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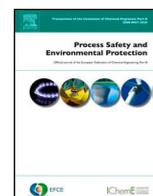


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Metals in e-waste: Occurrence, fate, impacts and remediation technologies



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ABSTRACT

Electronic waste (e-waste) is generated from the discarded electronic products. The generation of e-waste has increased significantly in the recent decades. Globally, the increased rate of e-waste generation is almost 2 metric tonnes (Mt) per year. It is estimated that about 74 Mt of e-waste will be produced in 2030. Therefore, e-waste can be a significant threat to the environment. Toxic metals (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 metals 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 different remediation technologies, the combination of biological, physical, and chemical processes shows relatively high removal efficiency; and they possess multiple advantages over other remediation technologies. Finally, this review also includes future outlook on e-waste management and remediation technologies.

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1. Introduction

Electronic wastes (e-wastes) are the waste of the discarded electrical appliances or electronic products like refrigerators, washing machines, fans, televisions, air conditioners, cell phones and computers (Rautela et al., 2021). 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. The global production of e-waste has become one of the significant issues due to the considerable demands of electronic products in human society (Fig. 1). Among different electrical consumables, the rapid growth of computing and communication devices are mostly responsible for the global boom of e-waste production (Mmereki et al., 2016). Other

reasons for the massive generation of e-wastes are (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 be increased by ~50% in 2030 (from 33.8 Mt in 2010–74.7 tonnes in 2030) (Fig. 2a). E-waste mainly contains metals (60%), plastics (15%) and metal-plastic mixture (5%) (Ari and Vidyadhar, 2016; Widmer et al., 2005). When the amount of toxic metals such as lead (Pb), mercury (Hg), arsenic (As), cadmium (Cd), selenium (Se), and chromium (Cr) exceeds the permissible level in the environment, it may pose risk and hazards to the human and the ecosystem by direct or indirect contact. Waste household equipment (about 45%), information and communications technology equipment (33.9%),

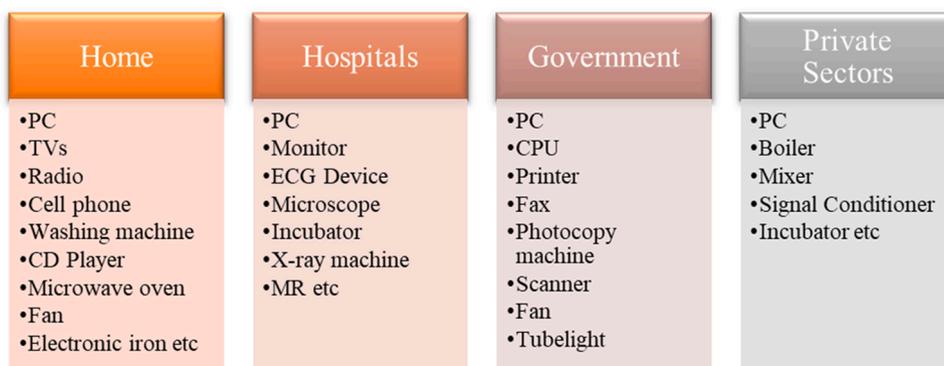


Fig. 1. Composition and major sources of e-waste.

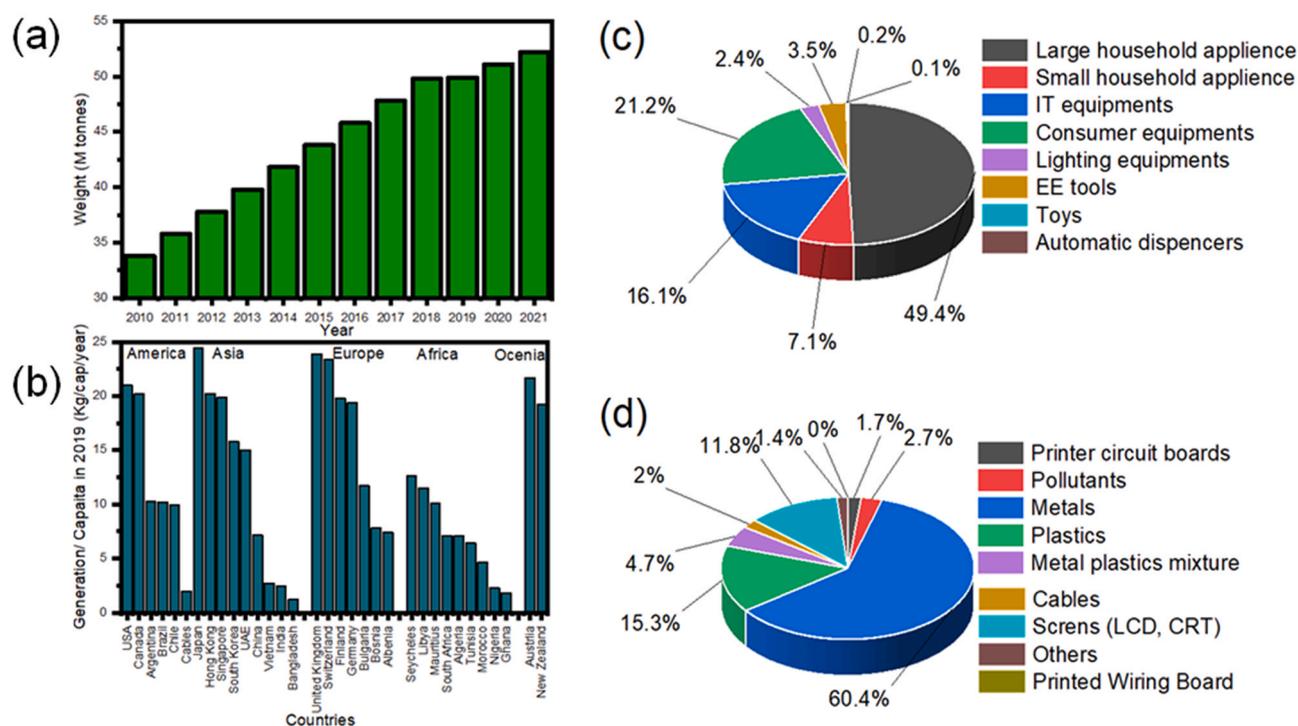


Fig. 2. Global e-waste generation scenario over the period. (a) Global quantity of e-waste generation (2020–2030 are estimated) (b) e-waste generation per capita in 2019 in different countries. (c) Composition and major sources of e-waste; average composition (c) and materials fractions of E-waste. (a–b) (data adopted from [Baldé et al., 2015](#); [Borthakur et al., 2019](#); [Forti et al., 2020](#)); (c–d) (data adopted from ([Kaya, 2016](#); [Vats and Singh, 2015](#); [Widmer et al., 2005](#)))

consumer electronics (13.7%) are the major contributor to e-waste ([Needhidasan et al., 2014](#); [Widmer et al., 2005](#)). Eight metals (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) ([Wang and Chen, 2006](#)).

The management of e-waste is one of the world's significant challenges as most e-wastes contain Metals and toxic substances ([Shittu et al., 2021](#)). 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 ([Andarani and Goto, 2014](#)). Due to rapid industrialization and the availability of high technology the production of e-waste has increased significantly. According to the International Solid Waste Association (ISWA), in 2019, approximately 53.6 metric tonnes (Mt) of e-waste was generated worldwide ([Fig. 2a](#)). In 2019, the generation of e-waste was 24.9 Mt (2.5 kg per capita) for Asia, 13.1 Mt (13.3 kg per capita) for America and 12 Mt (16.2 Kg per capita) for Europe ([Fig. 2b](#)). America and Europe are the second and third most significant contributor of e-waste generation in the world ([Andeobu et al., 2021](#)). In Asia, Japan, China and Singapore are the major producers of e-waste ([Li and Achal, 2020](#)). Whereas in Europe, U.K., Switzerland, Finland and Germany are producing almost the same quantities ([Fig. 2b](#)). Europe is recycling the highest amount of e-waste (5.1 Mt, 42.5%). On the other hand, most countries have a lower recycling rate of e-waste than the generation of e-waste. According to the statistics recycling portion of Asia is ~11.7%, while in America and Oceania, it is 9.4% and 8.8%, respectively ([Forti et al., 2020](#)). However, the actual amount of e-waste recycling may vary from region to region according to their annual income, management systems, policy and environmental laws ([Thakur and Kumar, 2021](#)). By the next ten years, the units of obsolete personal computers (PCs) in developing countries will increase by 400–700 metric tonnes (Mt) and in developed countries 200–300 Mt ([Yu et al., 2010](#)). According to Silicon Valley Toxics Coalition (SVTC) research in between 1994 and 2003, about 500 Mt PCs reached out of working

condition and became e-waste. More or less, 500 Mt PCs contain a massive amount of toxic metals (e.g., approximately 718,000 tonnes of Pb, 1363 tonnes of Cd, and 287 tonnes of Hg) ([Widmer et al., 2005](#)). 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 ([Mmereki et al., 2016](#)).

E-wastes deposited in landfills may contain metals are leached into the soil, ground water and surface water. Therefore, it may cause severe environmental hazards and human health issues ([Lin et al., 2022](#); [Long et al., 2021](#)). For example, a higher concentration of Cd causes chronic disease, kidney damage, bones and respiratory system diseases ([Pan et al., 2010](#); [Khan et al., 2020](#)). The neurodevelopment of young people can also be affected by Cr uptake ([Chen et al., 2010](#)). Diseases like cancer and skin allergies are caused by Ni electron gun of CRT (Cathode-ray tube) and in Ni-Cd batteries ([Padiyar, 2011](#)). Children are so much vulnerable to the exposure of metals, mainly Pb ([Briffa et al., 2020](#)). Exposure to even low Pb concentration can create different neurological problems such as delayed development, inattentiveness, irritability, hyperactivity, stunted growth and brain damage ([Islam et al., 2020](#)). The toxic effects of HM exposure have created worldwide attention to comply with the metals content (allowable limits) in the soil and crops ([Khalid et al., 2017](#)). Hence, proper treatment of e-waste is necessary and has become a crucial topic in solid waste management ([Song and Li, 2015](#)). Various treatment technologies of e-waste to recover metals are used, such as physical, thermochemical, pyro-metallurgical, hydrometallurgical, bio-metallurgical, and a combined method ([Chakraborty et al., 2022](#)).

Additionally, it is also necessary to deploy efficient and site-specific remediation methods, which will feasibly and efficiently remediate metalloids contaminated soils and surface water. For instance, during the last two decades, different soil remediation methods (e.g., bioleaching, phytoremediation, mycoremediation,

Table 1
E-waste sources, according to EU directives.

Category	Example	HMs content (%)	Ref.
Large household appliances	Refrigerators/freezers, washing machines dishwashers.	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)	(Li et al., 2019; Tian et al., 2012)
Small household appliances	Vacuum cleaners, kitchen machines.	Cu (18.8), Pb (4.79), Al (0.912) Cr, Cd, Ni (0.0051–0.0179)	(Kantarelis et al., 2011)
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)	(Cui and Zhang, 2008; Tian et al., 2012)
Consumer Equipment	Televisions, stereo equipment	Cu (10), Al (10) Pb (1.0) Ni (0.3)	(Hagelüken and Art, 2006)
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)	(Korfali et al., 2013; Miller and Harris, 2015)
Medical Devices (except all implanted and infected product)	Therapeutic, diagnostic and analytical equipment, massage devices, X-ray equipment, sterilizers.		

vitrification, earth-swap, soil flushing, solidification) have been developed (Chen et al., 2015; Fonti et al., 2016; Guo et al., 2006; Yaashikaa et al., 2022). These method's main objectives are to decline the total bioavailable amount of metalloids in soils and water and their frequent availability in the food cycle (Bhargava et al., 2012). Physical, chemical and biological methods or their combination (e.g. chemical, biological) are the conventional methods to remediate metalloids from contaminated soils sites.

Many investigations have been carried out on the generation of e-waste, 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 metals on both the environment and human are discussed. Furthermore, the current remediation scheme of metals generated from e-waste, their merits, drawbacks and gaps also reviewed critically. Future research directions and outlooks have also been suggested.

2. Major sources and metals content in E-waste

A significant amount of e-wastes is generated by small and large electrical and electronic equipment (EEE). Various sources of e-waste generation, composition of different material fractions and metals content of them are shown in Fig. 2c, d and Table 1 with their average percentages. The maximum amount of e-waste is generated from large household appliances (49%) and the average composition of metals in e-waste is more than 60%. Various types of metals that may present in e-waste. They are bulk elements (e.g., tin (Sn), copper (Cu), silicon (Si), iron (Fe) and aluminum (Al)); small amounts elements (e.g., cadmium (Cd) and mercury (Hg)); trace elements (e.g., 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 Fig. 2 (a, b). Most e-waste contains metals including Pb, Ni, Al, Cu, Fe, Pd, Au, and Ag. But the quantity of Fe, Cu, and Al are much higher than others element. In contrast, Au, Ag, and Pd presence is smaller than others, which are represented in ppm.(Fig. 3).

Personal computers (PCs) and mobile phones are the sources of a large quantity of total e-waste produced across the world. 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 and Pb) and precious metals (Au, Ag, and Pt). Recently produced PCB 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 S1 represents a comparative view of metal compositions in PCBs of personal computers (PCs) and mobile phones. The gold content in the cell phone is 5–10 times higher than gold ore. If this is multiplied with generated 150,000 tonnes of e-waste generated yearly, the amount becomes very attractive (Ayres, 1997). 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 Fig. 4. According to this figure, Pb, Fe, Ni and Cu are the dominant Metals in personal computers, mainly used in IDE cables, video cards, RAM and CPUs. Specifically, Pb is the dominant HM in RAM and CPUs. The total content of Pb in RAM and CPUs calculated approximately 57,000 and 27,000 mg dry kg⁻¹, respectively. On the other hand, in IDE cables, copper may consider as dominant metal (the total content of about 4400 mg dry kg⁻¹) (Komilis et al., 2013). Besides, more than 40 elements are found in mobile phones PCB, including hazardous metals (Be, Cd, As and Sb), the basic metals (Fe, Cu, Al, Ni, Zn and Sn), and precious metals (Au, Ag, Pt and Rd) which is about 19% of metals of its weight (Borthakur et al., 2019). Moreover, Al or Mg is used to produce the casing of mobile phones. However, in recent days, the use of metals is

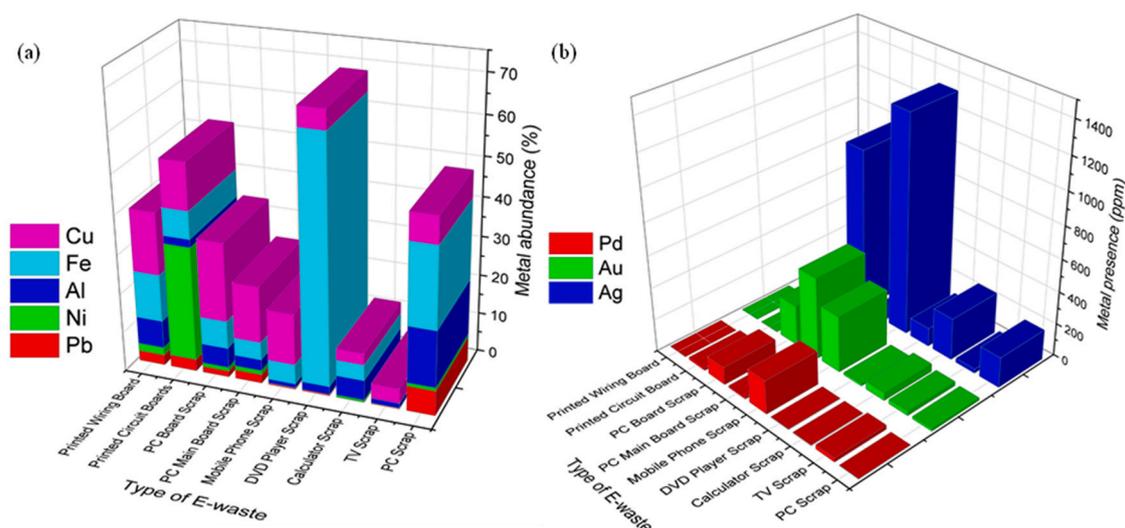


Fig. 3. Metals present in e-waste. (a) in percentage and (b) in ppm (Barbosa Jr et al., 2005; Clark and Norris, 1996; Jomova et al., 2011; Mishra and Rhee, 2010; Pan et al., 2010; Pant et al., 2012; Poon, 2008; Tsydenova and Bengtsson, 2011).

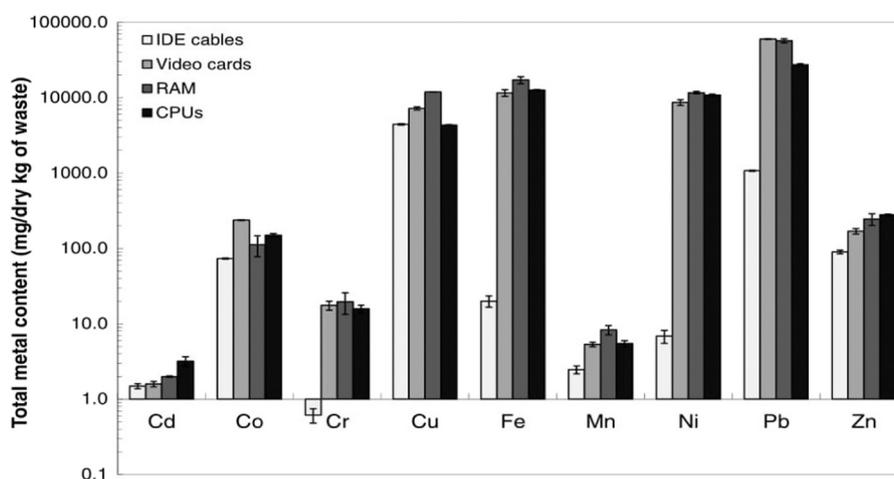


Fig. 4. Total heavy-metal content of the four PC components (IDE cables, Video cards, RAM, CPUs). (Komilis et al., 2013).

replaced by emerging materials such as carbon materials, organic materials, and many composites (Biswas et al., 2019; Biswas and Visell, 2019).

Though nowadays the use of Pb in electronic equipment is less, but other metals such Cd, Hg, Ni, Sn and some precious metals are excess amount in household apparatus, IT products and consumer equipment's. PCs and mobile phones are occupied a big portion in e-waste generation which contain Fe, Cu, Al with small amount of metals. Therefore, proper management system needs to adopt before dumping in environments.

3. Metals releases, biodegradation and bioavailability from e-waste

3.1. Metals released from e-waste

There are various sources from which metals enter into the environment. The intensive uncontrolled processing of e-waste has resulted in the release of large amounts of metals in the local environment and caused high metal concentrations in the surrounding air, dust, soils, sediments and plants (Song and Li, 2014). The common extraction processes of precious metals from e-waste, i.e.

strong acid leaching and open burning of dismantled components, have led to emit a huge amount of toxic metals and organic pollutants to the surrounding environment (Bi et al., 2010; Birloaga et al., 2013; Gullett et al., 2007). Metals are also released during the mining and extraction of different elements from their respective ores and return to the land through dry and wet deposition (Khaliq et al., 2014). Geologic locations, mainly e-waste recycling areas, have high metals releasing rates to the surrounding environment. Longtang and Guiyu in South China, are the example of the most famous e-waste recycling areas in the world where typical pollutants especially metals are released into local aquatic and terrestrial ecosystems, soils or sediments (Leung et al., 2008; Luo et al., 2011; Wong et al., 2007). Furthermore, rainfall type, intensity and pattern, temperature, wind and pH are the main factor that can regulate the metals release in the environment (Adesokan et al., 2016).

Developed countries have replaced massive amounts of obsolete electronic equipment's and home appliances with newer versions and cause a massive deposition after their lifespan in the environments by the various physical, chemical, physicochemical processes (e.g., dry and wet deposition). Approximately 70% of the metals in municipal solid waste landfills are estimated to come from electronics discards (Townsend, 2011). Uncontrolled burning, disassembly,

and disposal of untreated e-wastes cause a distribution of 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 (Fig. S1) shows the release of metals into the biosphere comprising air, soil, and aquatic environmental systems (Vincent, 2014). This is likely due to natural and anthropogenic processes and their accumulation in flora and fauna and the flow cycle of metals. Especially, people who are living in the e-waste recycling or processing area pose a significant risk to their health and nervous system (Awasthi et al., 2016).

Metals 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 concentration of metals such as Cu and Pb (Gullett et al., 2007).

The non-standard and crude e-waste recycling activities are the major sources of environmental pollutants (e.g., metals, polybrominated diphenyl ethers (PBDEs)) in e-waste recycling sites (Li et al., 2018a). A comparative study of soil pollution by He et al., (2017) investigated that Cd, Cu and Zn mainly polluted e-waste recycling areas. They showed that the coefficient of variation values of some metals were more than 30% for anthropogenic sources. Some important anthropogenic sources which significantly contribute to the metals 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 (He et al., 2005).

Toxic substances like Pb, Cd, and Hg leach into the soil and ultimately pollute the groundwater. 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 water resources, metals 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 metals can release. Metals are dispersed to many segments of the environment due to inappropriate e-waste management including soil, sediment, water, air, and plants, and they are found in significant quantities in e-waste recycling sites. Metals were significantly retained in the surface soil, obviating groundwater contamination (Wu et al., 2014). However, the concentration of metals above the threshold limit is a big concern of environmental pollution and may cause an ecological imbalance in the ecosystem. Also, released metals have severe health effects on the human and animals. Hence, immediate and proper handling of e-waste is necessary.

3.2. Bioavailability and accumulation e-wastes

3.2.1. Bioavailability of metals

Bioavailability is the ability of the pollutants to get into biological system and induce their effects. If e-waste is discarded improperly, the metals will then become bioavailable after going through different biodegradation processes such as metal oxidation which is very common resulting in e-waste metals oxidizing. That leads to corrosion process which significantly release metals in the

environment. Plants can subsequently ingest the metals as ions. (Damasceno et al., 2015). 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) (Bonnail et al., 2016; Fu et al., 2014; Roosa et al., 2016). Metals in soil, sediments, and water mainly come from E-waste have various species and chemistry (Table S2). Unlike organic chemicals, most metals cannot be easily metabolized into less toxic compounds because of their biodegradation ability. Once they are introduced into the aquatic environment, metals are redistributed throughout the water column, accumulated in sediments or consumed by biota (Frémion et al., 2016).

E-waste based metals can be bioavailable for soil. This is because their availability on soil depends on two factors such as (i) the metallic element that precipitates as positively charged ions (cations), and (ii) which makes up negatively charged components of salt (Dowd and Maier, 2000). 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 determine the metal availability in soils. Also, the extent of aeration, water content, temperature, and root exudates and microbial activities are considerable parameters that regulate the metal availability in soil (Masindi et al., 2015; Soleimani et al., 2018). The toxicity of metals within soils with high CEC is generally low, even at high total metal concentrations. When the soil pH is low (~3–6), the metal bioavailability increases typically due to its free ionic species compared to high soil pH (> 7), where it decreases metal bioavailability as a result of the formation of insoluble metal mineral phosphate and carbonate (Masindi, 2016; Masindi and Muedi, 2018). The mobility and bioavailability of certain metals in soils are usually in the order of Zn > Cu > Cd > Ni (Masindi, 2016; Soleimani et al., 2018). However, the concentration of metals 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 end up with human diets.

Sediments have been widely used as environmental indicators to assess metal pollution in natural water (Islam et al., 2015). The metals can react with various contents of the aquatic environment and can associate with multiple geochemical phases in the sediments (Morillo et al., 2004). Metal residues in contaminated habitats can bioaccumulate in aquatic ecosystems, aquatic flora and fauna (Hasan et al., 2016). 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 (Equeenuddin et al., 2013). Fig. 5 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 (Wojtkowska et al., 2016). 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. The association between solid particles and metals is also critical for the metal uptake into organisms via food ingestion (Förstner, 2006). The suspended solids accumulate insoluble metal compounds. But under certain conditions, the metal reached the interstitial water being dissolved. Metals 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 (Adams and Chapman, 2007).

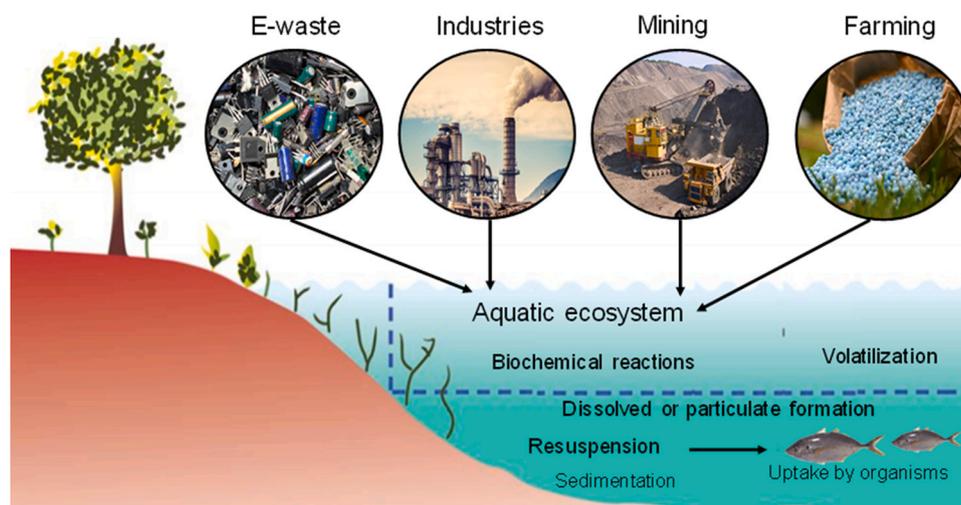


Fig. 5. HMs contamination sources influencing aquatic ecosystems.

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 (Rosado et al., 2016). E-waste released metals are bioavailable in soil by vermicomposting of organisms and water by bio-accumulation due to the impossible biodegradation of metals. The bioavailability of metals is responsible for several key variables and geochemistry.

3.2.2. Accumulation of metals in the environment

Metals accumulate in the environment and contaminate the food chains and soil due to their persistent nature (Ali et al., 2019a). Strong acid leaching and the open burning of dismantled components have 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 metals such as Cu, Cd, Hg, and Pb and organic contaminants (Alaee et al., 2003; Dopp et al., 2004; Gouin and Harner, 2003; Qjin et al., 2004; Yang et al., 2000). Metals 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. However, there are significant variations among different plant species in terms of metal accumulation ability (Robinson, 2009; Widmer et al., 2005). Soils are the major sink for metals released into the environment by the aforementioned anthropogenic activities. Once in the soil, metals are adsorbed by initial fast reactions (minutes, hours), followed by slow adsorption reactions (days, years). Therefore, they are redistributed into different chemical forms with variable bioavailability, mobility, and toxicity (Weidenhamer and Clement, 2007). The absorption of metals by plant roots is one of the main routes of entrance in the food chain (Jordao et al., 2006). Metals 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 (Khan et al., 2008). Metals 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 remain permanently in the marine

environment (Woo et al., 2009). When Metals 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 sink of metals and other pollutants in aquatic environments. A major fraction of the trace metals is introduced into the aquatic environment. These metals then become deposited with the bottom sediments. Accumulation of metals from one part to other part of the environment mainly occurred by the ecosystem where all the environmental components are interlinked with each other. Lastly metals are reached to the humans and all the living organisms through the food chain and causes harmful effects.

4. Impacts of heavy metals

Heavy metals are among the most common pollutants found in soil, sediment and wastewater. Heavy metals assert a toxicity threat to human beings and animals even at low concentrations. In dismantling and recycling process of disordered e-waste metals, brominated flame retardants and organic substances are released to the environmental media which is harmful for the local residents (Song and Li, 2015). Furthermore, e-waste contaminants can enter to the 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, acid disposal after hydrometallurgical processes and degraded e-wastes get mixed with the abiotic environment and then introduced into waters or onto soils. The dissolution or settling of airborne contaminants can also contaminate aquatic systems and water (Luo et al., 2006).

4.1. Soil

Among all the pollutants in soils, metals are of significant concern due to their inherent toxicity, bioaccumulation, persistence, and non-degradability (Liu et al., 2013). The most common metals found in the soil are Cu, Ni, Cd, Zn, Cr, As and Pb; where without As, most of them are e-waste based. The adverse effects of metals rely on soil properties, i.e. organic matter, clay contents, and pH (Speir et al., 1999). Metals obliquely affect soil enzymatic activities by shifting the microbial community, which usually synthesizes enzymes (Singh and Kalamdhad, 2011). Metals exhibit toxic effects on soil biota by affecting fundamental microbial movement and decreasing soil microorganisms number and activity. Conversely, long-term heavy metal effects can raise bacterial community fortitude and the tolerance of fungi such as arbuscular mycorrhizal fungi (de Mora et al., 2005). For example, Cd exhibits more toxicity to enzymes than Pb because of its greater dynamism and lower

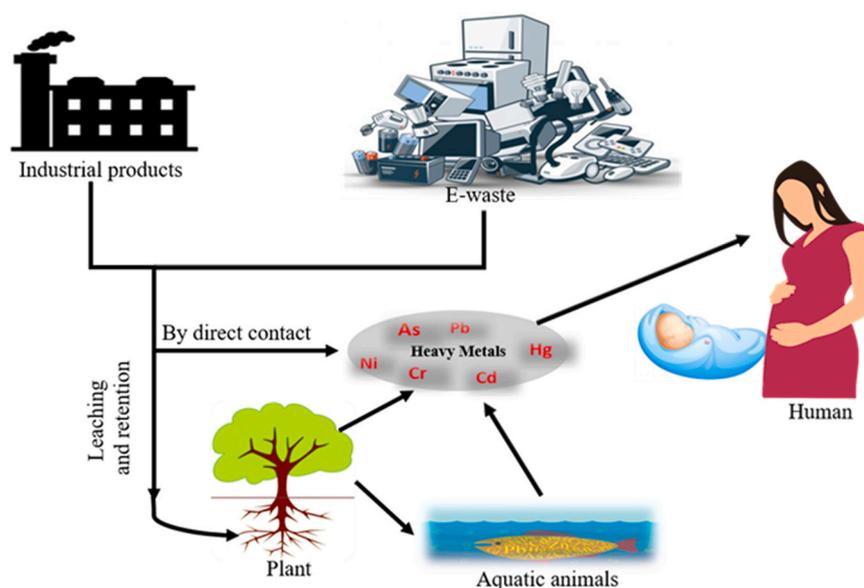


Fig. 6. Schematic diagram of heavy metal toxic exposure to the human system and abnormal growth and development. (modified from Vimalraj et al., 2017).

affinity for soil colloids (Pan and Yu, 2011). Cu avert β -glucosidase activity more than cellulose activity. Pb decreases the activities of the urease enzyme, catalase, inverses, and acid phosphatase mostly. 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 metals. The order of inhibition of urease activity commonly decreased according to the order $\text{Cr} > \text{Cd} > \text{Zn} > \text{Mn} > \text{Pb}$. The diversity and activity of soil microbes play significant roles in recycling plant nutrients, maintaining soil structure, detoxifying noxious chemicals, and controlling plant pests and plant growth communities are important soil quality indices (Wang et al., 2007b). 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 10–100 times less toxic than Cr^{6+} (Garnier et al., 2006). 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 (Ashraf and Ali, 2007). Uptake of metals by plants and subsequent accumulation along the food chain is a potential threat to animal and human health (Sprynsky et al., 2007). 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 (Bhattacharyya et al., 2008). Metals 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. The toxic effects of metals on soil, plants, living organisms and entire ecosystems are long-term. This reduce soil enzyme activity and cause the death of useful microorganisms and plants.

4.2. Water

Once an aquatic organism accumulates metals, they can be transferred through the food catena's upper classes (Ayandiran et al., 2009). When dispersed into aquatic ecosystems, metals and other contaminants stimulate the production of reactive oxygen species (ROS) that can spoil fishes and other aquatic organisms (Woo et al.,

2009). The consumption of fish containing elevated levels of metals is a concern because chronic exposure to metals 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 (Hall et al., 1997). These routes are through the food, non-food particles, oral consumption of water, gills and the skin. Once the pollutants are absorbed, they are transported by the blood to either a storage point or the liver for conversion and storage. If 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, an extrahepatic tissue (Ayandiran et al., 2009). In conclusion, metals contaminated water have adverse effects upon aquatic organisms and lastly damage the total aquatic ecosystem by the biological-magnification of heavy metals.

4.3. Human

The plant uptake of metals from soils at high concentrations may result in a severe health risk considering food-chain implications. The utilization of food crops contaminated with metals is a major food chain route for human exposure. Planting in contaminated soil represents a potential risk since the vegetal tissues can accumulate metals (Jordao et al., 2006). Metals become toxic when the body does not metabolize them and accumulates in the soft tissues (Masindi et al., 2018). Chronic level ingestion of poisonous metals has undesirable impacts on humans and the associated harmful impacts become perceptible only after several years of exposure (Ikeda et al., 2000; Khan et al., 2008). Metals toxicity exposure to the human system and abnormal growth and development is shown in Fig. 6. Zinc is considered relatively nontoxic, 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 diarrhea, bloody urine, vomiting, icterus (yellow mucus membrane), kidney failure and anemia, liver failure (Duruibe et al., 2007). 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 (Odum, 2016). Humans are reliant on

plants and animals for their survival, which puts them at risk if they consume metals-contaminated food.

5. Remediation technologies of metals released from e-wastes

Metals must be remedied from the environment, particularly heavy metals, in order to reduce their negative impacts. According to the characteristics of metal pollutants, remediation can be accomplished using biological, physical, chemical, or hybrid treatment procedures. The hybrid treatment techniques are gaining popularity among all of the technologies due to their high metal removal capability and environmentally benign behavior. As a result, new hybrid technologies are being created all the time to improve performance by merging independent processes. Following subsections represents the relevant remediation technologies of e-waste based metals.

5.1. Biological remediation processes

The biological treatment process is a self-purification process where microorganisms, plants, and animals are used to detoxify and remove the pollutants. The major advantages over conventional processes are this process does not produce toxic products and is cost-effective. The biological processes for metal remediation from groundwater or sub-surface of soil may appear by three mechanisms. They are adsorption, oxidation and reduction reactions, and methylation processes.

5.1.1. Bioleaching through microbial oxidation

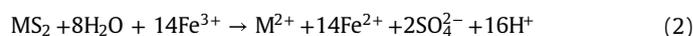
Bioleaching is an ex-situ/on-site remediation technology used globally in the metals contaminated site and environmental issues. In this biotechnology, microorganisms are used to solubilize metals and semi-metals from concentrates mainly used in the mining industry and bio-hydrometallurgy (Fonti et al., 2016). Various microorganisms engage in the bioleaching process where *Acidithiobacillus ferrooxidans* and *thiooxidans* have high removing performance (Table S3).

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, MS_2 = Insoluble metal sulfide and 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 metals inactive (Shahrabi-Farahani et al., 2014). The bacteria play an active role in the reaction by oxidizing Fe^{2+} to Fe^{3+} in the liquid phase via leaching, which is represented by the following chemical reactions 2 and 3 (Hocheng et al., 2014).



According to Rozas et al. (2017), 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 strain and work appropriately at neutral pH (7–8) and moderate temperature (30–40 °C) (Rozas et al., 2017). Pant et al. (2012) divided the leaching process into two parts; the first is the acid pre-leaching operation for 27 days and the second is the bioleaching operation for 280 days. At the end of these two operations, about 80% of Zn, 64% of Al, 86% of Cu, and 74% of 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 (Fonti et al., 2016). 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 metals removal.

5.1.2. Phytoremediation

Phytoremediation is a bioremediation technology used to exchange contaminants from a highly toxic to a less toxic form by accumulating, immobilizing, and transforming (Fig. S2) (Vidali, 2001). 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 (Khan et al., 2004). 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. *Cardaminopsis halleri*, *Bryophyllum. Pinnatum*, *Zea mays*, *Glycine max*, *Brassica junica*, *Brassica napus*, *Thlaspi caerulescens* plant species are mostly used with high efficiency in

different metals remediation (Table S4). According to Babu et al. (2013), the maximum removal capacity of Pb, Zn, As, Cd, Cu and Ni metals are 77%, 64%, 34%, 9%, 8% and 8% respectively at pH 6.5 by using *Alnus firma* with endophytic *Bacillus thuringiensis* GDB-1. It is recommended to study with the combination of two or more phytoremediation processes for better efficiency. Phytoremediation is mainly a natural process which mitigate the toxicity of metals from soil and water by accumulating, immobilizing, and

transforming. The uptake performance is depending on several factors and vary from plant species to species.

5.1.3. Microbial remediation

Microbial remediation of metals is very common and well-studied. Some metals such as Cr, Ca, Mg, Mn, Cu, Na, Ni and Zn are essential micronutrients for various metabolic 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+} and Hg^{2+} make inactive by binding with the sulfhydryl groups of enzymes (Sinha et al., 2009). 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.

Microorganisms play a fundamental role in bioremediation because of their high efficiency, easy operation, and lack of secondary pollution from contaminated soil, sediment, and wastewater (Chen et al., 2005; De et al., 2008). 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 (Vidali, 2001). *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 S5). Some microbes such as *Saccharomyces cerevisiae*, *Corynebacterium equi*, *Bacillus licheniformis*, and *Rhizopus arrhizus* can also remediate radioactive metals from the environment (Nakajima and Tsuruta, 2004). 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 (Sinha et al., 2009).

5.2. Physical remediation processes

5.2.1. Thermal remediation

Thermal treatment can significantly reduce the toxicity of metals at temperature (300–400) °C (Li et al., 2012). In this process, heating is done by using steam, microwaves, and infrared radiation to volatilize the pollutant (e.g., Hg, As) (Li et al., 2010). Shi et al. (2013) investigated the performance of the thermal remediation process for metal removal and they found that this process could remove 94% of Cd, 86% of Zn, 73.6% of Cr and 97% of Cu at 280 °C (Shi et al., 2013). A study conducted by Hseu et al. (2014) showed almost 99% removal of Hg by the thermos-gravimetric method at 550 °C for 1 h.

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 i.e., 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 (Navarro et al., 2013). By this vitrification method, Mn, Fe, Zn, Cu, Cr, Cd, Pb, Hg, As, and Se concentration are reduced by 91–100% using high temperature (more than 1300 °C) (Navarro et al., 2013). 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 (Nejad et al., 2018; Shi et al., 2013). This process's problems can be solved by characterizing pollutants and acquiring goodish consideration (Gullett et al., 2007). Therefore, thermal remediation methods have some potential but it is not sometimes not cost effective as it can create more problems (e.g., burning other chemicals and required sophisticated tools). This

process has up to 99% metals removal efficiency but the main problem is that this process pollutes the environment.

5.2.2. Ion-exchange

Different synthetic and polymeric cationic resins (e.g., purolite C100) used for the removal of metals from wastewater (Feng et al., 2000). The degree of ion exchange is 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 (Al-Enezi et al., 2004). The ion exchange process is more beneficial to remove Cd, Pb, Ni, Cr, Hg, Cu and Zn from water contaminants where the affinity of exchangers for ions are: $\text{Pb(II)} > \text{Cu(II)} > \text{Cd(II)} > \text{Zn(II)}$ (Dąbrowski et al., 2004). A study by Mier et al. (2001) reported high removal efficiency (> 95%) of Pb, Cd, and Cr using natural clinoptilolite. This process was carried out 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^{-1} , respectively (Shah and Devi, 1998). Moreover, natural zeolite is widely used to remove metals such as Zn, Co, Cu, Mn (Boros-Lajszner et al., 2018; Li et al., 2018b). However, pH sensitivity and non-selectivity of the exchangers reduce the process efficiency. Another disadvantage of this method is that metals are undesirable and swapped by other cations that are not harmful to the environment (Dąbrowski et al., 2004). In a recent study, macroporous ion-exchange resins (Amberlite IRA 743, Lewatit TP 208, and Lewatit TP 260) were applied for selective sorption of metals. From Cu^{2+} , Zn^{2+} , Ni^{2+} , Pb^{2+} , and Al^{3+} ions, hazardous Pb^{2+} ion was selectively extracted from the leached solution using a multi-elemental ion-exchange process (Nekouei et al., 2019).

This remediation process is applicable to remove metals from water, and not cover up high efficiency due to certain inconveniences. Ion-exchange is also a costlier method than other remediation processes. More investigations on hazardous ion-selective exchangers are required to get better results.

5.2.3. Adsorption by activated carbon

Activated carbons are extensively used for metals adsorption. Adsorption of metals ions from e-waste solution is a straightforward method through electrostatic interactions (Ali et al., 2019b). However, different factors, such as the surface area and porosity of adsorbent, metal ion complex, and pH. Also, the surface functionality and the size of adsorbing species have significant effects on the adsorption of metals in AC (Periyasamy et al., 2020). However, AC is a promising adsorbent as several previous research work experimented with the efficacy and factors affecting the adsorption process. A previous study showed that AC has better efficiency for As and Sb removal from Cu electro-refining solutions (Navarro and Alguacil, 2002). 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%) (Sahu et al., 2009a). Zinc chloride AC prepared from tamarind wood ash also has better removal efficiency for Pb and Cr (Sahu et al., 2010). A study by Ricordel et al. (2001) used AC prepared from peanut husks for the adsorption of Pb, Cd, Ni, Zn. AC adsorption mainly depends on particle size distribution, metal/AC ratio. However, ACs are very efficient adsorbent, but recovery of valuable metals is still challenging, the more comprehensive study should be done to determine methods and the most suitable desorbents for particular metals.

5.2.3.1. Metals 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. The nano-particles are utilized most frequently for the adsorption of metals via utilizing different

combinations of AC, carbon nanotubes (CNT), graphene, MnO, ZnO, TiO₂, MgO, and Fe₂O₃ (Tyagi et al., 2017). Nano-particles used as an adsorbent for metals removal should be nontoxic, possess high adsorption capacity, adsorb pollutants in less concentration (ppb), be easy to remove the adsorbed pollutants and should be recyclable for several times application (Anjum et al., 2019). For example, modified ZnO nano-adsorbent has unique micro/nanostructure compared to commercial ZnO, resulting in higher Cu (II) removal than unmodified ZnO (Singh et al., 2013). Moreover, various nano-assemblies were also used for removing various metals (e.g., Co²⁺, Ni²⁺, Cu²⁺, Cd²⁺, Pb²⁺, Hg²⁺, and As³⁺) at 30 °C and 24 h for better efficiency near to 100% (Singh et al., 2013). Kumar et al. (2013) reported that mesoporous hierarchical ZnO nano-rods have high removal efficiency of Pb²⁺ (160.7 mg/g adsorbed) and Cd²⁺ (147.25 mg/g adsorbed) from wastewater. It was reported that the modified Fe₂O₃ nano-particles shows a high affinity for removing different pollutants such as Cr³⁺, Co²⁺, Ni²⁺, Cu²⁺, Cd²⁺, Pb²⁺ and As³⁺ (Tyagi et al., 2017). Also, modified MnO has a good surface area (100.5 m² g⁻¹), showed higher adsorption of various metals such as As⁺, Cd²⁺, Pb²⁺ and Zn²⁺. Adsorption occurs on MnO, usually due to the inner-sphere formation mechanism by the ion-exchange process (Tyagi et al., 2017). Carbonaceous material, CNT shows excellent adsorption capacity of metals. Several studies reported the removal of Pb(II), Mn(II), Cu(II) by using MWCNTs (Tang et al., 2012; Tarigh and Shemirani, 2013). Graphene and functionalized graphene oxide show a very high efficiency for removing metals from wastewater (Dong et al., 2015; Santhosh et al., 2016). Nanomaterials have higher selectivity on the adsorption process and can adsorb very efficiently compared 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 metals using nanoscale zero-valent iron (nZVI) particles has been widely utilized and considered ideal candidates to remediate metals from e-waste (Morrison et al., 2002). For example, zeolite-aided nZVI nanoparticles have been utilized for the removal of Cd (II), Pb(II), and As (II) with an adsorption capacity of 48.63 mg/g, 85.37 mg/g and 11.52 mg/g, respectively (Li et al., 2018b). 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 (Gil-Díaz et al., 2017). Li and Zhang, (2007) 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 (Li and Zhang, 2007). Therefore, nZVI is a very potent material for the removal of many metals.

Nanoremediation is a promising technology due to its high metals removal efficiency (near 100%), advantages and acceptability over other processes. However, more focus should be given to the practical application on a large scale to determine the scalability of the nanomaterials applications for metals removal.

5.2.3.2. Sorption of metals by cellulosic materials and agricultural wastes. Cellulosic materials and agricultural wastes based materials have been applied to remove metals from water. Still, lower adsorption capacities were observed for pristine cellulosic materials, and therefore, modified cellulosic materials are commonly used for improving the removal capacities (Kamel et al., 2006). For example, O'Connell et al. (2008) investigated that the modified cellulosic material by halogenation, esterification, etherification or oxidation process had higher adsorption capacities. Similarly, a study by Sahu et al. (2009b) 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 (Wang et al., 2009). Various modified cellulose materials are used to remove various metals from contaminant sites (Table S6). 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 metals. These groups can make complexes or chelates with the metal ions, and sorption is occurred by complexation, adsorption, diffusion, chemisorption and ion exchange mechanisms. However, some signs of progress have been put forward on modifying the cellulosic materials more studies are required for suitable functionalization of these types of materials to improve the adsorption capacities cost-effectively.

5.2.4. Membrane technology

5.2.4.1. Microfiltration, ultrafiltration and reverse osmosis (RO). Microfiltration and ultrafiltration are very common physical methods for metal removal from water which have around 80% of efficacy except for As, Mo and Sb content because these metals remained a dissolved phase in an acidic medium (Arévalo et al., 2013). The ultrafiltration membrane has higher removal rates for metals such as Fe (92.14%) and Ni (61.90%) (Ortega Sandoval et al., 2019). Using microporous substances in filter systems can achieve 90–100% removal efficiencies for Cd, Cu, Pb, Ni, Cr, and Zn (Reddy Krishna et al., 2014). On the other hand, RO is also used for metal separation. For example, a comparative study Qdais and Moussa (2004) showed that the concentration of Cu²⁺ and Cd²⁺ was reduced to be about 3 ppm (99.4% removal efficiency) by RO process from water with an initial concentration of 500 ppm. The metal removal efficiency of RO is higher than nanofiltration (Qdais and Moussa, 2004). More feasibility analysis and evaluation of micro-ultrafiltration and RO techniques are required to obtain an overall better efficiency. Micro and ultra-filtration processes are two acceptable physical processes but both are expensive. However, ultra-filtration has higher removal efficiency than micro-filtration. More feasibility analysis and evaluation of these two process are required to obtain an overall better efficiency. On the other hand, RO process has better removal efficacy than other processes.

5.2.4.2. Membrane bioreactor (MBR) technology. MBR technology is being widely used for metal removal. Study shows that MBR offers 40–50% more efficiency in removing metals compared to conventional activated sludge process (Battistoni et al., 2007). The integrated system of electrically-enhanced MBR is most popular for its advantages and is suitable for wastewater treatment (Giwa et al., 2019). 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 (Wang et al., 2014). However, MBR with RO can remove metals but limited research has published focused on metals removal. Nevertheless, this technique may not be effective for low valence metal ions removal (Wang et al., 2014).

5.2.5. Solidification/stabilization (S/S)

In the solidification process, inorganic stabilisers or inorganic-organic-organic mixture amendments may be mixed with the contaminated sites for 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 (Guo et al., 2006).

Solidification technologies are not applicable for organic contaminants and some inorganic contaminants, such as oxyanions (e.g. Cr₂O₇²⁻, AsO₃⁻) and metals (e.g., Hg) because organic vapors are produced during mixing and heating. 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. Al-Wabel et al. (2015) was 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. (2009) 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⁻¹. However, this process performance is satisfactory compared to other physical process. This process is limited in soil and sediment remediation and low removal efficiency that is primarily dependent on the stabilizer performance and certain factors.

5.2.6. 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 metals more available for remediation from contaminated substances (Ron and Rosenberg, 2001). Surfactants such as sophorolipids and rhamnolipids are biological surfactants that can remove metals such as Cu, Ni, Zn, and Cd from the contaminated soil (Mulligan et al., 2001; Mulligan and Wang, 2006). Among them, rhamnolipid type I and type II with a surface tension of 29 mN/m have better metals removal capacity in liquid and foam form from contaminated soil. 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 metals by making complex compounds in the soil due to the lowering of interfacial tension (Mulligan and Wang, 2006). However, more study is required to assess the metals removal capacities by biosurfactants.

5.3. Chemical remediation processes

5.3.1. 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 metals to water and other environmental media is decreased (Zhou et al., 2004). 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) invasively 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 is 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 (Martin and Ruby, 2004). Several types of inorganic and organic amendments have often been used in the immobilization method to accelerate the attenuation of metal mobility and toxicity in soils. Lv et al. (2009) 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. (2015) was 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 metals remediation depending on specific site conditions. Furthermore, different immobilization technique using various amendments have various degree of removal efficiency of different metals (Table S7). This is a low performance remediation technique where chemicals are used to stabilize the metals from the contaminated soil, sediment and water. Among all amendments, CaCO₃ have high removal efficiency than others.

5.3.2. 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 (Sumner, 1988). Precipitation is one of the most conventional methods used to eliminate of metals from contaminated sources effectively. The mechanism is as follows:



Here M²⁺ 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 (Wang et al., 2005). Precipitation is a very convenient technique when the concentration of the metal(loid) ion is high and in the high pH soils in the presence of anions (e.g., SO₄²⁻, CO₃²⁻, OH⁻, and HPO₄²⁻) (Ok et al., 2011). A study by Aziz et al. (2008) 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%. A simple chemical remediation technique with low removal efficiency and only applicable to high concentration metal solutions.

5.3.3. 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 (Mulligan et al., 2001). The separation of the metals present in the soil occurs via electrophoresis, electric seepage, or electro-migration, resulting in a decrease in contamination (Yao et al., 2012). Electrokinetic remediation can reach more than 96% of metal removal efficiency for copper and zinc (Li et al., 1996). Rosestolato et al. (2015) 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 (Vocciante et al., 2016). For instance, by using KH₂PO₄ anolyte, Lee et al. (2016) 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, using 4–26 V as current, Ottosen et al. (2012) investigated the electrokinetic remediation method of Cu and Pb and found approximately 41% and 31% removal of Cu and Pb, respectively (Ottosen et al., 2012). Nejad et al. (2018) reported the reduction of 50–100%, Cd, Hg, Pb, Ni

Table 2
Metal removal efficiency with different chemical leaching method from the soil.

Leaching method	Reagents (mol/L)	Soil pH	Metal	Conc. ($\mu\text{g/g}$)	Removal efficiency (%)	Ref.
Batch 24 h, 12 steps, pH 7–8 (chelation)	EDTA (0.00275)	8.03	Cd	52	37	(Tejowulan and Hendershot, 1998)
			Cd	6.3	53	
			Cu	700	49	
			Pb	800	76	
			Zn	2650	84	
Batch 1 h, 1 step, pH 6 (chelation)	EDTA (0.1)	7.0	As	7540	13	(Moutsatsou et al., 2006)
			Cu	4100	41	
			Pb	64,195	44	
			Zn	55,900	38	
			As	1200	8	
Batch 22 h, 2 steps (chelation)	EDTA (0.25)	8.3	Cd	100	37	(Papassiopi et al., 1999)
			Pb	32,000	71	
			Zn	15,000	49	
			Pb	2730	65	
			As	1200	8	
Batch 1 h, 1 step, pH 4 (acid leaching + Cl^- complexation)	HCl + NaCl (5.5)	–	Cd	34	75–80	(Tampouris et al., 2001)
			Zn	3600	75–80	
			Pb	16,000	90–95	
			Pb	2730	65	
			As	1200	8	
Column: three steps with HCl(0.2 M) + CaCl_2 (1.9 M) at pH < 1 followed by 7 steps with CaCl_2 (2 M) at pH 6.5 (acid leaching + Cl^- complexation + ion exchange with Ca^{2+})	HCl + CaCl_2	5.6	Cd	34	75–80	(Tampouris et al., 2001)
			Zn	3600	75–80	
			Pb	16,000	90–95	
			Pb	2730	65	
			As	1200	8	
Batch 1 h, three steps, pH 4.5 (acid leaching)	HCl (1)	7.4	Pb	65,200	35	(Steele and Pichtel, 1998)
			Cd	52	56	
Batch 1 h, one step, pH 4 (acid leaching)	HCl (2)	7	As	7540	92	(Moutsatsou et al., 2006)
			Cu	4100	42	
			Pb	64,195	57	
			Zn	55,900	67	

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 (Virkutyte et al., 2002). 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 (Wang et al., 2007a).

5.3.4. 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 (Tampouris et al., 2001). The metals present in the soil is transferred from the contaminant to the liquid phase through ions exchange, precipitation, adsorption, and chelation. After that, metals are recovered from the leachate. The leachate mainly includes inorganic eluent, chelation agents, and surfactants. 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 (Alam et al., 2001). Most metals can form a stable composite with the ethylenediaminetetraacetic acid (EDTA) (among the other extractants) at a broad pH range.

Cd, Zn, Pb and Cr were removed by a 0.01 M Na_2EDTA 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 (Abumaizar and Smith, 1999). The efficiency of saponin on remediating HM contaminated soils was evaluated by Hong et al. (2002). 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. (2009) 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 2). To achieve a better understanding and maximum metals removal efficiency using the chemical leaching process, more exploration of the effect of leaching time and temperature and kinetic studies is 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 (Huang et al., 2011). 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.

5.4. Hybrid treatment processes

Various treatment processes discussed above used to remove metals 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 metals, hybrid or integrated processes work as a combination of two different methods.

5.4.1. Physico-chemical treatment technologies

5.4.1.1. Soil washing. The soil washing remediation technique is used to remove contaminates (metals) from soil particles, which is based on physical and chemical processes (Wuana and Okiyeimen, 2011). 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

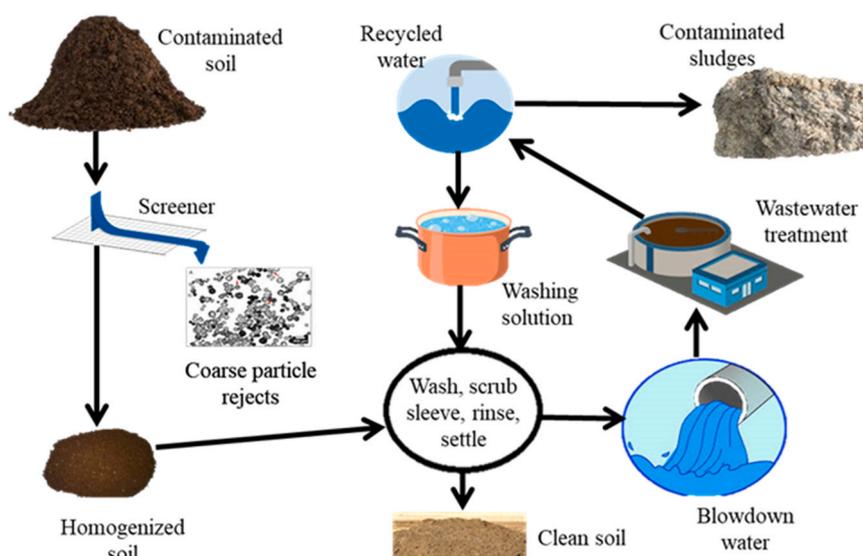


Fig. 7. A typical process diagram of soil washing for removing HMs contaminants. (modified from Liu et al., 2018).

results, a combination of both physical and chemical processes are necessary, as shown in Fig. 7.

Different chelating solutions are used in the soil washing process, such as EDTA, sodium persulfate and citric acid. Wuana et al. (2010) reported the removal efficiency of metals from soil using organic chelating acids in the order of EDTA > citric acid > tartaric acid with yields of metals in sequence Cu > Ni > Zn > Cd > Pb. 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 (Dermont et al., 2008).

5.4.1.2. Permeable reactive barriers (PRB). Permeable reactive trenches remove contaminants from contaminated water by degrading, precipitating, adsorbing or transforming processes.

5.4.1.2.1. 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 (Apak et al., 1998). It was found that red mud has high surface reactivity and the ability to remove metals from wastewater (Apak et al., 1998) and acid mine drainage (AMD) (Komnitsas et al., 2004). For example, Brunori et al., (2005) experimentally observed that the metal trapping ability of red mud was 35% of As from the initial concentration of 230 mg/L 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 (Huttenloch et al., 2001). Han et al., (2000) reported that granular AC 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 (Han et al., 2000). The activation of peat using an alkaline agent helped to increase the removal efficiency of metals. For example, NaOH-activated peat showed 100% removal of Cd, where the non-activated peat resulted in 75% removal of Cd (Fine et al., 2005). 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 (Roehl et al., 2005). Adsorption in PRB is a simpler and more efficient process for removing metals (almost 100%). The performance varies with the use of adsorbent in PRB.

5.4.1.2.2. 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 $Mg(OH)_2$, $MgCO_3$, $CaCl_2$, $CaSO_4$, and $BaCl_2$, 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 re-mobilized (Yin and Allen, 1999). Jun et al. (2009) investigated laboratory-scale A and B in PRBs for treating contaminated groundwater. Metals removal was 46.7–93.2% for reactor A and 58.7–99.6% 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 1 mg/L (Ahn et al., 2003; Kanel et al., 2006). This process is only applicable in water remediation where chemicals are used to fix and precipitate metals.

5.4.1.2.3. 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 (Barbaro and Barker, 2000; Fang et al., 2002). 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 (Seki et al., 2006). In PRB, this is a low-performance technology that relies on nutrients for contaminant biodegradation.

5.4.1.3. Ultrasonic leaching. This is the remediation technique of metals 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 (-1.5–2.0)) media is necessary (Dermont et al., 2008). 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 (Sharma et al., 2018). To remove Metals 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 (Huang et al., 2018). 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 metals removal (Pham et al., 2010, 2009). 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 (Huang et al., 2018). When ultrasonic leaching combined with an electrokinetic process, it improves metals removal efficiency, but this process is only applicable at low pH.

5.4.2. *Physiobiological methods*

5.4.2.1. Bio-electrokinetic method. This process is mainly suitable for soil remediation, where microbial and electrokinetic processes are used together to detoxify the contaminated soil. The integrated approach of bioleaching and electrokinetic remediation is advantageous because, at the same time, metals dissolved as an ionic solution and accumulate on the electrode (Peng et al., 2011). 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 (Peng et al., 2011). 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 (Zhu et al., 2013). 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.

5.4.2.2. 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 (Wang and Chen, 2009). *Agrobacterium* biomass was encapsulated with iron oxide nanoparticles for Pb adsorption, which showed an adsorption capacity of Pd of 197.02 mg/g (Tiwari et al., 2017). Various biosorbents with their uptake capacity and operating conditions for the bioremediation of metals are shown in Table S8. 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 (Rani et al., 2010). Biosorbents are primarily natural waste products that absorb metals in the presence of microorganisms, and their performance varies by microorganism species.

5.4.3. *Chemical-biological remediation approach*

Compared to individual processes, chemical and integrated biological processes have more efficiency for metal-containing wastewater treatment (Pradhan et al., 2017). For the remediation metals from contaminated sites, chemical–biological integrated treatment is considered to overcome a single method (Ahmed et al., 2016). Another study on the integrated chemical and biological treatment process could reduce 62.33% of total Cr(VI) using *Fusarium chlamydosporium* (Sharma and Malaviya, 2014). Metals removal from AMD

has been realized by selective sequential precipitation (bacterially produced H₂S helps bacterial production and the precipitation of the metals) of metals using this technique (Luptakova et al., 2012).

Although chemical-biological integrated treatment showed promising result on metal 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 (Lohner and Tiehm, 2009). However, selective recovery of metals with 99% removal efficiency is possible (Luptakova et al., 2012), also scaling up of this process is possible using a continuous system.

5.5. *Comparison of different technologies*

Each metals 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 metals from contaminated soil, sediment and water (Kim et al., 2010; Peng et al., 2011).

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, easy to operate and no secondary pollution (Alkorta et al., 2004; Archana and Jaitly, 2014; Sharma et al., 2018). The bioleaching process exhibited higher removal of metals (64–90%), but the limitation is time-consuming, whereas the composting process showed lower removal efficiency of metals (42–72%), and the public acceptance is also low (Table 3). Furthermore, microbial remediation and phytoremediation are long-term effective processes because they are harmless natural processes and 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 (Khalid et al., 2017). Compare to other technologies; the physical treatment technologies are widely used remediation process of metals because of low cost, simplicity, eco-friendly than other methods. But in many cases, physical methods showed inadequate removal of metals. Among all the technologies, thermal remediation (94–99%) and the use of biosurfactants (> 70%) are most effective in the remediation of metals 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 have high efficiency in removing metals. 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 3), 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 metals 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

Table 3
An overview of different treatment processes for HMs remediation.

Remediation Technology		Remediate Substances	Removal Substances	Removal Capacity	Advantages	Disadvantages	Acceptance	Reference
Biological Processes	Biobleaching	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	(Pant et al., 2012; Ren et al., 2009)
	Composting	Especially for soils	Mn, Fe, Zn, Cr, Cu	42–72%	Eco-friendly, self-stabilization process	Lengthy, less efficiency	Very low	(Chen et al., 2015; Taiwo et al., 2016)
	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.	Highest public acceptability	(Alkorta et al., 2004; Archana and Jaitly, 2014; Garbisu and Alkorta, 2001; Sharma et al., 2018)
	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	Only Hyperaccumulator plants are used. HMs can be released back after decomposition.	Very high public acceptability	(Archana and Jaitly, 2014; Chen et al., 2005; Sharma et al., 2018)
Physical Process	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	(Li et al., 2010; Navarro et al., 2013; Shi et al., 2013)
	Ion Exchange	Water, Co-contaminated soil	Pb(II), Hg(II), Cd(II), Ni(II), V(IV), Cr(III), Cu(II), Zn(II), As(III), As(V), Sb, Cr(VI)	40–99.5%	High removal capacity, simple and effective method, selective removal of ionic contaminant	Costly. Recharge or regeneration of materials	Medium to high public acceptability	(Dąbrowski et al., 2004; Shah and Devi, 1998)
Membrane & Filtration	Adsorption by Activated Carbon	Mainly water contaminant	Zn	89–97.95%	High removal capacity, eco-friendly	Highly dependent on particle size distribution, produce a large quantity of solid waste	High for water	(Navarro and Alguacil, 2002; Sahu et al., 2009a)
	Microfiltration & Ultrafiltration	Water		19–93%	Simple process for pretreatment of other processes, cost-effective	Applicable for specific particles, membrane clogging;	Low	(Guo et al., 2011; Michael-Kordatou et al., 2015; Racar et al., 2017)
	Reverse Osmosis (RO)	Water, Drinking water	Ionic contaminants	95–99%	High efficiency, produce high-quality water.	Only for liquid contaminants, costly method. Membrane fouling	High for water	(Pawlak et al., 2006; Volesky, 2003)
Solidification	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	(Cicek, 2003; Lin et al., 2012)
		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	(Al-Wabel et al., 2015; Lv et al., 2009)

(continued on next page)

Table 3 (continued)

Remediation Technology	Remediate Substances	Removal Substances	Removal Capacity	Advantages	Disadvantages	Acceptance	Reference
Replacement Method	Soil, Sludges, Sediments, water	All type of contaminants	–	Effective for small scale	Only applicable for small amounts of contaminants, highly costly	Very low: limited to highly contaminated soils	(RoyChowdhury et al., 2018; Sharma et al., 2018)
Sorption by Cellulosic materials & Agriculture Wastes	Water	Pb, Cd, Cr, As	71–96%	Eco-friendly, cost-effective, apply wastage substances	Applicable for some functional groups, pH dependent	Medium	(Amin et al., 2006; Sahu et al., 2009b; Wang et al., 2009)
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	
Chemical Processes	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	(Al-Wabel et al., 2015; Lv et al., 2009)
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	(Aziz et al., 2008; Summer, 1988)
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	(Khalid et al., 2017; Lee et al., 2016; Nejad et al., 2018)
Chemical Leaching	Soil, Sludge, 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	(Abumaizar and Smith, 1999; Li et al., 2009; Tokunaga and Hakuta, 2002)
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	(Anjum et al., 2019; Tyagi et al., 2017)
Hybrid Processes	Soil, Sludge	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	(Abumaizar and Smith, 1999; Wuana et al., 2010)
Permeable Reactive Barriers (PRB)	Water, Wastewater	Pb, As, Cd, Zn	30–70%	Cost-effective; high sorption capacity.	Sorbs cations with lesser ionic radii; depends on pH	Low	(Brunori et al., 2005; Santona et al., 2006)
Activated Carbon and peat		Cr, Cd	> 90%	High adsorption capacity; regeneration possible; acts better when coupled with microbes	More field-scale studies on inorganic and metal adsorption are needed	High	(Fine et al., 2005; Huttenloch et al., 2001)
Zeolites			40–80%			Medium	

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Table 3 (continued)

Remediation Technology	Remediate Substances	Removal Substances	Removal Capacity	Advantages	Disadvantages	Acceptance	Reference
Iron sorbents (ZVI and pyrite)		Cd, Cu, Ni, Cr, As	98%	Very high adsorbing capacity; hundreds of natural zeolites are available ZVI and pyrite are cheap; handling is easy	Selective adsorption capacity	High	(Ruggieri et al., 2008; Xenidis et al., 2010) (Bower et al., 2008; Sun et al., 2006)
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	(Ahn et al., 2003; Kanel et al., 2006)
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	(Janvis et al., 2006; Ludwig et al., 2009; Van Nooten et al., 2008)
Ultrasonic Leaching	Soil, Sludges, sediment	Zn, Pb, Cu and Cd	60–95%	High removal efficiency	An acidic solvent is needed, costly	Medium	(Huang et al., 2018)
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	(Xu et al., 2017)
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	(Rani et al., 2010; Tiwari et al., 2017; Wang and Chen, 2009)
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	(Giwa and Hasan, 2015; Hasan et al., 2014) (Chen et al., 2017)
Combination of Persulfate Oxidation and Chemical Washing	Soil, Sludges	Cu, Pb, Ni	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	(Abbas et al., 2017; Li and Yu, 2015)
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 sediment environment is required to reduce the production of methyl mercury and efflux	High	

pollutants for using toxic chemicals. To overcome this problem, additional 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 metals (Huang et al., 2012). Among the discussed hybrid technologies (Table 3), 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 bio-leaching and electrokinetics are the most effective technology because of their high uptake capacity of metals from contaminant soil, sediment or water (Xu et al., 2017). 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. Combining 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 (Racar et al., 2017). But these combined processes are mainly applicable for the remediation of metals 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 metals removal from electronic waste.

6. Gaps and future recommendations

The production of e-waste is increasing day by day. This resulted in the increase of heavy metal leaching from e-waste to the soil, sediment and water. Thus, an appropriate control measure is required on the production and disposal of e-waste. In addition, proper remediation technology is necessary to mitigate the contaminants from e-waste, especially metal present in the e-waste. The remediation techniques have some definite outcomes e.g.:

- (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.

However, 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 metals remediation are described in Table 3 with their performance, advantage, disadvantage and acceptability. The physical processes are not so effective, and the chemical processes have high metals removal efficiency but not eco-friendly. Recently, hybrid treatment technologies are most popular in metals remediation due to their effective performance over the single treatment processes. But the proper combination of the different methods is necessary for hybrid treatment. Recently, 3D printing technology and very thin layer metals oxides are fabricated into different devices for multiple applications. These are the very promising technologies to reduce the use of the materials. But, still they can become a further burden in future. Therefore, the proper disposal of such kinds of electronic wastes should follow before dumping them in the environment. Moreover, innovative new technologies should come forward for remediation of such kinds of pollutants. Finally, microplastics interactions with e-

waste based metal particles should be investigated in depth in the future so that they can be tracked and removed by using proper technologies.

In summary, each remediation approach has advantages and disadvantages over other alternatives. The best remediation technology is mainly determined by contaminated areas, pollutants concentrations, and other considerations. The development of new remediation technology is currently a major issue for researchers, that's why new hybrid technologies is growing all the time, and more study is needed in the future to achieve the best results.

7. Conclusion

Our research concludes that, by the incrementally increasing application and development of modern technologies each year, massive amounts of e-wastes are being produced continuously. Nevertheless, if we think about the positive side of modern technologies (e.g., advanced computing, robotics, lifesaving medical instruments, home appliance and so on), we cannot stop the emergence of modern technologies due to its necessity in daily life. However, we can establish effective e-waste recycling management systems by offering sustainable e-material consumption and production patterns and, where feasible, avoiding hazardous materials (e.g., utilizing biodegradable polymers) while manufacturing electrical equipment. Some metals 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 metals due to their complex formation and harmful environmental and animal impacts. Among all the previously used methods discussed in this review, biological and hybrid treatment processes have high removal capability and high public acceptability. Finally, it is important to explore new methods from other process objects based on similar characteristic for better performance than existing technologies.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.psep.2022.04.011](https://doi.org/10.1016/j.psep.2022.04.011).

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