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The definitive publisher version is available online at  
<https://doi.org/10.1016/j.psep.2022.04.011>

# **Mmetals in e-waste: Occurrence, fate, impacts and remediation technologies**

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## Abstract

E-waste is generated from the discarded electronic product. The generation of e-waste has increased significantly in the recent decade. Globally, the increased rate of e-waste generation is almost 2 Mt per year. It is estimated that about 74 M tonnes of e-waste will be produced in 2030. In 2019 about 17.4% of the e-waste globally generated was properly disposed of or recycled. The fate of the remaining 82.6% was not documented, which could be dumped without proper treatment or recycling. Therefore, e-waste can be a significant threat to the environment. Toxic heavy metals (HMs) (e.g., lead, mercury, nickel, and cadmium) are released to the environment from the e-waste and eventually enter into soil, sediment, groundwater, and surface water. The release of toxic metals in the environment causes adverse effects on human health, aquatic animals, and plants. Therefore, the proper management of e-waste is essential and becomes a major concern in the world. In this regard, this review provides a comprehensive summary of the occurrence, fate, and remediation of HMs generated from e-waste. The literature survey revealed that household electrical appliances are the primary source of e-waste, comprising approximately 50% of the overall production of e-waste. Among other remediation technologies (e.g., such as coagulation, media filtration), biological and hybrid treatment (e.g., the combination of biological, physical, and chemical processes) processes shows relatively high removal efficiency and possesses multiple advantages over other remediation technologies. However, this review concludes by providing an outlook of the future aspects of current remediation technologies for e-waste management.

## 1. Introduction

E-wastes are the wastes of the discarded electrical appliances or electronic products like refrigerators, washing machine, fan, television, air conditioners, cell phones and computers [1]. The global production of e-waste has become one of the significant issues due to the considerable demands of electronic products in human society. Among different electrical consumables, the rapid growth of computing and communication devices are mostly responsible for the global boom of e-waste production [2]. Other reasons for the massive generation of e-waste include (i) rapid advancement of information and communication industries, (ii) versatility of electronic equipment, (iii) rapid technological growth and modern innovations, and (iv) the declining flow of prices of electronic devices. It is estimated that the production of e-waste will increase by ~50% in 2030 (from 33.8 --- in 2010 to 74.7 – in 2030) (**Figure 1a**). E-waste mainly contains metals (60%), plastics (15%) and metal-plastic mixture (5%) [3, 4]. When the amount of toxic heavy metals (HMs) such as lead (Pb), mercury (Hg), arsenic (As), cadmium (Cd), selenium (Se), and chromium (Cr) exceeds the permissible level, then the e-waste is considered as hazardous [5]. Waste household equipment (about 45%), information and communications technology equipment (33.9%), consumer electronics (13.7%) are the major contributor to e-waste [6, 7]. Eight HMs (Pb, Cr, As, Zn, Cd, Cu, Hg, and Ni) are the most extensive and harmful for the environment according to the United States Environmental Protection Agency (USEPA) [8, 9]

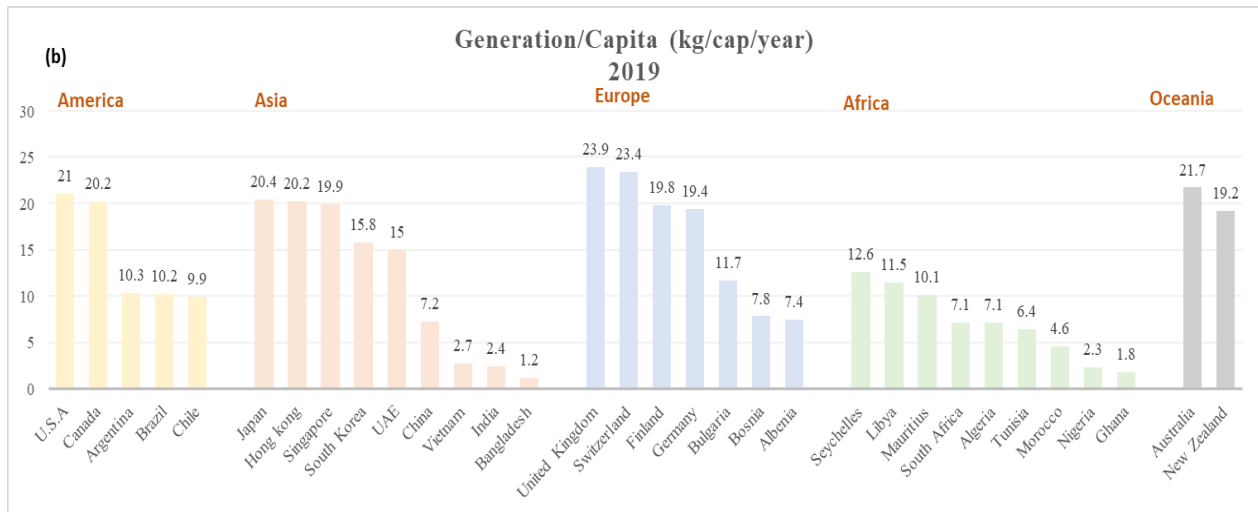
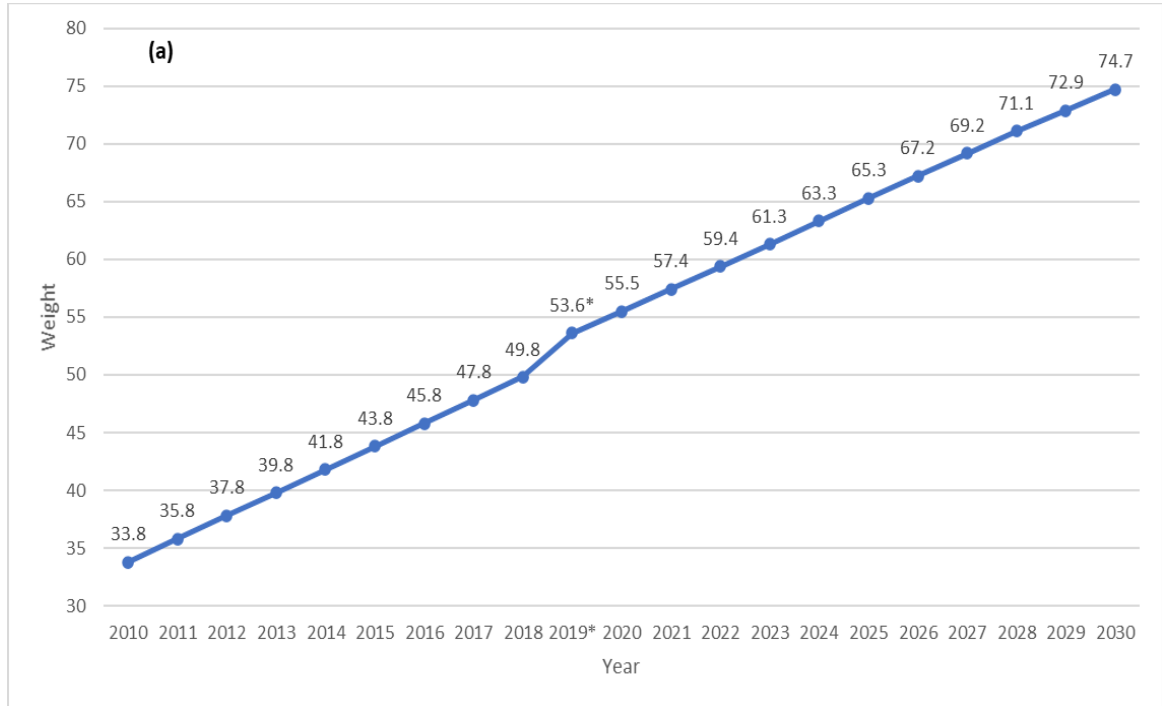
The management of e-waste is one of the world's significant challenges as most e-wastes contain HMs and toxic substances. The amount of e-waste represents a small portion of the overall municipal solid waste. However, the generation of e-waste depends on the consumption of e-items per capita and population [10]. Due to rapid industrialization, and the availability of high technology results in more e-waste production. According to the International Solid Waste Association, in 2019, approximately 53.6 M tonnes of e-waste was generated worldwide (**Figure 1a**). In 2019, the generation of e-waste was 24.9 M tones (2.5 kg per capita) for Asia, 13.1 M tones

(13.3 kg per capita) for America and 12 M tones (16.2 Kg per capita) for Europe (**Figure 1b**). America and Europe are the second and third most significant contribution to e-waste in the world. In Asia, Japan, China and Singapore are the major producers of e-waste. Whereas in Europe U.K, Switzerland, Finland and Germany produce an almost similar amount of e-waste (Figure 1b). Europe recycled the highest amount of e-waste 5.1 M tonnes (42.5%). On the other hand, most of the countries had a lower recycling rate of e-waste compared to the generation of e-waste. According to the statistics recycling portion of Asia 11.7%, while in America and Oceania is 9.4% and 8.8% respectively [11]. However, the actual amount may vary from region to region according to their annual income, management systems, policy and environments. By the next ten years, the units of obsolete PC in developing countries will increase by 400-700 M and in developed countries 200-300 M[12]. According to Silicon Valley Toxics Coalition research in between 1994 to 2003, about 500 M PCs reached out of working condition and became e-waste[13]. More or less, 500 M PCs contain a massive amount of toxic HMs (e.g., approximately 718,000 tonnes of Pb, 1363 tonnes of Cd, and 287 tonnes of Hg) [3]. Hence, e-waste generation and its management (e.g., treatment and disposal) have become a great concern to the waste management professionals, government and non-governmental organizations, municipalities and certain manufacturers [14].

HMs from waste electronic and electronic equipment (WEEE) are leached into the soil, groundwater and surface water. Therefore, it may cause severe environmental hazards and human health issues [15, 16]. For example, a higher concentration of Cr causes chronic disease, kidney damage, bones and respiratory system diseases [17]. The neurodevelopment of young people can also be affected by Cr uptake [18]. Diseases of heart rhythm, breathing and the risk of bladder cancer can be caused by As [19]. Diseases like cancer and skin allergies are caused by Ni (electron gun of CRT (Cathode-ray tube) and in Ni-Cd batteries) [20]. Children are so much vulnerable to the exposure of HMs mainly Pb. Exposure to even low Pb concentration can create different neurological problems such as delayed development, inattentiveness, irritability, hyperactivity, stunted growth and brain damage [21, 22]. The toxic effects of HM exposure have created worldwide attention to comply with the HMs content (allowable limits) in the soil and crops [23]. Hence, proper treatment of e-waste is necessary and has become a crucial topic in solid waste management [24]. Various treatment technologies of e-waste to recover metals are used, such as physical method, thermochemical method, pyro-metallurgical method, hydrometallurgical method, bio-metallurgical and a combined method.

Additionally, it is also necessary to deploy efficient and site-specific remediation methods, which will feasibly and efficiently remediate HM(loid)s contaminated soils and surface water. For instance, during the last two decades, different soil remediation methods (e.g., bioleaching, phytoremediation, mycoremediation, vitrification, earth-swap, soil flushing, solidification) have been developed [25-30]. The method's main objectives are to decline the total bioavailable amount of HM(loid)s in soils and water, and their frequent availability in the food cycle [31]. Physical, chemical and biological methods or their combination (e.g.chemical, biological) are the conventional methods to remediate HM(loid)s from contaminated soils sites.

Many investigations have been carried out on the generation of e-waste, potential recovery, environmental problems and remediation technologies. Up to now, there is no such kind of review that could address all of the issues together systematically. Henceforth, this review provides information on the overall scenario on the e-waste production, major sources and fate of e-waste. Besides, various impacts of HMs on both the environment and human are discussed. Furthermore, the current remediation scheme of HMs generated from e-waste, their merits, drawbacks and gaps also reviewed critically. Future research directions and outlooks have also been suggested.



**Figure 1.** Global e-waste generation scenario over the period. (a) Global quantity of e-waste generation (2020-2030 are estimated) (b) e-waste generation per capita in 2019 in different countries [11, 32, 33].

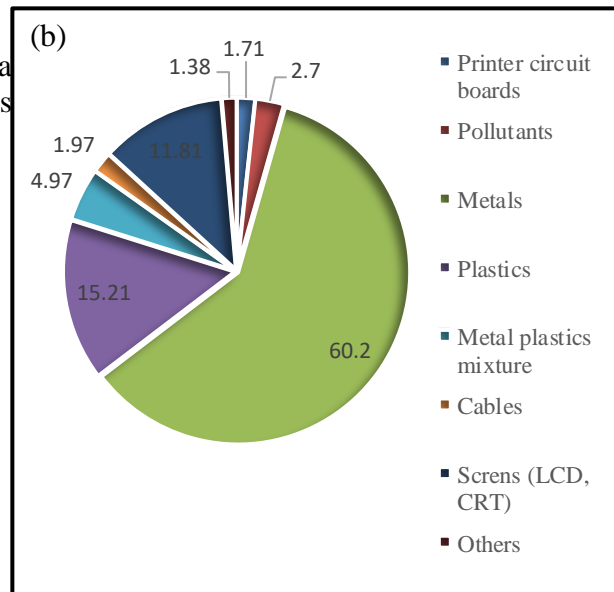
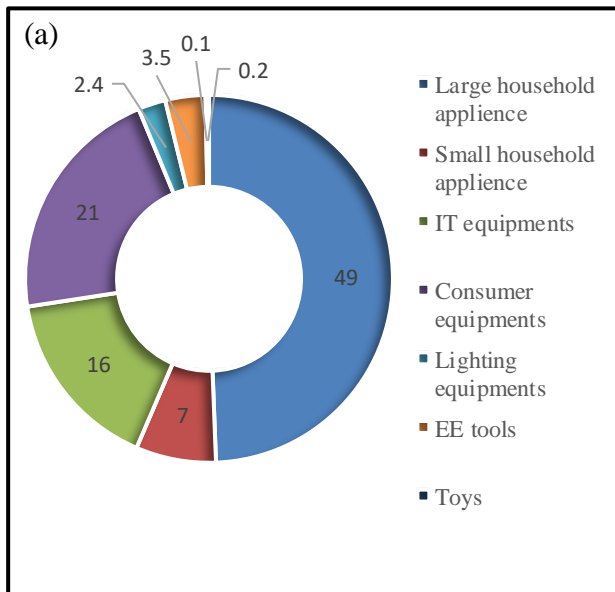
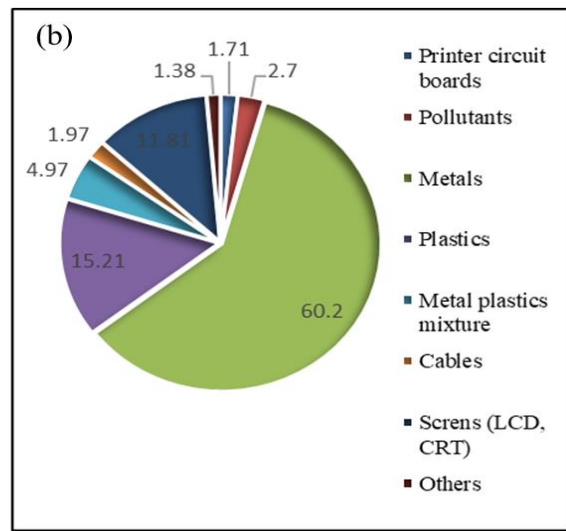
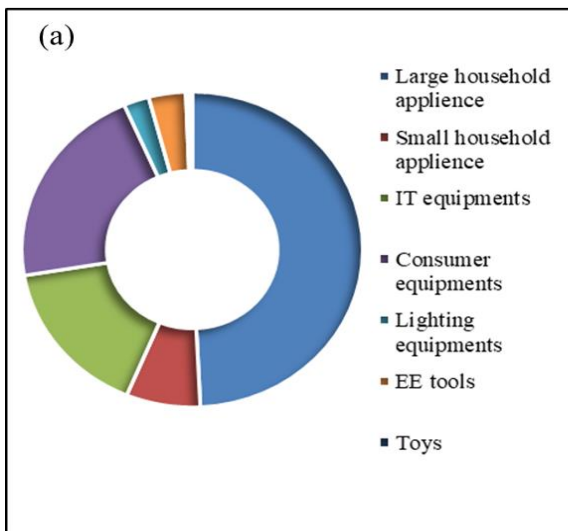
## 2. Major sources and HMs content in e-waste

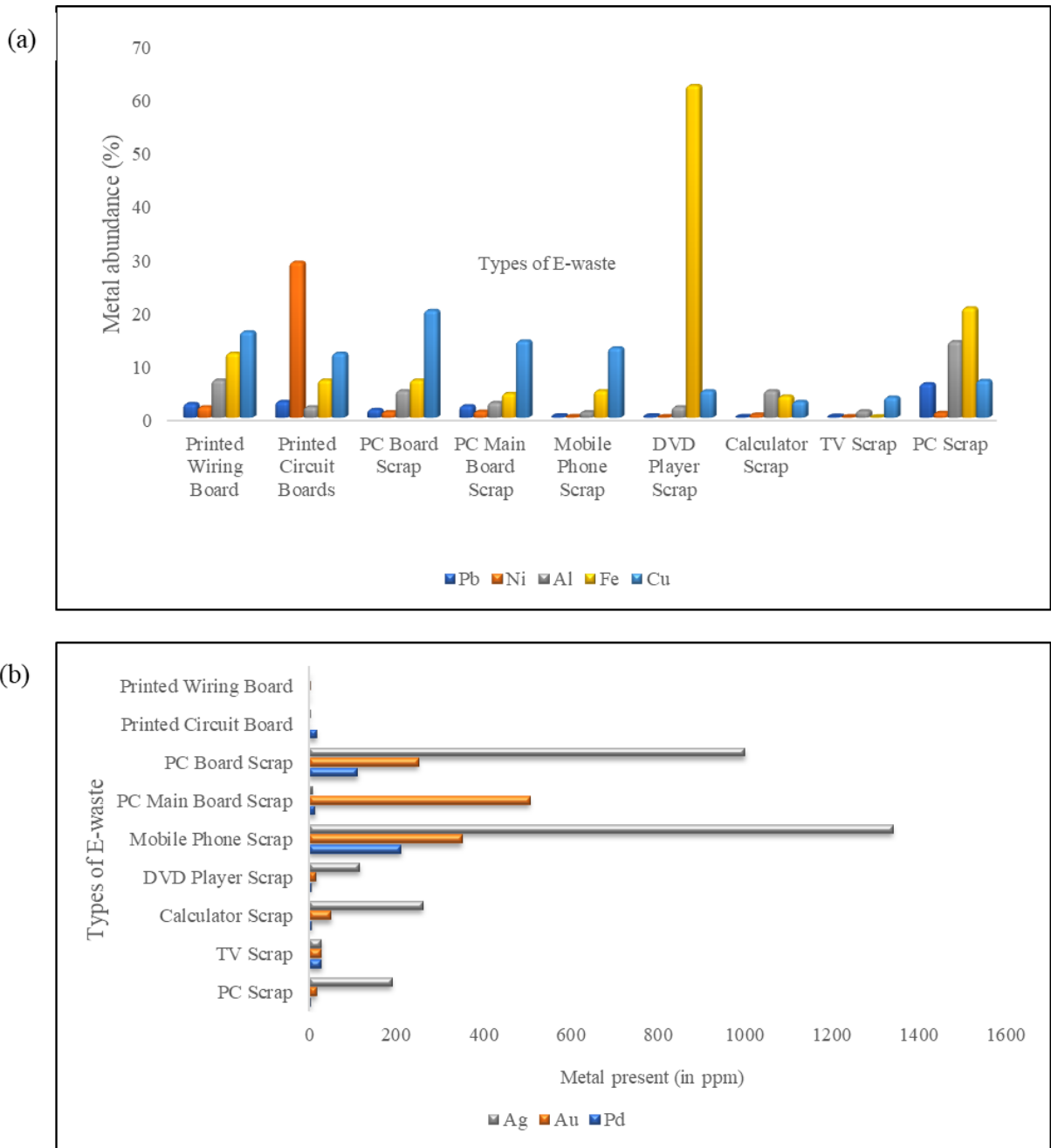
significant amounts of e-waste are generated by small and large electrical and electronic equipment. Various sources of e-waste are shown in **Figure 2a, b and Table 1.** the maximum amount of e-waste is generated from large household appliances (---%) and the average

composition of metals in e-waste is more than 60%. Various types of metals that may present in e-waste are listed below:

- i. Elements in bulk: tin (Sn), copper (Cu), silicon (Si), iron (Fe) and aluminium (Al)
- ii. Elements in small amounts: cadmium (Cd) and mercury (Hg),
- iii. Elements in trace amounts: germanium (Ge), gallium (Ga), barium (Ba), nickel (Ni), indium (In), vanadium (V), beryllium (Be), gold (Au), europium (Eu), titanium (Ti), ruthenium (Ru), cobalt (Co), palladium (Pd), manganese (Mn), silver (Ag), antimony (Sb), bismuth (Bi), selenium (Se), platinum (Pt), arsenic (As), lithium (Li) and boron (B).

The percentages of various types of metals present in e-waste are given in Figure 3 (a, b). Most e-waste contains HMs including Pb, Ni, Al, Cu, Fe, Pd, Au, Ag. But the quantity of Fe, Cu, and Al are much higher than others element. In contrast, Au, Ag, and Pd's presence is smaller than others, which are represented in ppm.





**Figure 3.** Metals present in e-waste. (a) in percentage and (b) in ppm [17, 36-43]

**Table 1.** E-waste sources, according to EU directives [44].

Category	Example	HMs content (%)	Reference
Large household appliances	Refrigerators/freezers, washing machines dishwashers.	Al (1.3-2), Sn (16.2), Cu (2-4.1).....	[45, 46]

		Al (1.3-2.0) ; Sn (1.6-2.0) , Cu (2.0- 4.1), Ag (0.0042–0.045), Pb (0.021–2.5) Cd (0.036–1.9)	
Small household appliances	Vacuum cleaners, kitchen machines.	Cu (18.8), Pb (4.79), Al (0.912) Cr,Cd, Ni (0.0051- 0.0179)	[47]
Information technology and telecommunication equipment	Computers, telephone, mobile phones, copying equipment, printers.	Cu (7.0-30) Al (1.41-14.17) Pb (1.20-6.29) Sn (1.0-3.15) Ni (0.85-2.5)	[46, 48]
Consumer Equipment	Televisions, stereo equipment	Cu (10), Al (10) Pb (1.0) Ni (0.3)	[49]
Electrical and electronic tools (except large scale stationary industrial tools)	Handheld drills, saws, screwdrivers.		
Toys, Leisure and sports equipment	Video games, sports computers, car racing, etc.	Pb (31-34), Cd (30-38) Hg (4.0-16) Cu (0.014) Sn (0.0039)	[50, 51]
Medical Devices (except all implanted and infected product)	Therapeutic, diagnostic and analytical equipment, massage devices, X-ray equipment, sterilizers.		

Personal computers and mobile phones are the sources of a large quantity of total e-waste produced across the world. It is estimated that about 3% of the total electronic scraps generated globally by weight is PCB [52]. Practically, most of the electronic equipment contains PCBs composed of the following types of materials:

- i. A laminate which is a non-conducting substrate;
- ii. Various recyclable metals (Cu, Al, Sn, Pb) and precious metals (Au, Ag, and Pt). Recently produced PCS boards may not contain Pb in their composition but may have other metals like Bi or Ag;
- iii. Various types of ceramic materials (can be reused or disposed of more appropriately)

**Table 2** represents a comparative view of metal compositions in PCBs of personal computers (PC) and mobile phones. The gold content in the cell phone is 5 to 10 times higher than gold ore. If this is multiplied with generated 150,000 tonnes of e-waste generated yearly, the amount becomes very attractive [53, 54]. Other metal such as copper content in PCBs is 10 -50 times higher than copper ore [53].

**Table 2.** Typical composition of metals in PCB of PC and mobile phone [55, 56]

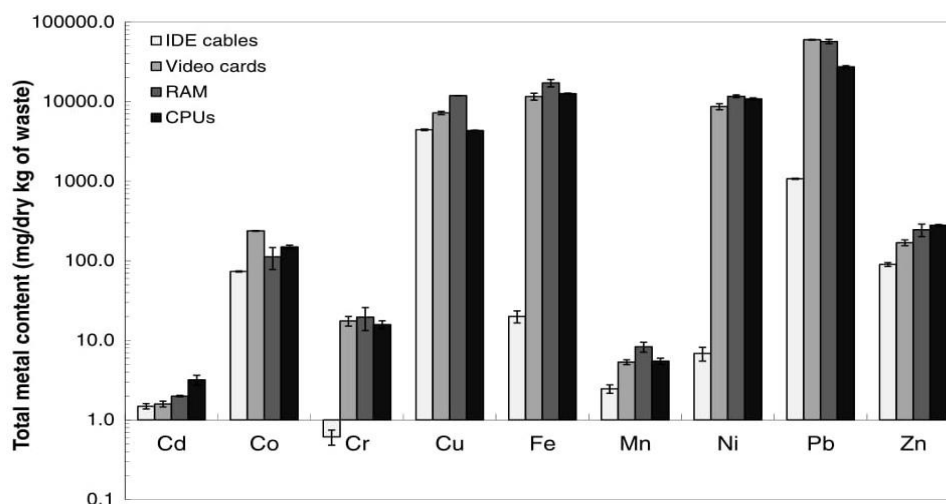
Metals Present in PCB	Weight Percentages (%)	
	Personal Computer	Mobile Phone
Cu	6.9287	33.5
Al	14.1723	1.41
Pb	6.2988	1.20
Zn	2.2046	1.92
Ni	0.8503	2.50



Fe	20.4712	2.32
Sn	1.0078	3.16
Au	0.0016	0.14
Pd	0.0003	0.03
Co	0.0157	0.02

The HM content of IDE cable (Integrated Drive Electronics Cable), video cards, RAM (random access memory), and CPU (Central Processing Unit) of PC is presented in **Figure 4**. Pb, Fe, Ni and Cu were found the dominant HMs in personal computers, mainly used in IDE cables, video cards, RAM and CPUs. Specifically, lead was the dominant HM in RAM and CPUs. The total content of Pb was approximately 57,000 and 27,000 mg dry kg<sup>-1</sup>, respectively. On the other hand, in IDE cables, copper was the dominant metal (the total content of about 4,400 mg dry kg<sup>-1</sup>) [57].

Besides, more than 40 elements contained in mobile phones PCBs, including hazardous metals (Be, Cd, As, Sb), the basic metals (Fe, Cu, Al, Ni, Zn, Sn), and precious metals (Au, Ag, Pt, Rd) which is about 19% of metals of its weight {Singh, 2018 #341}. Moreover, Al or Mg is used to produce the casing of mobile phones.



**Figure 4.** Total heavy-metal content of the four PC components (IDE cables, Video cards, RAM, CPUs) [57]

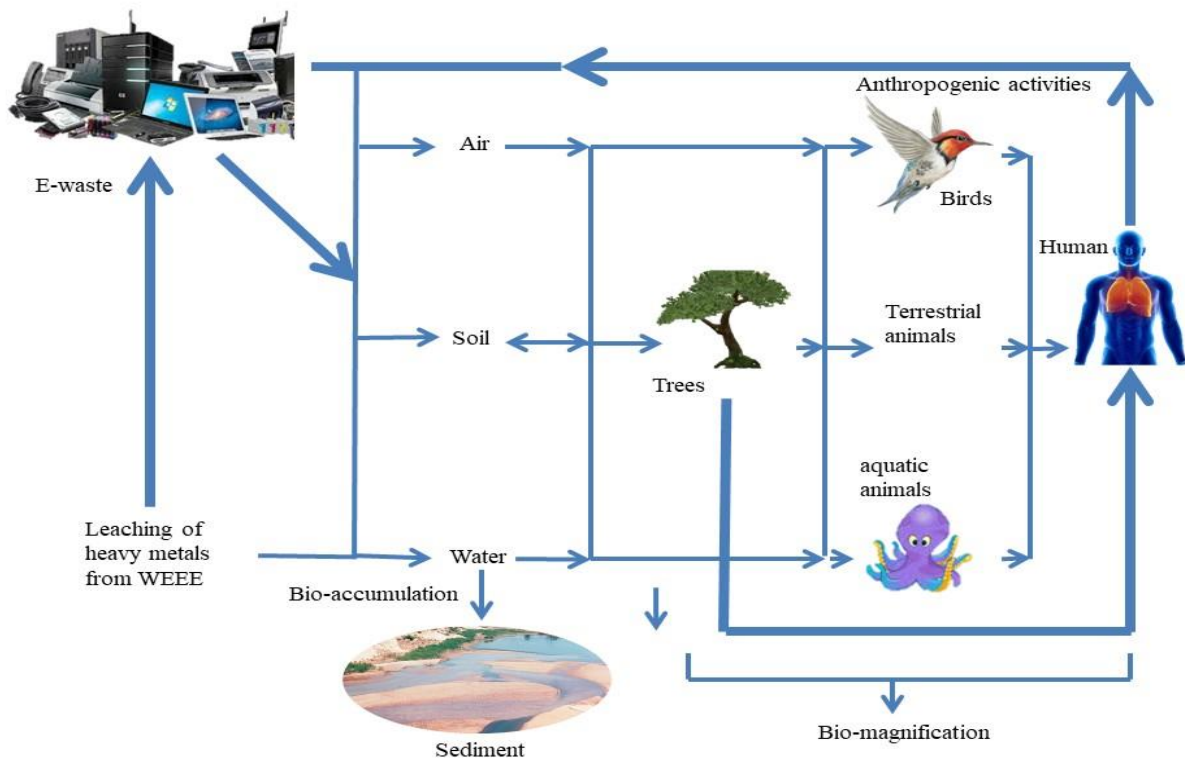
### 3. Heavy metals (HMs) releases, biodegradation and bioavailability from e-waste

#### 3.1. Heavy metals released from e-waste

There are various sources from which HMs enter into the environment. The intensive uncontrolled processing of e-waste has resulted in the release of large amounts of HMs in the local environment and caused high metal concentrations in the surrounding air, dust, soils, sediments and plants. HMs are released during the burning of e-waste, mining, and extraction of different elements from their respective ores and return to the land through dry and wet deposition [58]. The application of chemical fertilizers and combustion of fossil fuels, also

contribute to the anthropogenic input of heavy metals in the surrounding environment [59]. Geologic locations are responsible for the release of HMs into the environment. Furthermore, rainfall type, intensity and pattern, temperature, wind and pH are the main factor that can regulate the HMs release in the environment [60, 61].

Developed countries have replaced massive amounts of obsolete electronic equipment and home appliances with newer versions and cause a huge deposition after their lifespan in the environments by various physical, chemical and some physicochemical processes such as dry and wet deposition. Approximately 70% of the HMs in municipal solid waste landfills are estimated to come from electronics discards [62]. Uncontrolled burning, disassembly, and disposal of untreated e-wastes cause a distribution of heavy metals to the different environmental segments and create problems such as severe groundwater contamination, atmospheric pollution, or even water pollution either by immediate discharge or due to surface runoff. The ecological flow diagram presented in Figure 5 shows the release of HMs into the biosphere comprising air, soil, and aquatic environmental systems. This is likely due to natural and anthropogenic processes and their accumulation in flora and fauna and the flow cycle of HMs. Especially, people who are living in the e-waste recycling or processing area pose a significant risk to their health and nervous system [63, 64].



**Figure 5.** A systematic diagram of e-waste derived HMs affecting the environment and living systems (Reproduced from [65])

HMs mix with the earth's crust due to anthropogenic activities (such as mining and smelting operations, industrial production and use, and domestic and agricultural use of metals and metal-containing compounds) that causes severe human exposure. They are progressively accumulated in plants and terrestrial soil. Burning and incinerating electronic waste, industries, agriculture, wastewater, mining, and metallurgical processes and runoffs also lead to the release of pollutants into different environmental compartments. Atmospheric deposition is another possible pathway in the environment because the residual ash generated by the burning of e-waste, especially batteries and PCBs, contains high concentrations of heavy metals such as Cu and Pb [66].

The non-standard and crude e-waste recycling activities are the major sources of environmental pollutants (e.g., heavy metals, polybrominated diphenyl ethers (PBDEs) in e-waste recycling sites [67]. For example, heavy metals and PBDEs have become the main pollutants in the e-waste recycling sites in China (such as Guiyu, Qingyuan) near the riversides and worldwide [68, 69]. Anthropogenic processes of HMs have been noted to go beyond the natural fluxes for some metals. Metals naturally emitted in wind-blown dust are mostly from industrial areas. Some important anthropogenic sources significantly contribute to the HMs contamination in the environment, including automobile exhaust, which releases Pb; smelting releases As, Cu and Zn; insecticides that release As and burning of fossil fuels which release Ni, V, Hg, Se and Sn [70].

Toxic substances like Pb, Cd, and Hg leach into the soil and ultimately pollute the groundwater if e-waste is dumped. The polarity of water and hydrogen bonding enables water to dissolve, absorb, adsorb, or suspend many different compounds. Thus, water can easily acquire contaminants from its surroundings. Among the different types of pollutants affecting the water resources, HMs receive particular concern because of their substantial toxicity even at low concentrations.

Over the last two decades, the water used for acid-washing e-waste was directly discarded into the nearby stream. The unsalvageable e-waste after acid-washing can leach into the stream by rainfall and HMs can release. HMs were significantly retained in the surface soil, obviating groundwater contamination[72].

However, the concentration of HMs above the threshold limit is a big concern of environmental pollution and might trigger ecological imbalance in the ecosystem. Also, released HMs have severe health effects on human and animals. Hence, immediate and proper handling is necessary. In our following discussion (section. 6), we have reviewed the remediation technologies regarding this issue.

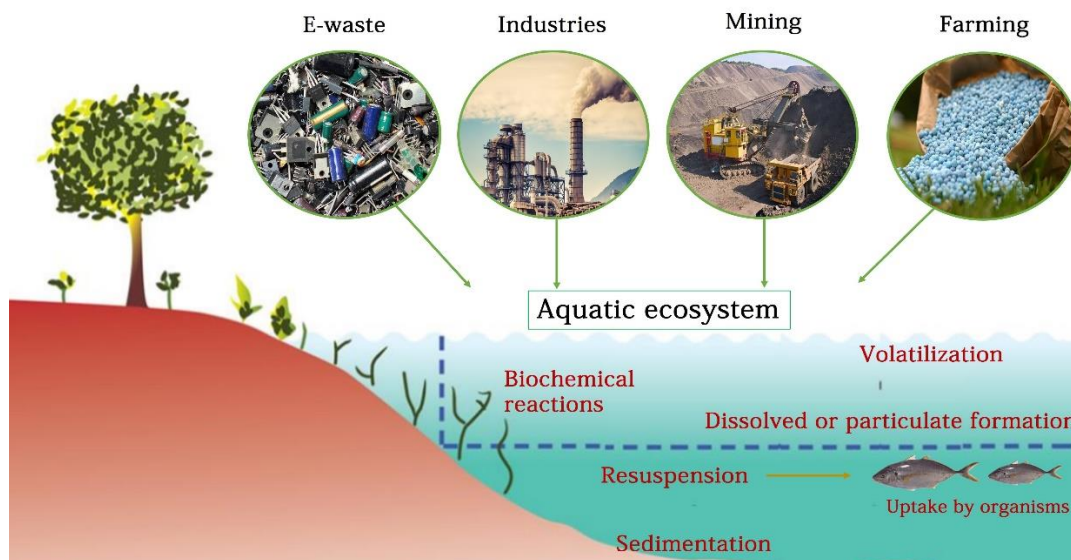
## **4.2. Bioavailability and Accumulation e-wastes**

### **4.2.1. Bioavailability of HMs**

Bioavailability is the capability of soil, sediments and waters to adsorb e-waste based HMs from the surrounding environments. Metal bioavailability is defined as the fraction of the total concentration of metal, which can accumulate in the body. The factors which control the bioavailability of metals are the organism biology (e.g., metals assimilation efficiency, feeding strategies, size or age, reproductive stage); metal geochemistry (e.g., distribution in water, sediment, suspended matters, and metal speciation); physical and chemical factors (e.g., temperature, salinity, pH, ionic strength, the concentration of dissolved organic carbon and total suspended solids) [73-76]. Heavy metals in soil, sediments, and water mainly come from E-waste have various species and chemistry (Table S1). Unlike organic chemicals, most metals cannot be easily metabolized into less toxic compounds, a characteristic of them being the lack of their

biodegradation ability. If once introduced into the aquatic environment, metals are redistributed throughout the water column, accumulated in sediments or consumed by biota [77].

E-waste based HMs can be bioavailable for soil. This is because their availability on soil depends on two factors, (i) the metallic element that precipitates as positively charged ions (cations) and (ii) another one, which makes up negatively charged components of salt [78]. Physico-chemical properties of soils, such as cation exchange capacity (CEC), organic matter, clay minerals, and hydrous metal oxides, pH and buffering capacity, redox potential and extent of aeration, water content, and temperature, together with root exudates and microbial activities determine the metal availability in soils [79, 80]. The toxicity of metals within soils with high CEC is generally low even at high total metal concentrations. When the soil pH is low, the metal bioavailability increases typically due to its free ionic species compared to high soil pH, where it decreases metal bioavailability as a result of the formation of insoluble metal mineral phosphate and carbonate [81, 82]. The mobility and bioavailability of certain metals in soils are usually in the order of  $Zn > Cu > Cd > Ni$  [79, 81]. However, the concentration of HMs within all components of the ecosystems varies considerably. The coexistence and persistence of metals in soils as multiple contaminants facilitate the entry and accumulation of these pollutants into food webs and ultimately ended with human diets.



**Figure 6.** HMs contamination sources influencing aquatic ecosystems

In the aquatic environment, sediments have been widely used as environmental indicators to assess metal pollution in natural water [84]. The HMs can react with various contents of the aquatic environment and can associate with multiple geochemical phases in the sediments [85]. Metal residues in contaminated habitats can bioaccumulate in aquatic ecosystems, aquatic flora and fauna [86]. Metal accumulation in sediments occurs through processes of precipitation of certain compounds. These compounds then bind with fine solid particles, associate with organic molecules, co-precipitation with Fe or Mn oxides or species bounded as carbonates according to the physical and chemical conditions between the sediment and the associated water column [87, 88]. **Figure 6** shows the sources of metal contamination affecting aquatic ecosystems. Many studies have shown that the free hydrated metallic ion is the most bioavailable form for Cu, Cd, Zn, and Pb [89, 90]. Thus, the importance of other chemical forms of dissolved metals and their complexes with suitable organic ligands having lower molecular weight should not be neglected.

It has been noticed that the presence of organic binders that increases the bioavailability of Cd in mussels and fish by facilitating the diffusion of the hydrophobic compound in the lipid membrane. The organic compounds of metals could be more bioavailable than the ionic forms [91]. For example, the mercurial organic compounds are lipid-soluble and penetrate the lipid membranes quickly, increasing the toxicity compared to mercuric chloride that is not lipid-soluble [92]. The adsorption on suspended solids affects the total concentration of metals present in the water body. The association between solid particles and metals is also critical for the metal uptake into organisms via food ingestion [93]. The suspended solids accumulate insoluble metal compounds. But under certain conditions, the metal reached the interstitial water being dissolved. HMs concentrations from sediments or suspended solids are much higher than in water, so a small fraction of them could be a significant source for bioaccumulation in planktonic and benthic organisms [94]. Other studies found that the bioavailability of metals in bivalve mollusks depends on sediment particle size due to their filter-feeding character. If the particles were coated with bacterial extracellular polymers or fulvic acids, the Cd, Zn, and Ag bioavailability would be significantly increased. In the overall case, metal-binding decreases the bioavailability of metals from the sediments [90, 95].

## **2.2. Accumulation of HMs**

HMs accumulates in the environment and consequently contaminate the food chains and soil due to their persistent nature [96]. Strong acid leaching and the open burning of dismantled components has led to the release of large quantities of toxic metals and organic pollutants into the surrounding environment. Studies have found that the air, surface water, groundwater, soil and river sediment of e-waste processing sites have been severely contaminated by HMs such as Cu, Cd, Hg, and Pb and organic contaminants [97-101]. HMs released from salvaging useful materials and from the uncontrolled open burning of electronic waste could penetrate the soils where vegetables and crops are grown by contaminated irrigation water and through direct deposition by air. Plants can easily take up these metals from the soil by their roots, transport them upwards to their shoots, and finally accumulate them inside their tissues. Although there are large variations among different plant species in terms of metal accumulation ability [102, 103]. Soils are the major sink for HMs released into the environment by the aforementioned anthropogenic activities. Once in the soil, HMs are adsorbed by initial fast reactions (minutes, hours), followed by slow adsorption reactions (days, years). Therefore, they are redistributed into different chemical forms with varying bioavailability, mobility, and toxicity [104]. The absorption of HMs by plant roots is one of the main routes of entrance in the food chain [105]. HMs accumulation in plants depends upon plant species, and the efficiency of different plants in absorbing metals is evaluated by either plant uptake or soil to plant transfer factors of the metals [106]. HMs are highly persistent, toxic in trace amounts and can potentially persuade severe oxidative stress in aquatic organisms. Thus, these contaminants are highly significant in ecotoxicology, and metals are not subject to bacterial degradation and hence remain permanently in the marine environment [107]. When HMs released into aquatic systems are generally bound to particulate matter; they eventually settle down and become sediments. Therefore, surface sediment is the most important reservoir or sinks of metals and other pollutants in aquatic environments. A major fraction of the trace metals introduced into the aquatic environment. These metals then become deposited with the bottom sediments [108].

## **5. Impacts of HMs**

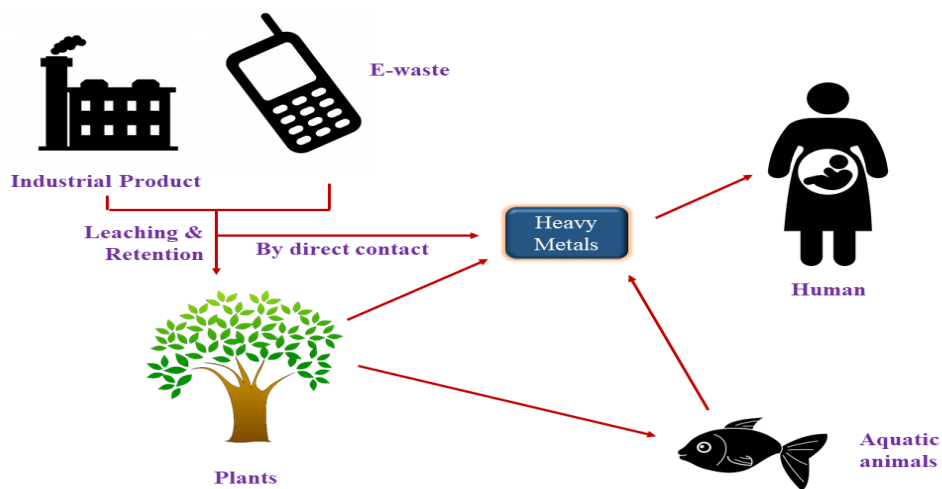
HMs are among the most common pollutants found in soil, sediment and wastewater. These metals assert a toxicity threat to human beings and animals even at low concentrations. E-waste contaminants can enter aquatic systems by leaching from dumpsites where they are processed, or unprocessed e-waste may have been deposited in the sediments through the water. Likewise, the disposal of acid after hydrometallurgical processes and degraded e-wastes get mixed with the abiotic environment and then introduce into waters or onto soils. The dissolution or settling of airborne contaminants can also result in the contamination of aquatic systems and water [109].

**Soil:** The most common types of HMs found in the soil are Cu, Ni, Cd, Zn, Cr, As and Pb [11]. The adverse effects of HMs rely on soil properties, i.e. organic matter, clay contents, and pH [110]. HMs obliquely affect soil enzymatic activities by shifting the microbial community, which usually synthesizes enzymes [111]. HMs exhibit toxic effects on soil biota by affecting fundamental microbial movement and decreasing soil microorganisms number and activity. Conversely, long-term HM effects can raise bacterial community fortitude and the tolerance of fungi such as arbuscular mycorrhizal fungi [112]. For example, Cd exhibits more toxicity to enzymes than Pb because of its greater dynamism and lower affinity for soil colloids [113]. Cu avert b-glucosidase activity more than cellulose activity. Pb decreases the activities of the urease enzyme, catalase, inverses, and acid phosphatase mostly. Phosphatase and sulfatase inhibit by As (V), but urease remains unaffected. Cd contamination has a nugatory effect on protease, urease, alkaline phosphatase, and arylsulfatase, whilst no significant impact was found in the case of invertase. Each soil enzyme exhibits a varied sensitivity to HMs. The order of inhibition of urease activity commonly decreased according to the order  $Cr > Cd > Zn > Mn > Pb$ . The diversity and activity of soil microbes conduct significant roles in recycling plant nutrients, maintaining soil structure, detoxifying noxious chemicals and controlling plant pests and plant growth communities are important indices of soil quality [114]. Chromium is common metal and present in soils as  $Cr^{3+}$  and  $Cr^{6+}$  characterized by distinct chemical properties and toxicities.  $Cr^{6+}$  is a potent oxidizing agent and is highly toxic, whereas  $Cr^{3+}$  is a micronutrient and a non-hazardous species 10 to 100 times less toxic than  $Cr^{6+}$  [115].  $Cr^{6+}$  has been reported to cause shifts in the composition of soil microbial populations and is known to cause detrimental effects on microbial cell metabolism at high concentrations [116]. In general, an increase in metal concentration adversely affects soil microbial characteristics, e.g. respiration rate, enzyme activity, which appears to be beneficial indicators of soil pollutions [117]. Uptake of HMs by plants and subsequent accumulation along the food chain is a potential threat to animal and human health [118]. Elevated Pb in soils may decrease soil productivity. A deficient Pb concentration may inhibit vital plant processes such as photosynthesis, mitosis, and water absorption with toxic symptoms of dark green leaves, wilting of older leaves, stunted foliage, and short brown roots [119]. HMs are potentially toxic and phytotoxicity for plants resulting in chlorosis, weak plant growth, yield depression, and may even be accompanied by reduced nutrient uptake, disorders in plant metabolism, and reduced ability to fixate molecular nitrogen in leguminous plants [120].

**Water:** Once an aquatic organism accumulates HMs, they can be transferred through the food catena's upper classes. Carnivores at the top of the food chain, including humans, get most of their HM burden from the aquatic ecosystem [121]. When dispersed into aquatic ecosystems, heavy metals and other contaminants stimulate the production of reactive oxygen species (ROS) that can spoil fishes and other aquatic organisms [107]. The consumption of fish containing elevated levels of metals is a concern because chronic exposure to HMs can cause health problems. Transport of metals in fish occurs through the blood, and the ions are usually bound to proteins.

The metals are brought into contact with the fish's organs and tissues and consequently accumulated to a different extent in different organs or tissues of the fish. There are five potential pathways for a pollutant to thrust into a fish [122]. These routes are through the food, non-food particles, oral consumption of water, and the skin. Once the pollutants are absorbed, they are transported by the blood to either a storage point or to the liver for conversion and storage. In case the pollutants are transformed by the liver, they may be gathered there or excreted in the gall or turned back into the blood for possible excretion by the gills or kidneys, or stored in fat, which is an extrahepatic tissue [121].

**Human:** The plant uptake of HMs from soils at high concentrations may result in a severe health risk considering food-chain implications. The utilization of food crops contaminated with HMs is a major food chain route for human exposure. Planting plants in contaminated soil represents a potential risk since the vegetal tissues can accumulate HMs [105]. HMs become toxic when the body does not metabolize them and accumulate in the soft tissues [123]. Chronic level ingestion of poisonous metals has undesirable impacts on humans, and the associated harmful impacts become perceptible only after several years of exposure [106]. HMs toxicity exposure to the human system and abnormal growth and development is shown in **Figure 7**. Zinc is considered relatively non-toxic, especially if taken orally, but an excess amount can cause system dysfunctions that impair growth and reproduction. The clinical indications of zinc toxicities have been reported as diarrhoea, bloody urine, vomiting, icterus (yellow mucus membrane), kidney failure and anaemia, liver failure [124]. Pb is thought to be physiological and neurological toxic to humans. Acute Pb inflammation may result in a dysfunction in the reproduction system, kidney, liver, and brain, resulting in sickness and death [125].



**Figure 7.** Schematic diagram of heavy metal toxic exposure to the human system and abnormal growth and development ( Reproduced from [126] )

## 6. Remediation technologies of HMs released from e-wastes

HMs remain in the environment as high, medium, or low in concentration which cannot remove easily. The remediation technologies used to mitigate their adverse effects on the

environment include biological treatment processes, physical treatment processes, chemical treatment processes and hybrid treatment processes.

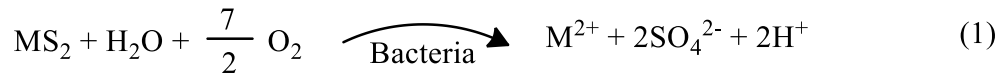
## 6.1. Biological remediation processes

The biological treatment process is a self-purification process where microorganisms, plants, and animals are used to detoxify the pollutants, the major advantages over conventional processes are this process does not produce toxic products and cost-effective [127]. The biological processes for HM remediation from groundwater or sub-surface of soil may appear by three mechanisms. They are adsorption, oxidation and reduction reactions and methylation processes [128]. According to Boopathy [129], nutrients are created by doing microbial cultures and biostimulation, which are used in HMs bioremediation such as land farming, composting, and bioreactors, venting by oxygen, using biofilters, bioaugmentation.

### 6.1.1. Bioleaching through microbial oxidation

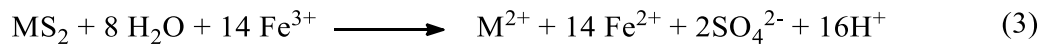
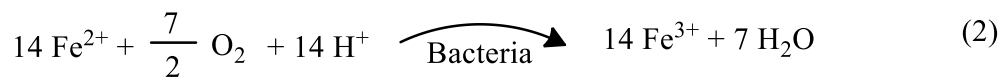
Bioleaching is an ex-situ/on-site remediation technology that is used globally in the HMs contaminated site and environmental issues. In this biotechnology, microorganisms are used to solubilize metals and semi-metals from concentrates mainly used in the mining industry and bio-hydrometallurgy [130]. Various types of microorganisms engage in the bioleaching process (**Table S2**) where *Acidithiobacillus ferrooxidans* and *thiooxidans* have high HMs removing performance.

Microbial oxidation involves both direct and indirect oxidation. In this process, microbes can easily attach to the metal salts and cause the dissolution of metals (Eq. 1):



Here,  $\text{MS}_2$  = Insoluble metal sulfide and  $\text{M}^{2+}$  = Free metal ion.

In the indirect oxidation method, microbes have interacted indirectly with the contaminants. In indirect bacterial leaching, elemental sulfur is oxidized by the sulfur-oxidizing bacteria and produces sulphuric acid. For this reason, the medium's pH is reduced and makes heavy metals inactive [131]. The bacteria play an active role in the reaction [132] by oxidizing  $\text{Fe}^{2+}$  to  $\text{Fe}^{3+}$  in the liquid phase via leaching, which is represented by the following chemical reactions.



According to Rozas et al. [133], an active strain (Hyhel-1; identified as *Bacillus* sp;) has a high leaching capacity and cost-effective performance to bio-leach copper (approximately 58.2%) from e-wastes. No acidic condition is needed for this stain and work appropriately at neutral pH (7-8) and moderate temperature (30-40 °C) [133]. Pant et al. [134] divided the whole leaching process into two parts; the first is the acid pre-leaching operation for 27 days and the second, the bioleaching operation for 280 days. At the end of these two operations, about 80% Zn, 64% Al, 86% Cu, and 74% Ni were leached out. Bioleaching is an eco-friendly and low-cost strategy for managing contaminated sediments, water, and soil in remediation technology. However, bioleaching efficiency mainly depends on several abiotic and biotic factors such as pH, oxidation/reduction potential, the concentration of the contaminants, growth substrates, temperature and oxygen [130]. Although this method has many advantages over the chemical and



physical processes, this method also has some disadvantages, such as a long time needed for bioleaching and low efficiency of HMs removal [27].

### 6.1.2. Phytoremediation

Phytoremediation (Figure S2) is a bioremediation technology used to exchange contaminants from a highly toxic to a less toxic form by accumulating, immobilizing, and transforming [135]. The capacity of “Phytoremediation technology” mainly depends on the plant’s ability to take up, store, or degrade pollutants and the characteristics of the pollutants present in the environment [136]. Some biological factors such as the interaction between plant and microbe’s uptake capacity of plant, displacement and tolerance mechanisms, and plant chelation ability also responsible for the performance of the phytoremediation process. These phytoremediation techniques are mainly applicable for the remediation of soil, sediment as well as water. But special care is necessary for this technique to get the best performance. *F Cardaminopsis halleri*, *Bryophyllum. Pinnatum*, *Zea mays*, *Glycine max*, *Brassica junica*, *Brassica napus*, *Thlaspi caerulescens* plant species are mostly used with high efficiency in different HMs remediation (Table S3). According to Babu et al., [137], the removal capacity of Pb, Zn, As, Cd, Cu, Ni metals by using *Alnus firma* was 77-10%, respectively. It is recommended to study with the combination of two or more phytoremediation processes for better efficiency [138].

### 6.1.3. Microbial remediation

Microbes (e.g., yeast, fungi or bacteria) use the contaminated substances as nutrients or energy sources for their growth [139]. Among HMs, some metals such as Cr, Ca, Mg, Mn, Cu, Na, Ni and Zn are essential micronutrients for various metabolic functions and redox functions. Other non-essential metals such as Cd, Pb, Hg, Al, Au, and Ag have no biological ineffectiveness. Still, they have harmful effects, and some of them, like  $Cd^{2+}$ ,  $Ag^{2+}$ ,  $Hg^{2+}$  make inactive by binding with the SH (sulfhydryl) groups of enzymes [140]. Microbial cell walls are mainly made with various functional groups such as carboxylate, hydroxyl, amino, and phosphate, which helps the microbes bind the metal ions in their cell wall. This binding is done by different bond interactions such as covalent bonding, electrostatic and van der Waals forces [141].

Microorganisms play a fundamental role in bioremediation because of their high efficiency, easy operation, and cannot produce any secondary pollution from contaminated soil, sediment, and wastewater [142, 143]. In the presence of hazardous chemicals, the microorganisms have high adaptation ability in any environment, such as at subzero temperatures, desert conditions, high aerobic and anaerobic conditions [135]. *Aspergillus niger*, *Bacillus subtilis*, *Penicillium canescens*, *Penicillium chrysogenum*, and *Pseudomonas* sp. *Rhizopus arrhizus* and *Yarrowia lipolytica* have high uptake capacity and cover maximum metals from their environments (Table S4). Some microbes such as *Sacchromyces cerevisiae*, *Corynebacterium equi*, *Bacillus licheniformis*, and *Rhizopus arrhizus* can also remediate radioactive metals from the environment [144]. Although the micro-remediation process has many advantages, the main limitation is that when the metals are bound to microbes, they can be released back into the environment soon after decomposing the microbes upon their death decay [140].

## 6.2. Physical remediation processes

The physical remediation process is a simple process where the HMs are removed physically from the environment without any chemicals and widely used for its advantages. Different types of physical treatment processes for HM remediation are described below.

### 6.2.1. Thermal remediation

Thermal treatment can significantly reduce the toxicity of HMs at temperature (300–400) °C [145, 146]. In this process, heating is done by using steam, microwaves, and infrared radiation (IR) to volatilize the pollutant (e.g., Hg, As) [147]. By thermal remediation process, 94% Cd, 86% Zn, 73.6% Cr and 97% Cu could be removed within a short time (please give time) at 280 °C [148]. A study conducted by Hseu et al., [149] showed almost 99% removal of Hg by the thermogravimetric method at 550 °C for 1h.

Vitrification is another thermal remediation process where vitreous materials are produced (usually an oxide solid) by entrapping and immobilizing the contaminant. In this process, high temperature (1700–2000 °C) is obtained by an electric current to melt the metals in the vitrified form and mainly used to treat organic and inorganic pollutants from contaminated soil and sediments [150]. By this vitrification method, Mn, Fe, Zn, Cu, Cr, Cd, Pb, Hg, As, and Se concentration is reduced by 91–100% using high temperature (more than 1300 °C) [150]. This remediation technology is a straightforward method for treating mixed contaminants. However, the limitations of this technique are- (i) only applicable for solid pollutants such as soils and sediments, (ii) very high temperature requires, (iii) produce toxic gases, and (iv) vitrified waste may need to be recycled again [148, 151]. This process's problems can be solved by characterising pollutants and acquiring goodish consideration [66, 152]. For example, laboratory simulated open burning practices of the e-waste recycling operations process used to assess the air emissions and leached ash characterization [66].

### 6.2.2. Ion-exchange

The ion exchange process is a versatile electroperturbation technique for removing various contaminants, including HMs. Different synthetic and polymeric cationic resins (e.g, purolite C100, amberlite IR120H ) used for the removal of HMs from wastewater [153, 154]. The degree of ion exchange influenced by different factors: i) size and valence of the metal ions, ii) concentration of the ions, iii) physical and chemical characteristics of ion exchangers and iv) temperature [155]. The ion exchange process is more beneficial to remove Cd, Pb, Ni, Cr, Hg, Cu, Zn from water contaminants where the affinity of exchangers for ions are: Pb(II) > Cu(II) > Cd(II) > Zn(II) [156]. A study by Mier et al. [157] reported high removal efficiency (> 95%) of Pb, Cd, and Cr using natural clinoptilolite. This process was done in a batch reactor for 18 h at the acidic condition. The removal capacities of the polyvinylpyridine resin for Zn, Cu, and Ni ions are 0.65, 0.51, and 0.59 mmol g<sup>-1</sup>, respectively [158]. Moreover, natural zeolite widely used to remove HM such as Zn, Co, Cu, Mn [159, 160]. However, pH sensitivity and non-selectivity of the exchangers reduce the process efficiency. Another disadvantage of this method is that HMs are undesirable and swapped by other cations that are not harmful to the environment [156]. In a recent study, macroporous ion-exchange resins (Amberlite IRA 743, Lewatit TP 208, and Lewatit TP 260) were applied for selective sorption of HMs. From Cu<sup>2+</sup>, Zn<sup>2+</sup>, Ni<sup>2+</sup>, Pb<sup>2+</sup>, and Al<sup>3+</sup> ions, hazardous Pb<sup>2+</sup> ion was selectively extracted from the leached solution using a multi-elemental ion-exchange process[161]. More investigations on hazardous ion-selective exchangers are required to get better results.

### 6.2.3 Adsorption by activated carbon

Activated carbons (AC) are extensively used for HMs adsorption. Adsorption of HMs ions from e-waste solution is a straightforward method through electrostatic interactions [162]. However, different factors, for example, the surface area and porosity of adsorbent, metal ion

complex, the solution pH. Also, the surface functionality and the size of adsorbing species have significant effects on the adsorption of HMs in AC[163]. However, AC is a promising adsorbent several previous research work experimented with the efficacy and factors affecting the adsorption process[163, 164]. A previous study showed that AC has better efficiency for arsenic (As) and antimony (Sb) removal from Cu electro-refining solutions [165]. The highest As(V) removal was observed with the highest ash content (coconut shell carbon 3%, peat-based extruded carbon 5%, coal-based carbon 5-6%), and on this AC have a high sorption capacity, probably 2860 mg/g As was adsorbed [166]. Pb adsorption from aqueous solution by this AC reported the maximum removal efficiency of 97.95% (experimental) and 134.22 mg/g (from Langmuir isotherm model) at pH 6.5. Tamarind wood mainly shows its better performance at pH 5.4 and investigated the high removal rate of Cr (>89%) [167]. Zinc chloride AC prepared from tamarind wood ash also has better removal efficiency for Pb and Cr [168]. A study by Ricordel et al. [169] used AC prepared from peanut husks (PHC) for the adsorption of Pb, Cd, Ni, Zn. PHC adsorption mainly depends on particle size distribution, metal/PHC ratio. Langmuir constant varied in the order of  $Pb^{2+} > Cd^{2+} > Ni^{2+} > Zn^{2+}$  and among them  $Pb^{2+}$  had the best attraction capacity to PHC. Lignite, peat charcoals [170], bio-char [171], and bone-char [172] were used in wastewater treatment to remediate HMs, which can be served as good substitutes for ACs. However, ACs are very efficient adsorbent, but recovery of valuable HMs are still challenging, the more comprehensive study should be done to determine methods and the most suitable desorbents for particular heavy metals.

#### **6.2.6. Sorption of HMs by cellulosic materials and agricultural wastes**

Cellulosic materials and agricultural wastes based materials have been applied to remove HMs from water, but lower adsorption capacities were observed for pristine cellulosic materials, and therefore, modified cellulosic materials are commonly used for improving the removal capacities [195]. For example, O'Connell et al. [196] investigated that the modified cellulosic material by halogenation, esterification, etherification or oxidation process had higher adsorption capacities. Similarly, a study by Martínez et al., [229] applied grape stalk, a by-product of wine production, and found sorption of Pb of 0.241 mMol/g, and Cd of 0.248 mMol/g at pH 5.5. Similarly, a study by Sahu et al. [197] used activated rice husk in a three-phase modified multi-stage bubble column reactor to treat contaminated water and reported 77.15% removal of Pb. Walnut hull has a better sorption capacity of Cr (VI) of 97.3% at pH 1.0 from contaminated water, and the adsorption efficiency depends on temperature, Cr(VI) concentration, adsorbent concentration, and electrolytes amount [198]. Various modified cellulose materials used to remove various HMs from contaminant sites (**Table S5**). A different functional group such as alcohol, carbonyl, amido, amino, acetamido and sulphhydryl present in the agricultural waste materials mainly responsible for the sorption of HMs. These groups can make complexes or chelates with the HM ions, and sorption is occurred by complexation, adsorption, diffusion, chemisorption and ion exchange mechanisms. Although some signs of progress have been put forward on modification of the cellulosic materials more study required for suitable functionalization of these types of materials to improve the adsorption capacities in a cost-effective manner.

#### **6.2.4. Membrane technology**

Over the past few decades, membrane technology received much attention because it works without the addition of chemicals requires relatively low energy. Different types of inorganic and organic membrane processes such as microfiltration, ultrafiltration, nanofiltration, reverse osmosis. Membrane distillation and forward osmosis are used to remove. In the membrane process, the removal of HM mainly depends on the membrane pore size and surface charge.

##### **6.2.4.1. Microfiltration and Ultrafiltration**

Microfiltration (MF) is a physical process where a contaminated fluid is passed through a special pore-sized membrane to separate the particles in a range of 0.1 to 10  $\mu\text{m}$  from process liquid. A MF process using DuPont's polyolefinic was developed to treat HMs containing groundwater, however, this filtration was suited for suspended solid removal less than 5000ppm. Further HMs removal can be achieved using the ultrafiltration process{Bharagava, 2017 #342}. Tyvek filter was used at the Palmerton Zn Superfund site in Palmerton, Pennsylvania to remove Zn. Their results showed 99% removal of suspended Zn solid{Andrew Hass, 2020 #343}.

An ultrafiltration (UF) technique is mainly necessary after the MF process for further cleanup [173]. In UF, a semipermeable membrane is used as a filter media and separate particles ranging from 0.1  $\mu\text{m}$  to 0.01  $\mu\text{m}$  from a contaminated liquid. Using MF and UF, the HMs removal efficiency was achieved up to 80% except for As, Mo and Sb content because these metals remained a dissolved phase in an acidic medium [174]. The UF membrane has higher removal rates for heavy metals such as Fe (92.14%) and Ni (61.90%) [175]. Using microporous substances in filter systems can achieve 90–100% removal efficiencies for Cd, Cu, Pb, Ni, Cr, and Zn [176]. More feasibility analysis and evaluation of MF and UF techniques are required to obtain an overall better efficiency.

##### **6.2.4.2. Reverse Osmosis (RO)**

RO retains 95–99% of the dissolved solutes (organic and inorganic) from the feed stream and produces high-quality water. Moreover, nonporous graphenes with functional groups (nitrogen, hydroxyl, boron) are used in RO process to improve process efficiency [177].

A pilot study byPawlak et al.[178] reported that the concentration of heavy metals (give the name) reduced from 60 ppb to 1 ppb, over an operation period of 30-days. A comparative study Qdais et al., [179] showed that the concentration of  $\text{Cu}^{2+}$  and  $\text{Cd}^{2+}$  was reduced to be about 3 ppm (99.4% removal efficiency) by RO process from water with an initial concentration of 500 ppm. The heavy metal removal efficiency of RO is higher than nanofiltration (NF) [179]. Using nanofiltration,  $\text{Cu}^{2+}$  and  $\text{Cd}^{2+}$  concentration were reduced to 13 ppm (97.4% removal efficiency) from wastewater where feed concentration was 500 ppm [179]. The advantages of the RO process over other traditional water treatment technologies are reducing the ionic contaminants, and organic pollutants, an eco-friendly approach, and no chemicals are needed here [178, 180].

##### **6.2.4.3. Membrane bioreactor (MBR) technology**

MBR technology is a combined process where membrane (microporous) and biological reactors (suspended activated sludge system) work together, which is widely used for wastewater treatment contaminated with insistent HMs [181].

Different processes such as i) entrapment of metal ions within sludge flocs, ii) adsorption of metal ions on the flocs surface, iii) diffusions of metals in the flocs structure and iv) biosorption

of metals ions involve in MBR to remove heavy metals from the contaminated water [182]. Precipitation of metal ions also can occur by changing pH in the reactor medium [183]. Study shows that MBR offers 40 to 50% more efficiency in removing HMs compared to conventional activated sludge process [184]. The removal HMs using MBR depends on solid retention time (SRT), suspended solids concentration and pH [185]. Nevertheless, metal liquor suspended solids enhanced the metal removal process creating more available biosorption. Moreover, biological solids that are mixed sludge completely retained in the bioreactor. Katsou et al.[186] found that average removal efficiencies accomplished by MBR 80%, 77%, 98% and 50% for Cu, Zn, Pb and Ni respectively from municipal wastewater. Similarly, in a laboratory-scale MBR, Mahmoudkhani et al. found average removal efficiency of 84%, 23% and 96% for Fe, Cu and Cd, respectively [187]. Furthermore, 50% As, 96% Ni, 95% Sb, 99% Cd and Pb were removed by MBR technology from water [188]. It has been observed that the addition of vermiculite in MBR can significantly increase the removal efficiency [185].

The major drawbacks in the MBR process, high cost associated with membrane fouling, aeration and excess sludge production. For achieving high quality-effluents, the combination of MBR (or OMBR (Osmotic Membrane Bioreactor) with RO is used. The RO process is mainly used in the OMBR dilution stage or post-treatment in the MBR process to re-concentrate the draw solution. To solve salinity in water remediation, recently OMBR is jointly used with microfiltration) and electrodialysis [302]. The integrated system of electrically-enhanced MBR (eMBR) is most popular for its advantages and is suitable for wastewater treatment [303]. Giwa et al. [304] investigated different eMBR with a new configuration of an anode-cathode-membrane module-cathode-anode (A-C-M-C-A) to treat wastewater. Using this integrated technology Dialynas et al.[305] achieved complete removal of Pb & Ni and 89% and 49% removal of Cr and Cu, respectively. In another study, AC carbon assisted MBR integrated with RO used for metal ions removal, maximum efficiency observed for Cd, Cu, Cr, and Pb; 72.0%, 75.6%, 27.2%, and 43.7% respectively[306]. However, MBR with RO can remove HMs but limited research has published focused on HMs removal. Nevertheless, this technique may not be effective for low valence metal ions removal [306].

### **6.2.5 Solidification/stabilization (S/S)**

Solidification is a fixation process that is widely used in remediation technology and more suitable for contaminated soil and sediment treatment [189]. In the solidification process, inorganic stabilisers or inorganic-organic-organic mixture amendments may be mixed with the contaminated sites for heavy metal immobilization. The organic stabilizers are mainly straw, leaves, xylogen, bark sawdust, bagasse, chitosan, poultry manure, rice hulls, sewage sludge, and inorganic binders are the clay, cement, fly ash, slag, calcium montmorillonite, Fe/Mn oxides, charcoal, zeolite and lime [190].

Solidification technologies are not applicable for organic contaminants and some inorganic contaminants, such as oxyanions (e.g.  $\text{Cr}_2\text{O}_7^{2-}$ ,  $\text{AsO}_3^-$ ) and metals (e.g. Hg) because organic vapors are produced during mixing and heating. To mitigate this problem, some pretreatment such as air stripping or incineration processes are necessary [191]. The solidification process mainly depends on factors, such as contaminants chemical composition, ambient temperature, and the amount of water in the contaminated matrix. These factors inhibit the binding between contaminants and binding materials. As a result, the stability of the matrix along with its strength is reduced [192].

Al-Wabel et al. [193] have been investigated the immobilization of Fe, Mn, Zn, Cd, Cu, Pb in maize plants depends on biochar rates and soil moisture levels. The result of decreasing metal concentration at soil moisture level 75% and 100% are 51.3 and 60.5% for Mn, 28 and 21.2% for

Zn, 53.2 and 47.2% for Cd, and 60 and 29.5% for Cu, respectively. Here, the removal amount is lower at the highest soil moisture level, and a better result was found without Fe metal. Also, Lv et al. [194] studied the remediation of Cd-contaminated soil with sodium bentonite in the solidification process. They found that the amount of Cd was reduced to 21.4, 27.6, 27.2, and 32.3% when the amount of sodium bentonite was 20, 30, 50, and 40 g kg<sup>-1</sup>.

#### **6.2.8. Metal removal by biosurfactants**

Biosurfactants are surface-active agents which are produced from biological systems, mainly from microorganisms. Surfactants are dissolved by their hydrophilic and hydrophobic groups and make HMs more available for remediation from contaminated substances [199]. Surfactin, sophorolipids and rhamnolipids, are biological surfactants that can remove HMs such as Cu, Ni, Zn, and Cd from the contaminated soil [200-202]. Among them, rhamnolipid type I and type II with a surface tension of 29 mN/m have better HMs removal capacity in liquid and foam form from contaminated soil [203]. Rhamnolipid remove 73.2% Cd and 68.1% Ni from the soil at pH 10. But this efficiency can be increased up to 11-15% when the rhamnolipid foam is used. The surfactants remove HMs by making complex compound in the soil due to the lowering of interfacial tension [200]. However, more study is required to assess the HMs removal capacities by biosurfactants.

### **6.3. Chemical remediation processes**

There are several chemical methods which are used for HMs remediation from the contaminated sites. They are described below.

#### **6.3.1. Immobilization techniques**

Chemical fixation or immobilization is a technique in which various chemical or reagents (e.g., amendments) are added into the contaminated sites to convert the toxic matters into hardly movable or insoluble substances. As a result, the migration of HMs to water and other environmental media is decreased [204]. This is a simple and very rapid process. Practically two types of techniques are used here, e.g., ex-situ and in-situ immobilization techniques. When the contamination of soil is high, it cannot remove, and its storage is connected with a high ecological risk (e.g., in the case of radionuclides) then the ex-situ technique is needed. This technique's main advantages are- (i) applicability is easy and rapid (ii) the functional and investing cost of this technique is comparatively low. The disadvantages are- (i) invasivity to the environment is high, (ii) solid wastes are generated (probably twice in volume after processing), (iii) the byproduct must be landfilled, (iv) danger of contaminants has occurred when the physicochemical conditions are changed, and (v) the stored wastes should be controlled carefully. On the other hand, fixing agent's amendments for unexcavated soil are applied for the in-situ technique. The technique has several advantages- low invasiveness, rapidity and simplicity, cost-effective, less waste production and high public acceptability. This in-situ immobilization also some demerits- (i) temporary solutions are used here, (ii) if physicochemical properties of soil are changed, pollutants may activate, (iii) only to the surface layer of soil (30–50 cm) reclamation process is applied, and (iv) permanent monitoring is essential [205, 206]. For accelerating the attenuation of metal mobility and toxicity in soils, several types of inorganic and organic amendments have often been used in the immobilization method. The amendments are applied mostly on clay, minerals, cement, organic composts, zeolites, phosphates and microbes [189]. Lv et al. [207] used the sodium bentonite immobilization technique to remediate Cd-contaminated soil and reported around 21.4% removal of Cd. Another study by Al-Wabel et al. [208] has been investigated the immobilization of Fe,

Cd, Zn, Mn, Cu, and Pb using biochar in maize plants. Both ex-situ and in-situ immobilization technologies are widely used in HMs remediation depending on specific site conditions. Furthermore, different immobilization technique using various amendments have various degree of removal efficiency of different HMs (**Table S6**).

### 6.3.2. Precipitation

Precipitation is a simple chemical process in which acid-base reactions are used for removing the soluble metal ions from the solution. Generally, after flocculation and sedimentation, precipitation is occurred [209]. Precipitation is one of the most conventional methods used to effectively eliminate of HMs from contaminated sources. The mechanism is as follows:



Here  $M^{2+}$  represents dissolved metal ions,  $OH^{-}$  represents the precipitant, and  $M(OH)_2$  represents insoluble metal hydroxide. Generally, in the pH range of 9–11, chemical precipitation occurs [210]. Precipitation is a very convenient technique when the concentration of the heavy metal(loid) ion is high and in the high pH soils in the presence of anions (e.g.,  $SO_4^{2-}$ ,  $CO_3^{2-}$ ,  $OH^{-}$ , and  $HPO_4^{2-}$ ) [211]. A study by Aziz et al. [212] reported that limestone has significantly removed more than 90% of metals (e.g., Cd, Ni, Cu, Zn, Cr, Pb) than crushed bricks (80%) and gravel (65%) in a continuous filtration process. The removal of the metals only by aeration and settlement was less than 30%.

### 6.3.3. Electrokinetic remediation

The electrokinetic process is a technique in which a low electric current is passed between a cathode and an anode where both the cathodes and anodes are embedded in the contaminated substances (e.g., soil, sediments). In addition to water, ions and small charged particles are transported between the electrodes. The mechanism involves- all cations moving towards negative, and all anions move towards positive and separated [201]. The separation of the HMs present in the soil occurs via electrophoresis, electric seepage, or electro-migration, resulting in a decrease in contamination [213]. More than 96% of metal removal efficiency can be reached for copper and zinc by electrokinetic remediation [214]. Rosestolato et al. [215] reported 60% removal of Hg from approximately 400 kg of contaminated soil. However, metal removal efficiency depends on the treatment duration, type of chemical used (anolyte) and which metal is to remediate [216]. For instance, by using  $KH_2PO_4$  anolyte, Lee et al. [217] reported that the removal efficiencies of As and Cu could increase by > 50 and ~20%, respectively. Furthermore, they have reported the inefficient removal of Pb and Zn (less than 20%). Moreover, the electrokinetic remediation efficiency was investigated by Juris et al. [218] for copper contaminated clayey soil using a graphite electrode. After 40 days of electrokinetic remediation, copper concentration in water was 0.05 and 0.085 mg L<sup>-1</sup>, representing a minimal quantity (around 0.02–0.03%) of the initial metal amount [219]. Using 4–26V as current, Ottosen et al. [220] investigated the electrokinetic remediation method of Cu and Pb and found approximately 41% and 31% removal of Cu and Pb, respectively [221]. Nejad et al. [151] reported the reduction of 50-100%, Cd, Hg, Pb, Ni removal of 80-100% and Cu removal of <50% using electrokinetic remediation method.

The electrokinetic remediation technique is economically effective because of its easy installation and operation [222]. However, the main limiting factor of this method is fluctuation in soil pH. In some cases, using ion-exchange membrane or complexant and adding buffer solutions in cathode and anode, soil pH can be controlled [223].

### 6.3.4. Chemical leaching

Chemical leaching is mainly a washing process in which the contaminated substances are washed using different reagents, freshwater and other fluids or gases [224]. The HMs present in

the soil is transferred from the contaminant to the liquid phase through ions exchange, precipitation, adsorption, and chelation. After that, HMs are recovered from the leachate. The leachate mainly includes inorganic eluent, chelation agents, and surfactants. The effects of various concentrations of sulfuric acid, phosphoric acid, hydrogen chloride, hydrogen fluoride, nitric acid on the extraction of As from the artificially polluted soil were investigated by Tokunage Hakuta [225]. They found phosphoric acid as the most promising reagent, resulting in 99.9% extraction of As with 9.4% acid concentration in 6 h. For removing As from contaminated soil, an eco-friendly and economical remediation method was studied at 40 °C and pH 6.0 with 300 mM phosphate concentration [226]. Most HMs can form a stable composite with the ethylenediaminetetraacetic acid (EDTA) (among the other extractants) at a broad pH range.

Cadmium, zinc, lead and chromium were removed by a 0.01 M Na<sub>2</sub>EDTA solution in different ratios from the soil. At soil to solution ratio of 1:5 at pH 5.1, the removal efficiencies of Pb, Zn, Cd and Cr were 67.8 %, 49.5 %, 89.3 % and 26.9 %, respectively [227]. The efficiency of saponin on remediating HM contaminated soils was evaluated by Hong et al. [228]. Various soil types (e.g., Andosol, Cambisol, Regosol) were washed with saponin in batch experiments where 90- 100% of Cd and 85-98% of Zn was removed. The efficiency of tea saponin on metal removal was investigated by Li et al. [229] and found that the removal of Pb, Cd, Zn, and Cu were 6.74%, 42.38%, 13.07%, and 8.75%, respectively when using 7 wt% tea saponin as the extractant which is eco-friendly than other chemical processes (**Table 4**). To achieve a better understanding and maximum HMs removal efficiency using the chemical leaching process need more exploration on the effect of leaching time and temperature and kinetic studies are required. Generally, organic leaching agents are used in the chemical leaching process are not effective; however, they are biodegradable. Apart from this, it is still challenging to recover valuable metals from organic compounds and chelating agents[230]. Though the chemical leaching process consumes chemicals, that should be another concern of environmental hazard. From an economic point of view, selective and low-cost chemical leaching processes are required.

**Table 4.** Metal removal efficiency with different chemical leaching method from the soil

Leaching method	Reagents (mol/L)	Soil pH	Metal	Conc. (µg/g)	Removal efficiency (%)	Ref.
Batch 1 h, 3 steps, pH 4.5 (chelation)	EDTA (0.0225)	7.4	Pb	65,200	56	[231]
			Cd	52	37	
Batch 24 h, 12 steps, pH 7–8 (chelation)	EDTA (0.00275)	8.03	Cd	6.3	53	[232]
			Cu	700	49	
			Pb	800	76	
			Zn	2,650	84	
Batch 1 h, 1 step, pH 6 (chelation)	EDTA (0.1)	7.0	As	7,540	13	[233]
			Cu	4,100	41	
			Pb	64,195	44	
			Zn	55,900	38	
Batch 22 h, 2 steps (chelation)	EDTA (0.25)	8.3	As	1,200	8	[234]
			Cd	100	37	
			Pb	32,000	71	
			Zn	15,000	49	
Batch 1 h, 1 step, pH 4 (acid leaching + Cl <sup>-</sup> complexation)	HCl + NaCl (5.5)	-	Pb	2,730	65	[235]
Column: three steps with HCl(0.2 M)	HCl + CaCl <sub>2</sub>	5.6	Cd	34	75-80	[224]



+ CaCl <sub>2</sub> (1.9 M) at pH < 1 followed by 7 steps with CaCl <sub>2</sub> (2 M) at pH 6.5 (acid leaching + Cl <sup>-</sup> complexation + ion exchange with Ca <sup>2+</sup> )			Zn	3,600	75-80	
			Pb	16,000	90-95	
Batch 1 h, three steps, pH 4.5 (acid leaching)	HCL (1)	7.4	Pb	65,200	35	[231]
			Cd	52	56	
Batch 1 h, one step, pH 4 (acid leaching)	HCl (2)	7	As	7,540	92	[233]
			Cu	4,100	42	
			Pb	64,195	57	
			Zn	55,900	67	

### 6.3.5. HMs removal using nanomaterial or nanoremediation

Nanoremediation is a new technology in which nano-sized particles (diameter < 100 nm) are utilized to remediate polluted water and soil. Currently, this technique is in its nascent stage [236]. The nano-particles are utilized most frequently in this technique for the adsorption of HMs: AC and carbon nanotubes (CNT), graphene, MnO, ZnO, and TiO<sub>2</sub>, MgO, and Fe<sub>2</sub>O<sub>3</sub> [237]. Nano-particles used as an adsorbent for heavy metals removal should be nontoxic, possess high adsorption capacity, adsorb pollutants in less concentration (ppb), easy to remove the adsorbed pollutants and should be recyclable for several times application [238]. For example, modified ZnO nano-adsorbent has unique micro/nanostructure compared to commercial ZnO, resulting in high removal of Cu (II) than unmodified ZnO [239]. Moreover, various nano-assemblies were also used for removing various HMs (e.g., Co<sup>2+</sup>, Ni<sup>2+</sup>, Cu<sup>2+</sup>, Cd<sup>2+</sup>, Pb<sup>2+</sup>, Hg<sup>2+</sup>, and As<sup>3+</sup>) at 30 °C and 24 h for better efficiency near to 100% [240]. Kumar et al. [241] reported that mesoporous hierarchical ZnO nano-rods have high removal efficiency of Pb<sup>2+</sup> (160.7 mg/g adsorbed) and Cd<sup>2+</sup> (147.25 mg/g adsorbed) from wastewater. Gupta et al. reported modified Fe<sub>2</sub>O<sub>3</sub> nano-particles shows a high affinity for removing different pollutants such as Cr<sup>3+</sup>, Co<sup>2+</sup>, Ni<sup>2+</sup>, Cu<sup>2+</sup>, Cd<sup>2+</sup>, Pb<sup>2+</sup> and As<sup>3+</sup> [242]. Also, modified MnO has a good surface area (100.5 m<sup>2</sup> g<sup>-1</sup>), showed higher adsorption of various HMs such as As<sup>3+</sup>, Cd<sup>2+</sup>, Pb<sup>2+</sup> and Zn<sup>2+</sup>. Adsorption occurs on MnO, usually due to the inner-sphere formation mechanism by the ion-exchange process [242]. Carbonaceous material, CNT shows excellent adsorption capacity of HMs. Several studies reported the removal of Pb(II), Mn(II), Cu(II) by using MWCNTs [243, 244]. Graphene and functionalized graphene oxide shows a very high efficiency for the removal of HMs from wastewater [245-247]. Nanomaterials have higher selectivity on adsorption process and able to adsorb very efficiently comparing to other materials, however, a large scale application of nanomaterials still challenging considering the production of friendly and inexpensive nanomaterials for the process.

The nanoremediation of HMs using nanoscale zero-valent iron (nZVI) particles has been widely utilized and considered ideal candidates to remediate HMs from e-waste [248]. For example, zeolite-aided nZVI nanoparticles have been utilized for the removal of Cd (II), Pb(II), and As (II) with the adsorption capacity of 48.63 mg/g, 85.37 mg/g and 11.52 mg/g, respectively [160]. The effectiveness of nZVI for immobilizing metal(loid)s in soil mainly depends on different factors such as soil properties, metal characteristics, other metal(loid)s presence, and the dose of nZVI [249]. Li et al. [250] investigated the applications of nanoscale nZVI for the removal of metal cations in water. They found the removal efficiency of 36.5% Cd(II), 71% Ni(II), 92.5% Zn(II), 97.5% Cr(VI), 99.7% Cu(II), 99.8% Ag(I) and 99.7% Pb(II) where 5 g/L nZVI were loaded

for all experiments and reaction time was 3 h [251]. Therefore, nZVI is a very potent material for the removal of many HMs. However, more focus should be given to the practical application on a large scale to determine the scalability of the nanomaterials applications for HMs removal.

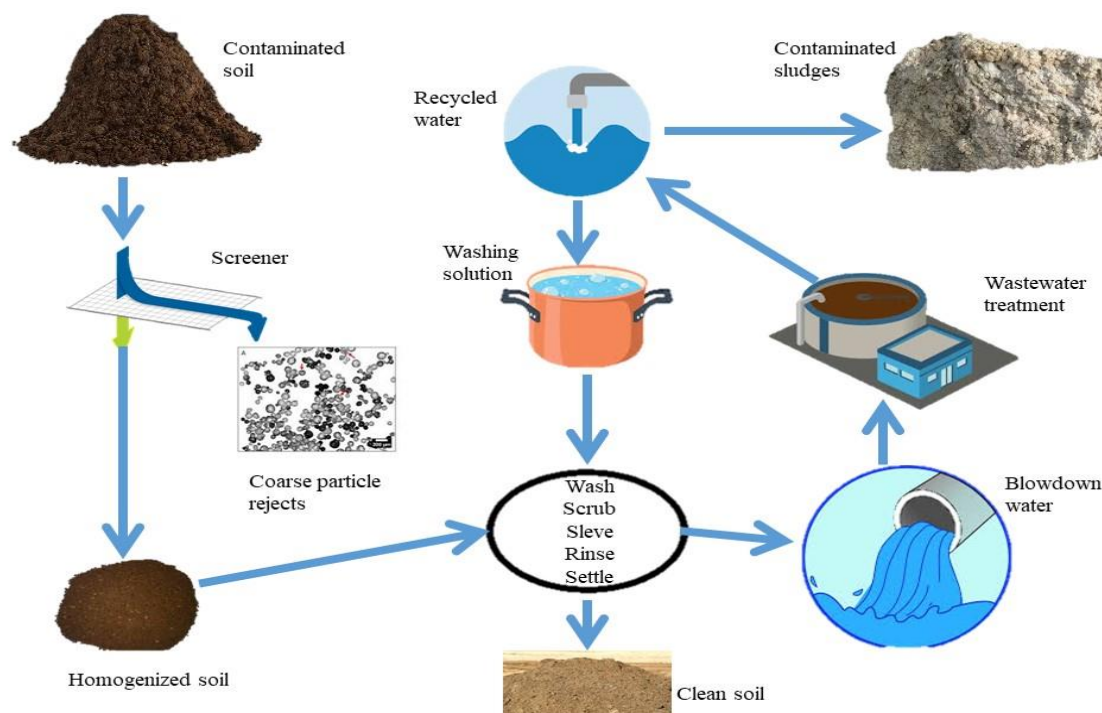
## 6.4. Hybrid treatment processes

Various treatment processes discussed above used to remove HMs for achieving better treatment efficiency. However, they might still face some difficulties like *in-situ* treatment failure and expensiveness. That's why the popularity of hybrid processes is increasing day by day because higher efficiency in various environmental matrices is gaining popularity. For achieving a synergistic and effective effort to remove HMs, hybrid or integrated processes work as a combination of two different methods [252].

### 6.4.1. Physico-chemical treatment technologies

#### 6.4.1.1 Soil Washing

The soil washing remediation technique is used to remove contaminants (HMs) from soil particles, which is based on physical and chemical processes [253]. The physical soil washing depends on various contaminated soil factors such as particle size, settling velocity, specific gravity, surface chemistry, and magnetic properties. Moreover, in chemical soil washing, the aqueous solution of acid, alkali, complexants, surfactants, or other solvents is used. However, to obtain effective results, a combination of both physical and chemical processes are necessary, as shown in **Figure 8**.



**Figure 8.** A typical process diagram of soil washing for removing HMs contaminants (Reproduced from [254])

Different chelating solutions are used in the soil washing process, such as EDTA, sodium persulfate and citric acid. Wuana et al. [255] reported the removal efficiency of HMs from soil

using organic chelating acids in the order of EDTA > citric acid > tartaric acid with yields of HMs in sequence Cu > Ni > Zn > Cd > Pb. Another study by Abumaizar and Smith [256] reported the removal efficiency of Cd, Zn, Cr, Pb was 70, 30, 25 and 10%, respectively, using the soil washing (column test) method at neutral pH. Ko et al. [257] found 63–75% removal of As, 59–60% removal of Zn, and 38–45% removal of Ni by using pilot-scale remediation of soil washing [257].

In summary, this process's merits are low acidic media, high removal capacity, cost-effectiveness and less requirement of washing liquid. But there are some drawbacks of this process such as (i) highly bound metal ions on soil particles, (ii) density of metal-contaminated soil particles, (iii) surface morphology, (iv) existence of contaminated metals in all type of soil particles, (v) different chemical forms of metals, and (vi) present of the high amount of humid in the contaminated sites [258].

#### **6.4.1.2. Permeable reactive barriers (PRB)**

The principle concept behind PRB is that in the flow path of a plume of contaminated water, a permanent, semi-permanent or replaceable reactive media is placed where water must move through the media. Permeable reactive trenches remove contaminants from contaminated water by degrading, precipitating, adsorbing or transforming processes [259].

##### **6.4.1.2.1 Sorption process in PRB**

Different technologies are used in the sorption process at PRB. Red mud at PRB is carried out from the bauxite during the Bayer process, containing fine particles of iron, aluminium, silica, cadmium, titanium oxides, and hydroxides [260]. It was found that red mud has high surface reactivity and the ability to remove HMs from wastewater [260] and acid mine drainage (AMD) [261]. For example, Brunori et al. [262] experimentally observed that the metal trapping ability of red mud was 35% of As from the initial concentration of 230 mg L<sup>-1</sup> and can be removed As up to 70%. Activated carbon and peat are mostly used in PRB because of their high adsorption capacity of organic and inorganic contaminants from water. They have a higher surface area and surface functionality [263]. Han et al. [264] reported that granular activated carbon (GAC) is more effective in removing Cr in PRB. The main advantage of GAC is that it can be regenerated by phosphate extraction and acid washing [264]. The activation of peat using an alkaline agent helped to increase the removal efficiency of HMs. For example, NaOH-activated peat showed 100% removal of Cd, where the non-activated peat resulted in 75% removal of Cd [265]. On the other hand, zeolites are mainly tectosilicate minerals with 3D aluminosilicate structure, which widely used in PRB because of their high ion-exchange, adsorbing, and sieving capacities [266].

##### **6.4.1.2.2 Chemical precipitation in PRB**

In PRBs, the reactive chemical agents can precipitate the contaminants by modifying the pH, and redox conditions of the contaminated solution. In this process, ferrous salts, phosphate, limestone, lime, fly ash, chemicals such as Mg(OH)<sub>2</sub>, MgCO<sub>3</sub>, CaCl<sub>2</sub>, CaSO<sub>4</sub>, and BaCl<sub>2</sub>, and zero-valent metals can be used as chemical reactive materials. Due to the change of environmental conditions, the immobilized contaminants and toxic degradation intermediates might be remobilized [267]. Jun et al. [268] investigated laboratory-scale A and B in PRBs for treating contaminated groundwater. HMs removal was 46.7 to 93.2% for reactor A and 58.7 to 99.6% for reactor B. The maximum adsorption capacity of BFS (Blast Furnace Slag) for As(III) remediation was 1.40 mg/g, where the initial concentration of As(III) was 1mg/L [269, 270].

##### **6.4.1.2.3 Biological barriers in PRB.**

For the microbial transformation from hazardous compounds, this technology is mainly used in engineered passive bioreactors. Many research studies have been investigated to change the redox conditions or provide substrates, which helps the natural bio-degradative systems [271-

274]. Biological reactive zones mainly depend on dissolved nutrients, injected nutrients, and the delivery of nutrients to support the biodegradation of contaminants, which passes through the barrier. Additionally, periodic replenishing of the media may become necessary. The efficiency of in-situ bio-barrier can be hampered by bio clogging, which decreases water saturation and hydraulic conductivity [275].

#### **6.4.1.3 Ultrasonic leaching**

This is the remediation technique of HMs where acidic solvent with contaminated substance is used during sonication and fragmentation. In this technique, high acidic (inorganic acids  $H_2SO_4$ ,  $HCl$ , and  $HNO_3$  help to maintain pH in between  $\sim(1.5-2.0)$ ) media is necessary [258]. In the ultrasonic leaching process approximately 95% of Cu, 82.2% of Zn and 87.3% of Pb were solubilized when the solution pH was 0.75 [276]. To remove HMs with better efficacy in the electrokinetic process, low-frequency ultrasound (generated by transducer) was necessary to activate the raw municipal solid waste incineration fly ashes [277]. In sonoelectrical process, ultrasound cleans the electrode surface, increase the mass transport, decrease the viscosity of pore fluid, improve the porosity and hydraulic conductivity, and eliminates the migration barriers during the electrokinetic process in HMs removal [278, 279]. In the ultrasonication test, 69.84% of Zn, 64.24% of Pb, 67.74% of Cu and 59.93% of Cd was removed at 45 °C for 30 min acoustic time correspondingly in sonoelectrical process leaching concentration of Zn, Pb, Cu and Cd were reduced by 85.92%, 98.22%, 88.53% and 98.34%, respectively, from the raw sample [277].

#### **6.4.2. Physiobiological methods**

##### **6.4.2.1 Bio-electrokinetic method**

This process is mainly suitable for soil remediation, where microbial and electrokinetic processes are used together to detoxify the contaminated soil [280]. The integrated approach of bioleaching and electrokinetic remediation is advantageous because, at the same time, HMs dissolved as an ionic solution and accumulate on the electrode [281]. The bio-electrokinetic method was applied to remove Cu and Zn, and it was found that this process can remove 78.61% of Cu and 99.11% of Zn [281]. Another study showed that combining bioleaching and Fenton-like reaction has better efficiency than the bioleaching and acidic leaching process. This combined process was used to treat samples under acidic conditions and the removal of Cu, Zn, Pb, and Cd reach up to 75.3%, 72.6%, 34.5% and 65.4%, respectively [282]. The combination of this technology is not well explored, however, this technology considered as a high efficient, feasible and rapid process for decontamination of sewage sludge.

##### **6.4.2.3 Immobilized biosorption**

Bacteria, yeast, fungi and algae are the common biomasses from activated sludge, so soil or fermented wastes have some functional chemical groups (e.g., carboxyl, amino, hydroxyl, phosphate, sulfhydryl and sulfate) responsible for biosorption. Comparing with the conventional techniques, this method has many merits, such as increased biosorption performance, higher stability and reusability [283]. *Agrobacterium* biomass was encapsulated with iron oxide nanoparticles for Pb adsorption, which showed an adsorption capacity of Pd of 197.02 mg/g [284]. **Tables 5** shows various biosorbents with their uptake capacity and operating conditions for the bioremediation of HMs. Other organisms (e.g., crustaceans, seaweeds, and moss) and agricultural waste products such as tea waste, whey, straw, exhausted coffee and defatted rice bran are used as biosorbents. Biosorption of Cu in *Bacillus sp.* (69.34% in immobilized cell and 44.73% in dead cell), Cd in *Pseudomonas sp.* (90.41% in immobilized cell and 86.66% in dead cell) and Pb in *Micrococcus sp.* (84.27% in immobilized cell and 79.22% in dead cell) were found in a comparative study [285].

**Table 5.** Effect of different experimental parameters in HMs remediation by biosorbents.

Heavy metals	Biosorbents	pH	Temp (°C)	Time (h)	C <sub>0</sub> (mg/L)	q <sub>m</sub> (mg/g)	Ref.
Cr	<i>Spirulina platensis</i> extract beads	2	24	24	100	41.2	[286]
Pb	SiO <sub>2</sub> nanoparticles immobilized <i>Penicillium funiculosum</i>	5	25	0.33	20–100	262.2	[287]
	<i>Arthrospira platensis</i> cells immobilized in sodium alginate	4	27	1	500	424	[288]
	<i>Halomonas BVR 1</i> immobilized alginate	8–10	27	2	75	9.68	[289]
	MNPs–Ca-alginate immobilized <i>P. chrysosporium</i>	5	35	8	200	176.33	[290]
Cu	Magnetic calcium alginate hydrogel beads(m-CAHBs)	2	27	6	259	159.24	[291]
Hg	<i>Phoenix dactylifera</i> biomass alginate	7	35	3	100	46.73	[292]
Cd	<i>Chlorella</i> –biochar immobilized complex	6	26	48	100	217.41	[293]
Ni	<i>E. coli</i> ATCC 29522-aided bentonite clay	5	37	24	100	58.82	[294]
Zn	Nano-Se immobilized <i>Phanerochaete chrysosporium</i>	6.5	30	96	40	13.9	[295]

### 6.4.3 Chemical-biological remediation approach

Compared to individual process, chemical and integrated biological process have more efficiency for heavy metal-containing wastewater treatment [296, 297]. For the remediation HMs from contaminated sites, chemical–biological integrated treatment is considered to overcome a single method [298]. A study by Ahmed et al. [298] showed 99.3% recovery of Cr (III) and 98.4% recovery of Cr(VI) from tannery waste by using a combined chemical and biological process. Another study on the integrated chemical and biological treatment process could reduce 62.33% of total Cr(VI) using *Fusarium chlamydosporium* [299]. HMs removal from AMD has been realized by selective sequential precipitation (bacterially produced H<sub>2</sub>S helps bacterial production and the precipitation of the metals) of metals using this technique [300].

Although chemical-biological integrated treatment showed promising result on HM remediation but suffer from several limitations such as (i) long acclimatization time, (ii) produce toxic by-product and (iii) changes in the biodegradable efficiency and generation of a large amount of sludge [301]. However, selective recovery of HMs with 99% removal efficiency is possible [300], also scaling up of this process is possible using a continuous system.

### 6.5. Comparison of different technologies

Each HMs remediation processes discussed above have its advantages and disadvantages in terms of removal efficiency, cost, environmental impact and operational flexibility. However, some of the problems could be overcome by employing an integrated process rather than a single process. The integrated processes have some advantages in terms of effectiveness, operation duration, environmental impact, adaptability, cost, versatility, large scale operation. The researchers reported integrated or combined treatment processes as an efficient treatment process for heavy metals from contaminated soil, sediment and water [281, 307].

The biological remediation processes consist of bioleaching, composting, phytoremediation, and microbial remediation as discussed earlier. All of the processes are used in modern science for their benefits such as low cost, no use of chemicals, environmentally friendly, high efficiency, ease to operate and no secondary pollution [276, 309, 310]. The Bioleaching process exhibited higher removal of HMs (64-90 %), but the limitation is time-consuming whereas the composting process showed lower removal efficiency of HMs (42-72%) and the public acceptance is also low (**Table 6**). Furthermore, microbial remediation and phytoremediation are long-term effective processes because they are harmless natural processes and also have several advantages over other bioremediation processes. Notwithstanding, bioremediation (Phyto/microbial remediation) processes also have some limitations, but these problems can be overcome or minimized by proper management and integrating them with other processes. An example is by using metal transporters, the production of antioxidative enzymes, and metal-detoxifying chelators of hyperaccumulator plants are increased [322]. Compare to other technologies the physical treatment technologies are widely used remediation process of HM because of low cost, simplicity, eco-friendly than other methods. But in many cases, physical methods showed poor removal of HMs and may not able to remove the HMs completely. Among all the technologies which are described above (**Table 6**), thermal remediation (94-99 %) and the use of biosurfactants (>70 %) are most effective in the remediation of HMs from solid contaminants (soil, sludge, sediment). On the other hand, MBR (60-99 %), RO (95-99 %), biosorption by cellulosic materials and agriculture wastes (>90 %) technologies are also effective.

Usually, most of the chemical technologies has high efficiency to remove HMs. However, the sludge or pollutants generated during the chemical remediation process requires further treatment. This process is also costly than other technologies. On the above discussed chemical technologies (**Table 6**), immobilization (21-60.5 %), and electrokinetic (50-100%) have high public acceptability due to their advantages over others process. Nanoremediation technology is an advanced and effective technology and has a high remediation capacity (60-100%) for HMs remediation, but it is applicable only on a small scale. These technologies are mainly applicable for controlling large plumes of contaminants in a large area. But the main problem of this process is to make secondary pollutants for using toxic chemicals. To mitigate this problem, further treatment processes are necessary after chemical treatment processes.

These days hybrid treatment process offers advantages over a single process due to higher efficiency in remediating HMs [340]. Among the discussed hybrid technologies (**Table 6**), PRB technology, bio-electrokinetic technology, ultrasonic leaching, immobilize biosorption, SMFCs and other combining methods have high removal capacity (>90 %), cost-effective and eco-friendly. And from all of them, bio-electrokinetic and the combination of bioleaching and electrokinetics are the most effective technology because of their high uptake capacity of HMs from contaminant soil, sediment or water [335]. In physio-chemical treatment technologies, PRBs have the most removal capacity and mainly suitable for groundwater remediation. This process is mainly depending on sorption processes, biological reduction processes and precipitation processes. The combination of some physical processes such as MBR with RO, MBR with the electrochemical process, or the combination of UF, NF and RO have better efficiency without using chemicals [315]. But these combined processes are mainly applicable for the remediation of HMs from liquid contaminants.

Eventually, all of the remediation technologies have some drawbacks with potentiality. Hitherto, coming novel technologies should put the top of the table to achieve the highest HMs removal from electronic waste.

**Table 6:** An overview of different treatment processes for HMs remediation.

	Remediation Technology	Remediate Substances	Removal Substances	Removal Capacity	Advantages	Disadvantages	Acceptance	Reference	
<b>Biological Processes</b>	Bioleaching	Soil, Sediments	Zn, Al, Cu, Cd, As, Pb, Ni	64-90 %	Low cost, No use of chemicals, Environmentally friendly, high Efficiency	Time-consuming, limited resources	Medium to high	[134, 268]	
	Composting	Especially for soils	Mn, Fe, Zn, Cr, Cu	42-72 %	Eco-friendly, self-stabilization process	Lengthy, less efficiency	Very low	[28, 308]	
	Phytoremediation	Soil, Water (small scale), Sediments	Pb, Zn, As, Cd, Cu, Ni, Cr, Mn	20-31,000 mg/kg	Eco-friendly, more aesthetically pleasing, cost-effective, easy to implement and maintain, does not require expensive equipment, disposal sites are not needed	Less efficient, Induce secondary pollution, contaminants may be transferred to the food chain, Time consuming. Only Hyperaccumulator plants are used.	Highest public acceptability	[276, 309-311]	
	Microbial Remediation		Mn, Ni, Zn, Cd, Cu, Au, Ag, Co, Hg, Pb, As, Th, U	0.27-11,00 mg/g	High efficiency, easy to operate, high adaptation power of microorganisms	HMs can be released back after decomposition,	Very high public acceptability	[140, 142, 276, 309]	
	Thermal Remediation	Soil, Sludge, Sediments	Hg, As, Cu, Cd, Fe, Ni, Zn, Pb	94-99 %	Short time, no chemicals are needed, high efficiency, reduce the radioactive wastes	Only applicable for solid contaminants, requires high temperature, costly, produce toxic gases.	Medium	[147, 148, 150]	
	Ion Exchange	Water, Co-contaminated soil	Pb(II), Hg(II), Cd(II), Ni(II), V(IV,V), Cr(III,VI), Cu(II), Zn(II)	40-99.5 %	High removal capacity, simple and effective method, selective removal of ionic contaminat	Costly. Recharge or regeneration of materials	Medium to high public acceptability	[156, 158, 312]	
	Adsorption by Activated Carbon	Mainly water contaminant	As(III), As(V), Sb, Cr(VI)	89-97.95 %	High removal capacity, eco-friendly	Highly dependent on particle size distribution, produce large quantity of solid waste	High for water	[165, 167]	
<b>Physical Process</b>	Membrane & Filtration	Microfiltration & Ultrafiltration	Water	Zn, COD, TC, DOC, IC	19-93 %	Simple process for pretreatment of other processes, cost-effective	Applicable for specific particles, membrane clogging;	Low	[313-315]
		Reverse Osmosis (RO)	Water, Drinking water	Ionic contaminants, Dissolved organic compounds	95-99 %	High efficiency, produce high-quality water.	Only for liquid contaminants, costly method. Membrane fouling	High for water	[178, 316]
		Membrane Bioreactor (MBR)	Wastewater, Drinking water	COD, BOD, SS, HM	20-99 %	Excellent effluent quality, good disinfection capability, shorter HRT, longer SRT, less sludge production, higher volumetric loading, reduced footprint and sludge production, process flexibility toward influent changes, and improved nitrification	Costly, membrane fouling, aeration, excess sludge and phosphorus production	High for water	[317, 318]
		Solidification	Soil, Sludges, Sediments	Fe, Mn, Zn, Cd, Cu, and Pb	21.2-60.5 %	In-situ simple and cost-effective method	Additives are necessary, low removal capacity, only applicable for solid contaminants and some HMs removal	Low	[193, 194]
	Replacement Method	Soil, Sludges,	All type of contaminants	-	Effective for small scale	Only applicable for small amounts of contaminants, highly costly	Very low: limited to	[276, 319]	



Chemical Processes			Sediments, water					highly contaminated soils	
	Sorption materials & Wastes	by Cellulosic & Agriculture	Water	Pb, Cd, Cr, As	71-96 %	Eco-friendly, cost-effective, apply wastage substances	Applicable for some functional groups, pH dependent	Medium	[197, 198, 320]
	Removal by Biosurfactants		Soil, Sludge, Water	Cu, Zn, Cd and Ni	68.1-72.3 %	Eco-friendly, cost-effective	Applicable for limited HMs removal, Only some microbes can produce surfactants	Low	
	Immobilization Techniques		Soil, Sludges	Fe, Mn, Zn, Cd, Cu, and Pb	21-60.5 %	Simple and very rapid process, investment and operational cost is comparatively low, acceptability of public is high	High ecological risk, a significant amount of solid wastes are generated, a toxic byproduct produced, permanent monitoring is necessary	High public Acceptability but harmful	[193, 194]
	Precipitation		Soil, Sludges, water	Cd, Ni, Cu, Zn, Cr, Pb	65-90 %	Nontoxic operations, cost-effective, simple process	Secondary waste generation, settling of sediments are prolonged and poor; different metal precipitates are aggregated, the sludge degradation rate is slow	Medium	[209, 212]
	Electrokinetic Remediation		Water, Soil, Sludges,	As, Cd, Cr, Co, Hg, Ni, Mn, Mo, Zn, Sb, Pb	50-100 %	Highly efficient in removing metals; applicable to a wide range of metals	The process depends on soil pore, water current density, grain size, ionic mobility, pH, contaminant concentration and total ionic concentration	Medium to High	[151, 321, 322]
	Chemical Leaching		Soil, Sludges, Water	Pb, Cd, Zn, As, Cu, Cr	26.9-98 %	Highly efficient, , Cost-effective.	Environmental risky, acids and toxic chemicals are needed	High but harmful	[256, 323, 324]
Hybrid Processes	Nano remediation		Water, Soil, Sediments	Cr, Co, Ni, Cu, Cd, Pb, As, Hg, V	60-100 %	New technology with high performance, the ability to adsorb pollutants in less concentration (ppb), non-toxic technology	Several times recycling is necessary, high cost.	Very High	[242, 325, 326]
	Soil Washing		Soil, Sludges	Cd, Zn, Cr, Pb, Cu, Ni, As	10-75 %	Working under low acidic conditions, less requirement of washing liquids, low cost, simplicity of the process	Low efficiency, chemicals are necessary, high amount of humic production, harmful for the environment, applicable for a small amount of soil/sludge	Medium to high public acceptability	[255, 256]
	Permeable Reactive Barriers (PRB)	Red mud	Water, Wastewater	Pb, As, Cd, Zn	30-70 %	Cost-effective; high sorption capacity.	Sorbs cations with lesser ionic radii; depends on pH	Low	[262, 327]
		Activated Carbon and peat		Cr, Cd and other heavy metals	> 90 %	High adsorption capacity; regeneration possible; acts better when coupled with microbes	More field-scale studies on inorganic and metal adsorption are needed	High	[263, 265]
		Zeolites		Cd, Cu, Ni, Cr, As	40-80 %	Very high adsorbing capacity; hundreds of natural zeolites are available	Selective adsorption capacity	Medium	[328, 329]
	Iron sorbents (ZVI and pyrite)		As(III), As(V), Hg	98 %	ZVI and pyrite are cheap; handling is easy	As gets released in the presence of silicate and phosphate in aquifer or soil	High	[330, 331]	

Chemical Precipitation in PRB		Cr, As, Cr, Ni, Pb, Mn, Se, Co, Cu, Cd, Zn, Ca, Mg, Sr and Al	47.7-99.6 %	A cheap method.	Highly sensitive to pH and presence of other organic materials, clogging of the barrier by metal hydroxides and carbonates	Medium	[268-270]
Biological Barriers in PRB		Fe, Ni, Zn, Al, Mn, Cu, U, Se, As, V, Cr	50-95 %	Removes both divalent and trivalent heavy metal species, able to treat mixtures of contaminants (nitrate, organic and heavy metals) together	A steady supply of nutrients should be provided to sustain the microbial population, PRB should provide C, N and P for the growth and reproduction of microbes	Medium to high	[332-334]
Ultrasonic Leaching	Soil, Sludges, sediment	Zn, Pb, Cu and Cd	60-95 %	High removal efficiency	An acidic solvent is needed, costly	Medium	[276, 277]
Bio-Electrokinetic	Soil, sludge, water	Zn, As, Pb, Cd	85-99%	Applicable to a wide range of metals, better efficiency, enhance the recycling utilization of HMs and eliminate their danger	Costly method, complex method	High	[276, 280, 335]
Immobilized Biosorption	Soil, Sludges, water, Industrial effluent	Cr, Pb, Hg, Zn, Cu, Cd, Ni	9.68-262.2 mg/g	Wastes are used as bio sorbent, high affinity and selectivity toward HMs	pH and temperature-dependent, difficulty in post-operation recovery	High	[283-285]
MBR with RO/Electrochemical Process	Water, Drinking water	COD, TP, TN, Ni, Fe, and Cr	80-99 %	High removal efficiency, less membrane fouling	More costly	Very High	[304, 336]
Combination of Persulfate Oxidation and Chemical Washing	Soil, Sludges	Cu, Pb, Ni, polychlorinated biphenyl, polycyclic aromatic hydrocarbons, polybrominated diphenylethers	59.4-97.1 %	High removal efficiency, cost-effective, high public acceptance	Various chemicals are needed, only applicable for solid contaminate substances and limited HMs	Very High	[337]
Sediment microbial fuel cells (SMFCs)	Sediments, Soil	Cr(VI), Cd(II), Cu(II), U(VI), V and Zn(II)	>90 %	Production of renewable energy, cost-effective	Mainly applicable for sediments, oxidized sediment environment is required to reduce the production of methyl mercury and efflux	High	[338, 339]

## 7. Gaps and future recommendations

The production of e-waste is increasing day by day. This resulted in the increase of heavy leaching from e-waste to the soil, sediment and water, which is a global problem. Thus, an appropriate control measure is required on the production and disposal of e-waste. Furthermore, remediation technology is necessary to mitigate the contaminants from e-waste, especially heavy metal present in the e-waste. The remediation techniques which exist for HMs remediation from their environments have some definite outcomes. Such as:

- (i) Complete or partial degradation of contaminants from the environment
- (ii) Removal of pollutants for further treatment or settlement
- (iii) Stabilization of contaminants from highly toxic to less toxic
- (iv) Separation of non-contaminated substances from the pollutants
- (v) Disposal of the polluted materials to the broader environment with restrictions
- (vi) Long term effectiveness of remediation technologies with better efficiency

The acceptance of different remediation technologies mainly depends on various factors such as area of contamination, cost requirement for remediation technology, quantity and quality of contaminants. The existing technologies used for HMs remediation are described in **Table 6** with their performance, advantage, disadvantage and acceptability. The physical processes are not so effective, and the chemical processes have high HMs removal efficiency but not eco-friendly. Recently, hybrid treatment technologies are most popular in HMs remediation due to their effective performance over the single treatment processes. But the proper combination of methods is necessary for hybrid treatment, which mainly depends on several factors. For example, when phytoremediation is used to combine several traditional remediation techniques (chelate assisted, microbial assisted, and transgenic plants), effective outcomes are achieved.

## 8. Conclusion

Including valuable metals, hazardous metals are increasing in the environment, which is alarming for future generations. However, we can not stop the emerges of modern technologies. It is expected that the developments of new technologies (for example, artificial intelligence technology, robotics, biomedical sensors and so on) will be continued in future generation also. Hence, it is impossible to ignore modern technologies; however, we can develop recycling management systems for e-waste by providing sustainable consumption and production patterns of e-material and avoiding using hazardous materials (e.g., using biodegradable plastics) where possible when producing electrical appliances. Some HMs used in electronics are highly toxic and harmful for living organisms and the environment. When they get into the soil sub-surface, sediments and water, then they form complexes. Thus, it is challenging to handle HMs due to their complex formation and their harmful environmental and animal impacts. Many techniques have been applied to remediate HMs from their environments. Among all methods discussed in this review, biological and hybrid treatment processes have high removal capability and high public acceptability. Furthermore, new concepts for HMs remediation are used as single or combined processes, and they are upcoming and promising technologies. Meanwhile, future studies of HMs remediation are not only limited to available technologies. It is important to explore new methods from other process objects based on similar characteristic for better performance than existing technologies.

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