.

4. Combination of Physical Separation and Chemical Extraction

The combined use of isolation of small particles and chemical leaching procedures offers a very beneficial implementation for decontaminating metal-affected soils. The standard approach is to use physical separation to concentrate particulate metal forms into smaller soil volumes, followed by chemical removal of this concentrated fraction to remove metals [7]. This dual approach capitalizes on the advantages of both physical and chemical processes, thereby improving soil remediation efficiency and effectiveness. Using hydroclassification and attrition scrubbing with water-based fluid, many soil washing processes are based on simple particle size separation. Separation of particle size is often used before chemical extraction, assuming that most of the metal contamination is contained in the fines. The practice of direct disposal is inappropriate since metal concentrations are usually high in the fine fraction. The fundamental principle is that fine soil fractions often contain a higher density of metal contaminants. Isolating these fine particles facilitates targeted chemical treatment, leading to increased efficiency and a reduction in the volume of soil needing chemical intervention. Size separation is essential because it aids in the control and reduction of the total volume of soil requiring thorough treatment.

Chemical extraction can be used to decontaminate the fine fractions and allow the recovery of metals in a saleable or concentrated form. This process involves the use of acids, chelating agents, or surfactants that dissolve metal contaminants, allowing their subsequent removal from the soil matrix. Besides facilitating the removal of metals from the concentrated fine fractions, this step also allows for the possible recovery of metals in a form that can be recycled or sold, so the treatment process becomes more economically viable. Soil washing systems may include other types of combinations depending on the properties of the soil matrix, the speciation of metals, and the metal type to be treated. The physical separation or chemical extraction combination may be reversed or a parallel cycle of physical separation or chemical extraction approaches may be involved. For instance, metals such as lead and cadmium, commonly present in fine particulate forms, are especially conducive to this integrated approach. Fine particle isolation, followed by chemical extraction, is highly effective at removing such contaminants. Some processes use chemically enhanced attrition scrubbing with chelating agents, acids or surfactants and later go through the hydrocycloning process to extract fine particles from the clean fraction [40]. Soil washing remediation technique aims to decontaminate the site completely by removing metals from the soil matrix. The primary objective of the extraction strategy is to retrieve metals for reusing and resale, but for projects lacking economic viability or technical feasibility for recovery and extraction processes, metal recovery is often not practicable. Soil washing is used in many cases to decrease metal concentrations to an acceptable level or to significantly decrease contaminated soil volume.

5. Conclusion

Soil washing is a technique that is distinctly suitable for metal-contaminated soil remediation. Most projects are focused on cost-effectiveness and well-established physical separation technologies in the industry of mineral processing. Soil washing can be an effective substitution for solidification and stabilisation and landfilling from an environmental and economic point of view. Soil washing provides benefits such as the ability to remove metals permanently from the soil and may in some cases allowance of metal recycling. Reduction of large volumes of contaminated soil and soil can be returned to the excavated site after being treated. Compared to other metal extraction methods, the process duration is typically short to medium. Nevertheless, the effectiveness of the soil washing process requires an exhaustive soil characterisation, an analysis of metal speciation and fractionation, and an understanding of the interaction of soil matrix with metal or contaminants.

Acknowledgments

The authors would like to acknowledge the team of Curtin University Malaysia for the technical support in this project.

Competing Interest

The authors declare no conflict of interest.

References

- [1] Chen, X.; Xia, X.; Zhao, Y.; Zhang, P. (2010). Heavy metal concentrations in roadside soils and correlation with urban traffic in Beijing, China. *Journal of Hazardous Materials*, *181*, 640‒646. [http://doi/org/https://doi.org/10.1016/j.jhazmat.2010.05.060.](http://doi/org/https:/doi.org/10.1016/j.jhazmat.2010.05.060)
- [2] Wu, Q.; Leung, J.Y.S.; Geng, X.; Chen, S.; Huang, X.; Li, H.; Huang, Z.; Zhu, L.; Chen, J.; Lu, Y. (2015). Heavy metal contamination of soil and water in the vicinity of an abandoned e-waste recycling site: Implications for dissemination of heavy metals. *Science of The Total Environment*, *506*‒*507*, 217‒225. [http://doi/org/https://doi.org/10.1016/j.scitotenv.2014.10.121.](http://doi/org/https:/doi.org/10.1016/j.scitotenv.2014.10.121)
- [3] Ashraf, S.; Ali, Q.; Zahir, Z.A.; Ashraf, S.; Asghar, H.N. (2019). Phytoremediation: Environmentally sustainable way for reclamation of heavy metal polluted soils. *Ecotoxicology and Environmental Safety*, *174*, 714‒727. [http://doi/org/https://doi.org/10.1016/j.ecoenv.2019.02.068.](http://doi/org/https:/doi.org/10.1016/j.ecoenv.2019.02.068)
- [4] Treatment Technologies for Site Cleanup: Annual Status Report, Twelfth Edition. (accessed on 1 February 2024) Available online: [https://www.epa.gov/remedytech/treatment-technologies-site](https://www.epa.gov/remedytech/treatment-technologies-site-cleanup-annual-status-report-twelfth-edition)[cleanup-annual-status-report-twelfth-edition.](https://www.epa.gov/remedytech/treatment-technologies-site-cleanup-annual-status-report-twelfth-edition)
- [5] Dermont, G.; Bergeron, M.; Mercier, G.; Richer-Laflèche, M. (2008). Soil washing for metal removal: A review of physical/chemical technologies and field applications. *Journal of Hazardous Materials*, *152*, 1‒31. [https://doi.org/10.1016/j.jhazmat.2007.10.043.](https://doi.org/10.1016/j.jhazmat.2007.10.043)
- [6] Wills, B.A. (2008). Wills' Mineral Processing Technology: An Introduction to The Practical Aspects of Ore Treatment and Mineral Recovery, $7th$ Ed.; Butterworth-Heinemann (Elsevier): Burlington, Massachusetts, USA.
- [7] Contaminants and remedial options at selected metal-contaminated sites. Technical resource report. (accessed on 1 February 2024) Available online: [https://nepis.epa.gov/Exe/ZyPURL.cgi?Dockey=100026C0.TXT.](https://nepis.epa.gov/Exe/ZyPURL.cgi?Dockey=100026C0.TXT)
- [8] Mercier, G.; Duchesne, J.; Blackburn, D. (2001). Prediction of Metal Removal Efficiency from Contaminated Soils by Physical Methods. *Journal of Environmental Engineering, 127,* 4. [https://doi.org/10.1061/\(ASCE\)0733-9372\(2001\)127:4\(348\).](https://doi.org/10.1061/(ASCE)0733-9372(2001)127:4(348))
- [9] Shim, H.Y.; Lee, K.S.; Lee, D.S.; Jeon, D.S.; Park, M.S.; Shin, J.S.; Lee, Y.K.; Goo, J.W.; Kim, S.B.; Chung, D.Y. (2014). Application of electrocoagulation and electrolysis on the precipitation of heavy metals and particulate solids in washwater from the soil washing. *Journal of Agricultural Chemistry and Environment*, *3*, 130.
- [10] Cho, K.; Kim, H.; Purev, O.; Choi, N.; Lee, J. (2022). Physical Separation of Contaminated Soil Using a Washing Ejector Based on Hydrodynamic Cavitation. *Sustainability*, *14*, 252. [https://doi.org/10.3390/su14010252.](https://doi.org/10.3390/su14010252)
- [11] Gómez-Sagasti, M.T.; Anza, M.; Hidalgo, J.; Artetxe, U.; Garbisu, C.; Becerril, J.M. (2021). Recent Trends in Sustainable Remediation of Pb-Contaminated Shooting Range Soils: Rethinking Waste Management within a Circular Economy. *Processes*, *9*, 572. [https://doi.org/10.3390/pr9040572.](https://doi.org/10.3390/pr9040572)
- [12] Mulligan, C.; Yong, R.; Gibbs, B. (2001). An evaluation of technologies for the heavy metal remediation of dredged sediments. *Journal of Hazardous Materials*, *85*, 145‒163. [http://doi/org/10.1016/S0304-3894\(01\)00226-6.](http://doi/org/10.1016/S0304-3894(01)00226-6)
- [13] Pasciucco, F.; Pecorini, I.; Di Gregorio, S.; Pilato, F.; Iannelli, R. (2021). Recovery Strategies of Contaminated Marine Sediments: A Life Cycle Assessment. *Sustainability*, *13*, 8520. [https://doi.org/10.3390/su13158520.](https://doi.org/10.3390/su13158520)
- [14] Luttrell, G.; Westerfield, T.C.; Kohmuench, J.N.; Mankosa, M.J.; Mikkola, K.A.; Oswald, G. (2006). Development of high-efficiency hydraulic separators. *Minerals and Metallurgical Processing*, *23.* 33‒39. [http://doi/org/10.1007/BF03403333.](http://doi/org/10.1007/BF03403333)
- [15] Priya, A.K.; Muruganandam, M.; Ali, S.S.; Kornaros, M. (2023). Clean-Up of Heavy Metals from Contaminated Soil by Phytoremediation: A Multidisciplinary and Eco-Friendly Approach. *Toxics*, *11*, 422. [https://doi.org/10.3390/toxics11050422.](https://doi.org/10.3390/toxics11050422)
- [16] Hu, B.; Huang, L.; Yang, B.; Xie, X.; Tong, X.; Zhang, X.; Sun, X. (2022). Flotation Performance and Adsorption Mechanism of a Novel Chelating Collector for Azurite. *Minerals*, *12*, 441. [https://doi.org/10.3390/min12040441.](https://doi.org/10.3390/min12040441)
- [17] Du, Y.; Tong, X.; Xie, X.; Zhang, W.; Yang, H.; Song, Q. (2021). Recovery of Zinc and Silver from Zinc Acid-Leaching Residues with Reduction of Their Environmental Impact Using a Novel Water Leaching-Flotation Process. *Minerals*, *11*, 586. [https://doi.org/10.3390/min11060586.](https://doi.org/10.3390/min11060586)
- [18] Cauwenberg, P.; Verdonckt, F.; Maes, A. (1998). Flotation as a remediation technique for heavily polluted dredged material. 1. A feasibility study. *Science of The Total Environment*, *209*, 113‒119. [https://doi.org/10.1016/S0048-9697\(98\)80102-2.](https://doi.org/10.1016/S0048-9697(98)80102-2)
- [19] Rikers, R.A.; Rem, P.; Dalmijn, W.L. (1998). Improved method for prediction of heavy metal recoveries from soil using high intensity magnetic separation (HIMS). *International Journal of Mineral Processing*, *54*, 165‒182. [https://doi.org/10.1016/S0301-7516\(98\)00017-9.](https://doi.org/10.1016/S0301-7516(98)00017-9)
- [20] Orjuela-Abril, S.; Torregroza-Espinosa, A.; Duarte-Forero, J. (2023). Innovative Technology Strategies for the Sustainable Development of Self-Produced Energy in the Colombian Industry. *Sustainability*, *15*, 5720. [https://doi.org/10.3390/su15075720.](https://doi.org/10.3390/su15075720)
- [21] Lackner, M.; Hribernig, T.; Lutz, M.; Plank, M.; Putz, K. (2022). Extraction of aged hydrocarbons from contaminated soil using plant-oil-in-water emulsions combined with oil/water separation by reusable non-wovens. *Applied Sciences*, *12*, 6179. [https://doi.org/10.3390/app12126179.](https://doi.org/10.3390/app12126179)
- [22] Liu, Y.; Li, C.; Omar, R.B.; Shi, X.; Zhang, H.; Faiz, N.N. (2021). Sediment Sources and Dispersion on the Western Sunda Shelf, Malay Peninsula, Southern South China Sea. *Water*, *13*, 2823. [https://doi.org/10.3390/w13202823.](https://doi.org/10.3390/w13202823)
- [23] Ku, J.; Wang, K.; Wang, Q.; Lei, Z. (2024). Application of Magnetic Separation Technology in Resource Utilization and Environmental Treatment. *Separations*, *11*, 130. [https://doi.org/10.3390/separations11050130.](https://doi.org/10.3390/separations11050130)
- [24] Choi, Y.; Rhee, S.-W. (2017). Evaluation of Energy Consumption in the Mercury Treatment of Phosphor Powder from Spent Fluorescent Lamps Using a Thermal Process. *Sustainability*, *9*, 2013. [https://doi.org/10.3390/su9112013.](https://doi.org/10.3390/su9112013)
- [25] Marino, M.; Brica, R.; Neale, C. (2006). Heavy metal soil remediation: The effects of attrition scrubbing on a wet gravity concentration process. *Environmental Progress*, 16, 208–214. [http://doi/org/10.1002/ep.3300160318.](http://doi/org/10.1002/ep.3300160318)
- [26] Yarlagadda, P.S.; Matsumoto, M.R.; VanBenschoten, J.E.; Kathuria, A. (1995). Characteristics of Heavy Metals in Contaminated Soils. *Journal of Environmental Engineering*, 121, 276–286. [http://doi/org/10.1061/\(ASCE\)0733-9372\(1995\)121:4\(276\).](http://doi/org/10.1061/(ASCE)0733-9372(1995)121:4(276))
- [27] Dahlin, C.L.; Williamson, C.A.; Keith Collins, W.; Dahlin, D.C. (2002). Sequential Extraction Versus Comprehensive Characterization of Heavy Metal Species in Brownfield Soils. *Environmental Forensics*, *3*, 191‒201. [https://doi.org/10.1006/enfo.2002.0090.](https://doi.org/10.1006/enfo.2002.0090)
- [28] Tessier, A.P.; Campbell, P.; Bisson, M. (1979). Sequential Extraction Procedure for the Speciation of Trace Metals. *Analytical Chemistry*, *51,* 844–851[. http://doi/org/10.1021/ac50043a017.](http://doi/org/10.1021/ac50043a017)
- [29] Isoyama, M.; Wada, S.-I. (2007). Remediation of Pb-contaminated soils by washing with hydrochloric acid and subsequent immobilization with calcite and allophanic soil. *Journal of Hazardous Materials*, *143*, 636‒642[. https://doi.org/10.1016/j.jhazmat.2007.01.008.](https://doi.org/10.1016/j.jhazmat.2007.01.008)
- [30] Tejowulan, R.S.; Hendershot, W. (1998). Removal of trace metals from contaminated soils using EDTA incorporating resin trapping techniques. *Environmental Pollution*, *103*, 135‒142. [http://doi/org/10.1016/S0269-7491\(98\)00080-3.](http://doi/org/10.1016/S0269-7491(98)00080-3)
- [31] Zhang, M.; Yang, S.; Zhang, Z.; Guo, C.; Xie, Y.; Wang, X.; Sun, L.; Ning, Z. (2023). Development and Application of an Integrated Site Remediation Technology Mix Method Based on Site Contaminant Distribution Characteristics. *Applied Sciences, 13*, 11076. [https://doi.org/10.3390/app131911076.](https://doi.org/10.3390/app131911076)
- [32] Tampouris, S.; Papassiopi, N.; Paspaliaris, I. (2001). Removal of contaminant metals from fine grained soils, using agglomeration, chloride solutions and pile leaching techniques. *Journal of hazardous materials*, *84*, 297‒319. [http://doi/org/10.1016/S0304-3894\(01\)00233-3.](http://doi/org/10.1016/S0304-3894(01)00233-3)
- [33] Abumaizar, R.J.; Smith, E.H. (1999). Heavy metal contaminants removal by soil washing. *Journal of Hazardous Materials*, *70*, 71‒86. [https://doi.org/10.1016/S0304-3894\(99\)00149-1.](https://doi.org/10.1016/S0304-3894(99)00149-1)
- [34] Lim, T.-T.; Chui, P.-C.; Goh, K.-H. (2005). Process evaluation for optimization of EDTA use and recovery for heavy metal removal from a contaminated soil. *Chemosphere*, 58, 1031–1040. [https://doi.org/10.1016/j.chemosphere.2004.09.046.](http://doi/org/https:/doi.org/10.1016/j.chemosphere.2004.09.046)
- [35] Ehsan, S.; Prasher, S.O.; Marshall, W.D. (2006). A washing procedure to mobilize mixed contaminants from soil: II. Heavy metals. *Journal of Environmental Quality*, *35*, 2084‒2091. [http://doi/org/10.2134/jeq2005.0475.](http://doi/org/10.2134/jeq2005.0475)
- [36] Mulligan, C.N.; Yong, R.N.; Gibbs, B.F. (1999). On the use of biosurfactants for the removal of heavy metals from oil-contaminated soil. *Environmental Progress*, 18, 50–54. [https://doi.org/10.1002/ep.670180120.](https://doi.org/10.1002/ep.670180120)
- [37] Mulligan, C.N.; Yong, R.N.; Gibbs, B.F.; James, S.; Bennett, H.P.J. (1999). Metal Removal from Contaminated Soil and Sediments by the Biosurfactant Surfactin. *Environmental Science & Technology*, *33*, 3812‒3820[. http://doi/org/10.1021/es9813055.](http://doi/org/10.1021/es9813055)
- [38] Pianowska, K.; Kluczka, J.; Benke, G.; Goc, K.; Malarz, J.; Ochmański, M.; Leszczyńska-Sejda, K. (2023). Solvent Extraction as a Method of Recovery and Separation of Platinum Group Metals. *Materials*, *16*, 4681. [https://doi.org/10.3390/ma16134681.](https://doi.org/10.3390/ma16134681)
- [39] Mystrioti, C.; Papassiopi, N. A (2024). Comprehensive Review of Remediation Strategies for Soil and Groundwater Contaminated with Explosives. *Sustainability*, *16*, 961. [https://doi.org/10.3390/su16030961.](https://doi.org/10.3390/su16030961)
- [40] Ko, I.; Lee, C.-H.; Lee, K.-P.; Lee, S.-W.; Kim, K.-W. (2006). Remediation of soil contaminated with arsenic, zinc, and nickel by pilot-scale soil washing. *Environmental Progress*, 25, 39-48. [http://doi.org/10.1002/ep.10101.](http://doi.org/10.1002/ep.10101)

© 2024 by the authors. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).