Review Article

Technologies for Remediation of Heavy Metals in Environment and Ecosystem: A Critical Overview of Comparison Study

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ABSTRACT

Ecosystem contamination is a global issue that is directly related to the advancement of contemporary society. Because of its effects on both human health and the natural environment that's why heavy metal pollution of the environment is currently a serious environmental issue that affects all of humanity. Heavy metal pollution receives special attention because it frequently presents the greatest remedial challenges. Certain technologies, usually based on physical, chemical, thermal and biological methods have long been used to remove heavy metals from contaminated environments to a level that is safe and acceptable. The kind and properties of the heavy metals, the degree of pollution and the combination of contaminants in the environment all have a significant impact on how effective these techniques are. The environment is also at risk from a few conventional technologies. Thus, development and research are needed for effective environmentally friendly methods that are based on natural materials or soil elements. With a focus on novel approaches to remediation techniques, this manuscript will give an overview of recent exploration and research, attempts to evaluate the effectiveness of remediation and advancements in decontamination technologies that can be used to remove heavy metals from contaminated areas. Additionally, a summary of the causes, consequences and technologies for cleanup are provided for heavy metals.

Keywords: Environment, Ecosystem, Ecosystem Contamination,Heavy Metals, Remediation, Technologies

1. Introduction

Heavy metals are regarded as significant environmental pollutants due to their high density and high toxicity even at low concentrations [1-2]xx. The term "heavy metal" generally refers to metallic elements with an atomic weight higher than that of Iron (Fe) (55.8 g mol^{-1}) or a density greater than 5.0 g cm^{-3} and these metals are naturally present in the environment [1, 3]. However, some metals with an atomic weight lower than that of Fe, for example, Cr, and others that are considered metalloids such as Arsenic (As) and Selenium (Se) are also commonly referred to as heavy metals [3-4]. Heavy metals can play a role as micronutrients such as Cu, Fe, Mn, Mo, Zn and Ni but they can also be toxic to humans, e.g., Hg, Pb, Cd, Cu, Ni and Co depending on the exposure levels [3, 35]. according to the United States Environmental Protection Agency (USEPA) compilation of eight heavy metals namely, lead (Pb), chromium (Cr), arsenic (As), zinc (Zn), cadmium (Cd), copper (Cu), mercury (Hg) and nickel (Ni) are listed to be the most widespread heavy metals in the environment [2, 4]. According to the coordination chemistry of heavy metals, the above-mentioned heavy metals are also categorized as class B metals that are non-essential (highly toxic) trace elements [5]. Broad classification of heavy metals with examples is tabulated in Table 1. Heavy metals constitute an ill-defined group that is most commonly found at contaminated sites [6]. They are characterized by their long persistence in the natural environment leading to serious health consequences in humans, animals and plants even at very low concentrations such as 1.0 or 2.0 μg in some cases [2, 4, 7]. A wide array of toxic heavy metals like Cr, Cd, Hg, Pb, etc., disposed of by industries will remain non-

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degradable and contaminate the soil and water to a greater extent [8-10]. Because of the high propensity nature of the heavy metals, they tend to accumulate in various environmental matrices, resulting in misleadingly higher concentrations than the prescribed average safety levels [11-14]. According to the Comprehensive Environmental Response Compensation and Liability Act, USA, the maximum permissible limit of heavy metals in aqueous medium is as follows, Cr-0.01 mg/L, Ar-0.01 mg/L, Cd-0.05 mg/L, Hg-0.002 mg/L, Pb-0.015 mg/L and Ag-0.05 mg/L respectively [2, 15-17]. If the heavy metal concentration exceeds those recommended, it can be a major source of many human life-threatening complications such as atherosclerosis, cancer, Alzheimer's disease, Parkinson’s disease, etc [2, 18].

Remediation activities aim to transform unusable property into available use and conserve land resources to improve environmental conditions in the contaminated site and around it as well as to reduce the risk to humans and the environment [19]. Remediation means actions taken to clean up, mitigate, correct, abate, minimize, eliminate, control contain and prevent a release of a contaminant into the environment to protect human health and the environment,these cleanup technologies can be applied on or off-site, utilizing three kinds of remediation treatments such as electrical, biological and chemical techniques [20-22]. This article aims to precise and evaluate the recent technologies applied in the remediation of heavy metals contaminated soils in literature to gain the suitable technique that should be applied in contaminated soils and to minimize the hazards of such toxic metals under Egyptian conditions [2, 23].

Table: 1. Classification of heavy metals with example

Class of heavy metal	Example	**Reference**
1. Macro-nutrient elements	Cobalt, Iron	[2, 3]
2. Micro-nutrient	Copper, Nickel, Chromium, Manganese,	[2, 3]

elements	Molybdenum	
3. Highly toxic elements	Mercury, Cadmium, Lead, Silver, Gold, Palladium, Bismuth, Arsenic, Platinum, selenium, Tin, Zinc.	[2, 4]
4. Precious elements	Platinum, Silver, Gold, Ruthenium.	[2, 6]
5. Radionuclides	Uranium, Thorium, Radium, Cerium, Praseodymium.	[2, 7]

2.0. Metals as environmental pollutants

Heavy metals are naturally occurring elements that are found throughout the earth's crust. Heavy metal pollution is caused as a result of both natural and anthropomorphic activities like mining, smelting, industrial production and the use of metals and metal-containing compounds for domestic and agricultural applications [2, 23]. These sources were reported to contribute to human exposure and environmental contamination by various researchers. Toxicological properties of heavy metals are characterized by the persistence of metal (long half-life), soil residence time (>1,000 years), chronic and sub-lethal effects of the metal, bioaccumulation, biomagnification, teratogenic and carcinogenic properties of the metal [2, 24].

2.1. Heavy Metals Distribution in Environment

2.1.1. Natural Sources

2.1.1.1. Heavy Metals in Soils

Rocks disintegrate into fine particles or soil by the influence of ice, water, temperaturexetc. The soil matrix is a major reservoir or transporting media for heavy metals because the soil and heavy metals associations have rich and diverse binding characteristics [2, 24]. Metals do not biodegrade like organic pollutants rather they bioaccumulate in the environment. Soil matrix may adsorb, oxidize, exchange, catalyze, reduce or precipitate the metal ions [2, 25]. These processes depend on several factors such as pH, water content, temperature, particle size distribution, nature of metal and the clay content. This composition will determine the mobility,

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solubility and toxicity of heavy metals present in the soil [25]. Generally, the minerals are dissolved by interacting with carbonic acid and water [2, 26]. The insoluble minerals are dispersed into fine particles. Soils are contaminated by metals and metalloids from metal wastes, gasoline, animal manure, sludge, wastewater irrigation, atmospheric deposition, etc [26]. The most common heavy metals found in soils are Pb, Cr, Zn, Cd, and Hg. Due to bioaccumulation and biomagnification, these metals decrease crop production and affect the food chain [2]. The soil concentration ranges and regulatory guidelines for some heavy metals are given in **Table 2.**

Table: 2. Concentration ranges and regulatory guidelines of heavy metals

Metals	Soil concentration range (mg/kg)	Regulatory limits (mg/kg)	Reference
1. Pb	1.00-69000.00	600.00	[2, 22]
2. Cd	0.10-345.00	100.00	[2, 22]
3. Cr	0.05-3950.00	100.00	[2, 23]
4. Hg	<0.01-1,800.00	270.00	[2, 24]
5. Zn	150.00-500.00	1500.00	[2, 24]

The heavy metals present in the soil become contaminants due to the following reasons such as rapid generation via man-made cycle, direct exposure of mine samples due to transportation from mines to environmental locations high metal disposal etc. [4, 26]. The heavy metal balance in the soil can be expressed in the form of the equation shown below.

$$Mtotal=(Mp+Ma+Mf+Mag+Mow+Mip)-(Mcr+Ml) \quad ---------- \quad (1)$$

where “M” is the heavy metal, “p” is the parent material, “a” is the atmospheric deposition, “f ” is the fertilizer sources, “ag” is the agrochemical sources, “ow” are the organic waste sources, “ip” are other inorganic pollutants, “cr” is the crop removal, and “l” is the losses by leaching, volatilization [4, 26].

2.1.1.2. Heavy Metals in Water

Metal composition in surface water like rivers, lakes, ponds? etc. is influenced by the type of soil, rock and water flow. Metals present on the surface of the soil are carried out from its path which ends up in sewage and reservoirs [2]. The rainwater gets contaminated while passing through the atmosphere. Water sources get contaminated by the flow of various industrial effluents into it [2, 26]. Groundwaters are contaminated by landfill leachates, deep well liquid disposal, industrial wastes, etc. Factors such as temperature, pH, living organisms, cation exchange, evaporation, absorption etc., will also influence the metal composition in the water. This has urged various researchers to develop many technological processes of remediation to bring these contaminant levels within the regulatory limit in the environment **in Table 3.**

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Table: 3. Indian & European standards (EU) for heavy metals in soil, food and drinking water

Heavy metal	Soil (µg/kg)	Food (mg/kg)	Water (mg/L)	EU standards soils(µg/g)	Reference
Cd	3.0-6.0	1.50	0.01	3.0	[2, 10]
Cr	-	20.00	0.05	150.0	[2]
Cu	135.0-270.0	30.00	0.05	140.0	[2]
Fe	-	-	0.03	-	[2, 10]
Ni	75.0-150.0	1.50	-	75.0	[2]
Pb	250.0-500.0	2.50	0.10	300.0	[2]
Zn	300.0-600.0	50.00	5.00	300.0	[2, 11]
As	-	1.10	0.05	-	[2]
Mn	-	-	0.10	-	[2, 11]

2.1.1.3. Heavy Metals in the Atmosphere

Heavy metals are released into the atmosphere as gases and particulates by surface erosion and colloid loss. Sources of heavy metals in the atmosphere include mineral dust, sea salt particles, volcanic eruptions and forest fires [2, 26]. Other than these natural sources, heavy metal air pollution can also originate from various industrial processes that involve the formation of dust particles, e.g., metal smelters and cement factories [2, 25]. Volatile metals such as Se, Hg, As and Sb are transmitted in gaseous and particulate form in the atmosphere. Metals such as Cu, Pb and Zn are transported in particulate form [2. 26]. The presence of heavy metals depends upon several site-specific factors such as the quantity and characteristics of the industrial pollutants, environmental sensitivity, potential for environmental release, proximity of these heavy metals in humans and its effect on their health [5].

2.1.1.4. Anthropogenic Sources of Heavy Metals

Heavy metals are released into the environment by various anthropogenic activities. The introduction of heavy metals due to continuous input of pesticides and fertilizer for food production is transported to surface water by infiltration [6, 26]. Zn and Cd are commonly present in phosphate fertilizers and the input of these fertilizers is directly proportional to the concentration of heavy metals [6-7]. In addition to Zn and Cd, pesticides used in agriculture have elements such as Hg, As and Pb. Though metal-based pesticides are no longer in use, the earlier unregulated pesticide application has led to increased accumulation of heavy metals in various environmental matrices [7, 26]. Added to these, various industrial activities such as mining, coal combustion, effluent streams and waste disposal have increased the heavy metal contamination in the environment [2, 8] and the sources are illustrated in Fig. 1.

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The sources of heavy metals are summarized in **Fig. 1**.

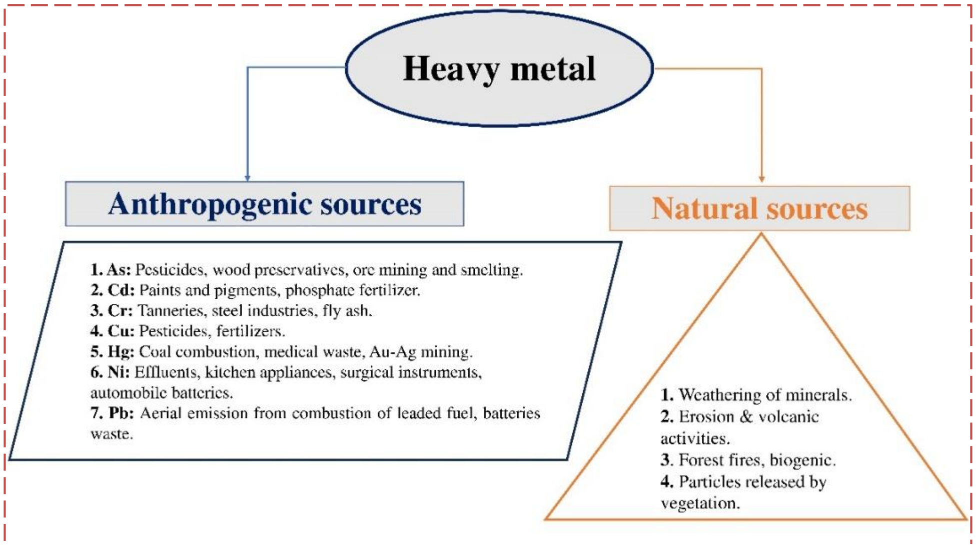

Fig. 1. Anthropogenic and natural sources of heavy metal

2.2. Effects of Heavy Metals in Environment

Heavy metals are notable contaminants because they are toxic, non-biodegradable in the environment and easily accumulated in living organisms. The presence of heavy metals released from various sources is either directly or indirectly released into the environment which affects humans, animals and plants [7, 26]. These heavy metal contaminants in soil were also reported to affect the ecosystem by disturbing the food chain, reducing the food quality due to phytotoxicity and loss of soil fertility etc. [26]. The main pathways of exposure are through inhalation, ingestion and dermal contact. Due increased risk of human exposure to heavy metals leads to serious health implications and environmental deterioration Hence, these metals are categorized as systemic toxicants that can induce adverse health effects in Table 4 in humans that include cardiovascular diseases, developmental abnormalities, neurologic and neurobehavioral disorders, diabetes, hearing loss, hematologic and immunologic disorders and various types of cancer [8, 28].

Table 4. shows a brief list of their adverse health effects and their application

Element	Uses	Adverse health effects	Reference
Cd	Automobile exhaust	Respiratory, cardiovascular and renal effects.	[2, 7]
Cr	Pesticides, detergent.	Mental disturbance, cancer, ulcer and hyperkeratosis.	[2, 7]
Cu	Most uses are based on electrical conductor properties.	Anaemia and other toxicity effects are induced indirectly through interaction with other nutrients.	[2]
Pb	Batteries, alloys.	Neurotoxic.	[2, 8]
Ni	Batteries, Catalysts, Electronics.	Skin allergies, lung fibrosis and diseases of the cardiovascular system.	[2, 8]
Zn	Fertilizers, plastics, pigments.	Abdominal pain, nausea, vomiting, diarrhoea, gastric irritation, headache, irritability, lethargy, anaemias.	[2]

3.0. Remediation Technologies

3.1. Criteria for Selection of Remediation Methods

The selection of acceptable technology is based on the key issue which is the most appropriate technology to be used. A variety of methods are proposed for soil and groundwater remediation ranging from biological to advanced complicated engineering techniques [1, 28]. The technologies may be specific to the site and the contaminant or contaminant class. Considering the concept of sustainable development remediation technologies have to correspond to the principle of low energy and resource use, low waste production, and minimized footprint. In other words, the technologies have to have innovative characteristics [1, 29].Decisive criteria for screening of remediation methods include such as short-term or long-term effectiveness to reach remediation goals, effectiveness of contaminant reduction in site, reduction of contaminant toxicity and cost-effectiveness of remediation [1, 29].

3.2. Classification of Remediation Technologies

3.2.1. Based on Type of Application

Remediation technologies can be divided into two categories such as in-situ and ex-situ remediation methods as well as on-site and off-site technologies [9, 28] expressed in Table 5. On-site remediation is carried out on the contaminated site using in situ or ex-situ approaches. In the off-site treatment process contaminated soil or groundwater is removed from the excavated site (ex situ approach) [2, 28]. It demands the transportation of the contaminated soil or groundwater to the treatment facilities. An in-situ method means that a contaminated substance in soil or groundwater is treated in the place where the contamination is determined. In-situ technologies are used for the treatment of unexcavated soil or unextracted groundwater that remains relatively undisturbed after the cleanup process [29]. The in-situ treatment approach demands detailed information on the physical chemical and biochemical characteristics of pollution as well as on geological hydrogeological and other conditions of the contaminated area. In-situ remediation increases the safety of the personnel involved in the process of treatment and it is more economical and disrupts the site less. Ex-situ technologies are applied to excavated soil or extracted groundwater. Ex-situ treatment can be carried out on or off-site [1, 2]. The following in-situ and ex-situ remediation technologies are used for the reduction of heavy metal contamination levels.

Table 5 Classification of remediation

Classification of Remediation		
a) in-situ and ex-situ technologies	b) in-situ technologies	c) ex-situ technologies
bioremediation, stabilization/solidification, and separation/concentration [10].	soil flushing, electrokinetics, barriers/treatment walls, chemical treatment, soil amendments and phytoremediation [10].	soil washing [10].

3.2.2. Technologies Based on the Used Processes

Technologies based on the used processes include biological separation, physical separation, chemical separation and thermal separation.

In bioremediation technologies, microbiological metabolism is used to transform or degrade soil or groundwater contaminants into harmless substances. The latter can be carbon dioxide, water, fatty acids and some others. Physical and chemical treatment technologies are based on the physical or chemical properties of the contaminants as well as on the contaminated media to chemically convert, separate, or contain the contamination [10-12].

Physical separation treatment involves the phase transfer of metal contaminants from the contaminated media by exploiting differences in certain physical characteristics between metal bearing and soil particles considering their size, density, magnetism and hydrophobic surface properties [11-14]. Physical separation is the concentration technique of metal-bearing particles applied in the mining and mineral processing industry but in soil remediation, it indicates separation of metal particles from soil. The use of physical separation technologies is exactly depended on the sorption capacity of heavy metal chemical forms in soil, their concentration level and soil properties [1, 29]. Thereby these technologies are mostly applied in anthropogenic-influenced industrial areas with high heavy metal concentrations in anthropogenic soils [12, 30]. Ex-situ technologies include soil excavation and following separation based on the physical properties of the particles. These systems are useful in two situations [1]. when the pollutant is present in the form of discrete particles in the soil and another one when the pollutant is concentrated in specific particle-size fractions, as in common for trace elements in the fine fraction of soil [1, 12-13]. The classification of remediation technologies for heavy metal is summarized in Fig. 2.

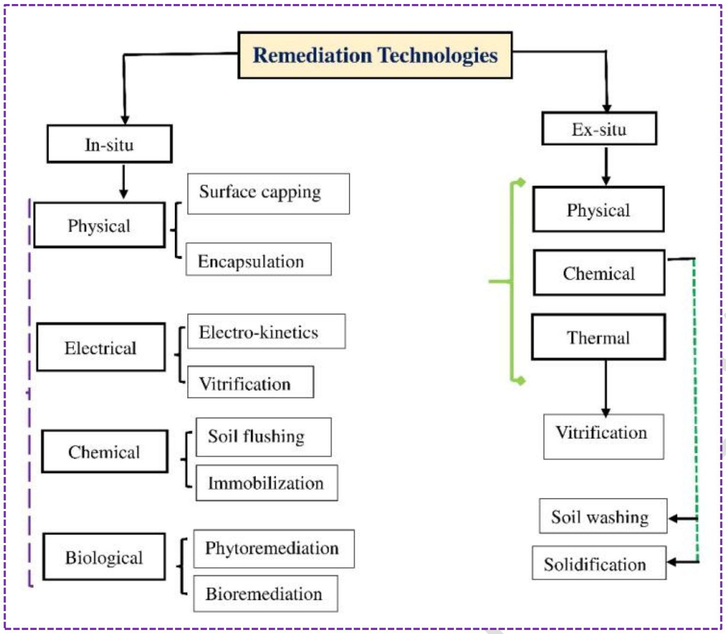

Fig. 2. Classification of remediation technologies

The thermal processes include contaminant properties such as the volatility, burning, decomposing, destructing and some others using heat. Some classifications add the vitrification to the S/S (stabilization/solidification) sub-category because of the similar result at the end of the vitrification process [13].

However, the process is different in comparison with the classical S/S technique. Vitrification involves the insertion of electrodes into the soil which must be able to carry a current and then solidify as it cools. In-situ vitrification employs electrical power to heat and melt contaminated soil [10, 30]. Organic contaminants are destroyed through pyrolysis while volatile metals may evolve in off-gases, necessitating off-gas treatment. Vitrification is applicable for soils contaminated with heavy metals, organic contaminants with high sorption coefficients and radioactive materials as well [13]. However, effectiveness is reduced in soils with high organic matter, high moisture content or soils containing large metallic objects (e.g., pipes or drums). As an alternative, in-situ soil heating decontaminates soils through vaporization, steam distillation and stripping and may be performed through power line frequency heating or radiofrequency heating [13]. During the vitrification process, toxic gases can be also produced. The full-scale application exists for As, Pb and Cr contaminated soils. Mixed wastes can also be treated using vitrification. High clay and moisture contents and debris can affect the efficiency of the process. of a large volume [13, 30].

3.3. Modern technologies for the remediation of heavy metals

Modern technologies for the remediation of heavy metals are classified into four types such as electro-kinetic remediation, chemical remediation, bioremediation and phytoremediation.

3.3.1. Electro-kinetic Remediation

Electro-kinetic (EK) remediation is an in-situ process in which an electrical field is created in a soil matrix by applying a low-voltage direct current (DC) to electrodes placed in the soil [14]. As a result of the application of this electric field, heavy metal contaminants may be mobilized, concentrated at the electrodes and extracted from the soil [14].Electro-kinetic remediation field setup with a well is constructed, filling electrolyte, inserting anode (+) & cathode (-), applying electric field, electro-osmosis and electrolysis of water, desorption of metal ions by acidic front, electro-migration and precipitation or dissolution of metal ions and pumping and storing of exhausted electrolytes [14, 29] in Fig. 3.

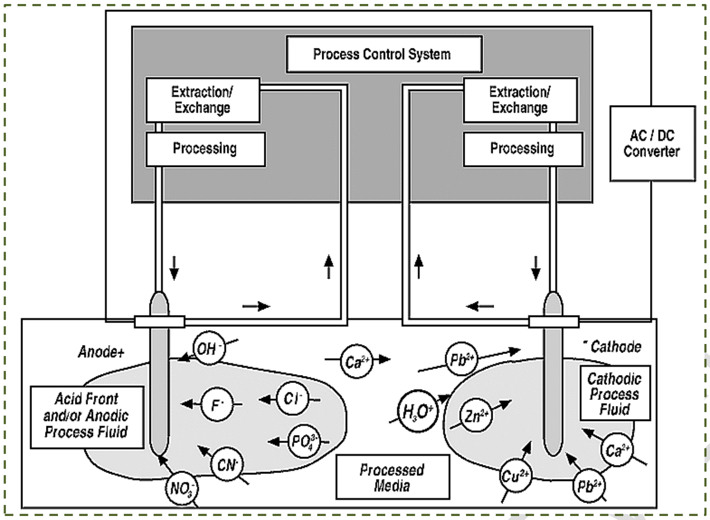

Fig. 3. A Flow diagram of the electro-kinetic remediation process.

The electrodes can be placed in either a vertical or horizontal array. When DC is applied to the electrodes an electrical field develops between the anodes and cathodes. The application of the electric field has several effects on the soil, water and contaminants [15, 30]. These effects include electro-migration, electro-osmosis, changes in the pH of the system and electrophoresis [16, 31]. Electro-migration refers to the movement of cations and anions under the influence of the electrical field. These ions concentrate in the solutions near the electrodes or may undergo reactions at the electrodes which may plate the metals onto the electrodes or liberate gaseous compounds [16, 30]. Electro-osmosis is the movement of liquid-containing ions relative to a stationary charged surface or a bulk transport of water which flows through the soil as a result of the applied electrical field. pH changes occur under the influence of the current as a result of electrolysis reactions at the electrodes. Oxidation of water occurs at the anode and generates hydrogen (H+) ions; H+ ions generate an acid front, which migrates to the cathode [16].

Electrophoresis is the movement of a charged particle in a liquid, caused by an electric field In contrast, the reduction of water occurs at the cathode and generates hydroxyl (OH-) ions in equation 3 which migrate as a base front towards the anode [16].

$2H_2O - 4e^- \longrightarrow O_2 + 4H^+$ (Equation 2)
$4H_2O + 4e^- \rightarrow 2H_2 + 4OH^-$ (Equation 3)

The transport of the H+ ions is approximately two times faster than the OH- ions [16]. Thus, the acid front moves at a greater rate than the base front [17, 31]. Unless the transport of the proton (H+ ion) is retarded by the soil buffering capacity, the soil between the electrodes will be acidified. This acidification results in the solubilization of contaminants due to desorption and dissolution of species from the soil [17]. Once contaminants are present in ionic form in the soil pore fluid, they migrate to the electrode opposite in polarity under the applied electric field or via electro-osmosis, leading to their extraction from the soil at the electrodes. Extraction and removal of the contaminants may be accomplished by electro-deposition, precipitation or co-precipitation at the electrode [17, 31]. EK remediation heavy metal extraction rate and efficiency are dependent upon many subsurface characteristics such as soil type and grain size, contaminant concentration, ionic mobility, total ionic concentration, types of contaminant species and their solubility, etc. Additional complications with the application of EK remediation can arise from the presence of organic contaminants and possibly the organic material in the soil [14].

Table 6. Advantages and disadvantages of electro-kinetic remediation

Causes	Description
Advantages	In-situ and passive technique. Solar-driven and low-cost. Reduction of soil erosion and dispersal of dust and contaminants in soil. Easy harvesting of the plants [14].
Disadvantages	Limited practical experience and therefore not accepted by many regulatory agencies. The toxicity and bioavailability of degradation products remain largely unknown. Treatment is slower than the traditional physico-chemical

	techniques. Contamination may spread through the food chain if accumulator plants are ingested by animals. The area to be decontaminated must be large enough to allow the application of cultivation techniques [14].

The advantages and disadvantages of electro-kinetic remediation are illustrated in Table 6.

3.3.2. Chemical Remediation

Chemical processes include reduction of the bioavailability/mobility of heavy metals as well as other contaminants upon chemical reactions with specific reagents. Technologies for heavy metal remediation is based on precipitation, oxidation-reduction and organic-metal complexion reactions that are well-known and widely studied techniques in chemistry, for example, chemical technology, environmental, analytical and radio-analytical chemistry [31]. Theuse of chemical processes have a drawback conjugated with the possible side effects on the environment that has to be taken into consideration in the choice of chemical reagents in treatment process. Dissolved heavy metals in groundwater can be precipitated out of the solution in various insoluble compounds [1, 31]. The most common heavy metal precipitates are hydroxides, sulphides, carbonates, phosphates, oxalates and some others. Solubility product constant (Ksp) of the insoluble compound characterizes the perfection of the precipitation reaction.Using, phosphate-containing precipitating reagents U, Pu, and heavy metals (Cu, Cd, Pb and Zn) are transformed into low-solubility phosphate mineral phase in which they are not bioavailable [1, 31]. Effectiveness is provided due to the extremely low Ksp value, for example, the solubility product constant of U-phosphate (autunite) is Ksp=10.0-49.0 and of Pb apatite (pyromorphite) is Ksp=10.0-80.0 [18]. The lower the solubility product constant, the moreinsoluble the precipitated compound is. Precipitation of insoluble substances causes two other processes such as co-precipitation and sorption that occur simultaneously with the formation of precipitates [30]. Thereby heavy metals that stand in aqueous media in minor and microconcentrations are also concentrated in the solid phase. Precipitation, co-precipitation and sorptionare spontaneous processes with the tendency of disorder (entropy) decreasing, i.e., contaminantsarrive in the solid phase [1]. Chemical treatment by reductive as well as oxidative mechanisms may be used to detoxify or decrease the mobility of metal contaminants [19]. This method is commonly used for wastewater treatment [19, 31]. Oxidation reactions which detoxify, precipitate or solubilize metals involve the addition of potassium permanganate, hydrogen peroxide and hypochlorite or chlorine gas. Neutralization reactions are performed to adjust the pH of acidic or basic soils. Reduction reactions are induced through the addition of alkali metals such as sodium, sulphur dioxide, sulphite salts and ferrous sulphate [20]. Sometimes chemical treatment is used to pretreat the soil for solidification or other treatments. For example, chemical reduction of Cr (VI) is performed during solidification/stabilization (S/S). Oxidation is less commonly used with S/S. These reactions are, however, not specific and there is a risk of converting other metals into more toxic or mobile forms. Arsenic is most applicable for chemical oxidation since As (V) is less toxic than As (III) [21]. Co-precipitation of high concentrations of As (V) and Fe (III) forms $FeAsO_4$ while low concentrations of As (V) co-precipitate with $Fe(OH)_2$ with high concentrations of Fe (III) to form arsenic ferrihydrite, a product that is resistant to acid and neutral leaching. Mercury, lead, selenium and silver are also applicable for reduction. These chemical treatments can be performed in situ by injection into the groundwater but have the potential to introduce further contamination [20, 32]. Advantages are fast and easy applicability, most of the metals can be removed. Relatively low costs and covers a broad spectrumof inorganic pollutants [14]. On the other hand, the disadvantages are a large amount of sludge is produced and disposal problems. Temporary solutions and permanent monitoring are necessary. It has the potential to introduce further contamination [14].

3.3.3. Bioremediation Techniques

Biological technologies for remediation take advantage of the pathways developed by micro-organisms to protect themselves from oil products and metals. Common protection mechanisms include oxidation, reduction, sorption and methylation [32]. At present biotechnologies that incorporate these mechanisms are in an advanced stage of development for the remediation of organic compounds but experience is limited for inorganic contaminants [14-16] are explained in Table 7.

Table 7. Advantagesand disadvantages of bioremediation techniques

Advantages	Disadvantages
Bioremediation takes advantage of the natural ability of microorganisms to extract chemicals from water, soil and sediment using energy from sunlight. It is a cost-effective technique compared to other physiochemical treatment methods. Less energy is required as compared to other technologies. Often little to no residual treatment [14].	The process of bioremediation is slow. Treatment time is typically longer than that of other remediation technologies. It does not remove all quantities of contaminants from the polluted site. Bioremediation is not useful for the treatment of inorganic contaminants. For in-situ remediation, the site must have soil [14].

3.3.4. Phytoremediation

Phytoremediation is a bioremediation process that uses various types of plants to remove, transfer, stabilize or destroy contaminants in the soil and groundwater. There are several different types of phytoremediation mechanisms [22].

Table 8. Mechanisms of phytoremediation

Types	Mechanisms
I. Rhizosphere biodegradation	In this process, the plant releases natural substances through its roots, supplying nutrients to microorganisms in the soil. The microorganisms enhance biological degradation [22].
II. Phyto-stabilization	Chemical compounds produced by the plant immobilize

	contaminants rather than degrade them [22].
III. Phyto-accumulation also called phytoextraction	Plant roots absorb the contaminants along with other nutrients and water. The contaminant mass is not destroyed but ends up in the plant shoots and leaves. This method is used primarily for wastes containing metals[22].
IV. Hydroponic Systems for Treating Water Streams (Rhizofiltration)	Rhizofiltration is similar to phyto-accumulation but the plants used for cleanup are raised in greenhouses with their roots in water. This system can be used for ex-situ groundwater treatment. Groundwater is pumped to the surface to irrigate these plants. Hydroponic systems utilize an artificial soil medium such as sand mixed with perlite or vermiculite. As the roots become saturated with contaminants, they are harvested and disposed of [22].
V. Phyto-volatilization	Plants take up water containing organic contaminants and release the contaminants into the air through their leaves[22].
VI. Phyto-degradation	Plants metabolize and destroy contaminants within plant tissues [22].
VII. Hydraulic Control	Trees are indirectly remediated by controlling groundwater movement. Trees act as natural pumps when their roots reach down towards the water table and establish a dense root mass that takes up large quantities of water. A poplar tree, for example, pulls out of the ground 30.00 gallons of water per day and cottonwood can absorb up to 350.00 gallons per day [22].

The main advantages are in-situ and ex-situ, amenable to a variety of organic and inorganic compounds, suited to remediation of large areas of soil, costs are effective compared to conventional methods, easy to implement and maintain and accepted by the public, conserves natural resources and is environmentally friendly [14, 32]. In another way, the disadvantage is takes several years to remediate a contaminated site, not as effective for sites with high contaminant concentrations, is slower than conventional methods and the toxicity and bioavailability of biodegradation products are not known. It doesn’t work in the winter, disposal of contaminants accumulated in plants after harvesting-pollution again [14, 33]. These above technologies without bioremediation are considered to be inefficient because of expensive cost. Nowadays it is the most applicable method for the remediation of heavy metals.

3.4. Bioremediation Method

Bioremediation can be defined as the ability of certain biomolecules or types of biomasses to bind and concentrate selected ions or other molecules present in aqueous solutions. Bioremediation using microorganisms shows great potential for future development due to its environmental compatibility and possible cost-effectiveness [23]. A wide range of microorganisms, including bacteria, fungi, yeasts and algae can act as biologically active methylators which canat least modify toxic species [24, 34]. Many microbial detoxification processes involve theefflux or exclusion of metal ions from the cell which in some cases can result in highlocal concentrations of metals at the cell surface where they can react with biogenic ligands and precipitate [25]. Although microorganisms cannot destroy metals, they can alter their chemical properties via a surprising array of mechanisms [23, 35]. In bioremediation by micro-organisms typically employing one type of organism or a consortium of microorganisms, highly toxic chemicals are converted into less toxic chemicals by biological means. The technology makes use of the metabolic potential of microorganisms to clean up contaminated environments and has been proposed as an attractive alternative owing to its lower cost and higher efficiency compared with other physicochemical methodologies. Microorganisms can decompose or transform hazardous substances into less toxic metabolites or degrade them to nontoxic end products [36]. Microorganisms can also survive in contaminated habitats because they are metabolically able to exploit contaminants as potential energy sources [23]. Metals are used in bioremediation processes employing microorganisms and chromium, copper, cadmium and lead together account for 70.0% of applications, although nickel and zinc are also used [26, 37]. Other metals that are used to a lesser extent include arsenic and mercury [27, 38].

3.5. Types of Micro-organisms Used in Bioremediation

Several studies have shown that many organisms such as prokaryotes and eukaryotes have a natural capacity to biosorb toxic heavy metal ions. Examples of microorganisms studied and strategically used in bioremediation treatments for heavy metals include the following Table 9 [26, 39]. Recent studies have demonstrated the ability of certain fungi such as Aspergillus, Penicillium and some yeasts such as S. cerevisiae to remove heavy metals from certain environments. The species Escherichia coli, Bacillus subtilis and Staphylococcus aureus have also been used for the removal of heavy metals from water bodies [23, 40].

Table 9. shows the ability of typical microorganisms (algae, bacteria, fungi and yeasts) to remove heavy metals from certain environments.

Microorganism	Type	Metal	Reference
Algae	Ascophyllum nodosu., Chlorella pyrendoidosa. Spirogyra spp. and Spirullina spp.	Pb, Cu, Cd, Zn. U. Cr, Cu, Fe, Mn, Zn.	[26]
Bacteria	Pseudomonas veroni. Sporosarcina ginsengisoli.	Cd, Zn, Cu. As	[26]
Fungi	Aspergillus, Mucor, Penicillium and Rhizopus. Aspergillus niger, Aspergillus foetidus and Penicillium simplicissimum.	Cd, Cu, Fe. Ni, Co, Mo, V, Mn, Fe, W, Zn.	[26]
Yeast	Candida tropicalis. Saccharomyces cerevisiae	Cd, Cr, Cu, Ni, Zn. Cr, Ni, Cu, Zn.	[26]

4.0. Chemistry of Micro-organism

The structure of the cell wall of a microorganism contains various macromolecules such as polysaccharides and proteins with a high number of charged functional groups including carboxyl, imidazole, sulfhydryl, thioether, phenol, carbonyl, amide, ester sulfate, amino and hydroxyl group [3]. The positively charged metal present in the solution gravitates toward these functional groups and adsorption occurs [27, 40]. The form in which microorganisms are cultivated can influence the cell wall composition and this can be exploited to improve theadsorption capacity of the microorganisms. Bacteria can remove heavy metals fromwastewater via functional groups such as ketones, aldehydes and carboxyl groups present in their cell walls and thereby produce less chemical sludge [27]. Both gram-positive and gram-negative bacteria are used for the uptake of metals. Green, red and brown algae are also used as biosorbents. Some functional presents in bacteria such as uronic acid of carboxyl groups and sulfate groups, xylans, galactans and alginic acid are capable of performing ion exchange. The advantage of using algae as biosorbents is that they generally do not produce toxic substances, unlike other microorganisms such as bacteria or fungi [3]. Fungi and yeasts are also used for the adsorption [3, 40]. The biggest advantage of fungi is highly variable ranging in size from mushrooms to microscopic moulds. They are easy to grow and produce asubstantial biomass. The cell walls of fungi are rich in polysaccharides and glycoproteinswhich contain, for instance, amine, imidazole, phosphate, sulfate, sulfhydryl and hydroxylgroups [27]. However, the cell walls of yeasts contain a micro-fibrillar structure composedof more than 90.0 % polysaccharides. The main groups present in these walls are amine,hydroxide, carboxyl, sulfate and phosphate groups [3, 41].

4.1. Mechanisms Associated with Bioremediation by Micro-organisms

Bioremediation can be separated into two categories, bio-sorption and bio-accumulation. How microorganisms interact with heavy metal ions is partially dependent on whether they are eukaryotes or prokaryotes, wherein eukaryotes are more sensitive to metal toxicity than prokaryotes [3, 28]. The possible modes of interaction are active extrusion of metal, intracellular chelation (in eukaryotes) by various metal-binding peptides and transformation into other chemical species with reduced toxicity [28]. For bioremediation to be effective, microorganisms must enzymatically attack the pollutants and convert them to harmless products. Bacteria and higher organisms have developed mechanisms associated with resistance to toxic metals and rendering them innocuous [28, 42]. Several microbes, including aerobes, anaerobes and fungi are involved in the enzymatic degradation process. Most bioremediation systems are run under aerobic conditions but anaerobic conditions make it possible for microbial organisms to degrade otherwise recalcitrant molecules [3, 28]. **Fig. 4.** shows the major groups of microorganisms commonly used for bioremediation of metals which include bacteria, microalgae, fungi and yeast.

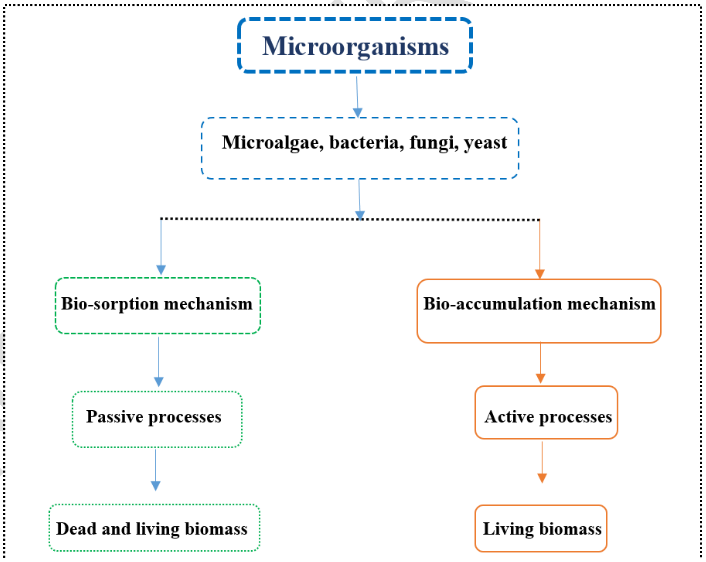

Fig. 4. Microorganisms employed in the bioremediation and processes/mechanisms involved in the case of dead and living biomass**.**

Both living and dead biomass can occur for biosorption because it is independent of cell metabolism. On the other hand, bioaccumulation includes both intra- and extracellular processes where passive uptake plays only a limited and not very well-defined role. Therefore, living biomass can only occur for bioaccumulation [29].

4.2. Mechanism of Bio-accumulation

The process of metal accumulation on the cell surface is dependent on the metabolic activity of the microorganism as well as the characteristics of the cell surface and it is known as bioaccumulation [29].The pathway via which the metal binds to a specific site of the biomass is of great importance to the efficiency of a bioremediation process [3, 43]. For example, the ingestion of sediments by microorganisms is considered a principal route of exposure to metals although free metal ions in sediment pore waters are generally considered to be the most bioavailable form of metals. Thus, metal accumulation is affected by the feeding behaviour of microorganisms.After the ingestion of heavy metals, a process of metal excretion and/or detoxification begins toavoid potential toxic effects. However, microorganisms will not suffer the toxic effects of thepresence of metals when they are stored in detoxified forms [30, 43]. Moreover, the metal–biomassinteraction is dependent on the type of metal that can bind to oxygen-containing or Sulphar and nitrogen-containing ligands. Although this may be a simple overview of the mechanisms involved, it can act as a starting point for proposing new approaches related to the efficiency of metaluptake by microorganisms [44]. Otherwise, microorganisms can synthesize metal-binding proteins such as MTs or PCs, and the proteins are strongly related to the capacity of metal adsorption, accumulation and resistance [3, 30]. In particular, metalloproteins are a large group of these proteins which play an important role mainly in regulating the number of metals within the cells.

Metal binding proteins present outside of the cell membrane attract metal ions that exist in the solution and assist the transport to the cytosol where metallochaperones (specialized protein chelators) transfer metals to the appropriate receptor protein [3, 30]. The binding sites of the metal binding proteins have been improved to other proteins, such as heterologous metalloproteins by using genetic techniques. The technique of changing the proteins on the cell surface, into heterogeneous ones by using recombinant DNA has emerged as a novel approach to enhance the capacity of adsorption. Both bacteria and yeasts have been investigated for this purpose [23, 30].

4.3. Mechanism of Bio-sorption

Bio-sorption is a passive adsorption mechanism that is fast and reversible. The metals are retained using physicochemical interaction such as ion exchange, adsorption, complexion, precipitation and crystallization between the metal and the functional groups present on the cell surface. Several factors can affect the biosorption of metals such as pH, ionic strength, biomass concentration, temperature, particle size and presence of other ions in the solution [3, 30] in Fig. 5.

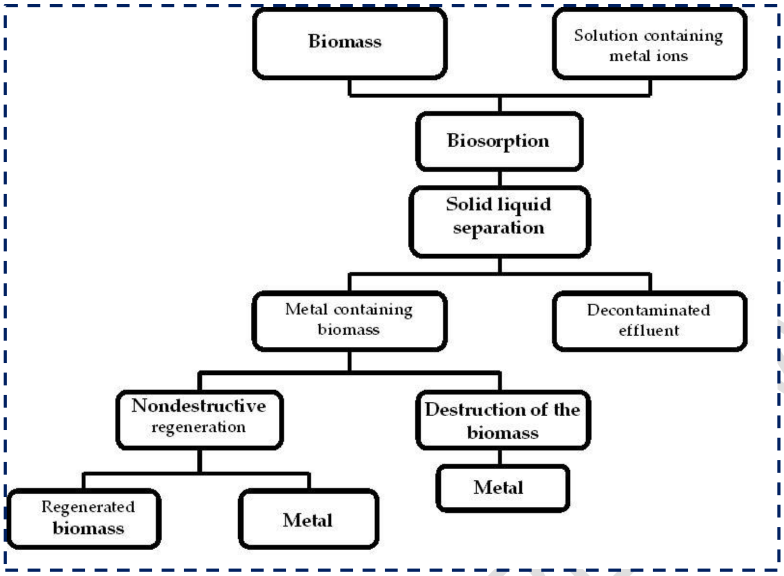

Fig. 5. Schematic diagram of bio-sorption process.

Micro-organisms can retain metals; it is necessary to define and to understand the functional groups responsible for the adsorption phenomenon [30]. Bio-sorption bydead biomass or by some molecules or their active groups is a passive process based mainly on the affinity between the biosorbent and the sorbate. In this case, the metal is sequestered by chemical sites naturally present in the biomass [30, 45]. The diagram in Fig. 5 illustrates the main steps in this process. In most cases, the biosorption process is rapid and takes place under normal temperature and pressure. After the process of phase separation, a biomass “charged” with metal ions and an effluent free of contamination is obtained. Two paths can be followed to deal with the “contaminated” biomass, the one of greatest interest being biosorbent regeneration and metal recovery [30, 46]. This process is the most attractive becausebiomass can be used for the removal of other metal species from other contaminated effluents.The other option is the destruction of the biomass which offers no possibility of reuse [30, 47]. Ion exchange is an important concept in biosorption because it explains many of the observations made during heavy metal uptake experiments [30]. In this context, the term ion exchange does not explicitly identify the mechanism of heavy metal binding to biomass and electrostatic or London–van der Waals forces should be considered as the precise mechanism of chemical binding such as ionic and covalent bonds [30]. Fig. 6 provides a schematic representation of an ion exchange mechanism for a biosorbent material where “Me” represents a metal with valence +2.

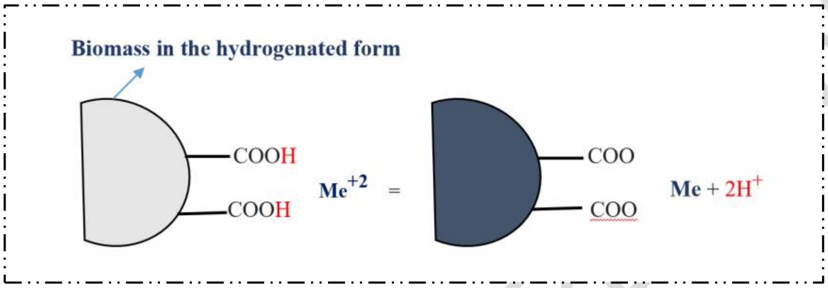

Fig. 6. Schematic diagram of an ion exchange mechanism.

After the metal removal from aqueous solutions by the biomass, the recovery of the metal is an important issue. This can be achieved through a metal desorption process [30, 48].

Table 10. Comparison between Bio-sorption and Bio-accumulation

Characteristics	Bio-sorption	Bioaccumulation	Reference
Cost	Usually, low. Biomass can be obtained from industrial waste. The cost is associated mostly with the transportation and production of biosorbents.	Usually, high. The process occurs in the presence of living cells that have to be supported.	[34]
pH	The solution pH strongly affects the sorption capacity of heavy metals. However, the process can occur within a wide pH range.	Significant changes in pH can strongly affect living cells.	[34, 49]
Selectivity	Poor. However, this can be increased by modification/biomass transformation.	Better than in the case of bio-sorption.	[34]
Rate of removal	Most mechanisms occur at a fast rate.	Slower rate than in the case of bio-sorption because intercellular accumulation takes a long time.	[34]

Regeneration and reuse	Biosorbents can be regenerated and reused in many cycles.	Reuse is limited due to intercellular accumulation.	[34]
Recovery of metals	With an adequate eluent, the recovery of heavy metals is possible.	Even, if possible, biomass cannot be used for other purposes.	[34]
Energy demand	Usually, low.	Energy is required for cell growth.	[34]

Table 11. Comparison between bioremediation and phytoremediation process

Bioremediation	Phytoremediation
Use of naturally occurring deliberately introduced micro-organisms to consume and break down environmental pollutants to clean a polluted site. Method of removing contaminants from ecosystems and used mainly for organic pollutants. Mainly use microbes. Can be either in-situ or ex-situ. More ecologically friendly.[32, 50].	A process of decontaminating soil or water by using plants and trees to absorb or break down pollutants. A type of bioremediation and amenable to a variety of organic and inorganic compounds. Depends on plants. Mainly an in-situ process. Cost-friendly.[32, 50].

Table 12 Comparison between bio-remediation and chemical remediation

Bioremediation method	Chemical remediation method
It is a cost-effective technique compared to other physiochemical treatment methods. Bioremediation is not useful for the treatment of inorganic contaminants. Often little to no residual treatment. More ecologically friendly[32].	It is a costly method. Covers a broad spectrum of inorganic pollutants. Large amounts of sludge are produced. It has the potential to introduce further contamination[32].

Table 13 Comparison between bioremediation and electro-kinetic remediation method

Bioremediation method	Electro-kinetic remediation method
In-situ and ex-situ. Both active and passive techniques. Fast process. It is eco-friendly.	In-situ. Passive technique. Treatment is slower than the traditional physico-chemical techniques. Contamination may spread through the food chain. The toxicity and

Pollutants can be determined. It can be applied anywhere. High acceptance by the public. [33, 51].	bioavailability of degradation products remain largely unknown. The area to be decontaminated must be large enough to allow the application of cultivation techniques. Limited practical experience and therefore not accepted by many regulatory agencies.[33, 52].

Different types of comparison of technologies are compensated in Table 11 to Table 14.

Conclusion

Effective cleanup techniques are required because heavy metal pollution of the environment is a global issue. For polluted sites to be effectively cleaned up, a variety of remediation approaches are employed. We evaluated the efficacy of many remediation strategies that are frequently employed to clean up contaminated sites in this review. Heavy metal cleanup is now an intricate and challenging task due to the associated costs and technical challenges. For the cleanup of heavy metal-polluted sites, the majority of conventional remediation approaches are ineffective. Although physically remediating polluted soil can remove heavy metals completely, these approaches are expensive and disruptive. These techniques are limited to tiny soil areas. Chemical cleanup techniques are quick, easy to use, widely accepted and reasonably priced. But because they can't discharge more pollutants into the environment, these cleanup techniques aren't environmentally benign. Large-scale heavy metal cleanup is made possible by bioremediation, a safe, least harmful, environmentally friendly and economical remediation method. However, more basic and field-based research is still required in this area. The growth environment, plant tolerance to metals and soil solubility of metals all affect how effective phytoremediation is. Although phytoextraction takes longer, it works well for low-to-moderate concentrations of heavy metals. It is possible to combine bioremediation with several other conventional remediation methods to good effect. Plants benefit from the protection that soil microbial interaction provides from heavy metal toxicity and increased absorption of metals

Comment [a7]: The review work

through the production of different compounds. An interdisciplinary approach integrating the work of soil chemists, plant biologists, geneticists, microbiologists and environmental engineers is necessary for effective bioremediation as the process typically deals with heterogeneous and multiphasic environments, such as soils. The study's findings indicate that the primary determinants of the suitability and choice of remediation methods are the associated expenses, the duration needed, the efficacy over an extended period, the degree of widespread acceptance and the site's high and multi-metal contamination.

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