

1 **Recovering metal(loids) and rare earth elements from closed landfill sites**
2 **without excavation: leachate recirculation opportunities and challenges**

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10 **Abstract:** Metal(loids) and Rare Earth Elements (REE) ('metals') are naturally scarce and
11 economically high-value. They are used in a wide range of products, with demand continuing to
12 grow. Metal-bearing wastes are a secondary source of raw material that can meet this demand by
13 providing a previously unconsidered low impact supply source. In particular, landfill leachate
14 emerges as a significant potential resource as it contains high concentrations of metalloids, metal
15 ions and RRE. However, realising a profitable return on investment for leachate processing is a
16 tremendous challenge due to a relatively low recovery rate, variation in the multi-element value
17 and the effect of other chemicals (organic and inorganic) in these complex mixtures. There is a
18 need to better understand the mechanisms and potential applicability of investigation and extraction
19 methods for optimising metals recovery from leachate. This paper addresses this need by providing
20 a systematic review of the critical factors and environmental conditions that influence the
21 behaviour of metals within the landfilled waste. The paper provides a synthesis of how these may
22 affect leachate recirculation efficiency for recovery in the context of a range of opportunities and
23 challenges facing circular economy practitioners.

24 **Keywords:** circular economy, non-intrusive investigation, organic compounds, mobility, metal
25 recovery

26 **1 Introduction**

27 Metals, metalloids and REE, collectively termed ‘metals’, are finite natural resources with
28 increasing demand. It is vital to find alternative sources to ensure supplies of these metals,
29 especially for new technologies such as electric vehicles, renewable energy generation and
30 battery storage (Jowitt et al., 2020). Several recent studies reported that a worthwhile amount of
31 valuable secondary raw materials is available within closed landfill sites (Gutiérrez-Gutiérrez et
32 al., 2015; Dino et al., 2017; Krook et al., 2018; Särkkä et al., 2018; Esguerra et al., 2019; Parrodi
33 et al., 2019; Wagland et al., 2019). There are around 21,000 closed landfill sites across England
34 and Wales (DEFRA, 2021; ENDS Report, 2021) and somewhere between 150,000 and 500,000
35 landfill sites within Europe, with an estimated 90% of them being non-sanitary (Jones et al.,
36 2018). Closed landfills represent a significant opportunity across Europe and the UK to recover
37 value from waste materials as they provide a previously unconsidered localised long-term storage
38 deposit of secondary raw materials similar to traditional metal mineral resources. Enhanced
39 Landfill Mining (ELFM) address the combined and integrated valorisation of distinct landfilled
40 urban waste streams as both materials (Waste-to-Materials, WtM) and energy (Waste-to-Energy,
41 WtE) while meeting the most stringent ecological and social criteria (Jones et al., 2013). ELFM
42 provides an opportunity for combined resource recovery and reclamation of land while mitigating
43 future environmental liabilities and remediation costs through excavation innovative
44 transformation technologies (Jones et al., 2013; Esguerra et al., 2021; Vollprecht et al., 2021).
45 However, existing technologies and good practices are unable to demonstrate the economic
46 viability of such schemes. This is partly because mining processes for recovering material result
47 in high capital costs. There are also important uncertainties about the actual abundance and
48 concentration of suitable waste materials in landfill environments, which need to be known early
49 on in the lifecycle of a metal’s recovery project.

50 Two essential by-products of waste disposal by landfills are leachate and landfill gas. The former
51 is formed when rainwater infiltrates and percolates through the degrading waste, the latter by
52 microbial degradation of biodegradable waste materials under anaerobic conditions (Chu, 2008).
53 Effective treatment methods are required as leachate contains trace chemicals, contaminating
54 groundwater, surface water and soil, potentially polluting the environment and harming human
55 health (Brennan et al., 2016). However, ambitions for the landfill management should go beyond
56 protecting human health and the environment, with conservation of energy and recovery of
57 natural resources high on the agenda. Landfill leachate comprises recoverable metals, organics,
58 phosphorus, ammonia, and water (Iskander et al., 2017; Kurinawan et al., 2021). The presence of
59 recoverable metals means that landfill leachate can be of great importance as an alternative to
60 conventional mineral exploration as the sediment of the leachate showed presence of REEs
61 content was more than twice the content in landfilled waste (Gutiérrez-Gutiérrez et al., 2015), but
62 also it can negate the need for full-scale landfill mining.

63 There are four objectives of this review paper: i) to give an overview of the properties and metals
64 content in landfill leachate to gain insight into the opportunities for metal recovery from leachate;
65 ii) to explore the knowledge on various factors affecting metals solubility; iii) to evaluate the
66 efficiency of recirculation for increasing metal recovery rates and; iv) to discuss the opportunities
67 for metal recovery from leachate, analyse the challenges associated with the recovery, and present
68 the perspectives for future research and technology development to maximise the benefits of
69 metals recovery from closed landfill leachate.

70 **2 LANDFILL LEACHATE PROPERTIES AND METALS CONTENT**

71 Leachate can be formed as a result of chemical and biochemical processes within the landfill. There
72 is nonuniform and intermittent percolation of moisture through the solid waste in the landfill due

73 to leachate generation (Hughes et al., 2013; Edokpayi et al., 2018). Several factors influence
 74 leachate composition, such as the age of landfill, depth of the waste in the landfill, location of the
 75 site, and weather condition of the landfill site; another critical factor affecting leachate composition
 76 is the composition of the waste deposited in the landfill (Jang and Townsend, 2003; Kalčíková et
 77 al., 2011; Moody et al., 2017). Generally, the waste composition is categorised as organic (food
 78 and garden waste), paper, plastic, glass, metals, etc. Table 1 shows the composition of global waste.

79 **Table 1.** Waste composition range

Component	Range (%)	References
Organic waste	15-58	Tapia, 2009; European Commission, 2016; Abdel-Shafy and Mansour, 2018; Kaza et al., 2018
Paper and cardboard	16-27	
Glass	2-4.5	
Plastic	9-20	
Wood	3-7	
Metal	3-9.2	
Textile	3-9	
Rest	3-18	

80
 81 A wide variety of metal contents is collected into the leachate as it drains through the pile of waste
 82 in the landfill (Eggen et al., 2010; Edokpayi et al., 2018). Leachate is also rich in ammonia and
 83 inorganic components such as iron, chlorine, sulphate, and metals (Öman and Junestedt, 2008).
 84 Inorganic compounds may consist of potentially harmful elements such as Pb, Hg, and As in pure
 85 form or combined with other elements (Jan et al., 2015). As leachate contains a wide range of
 86 metals, it can be considered a great potential for metal recovery. However, there are also challenges
 87 associated with the method to meet the need for sustainable leachate management that maximises
 88 valuable metal recovery. The key challenge on metal recovery from leachate is the low
 89 concentration of metals which is often affected by landfill age and type (Table 2).

90

91 **Table 2.** The concentration range of chemical constituents of landfill leachate determined from
 92 available literature

Parameter	Concentration range (mg/l)	Parameter	Concentration range (mg/l)	References
Alkalinity (as CaCO ₃)	0-20,850	Nitrogen (Ammonia)	0-1,250	Kjeldsen et al., 2002
Aluminium	0-2	Nitrogen (Nitrate)	0-9.8	Kjeldsen et al., 2002
Antimony	0-3.19	Nitrogen (Nitrite)	0-1.46	Christensen et al., 2001
Arsenic	0-0.04	Nitrogen (Organic)	0-1,000	Christensen et al., 2001
Barium	0-2	Nitrogen (Total Kjeldahl)	1-100	Christensen et al., 2001
Beryllium	0-0.36	Nickel	0-7.5	Christensen et al., 2001
BOD ₅	0-4,000	Phenol	0.17-6.6	Akinbile et al., 2012
Boron	0.5-10	Phosphorus (Total)	0-234	Kjeldsen et al., 2002
Cadmium	0-0.01	Phosphate	1-10	Christensen et al., 2001
Calcium	100-1,000	pH	4.5-9	Adamcová et al., 2016
Chloride	20-2,500	Potassium	0.16-3,370	Akinbile et al., 2012
Chromium	0-0.05	Selenium	0-1.85	Adamcová et al., 2016
Cobalt	0-7.58	Silicon	0-12	Kjeldsen et al., 2002
COD	150-6,000	Silver	0-1.96	Christensen et al., 2001
Conductivity (µmho/cm)	480-72,500	Sodium	0-8,000	Kjeldsen et al., 2002
Copper	0-9.9	Thallium	0-0.32	Adamcová et al., 2016
Cyanide	0-6	Tin	0-0.16	Adamcová et al., 2016
Fluoride	0.1-1.3	TDS	0-42,300	Akinbile et al., 2012
Hardness (as CaCO ₃)	400-2,000	Titanium	0-1.5	Christensen et al., 2001
Iron	0-5,500	TSS	140,900	Kjeldsen et al., 2002
Lead	0-5	TOC	335,000	Adamcová et al., 2016
Magnesium	16.5-15,600	TVA (as Acetic acid)	0-19,000	Akinbile et al., 2012
Manganese	0.05-1,400	Turbidity	40-500	Adamcová et al., 2016
Mercury	0-3	Sulphate	0-300	Adamcová et al., 2016
Organic halides	0.32-3.5	Zinc	0-1,000	Christensen et al., 2001
Benzene	0.1-0.6	Phenols	0-4	Christensen et al., 2001
Ethylbenzene	0-4.9	Toluene	0-3.2	Akinbile et al., 2012

93 Note: The grey shaded cell indicates metals and metalloids

94 The leachate produced in young landfills (< 5 years old) contains a substantial number of organic
 95 compounds derived from biodegradable organic water materials, which undergoes rapid anaerobic
 96 fermentation within confined landfills. As a result, volatile fatty acids (VFAs) are produced, e.g.
 97 acetic, propionic, iso-butyric, n-butyric, iso-valeric, and n-valeric acid. It is very well known that
 98 organic acids, such as VFAs, may play an essential role in the mobilisation of metals through either
 99 the formation of soluble ligand: metal complexes or a decrease of pH (Molaey et al., 2021). Thus,
 100 VFAs are considered valuable substrates for metal dissolution, increasing the release of metals in
 101 landfill environments. Young leachate is characteristic of its high content of biodegradable organic

102 matter. BOD (Biochemical oxygen demand), COD (Chemical oxygen demand), and BOD/COD
 103 ratio act as indicators of microbial activities and organic pollution. BOD/COD describes the
 104 biodegradability level of materials by which organic matter containing leachate is readily broken
 105 down in the environment (Samudro and Mangkoedihardjo, 2010). Therefore, young leachate shows
 106 a high BOD/COD indicator.

107 As landfill age increases, the BOD/COD ratio in leachate decreases (Table 3). This is due to the
 108 decomposition of the majority of biodegradable compounds and small quantity changes of less
 109 degradable organic matter at the same time that acidic conditions begin neutralise (Talalaj, 2015).
 110 As a consequence, the higher pH condition results in decreasing metal release by complexation and
 111 precipitation (Zhang et al., 2018). Older leachate from the methanogenic phase is partially
 112 characterised by the lower concentration of VFAs. As the content of VFAs and other readily
 113 biodegradable organic compounds in the leachate decreases, the organic matter (OM) in the
 114 leachate becomes dominated by refractory compounds, such as humic acid (HA) and fulvic acid
 115 (FA), which are known to bind metals to their hydroxyl and carboxyl groups, and either mobilise
 116 metals or delay their release (Leung and Kimaro, 1997; Bozkurt et al., 2000; Kochany and Smith,
 117 2001; Klavinsa et al., 2006; Gutiérrez-Gutiérrez et al., 2015). The humic substances (HS) give a
 118 dark colour with increasing pH due to the dissociation of protons (Stevenson, 1994). The decrease
 119 in VFAs increases pH; consequently, metals have a relatively low concentration in older landfill
 120 leachate as the solubility of metals is decreased with increasing pH.

121 **Table 3.** Selected characteristics of leachate according to landfill age

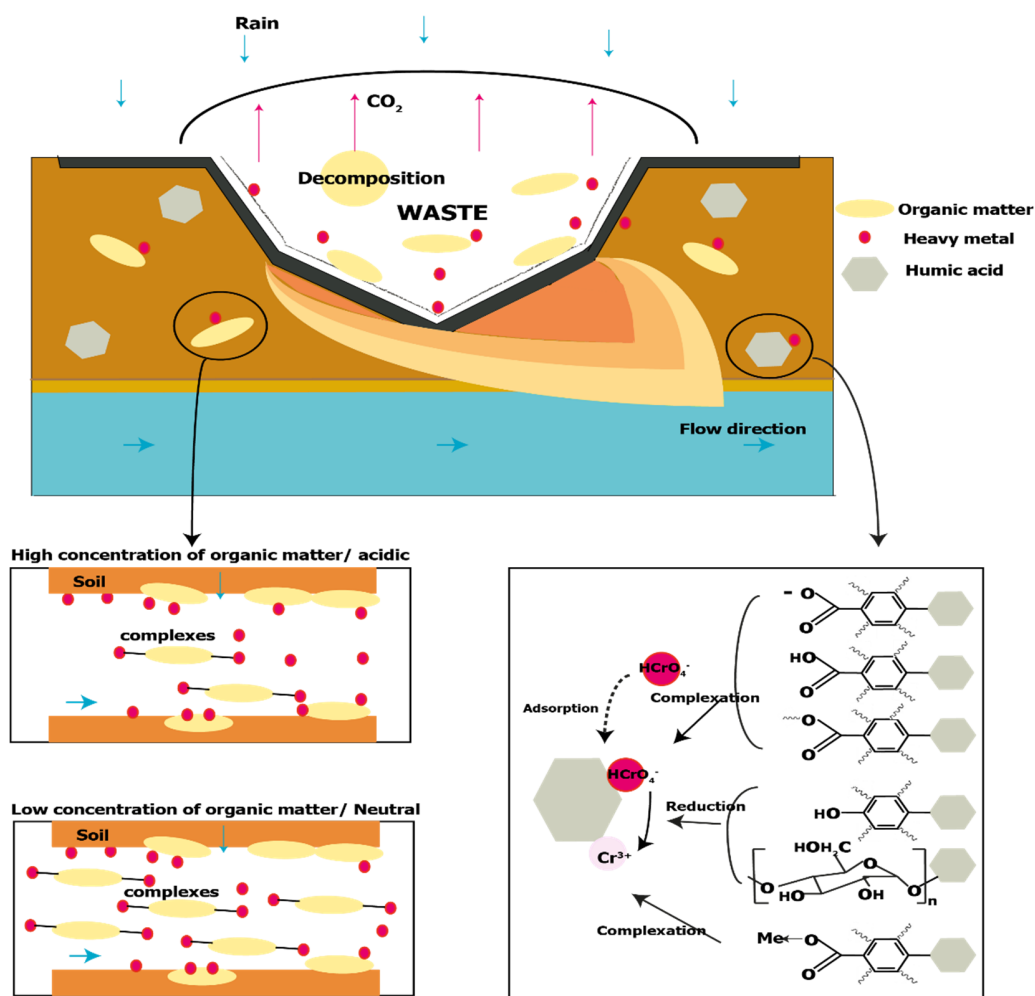
Parameter	Young	Intermediate	Old	References
Age (years)	<5	5-10	>10	Renou et al., 2008
pH	<6.5	6.5-7.5	>7.5	Bhalla et al., 2013
COD (mg/L)	>10,000	4,000-10,000	<4,000	Bhalla et al., 2013
TOC/COD	<0.3	0.3-0.5	<0.5	Abbas et al., 2009; Zhou et al., 2010

BOD ₅ /COD	>0.3	0.1-0.3	<0.1	Bhalla et al., 2013
Organic compounds	80% VFA	5-30% VFA + humic and fulvic acids	Humic and fulvic acids	Bhalla et al., 2013
Heavy metals (mg/l)	Low medium >2	Low <2	Very low <2	Renou et al., 2008
Molecular size distribution	Over a broad range-high fraction of low molecular weight organics	N/A	Over a narrow range-high fraction of high molecular weight organics	Abbas et al., 2009; Zhou et al., 2010
Biodegradability	Important	Medium	Low	Bhalla et al., 2013

122

123 3 PHYSICO-CHEMICAL FACTORS AFFECTING METALS SOLUBILITY

124 Several factors affect metals solubility within solid waste deposits (Fig 1). Important processes
125 include abiotic redox processes, dissolution/precipitation of minerals, sorption, ion exchange,
126 organic matter biodegradation, and complexation. The resulting matrix redox changes strongly
127 influence both the inorganic and organic biogeochemistry of the landfill and therefore influencing
128 the behaviour and fate of metals within landfills (Christensen et al., 2001). Gaining insights into
129 the geochemistry of landfill is therefore needed to better understand the solubility of metals and
130 predict metals recovery.



131

132 **Figure 1.** Processes occurring in landfills which affect metal solubility

133 **3.1 Weathering and natural attenuation**

134 The dissolution of metals can occur during natural events, such as weathering and natural
 135 attenuation. During weathering processes, a broad range of physical and chemical reactions such
 136 as hydrolysis, precipitation, pH neutralisation, oxidation/reduction of metals, sorption, and
 137 complexation will change the overall characteristics of metals (Chimenos et al., 2003; Poletini et
 138 al., 2004; Saffarzadeh et al., 2011; Takahashi and Shimaoka, 2012). Natural attenuation can be
 139 defined as a process by which the concentration of leachates is reduced to an acceptable level by
 140 natural processes. It can both mobilise and immobilise metals (Beaven et al., 2013). Based on the

141 definition, *in-situ* natural attenuation mechanisms are identified as physical (diffusion, sorption,
142 dispersion, dilution, and volatilisation), chemical (precipitation, adsorption, ion exchange, redox
143 reaction) and biological (biodegradation) processes. For this reason, it is desirable to be able to
144 predict how the metals in the landfill environment will behave over time when exposed to the
145 weathering effects of infiltrating rainwater and the atmosphere. The effect of weathering on metals
146 solubility is likely to be significant as pH is a dominant parameter in metals solubility and
147 complexation (Stumm and Morgan, 1981). The redissolution of their respective hydroxide mainly
148 causes the release of metals as pH is controlled by the solubility of $\text{Ca}(\text{OH})_2$. Therefore, weathering
149 reactions leads to a decrease in pH (Chimenos et al., 2003).

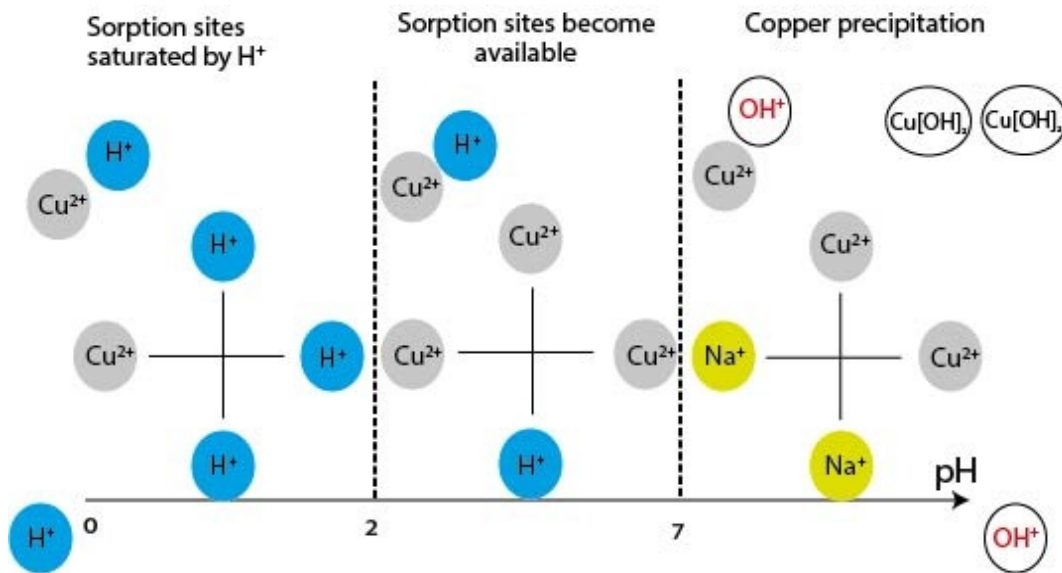
150 **3.1.1 Oxidation**

151 Several studies have been shown the formation of Fe/Al-(hydrate) oxides and calcite by weathering.
152 It indicates that metals release may be controlled by sorption processes caused by weathering
153 (Zevenbergen and Comans, 1994; Meima et al., 1997a; Meima and Comans, 1999; Saffarzadeh et
154 al., 2011; Takahashi and Shimaoka, 2012). Saffarzadeh *et al.* (2011) proposed the following order
155 based on their direct metal uptake capacity: Fe-hydrate > Al-hydrate > calcite. Calcite is not
156 adequate for direct metals sorption; however, they play a crucial role in buffering the system, pH
157 neutralisation; consequently, it minimises metal leaching. Thus, weathering is expected to result in
158 a reduced metal solubility in the long term (Meima and Comans, 1999).

159 **3.1.2 Sorption and precipitation**

160 Temporal studies of metal mobility in soils show that mobility decreases over time, suggesting that
161 a high proportion of metals within Municipal solid waste (MSW) which consists of everyday items
162 we use and then throw away, are insoluble (Peters and Shem, 1993; Aucott, 2006). The reasons for
163 the reduced mobility of metals in soil include sorption on soil particles and particularly to HS,
164 precipitation under anaerobic conditions, adsorption, and chelation with inorganic and organic

165 ligands in landfills (Bozkurt et al., 2000). Christensen *et al.* (2001) reported that metals in landfills
 166 do not constitute a significant pollution problem due to strong attenuation by sorption and
 167 precipitation (Fig 2).

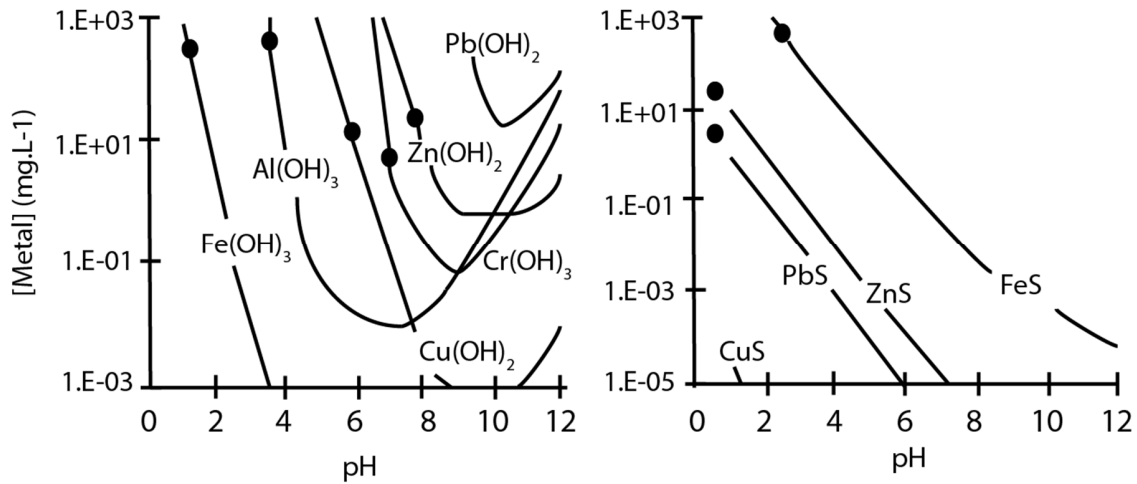


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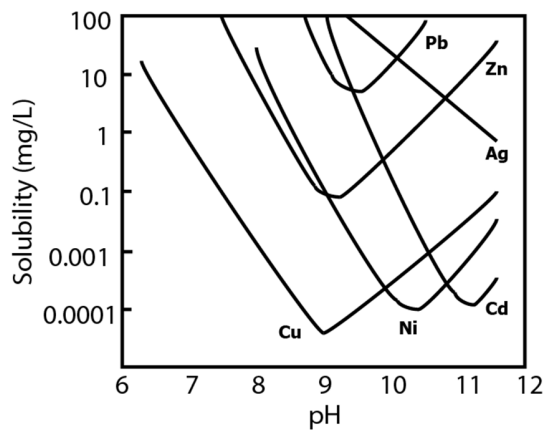
169 **Figure 2.** Copper sorption mechanisms (Adapted from Abbar et al., 2017)

170 In general, inorganic content of leachate ranges between 1 and 2000 mg/L. By raising the pH
 171 value, metallic hydroxide compounds become insoluble and precipitate from the solution.
 172 Alkaline conditions promote metal precipitation and adsorption, depending on the metal
 173 speciation (Lukman et al., 2013). Fig. 3 shows the solubility curves of selected metal ions and
 174 their respective solubility versus pH. Cu and Ni have a similar curve, albeit that the minimum
 175 solubility of Ni occurs at approximately pH 10.5 and the minimum solubility of Cu occurs at pH
 176 9. Zn is amphoteric, being soluble in both acid and alkaline conditions. Cu and Zn readily form
 177 metallic complexes with ammonia. These metal complexes remain highly soluble at the higher
 178 pH values, prohibiting respective metal hydroxide precipitation. Cu sulphide is insoluble, and the
 179 presence of sulphide precipitates Cu as it dissociates from the ammoniacal complex. Precipitate

180 in landfill environments strongly relates to organic decomposition and the formation of
 181 microorganisms during the process of methanogenesis (Li et al., 2015). According to Fig 3,
 182 precipitation is unlikely to occur in strongly acidic conditions except for Fe, Al, Pb and Zn.



183



184

185 **Figure 3.** Solubility of metal hydroxides as a function of pH (Marchioretto et al., 2005)

186 Marchioretto *et al.* (2005) reported that when Fe and Al are present in landfill leachate, adsorption
 187 and co-precipitation may occur between Cr, Pb, and Zn with Fe(OH)₃ and Al(OH)₃ as pH increases.
 188 The leachability of metals is also influenced by the chemical and physical affinity of metal ions
 189 and various waste materials under landfill conditions (Ward et al., 2005). Sulphates in waste are
 190 reduced to sulphide that forms insoluble precipitates with most metals or containing amino acids

191 during anaerobic (Christensen et al., 2001). Dissimilatory microbial sulphate reduction is when
192 certain bacteria use sulphate as the electron acceptor in the oxidation of organic matter. However,
193 Cr does not form an insoluble sulphide; it is only precipitated out in the form of hydroxide.
194 Sulphides of the metals are more difficult to dissolve, both in oxidising and reducing environments
195 (Hammack and Edenborn, 1992).

196 **3.1.3 Carbonation and redox**

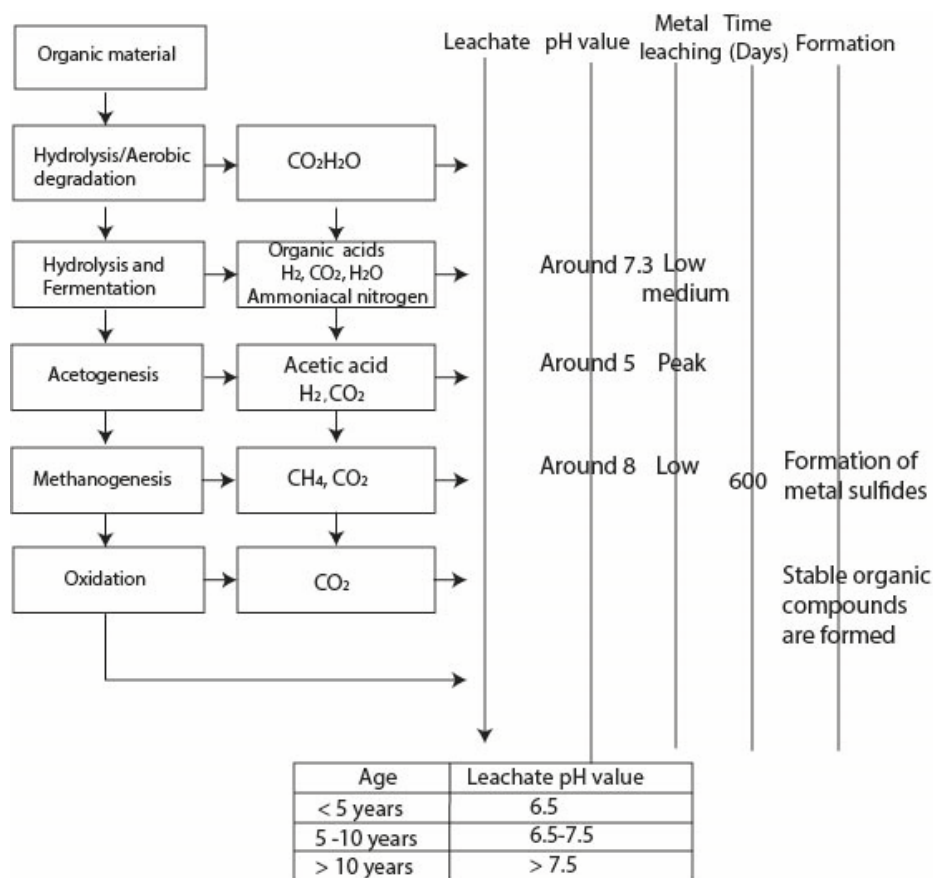
197 Carbonates are also capable of forming precipitates with metals and are abundant in landfill
198 leachate. Nevertheless, the solubility of metal carbonates is generally high (Christensen et al.,
199 2000). Metal precipitates of carbonate will dissolve, where the carbonate release will buffer the pH
200 value as the pH decreases, which is called the humic phase (Kjeldsen et al., 2010). As attenuation
201 mechanisms affect metal concentration and stability, it should be considered the metal adsorption
202 and precipitation-pH relationship for recovering metals with high concentration from an economic
203 point of view. The redox potential influences precipitation and should be considered when
204 considering metal solubility. Redox potential is a measure of the propensity of a chemical or
205 biological species to either acquire or lose electrons through ionisation (Lu and Marshall, 2013).
206 Various parameters in landfill leachate can reflect transformations in redox potential. For example,
207 as sulphate is reduced, their concentrations decrease. An increase in redox potential effects on the
208 oxidation of reduced sulphur compounds to SO_4^{2-} . Oxidation of metal sulphides takes place,
209 leading to metals release. The redox conditions in landfill leachate affect metal-organic interactions
210 through the organic ligands, as organics are sensitive to redox conditions (Merian and Clarkson,
211 1991). Abundant OM tends to have low redox potential values. The speciation of metals, which is
212 related to their mobility, is dependent on pH, redox, and organic compounds (Baun and Christensen,
213 2004). Each of the oxidation states has different metal complexation constants, and organic
214 compounds may mobilise it to an extent critically dependent upon the redox conditions (Herbert et

215 al., 1993). For example, Tingzong *et al.* (1997) found that Pb was bound to iron and manganese
216 hydroxide under oxidised conditions. As the landfilled waste shifted to reducing conditions, Pb
217 was leached out. Chuan *et al.* (1996) also reported that the solubility of Pb, Cd and Zn in soils
218 increased when redox potential decreased, and this was due to the dissolution of Fe-Mn ox-
219 hydroxides under reducing conditions resulting in the release of metals. In contrast, Sims and
220 Patrick (1978) found that soluble Zn decreased at low redox potential, which may be caused by
221 different environmental conditions and soil types. Also, Kamon *et al.* (2002) found that low redox
222 potential and alkaline conditions induced by anaerobic respiration in landfill sites tend to prompt
223 immobilization of Zn but a mobilization of Iron. Overall, redox potential strongly affects the
224 behaviour of metals in leachate even though there uncertainty remains regarding to what degree
225 such as different environmental conditions.

226 **3.2 Organic matter decomposition and metal leachability**

227 As landfill age increases, the leachate passes through successive stages of organic substance
228 decomposition, which influences metal leachability. Metal leachability is highest when
229 hydrolysis, fermentation and acetogenesis dominate due to an accumulation of VFA and a pH
230 decrease (Fig 4). The primary acids formed during fermentation are acetic acid (CH_3COOH),
231 propionic acid ($\text{CH}_3\text{CH}_2\text{COOH}$), butyric acid ($\text{CH}_3\text{CH}_2\text{CH}_2\text{COOH}$) and ethanol ($\text{C}_2\text{H}_5\text{OH}$).
232 Carboxylic acids act as chelating agents, and there may be an increase in carboxylic functional
233 groups on humic compounds due to the waste being oxidised (Kjeldsen *et al.*, 2010). Qu *et al.*
234 (2008) demonstrated that metals in leachate are bound to organic substances such as fatty acids,
235 FAs and HAs. The fatty acids, FAs, and HAs content in leachates decrease as landfill age
236 increases (Fan *et al.*, 2006; He *et al.*, 2006; Qu *et al.*, 2008). The fatty acids are accumulated
237 during the acid phase of the waste stabilisation (Christensen and Kjeldsen, 1989). FA

238 predominates in young unstable leachates, and its concentration decreases as landfill age
239 increases. The HA-forming processes are dependent on microbial degradation of OM, and the HA
240 increases with the age of leachate, eventually reducing due to the leachate becomes more stable
241 and diluted (Artiola-Fortuny and Fuller, 1982). HA has more carboxylic groups than FA and
242 contains bands of aromatic C=C (Gustafsson and Berggren, 2005; Shirshova et al., 2006). The
243 binding capacities of HS to metals within leachate and solid waste may imply that the solubility
244 of HS strongly influences the mobility of metals (Qu et al., 2008). To the best of our knowledge,
245 no work has been reported on the effects of organic matter decomposition on metal release in
246 landfill environments. It is expected that different metals have different impacts on the
247 decomposition processes of organic matter. Further research is required on the role of organic
248 matter degradation on the release of individual metals.



249

250 **Figure 4.** Metal leaching Process in a landfill and leachate pH value (Adapted from Zainol et al.,
 251 2012, Adhikari et al., 2014)

252 **3.3 Chelation and complexation with organic substances**

253 Most metal ions bind to neutral molecules in different oxidation states called a ligand, defined as
 254 an ion or molecule that binds to a central metal atom to form a complex (both organic; carboxylic
 255 acids, amino acids, HAs and inorganic) (Table 4). Ligands lead to the formation of metal complexes
 256 and metal chelates (Fig 5). Complexation with organic ligands is known to influence the mobility
 257 of metal by either increasing or decreasing its sorption on mineral surfaces. Many organic
 258 substances have been commonly identified in leachates worldwide (Details in supplementary data
 259 Table 1) (Paxéus, 2000; Staley et al., 2006; Zhang and Zhang, 2009).

260 **Table 4.** Mechanisms of adsorption for organic compounds in soils (adapted from Sposito, 1984)

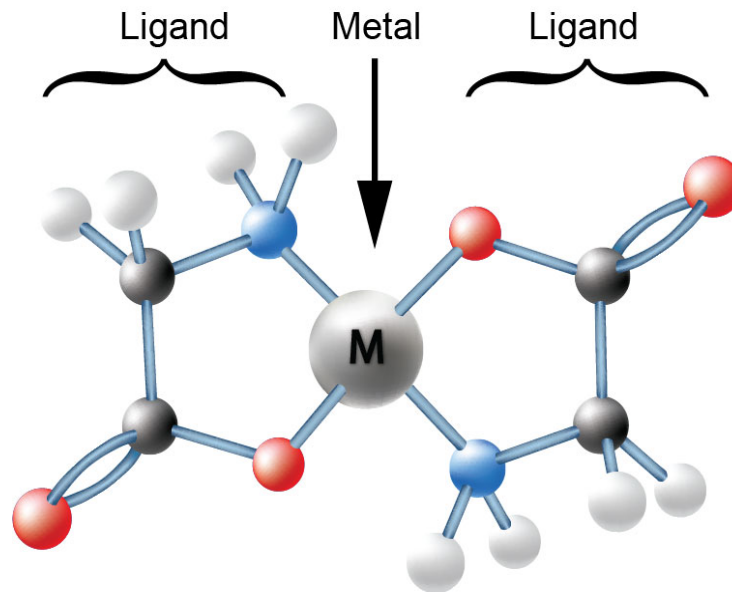
Mechanisms	Principal organic functional group
Cation exchange protonation	amines, ring NH, heterocyclic N amines, ring NH, Heterocyclic N, carbonyl, carboxylate
Anion exchange water bridging	carboxylate amino, carboxylate, carbonyl alcoholic OH
Cation bridging	carboxylate, amines, carbonyl, alcoholic OH
Ligand exchange hydrogen bonding	carboxylate amines, carbonyl, carboxyl, phenylhydroxyl
Van der waals bonding	uncharged, nonpolar organic functional groups

261

262 Among the organic ligands, HS are the main organic compounds present in landfill leachate (Zhou
 263 et al., 2015). HS are the main component of soil OM or humus, most of which combine with the
 264 inorganic constituents in the soil (Pettit, 2004). HS have several functional chemical groups
 265 (carbonyl, hydroxyl carboxylic acid, phenolic ring, and quinone), which may combine with ions
 266 such as Fe^{3+} , Mg^{2+} , and Ca^{2+} and form chelate complexes (Fig 6); thus change the solubility of
 267 metals (Tipping et al., 2002). Generally, the potential for complex formation between metals and
 268 organics increases with pH alkalisation (Hummel et al., 2000). Farrah and Pickering (1997a)
 269 showed that the proportions of metal bound as hydroxyl complexes increase at pH 5 and above
 270 although the capacity for complexation shows no dependence on pH (Antelo et al., 2000). Instead
 271 the metal-organic interaction depends on the stability of complex formation and metal
 272 concentration. Esakku *et al.* (2003) reported higher stability constants for Cu complexes with OM
 273 and that these lead to higher Cu content in the organic fraction.

274 Phenolate, amino, and carboxylate groups enhance the formation of metal complexes at high pH,
275 thus become increasingly stable at higher pH levels (Rieuwerts et al., 1998). Carboxylic and
276 hydroxyl functional groups show acid-base behaviour.

277

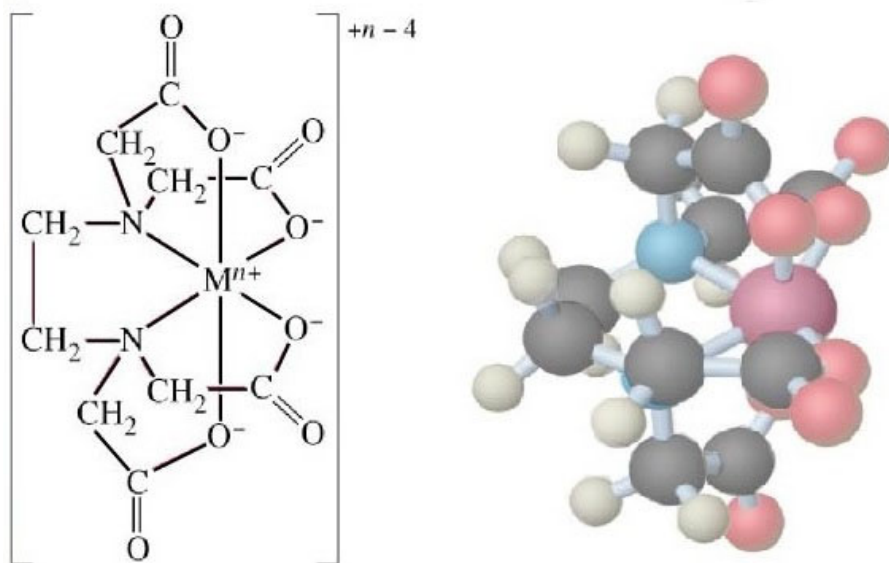


278

279 **Figure 5.** An amino acid chelates (Albion Technology, 2021)

280 At low pH, hydrogen ions compete with the metal ions for these sites, and as pH increases, less
281 hydrogen ions are present and complex site availability for metal ions increases (Scott et al., 2005).

282 There are challenges to understanding the complexation of different metal ions, e.g. i) organic
283 compound functional groups influence the type of reaction it has with metals; and ii) the length of
284 hydrocarbon chain length in carboxylic acid increases its metals adsorption capacity but decreases
285 its stability as complex (Abollino et al., 2003).



286

287 **Figure 6.** Metal ion complexation and chelate effect (Adapted from Tsezos et al., 2006)

288 Soil organic matter can influence the mobility and speciation of metal, where complexation
 289 reactions modify its accumulation potential (Kennou et al., 2015). For example, when organic
 290 materials, rich in soluble organic carbon and a large proportion of FAs are applied to soil, metal
 291 mobility increases due to the formation of soluble metal-organic complex (Pérez-Esteban et al.,
 292 2014). In contrast, when a chelating agent binds to a metal ion in more than one place
 293 simultaneously, chelated compounds become more stable (Pohlmeier, 2004). It has long been
 294 recognised that complexation may lead to increase metal solubility or decrease adsorption
 295 (Cavallaro and McBride, 1978; Bradl, 2004; Güngör and Bekbölet, 2010; Ahmed et al., 2019).
 296 Similar observations have been shown to occur within the landfilled waste. A variety of organic
 297 compounds can be expected in the leachates, which afford the potential for metal-organic
 298 interactions through the organic ligands. Previous studies established that dissolved organic matter
 299 (DOM) in MSW has a high affinity for metals, especially for Cu and Pb (Christensen et al., 1996;

300 Christensen et al., 2000; Huo et al., 2008). Most insoluble metals are present in their refractory
301 chemical form, i.e. PbSO₄. Over time the oxidation/ reduction of these metals to a soluble form
302 through complexation appears likely (Takahashi et al., 2010). If insoluble metal-DOM complexes
303 are formed, the mobility of the metals in question and the DOM to which they are complex
304 decreases. Metal mobility is less clear when soluble complexes are created with DOM (Jansen et
305 al., 2003). On the one hand, it may increase because, i) the mobility of DOM is affected by its
306 functional groups and adsorption to soil particles (Kaiser et al., 1997); ii) binding to DOM prevents
307 immobilisation by precipitation of inorganic metal complexes. On the other hand, the mobility of
308 soluble metal-DOM could decrease complexes when they bind to soil particles through cation
309 bridging (Guggenberger and Zech, 1993).

310 The leachability of metals could be enhanced through ligand complexation where organic acids
311 such as carboxylic acids and phenols, formed during the decomposition of organic compounds,
312 decrease pH. The pH determines the number of acidic functional groups on deprotonated DOM,
313 which increases the availability of sorption sites for binding metals (Stevenson, 1994).

314 Jensen *et al.* (1999b) determined organic complexes of heavy metals in landfill leachate polluted
315 groundwater in the Vejen landfill. They found that organic complexes made up a significant part of
316 the total content of heavy metals: Cd 85%, Ni 27-62%, Zn 16-36%, Cu 59-95%, and Pb 71-91%.
317 Kalis *et al.* (2006) found that the metal-humic acid complexes become the dominant complexed
318 species when humic acid is present. Yu *et al.* (2018) reported that most of the complexes between
319 Cd and the HS would be insoluble, and the complexation could contribute significantly to the
320 reduction in the concentration of Cd in soil solution. Van Ginneken *et al.* (2001) discovered that
321 the stability of chelated metals and noncyclic metal complexes depends on several factors,

322 including pH, metal oxidation state, and ionic strength. Organic-metal complexes are increasingly
323 stable at higher pH levels due to the ionization of functional groups (Jones and Jarvis, 1981;
324 Rieuwerts et al., 1998). Conversely, organic acids present in the dissolved organic carbon (DOC)
325 may act as chelating agents, enhancing the mobilisation of metals (Christensen et al., 1996).

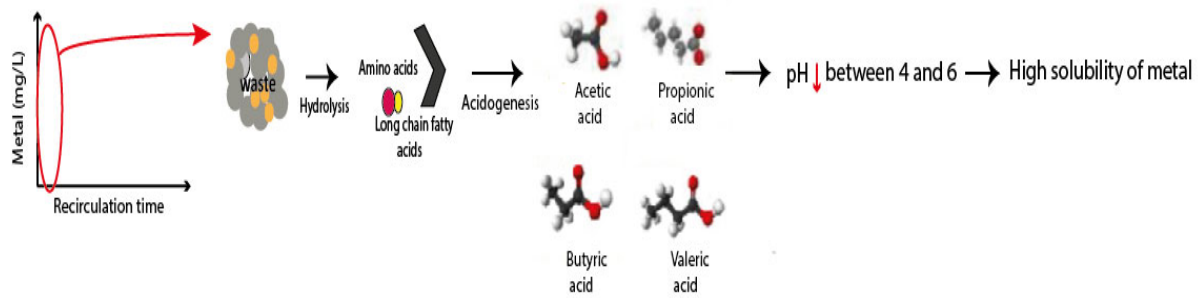
326 Complexing behaviour significantly influences metal attenuation as it affects their mobility and
327 saturation indices (Qu et al., 2019). As metal-organic complexation plays a critical role in the
328 mobility of metals in landfill environments, lab and field experiments would be required to
329 establish the relationship between complexing characteristics and observed metals leaching,
330 performed under varying environmental conditions to optimise metals recovery.

331 **4 LEACHATE RECIRCULATION STRATEGY FOR METAL RECOVERY**

332 Leachate recirculation within landfills has been widely used for a range of purposes since the 1970s,
333 including leachate management, enhanced landfill gas generation or recovery, and improved
334 landfill sustainability (EPA, 2009). Leachate recirculation is a process where leachate is re-
335 introduced into the landfill through an artificial recharge system (White et al., 2011). This technique
336 aims to encourage saturation to stimulate the degradation processes, leading to more rapid
337 stabilisation of the landfill (Scott et al., 2005). However, leachate recirculation can also increase
338 the chloride content. Chloride contents may also be an important controlling factor for metal release.
339 Chloride affects the behaviour of metals by binding the metals on humic acids and the adsorption
340 of metals, such as the adsorption of Cd on iron hydroxides or their desorption mobility (Guevara-
341 Riba et al., 2005; Begeal, 2008; Damikouka and Katsiri, 2020). The ionic forms of Cu and Cd can
342 form metal compounds with the anions such as CuCl_2 , CdCl_2 or CuSO_4 , leading to chloro-
343 complexation and the formation of dissolved metal-chloride compounds increase the mobility of

344 metals (Kirkelund et al., 2010; Damikouka and Katsiri, 2020). This implies that leachate
345 recirculation affects metal recovery. Leachate recirculation can significantly influence metal
346 behaviour and fate within waste matrices (Ledakowicz and Kaczorek, 2004). For example, Yao *et*
347 *al.* (2014) found that leachate recirculation contributed to faster stabilisation of the landfill and
348 reduced leachability of Cu and Zn from the landfill. He *et al.* 2007 have also shown that
349 recirculating leachate, which is by the sequential reactors, in landfills in the early stage allowed
350 methanogenesis to be reached much earlier and that this was accompanied by a reduction of total
351 metals released from landfills. In contrast, Qu *et al.* (2008) demonstrated that the initial stage of
352 leachate recirculation had low leachate pH (5-6) and highly VFA levels (acetate 4500-700 mg/L,
353 propionate 1450-2950 mg/L and butyrate 4500-7200 mg/L) due to the acidification stage, in
354 resulting the concentration of the metals was high at this stage. Bilgili *et al.* (2007) showed that the
355 release of metals can significantly increase at the beginning of leachate recirculation as in the early
356 stages of the waste degradation, pH of the leachate is low contributing to higher solubility of metals
357 and dissolution into leachate (Fig 7).

358 As stated previously, due to the low concentrations of metals, the way to make it economically
359 viable in recovering metals is to maximise metal concentrations. Recirculating leachates will
360 accentuate the potential for increased metal mobility within and from the landfill when oxidised
361 conditions are introduced. Leachates have been shown to have an increased capability to enhance
362 metal mobility when oxidised (Mårtensson et al., 1999). It implies that leachate recirculation
363 provides higher extractable metals in the initial leaching phase.



364

365 **Figure 7.** Leachate recirculation strategy for metal recovery

366 Therefore, regarding the critical challenges in metal recovery in leachate, this suggests that where
 367 leachate recirculation is applied with combining organic matter such as HA and pH could increase
 368 the economic feasibility.

369 **5 Opportunities for metals recovery from landfill leachate**

370 Leachate contains a wide range of metal such as Cu, Zn, Cr, Cd in addition to REEs (Kjeldsen et
 371 al., 2002). Therefore, leachate in landfill sites has a great potential for metal recovery. There are
 372 several ways for recovering metals from waste, such as recycling, physico-chemical, thermo-
 373 chemical, pyro-metallurgical, hydro-metallurgical, bio-metallurgical, landfill-mining methods
 374 (Wang et al., 2017). Due to the lack of economically and operationally feasible primary resources
 375 for the production of metals, many countries are forced to depend on recycling metals from
 376 secondary sources such as industrial residues and end-of-life products. Table 5 shows various
 377 methods to recover metals from waste streams and leachate, suggesting the possibility of recovery
 378 metals in landfill leachate. Despite published research works on recovering metals from leachate,
 379 studies are scanty in an application towards the recovery of metals from real landfill leachate in the
 380 site. The main problem to recover metals from leachate is further compounded for metals that exist

381 at low concentrations (Table 2), which can be affected by many factors such as landfill age, type,
382 and chemical and physical mechanism in landfill environments. Also, the strong acidic condition
383 can affect the environment; thus, it may later pose harmful risks to the environment if not managed
384 well. The concentration of metals varies widely, and REEs has a very low concentration (1 or 2
385 $\mu\text{g/g}$ waste), which is limiting the economic viability of the recovery. Relatively, high
386 concentrations of over 1% are needed to approach the cost-effective recovery metals (Umeda et al.,
387 2011). Therefore, process optimisation is important to maximise the concentration of metals or co-
388 extraction of other added-value materials such as nitrogen and phosphorus, improving the process's
389 cost-effectiveness and efficiency. By understanding the processes, including organic-metal
390 interaction in leachate, recovering metal can achieve higher average productivity.

391 Barriers remain in recovering valuable materials present due to the unknown concentrations and
392 distributions of metals in landfills and not meet the reasonable financial level. Metal recovery from
393 leachate has not been investigated before, but prior studies have demonstrated that metals can be
394 recovered from wastewater and aqueous solutions. However, the methods remain limited; for
395 example, physicochemical methods are energy and capital intensive due to the costs of chemicals,
396 oxidants, and membranes; the biological treatment process is limited by treatment effectiveness
397 and energy requirement (Ahn et al., 2002; Kargi and Pamukoglu, 2003).

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403 **Table 5.** Metals recovery from wastewater and liquid solution

Source	Method	Characteristics	Effects	Reference
Leachate from landfill waste	Bioelectrochemical systems (BES)	·BES employs biological and electrochemical reactions to recovery resources from a wide range of substrates.	·The electrical conductivity of leachate makes it favorable for electricity generation, and it contains a high concentration of ammonium nitrogen, which may be recovered for agricultural application. ·Metal also may be recovered by the modified microbial electrolysis cells.	Iskander et al., 2016
Sulfate leachate of cathode scrap of Li-ion batteries	Liquid-liquid extraction	1) The cathode scraps undergo heat treatment to completely liberate the cathodic materials from aluminum foil. 2) Solubilise Co, Li, Fe, Mn, Ni, and Al by leaching the cathodic materials in sulphuric acid in the presence of H ₂ O ₂ . 3) Oxidative precipitation of Mn from liquor with KMnO ₄ and extraction of Al and Fe using D2EHPA. 4) Treat to recover the metals.	·Focuses on selective recovery of Co, Ni, and Li from the sulphate leachate of cathode scarp generated during the manufacture of Li ion batteries. ·High-purity Co in a solution can be recovered by solvent extraction using the sodium salt of PC-88A. ·The metals extraction efficiency and separation factor depend upon the extractant concentration and the equilibrium pH of the aqueous phase.	Nguyen et al., 2014
Solution and leachate derived from electronic scrap	Biogas	·Precipitates metals from solution using the off-gas ·Recovery Au, Pd, and Ag from leachate derived from electronic scarp; safe microbiologically	·Au was recovered from electronic scrap leachate with selectivity against Cu using biogas as they could partially separate Au from Cu. ·In acidic conditions, Au and Cu are removed rapidly and separated from the liquor. ·The solid Pd and Ag will not easily be separated in water via biogas. ·Amines must be avoided for recovering metals using biogas.	Macaskie et al., 2007

End of life electronic wastes	Hydrometallurgical process	<ul style="list-style-type: none"> ·Ferric sulphate concentration range (at 1:10 and 1:5 Cu to sulphide molar ratio 1) filled up by using N₂ gas for anaerobic conditions 2) add of 10 ml of Na₂S·9H₂O solution 	<ul style="list-style-type: none"> ·Effect of Fe³⁺ on leaching of Cu and selective recovery of Cu from the polymetallic leachate. ·Lixiviant concentration and pH were the important parameters in CuSO₄ precipitation. ·The precipitation mostly occurs in the acidic pH range (0.5 to 1.5). ·CuSO₄ can be further pyro/hydrometallurgical processed to produce Cu metal. 	Sethurajan and Hullebusch, 2019
Aqueous solutions	Biosorption	<ul style="list-style-type: none"> ·The phosphorylation yeast cells were used in Cu adsorption experiments with 0.1 M HCl, which is strongly influenced by the pH of the solution. ·Recovering metals from aqueous solutions. The biosorption of metals is a complex process affected by the adsorbent, the types, and the concentrations of metals in the solution. 	98% of the Cu ions adsorbed to phosphor cells could be recovered by treating the cells with HCl.	Ojima et al., 2019
Sulfuric acid leaching liquor of spent Li-ion batteries	Hydrometallurgical process	<ul style="list-style-type: none"> ·Needs to refine the residues into a purer form such as salts, hydroxides and metals. 1) selective precipitation method by adding dimethylglyoxime (DMG, C₄H₈N₂O₂) reagent 2) extraction using cobalt loaded phosphoric acid (D2EHPA) 3) Separation and recovery of metal (Ni, Mn, Co, and Li) from sulfuric acid leaching liquor 	High purities of Co and Li were recovered as CoC ₂ O ₄ ·2H ₂ O and Li ₂ CO ₃ .	Chen et al., 2015
Acid mine leachate	Sequential precipitation	<ul style="list-style-type: none"> 1) Sequential precipitation; add a sodium hydroxide solution of 5 M 2) Selective dissolution; pre-concentrates of the valuable elements were re-dissolved into solution 3) Oxalic acid precipitation; 	·95% of the Cu and Zn were recovered from the residual liquid using Na ₂ S at pH 2 and 3.	Zhang and Honaker, 2020

		dissolving 8 g oxalic acid dehydrate in 50 ml deionised water using an ultrasonic batch 4) Na ₂ S precipitation; 1 M Na ₂ S REEs, Cu, Zn Ni and Co recovery from an acid mining leachate. The sample was collected from a coal preparation plant.	·The optimise the oxalic precipitation for the REE recovery is using a solution pH of 1.2	
Sludge	Precipitation	·Add 3 mol dm ⁻³ NaOH, ·The recovery process of heavy metals from polluted sludge leachate with biosurfactant elution by batch and column experiments.	The recovery efficiency of heavy metals (Pb, Ni, and Cr) reached over 90% by the precipitation method with pH 10.9.	Gao et al., 2012
Wastewater	Cementation	·Add Fe, Al, and Zn metallic powders into 250 mL of wastewater ·Stir continuously with a magnetic stirrer	·Cu, Au, and Pd can be recovered by using Fe and Al powder. ·Precious metals can be effectively recovered by combining processes (cementation, neutralisation and reduction)	Umeda et al., 2011
Wastewater	Photoelectrochemical cell	·A stock solution was prepared by dissolving the metal salts into deionised water ·The photoanode and the Pt strip cathode were connected with a commercial Cu wire ·The photoanode was irradiated with a UV lamp	·Heavy metals were recovered by mechanical scratching of the cathode surface.	Wang et al., 2017
Wastewater	Electrochemical reactor	·Prepare solutions using deionised water ·Determining the quality characteristic to be optimised. ·Identification of the noise factors and test conditions. ·Identification of the control parameters.	·The highest efficiencies were obtained for Pb and Cu recovery from diluted solution: 75.8 % and 89.9 %	Kaminari et al., 2007

405 Bioelectrochemical systems (BES) is an environmental strategy that employs biological and
406 electrochemical reactions to generate electricity and recovery resources from a wide range of
407 substances. Organic compounds in BES tends to produce electricity and other value-added
408 compounds by oxidising microorganisms. However, a high concentration of metals in landfill
409 leachate can be recovered through BES, and the reduction in leachate volumes can be achieved
410 using osmotic processes integrated with BES. Also, hydrometallurgical processes have gained
411 considerable attention as they show effectiveness in the extraction of metals (Gunarathne et al.,
412 2020). Hydrometallurgical metal recovery is typically performed in three main stages: metal
413 dissolution, concentration and purification, and metal recovery (Gupta,2006). Thus, further
414 research should be studied to take a circular approach, recovering metals from landfill leachate
415 using BES after enhancing metal concentration through leachate recirculation or
416 hydrometallurgical processes using less toxic chemical solvents to be used as leaching agents and
417 assist of acids and pH value.

418 **6 Conclusion**

419 The literature review on landfills has shown a metals recovery opportunities in landfill leachate
420 using factors influencing metal mobility in landfill environments. Landfill leachate is a significant
421 potential resource in landfill as it contains a large variety of dissolved extractable metals. Metal's
422 recovery opportunities may increase by several factors, influencing metal mobility in landfill
423 environments as an excellent challenge for metal recovery from leachate is the low concentration
424 of metals. Younger landfill leachate (>5 years old) has higher organic matter content due to the
425 generation of dissolved and solubilised organic matter, consequently increasing metal release.
426 Therefore, it implies that metal recovery may be effective in younger landfill leachate. Physio-
427 chemical processes affect soluble metal concentration, which is critical to predicting metal recovery

428 as they can govern the mobility of metals. More knowledge is required concerning the complexes
429 in leachate in general and specifically on the importance of the organic matter in leachate, which
430 led to an increase in the metal release rate. This article has also identified the gaps and has indicated
431 that further efforts are required concerning leachate recirculation. It may ensure that maximising
432 the concentration of metals from landfill leachate will be the economic feasible. Although mature
433 technological advances provide opportunities for recovering metals from landfill leachate,
434 significant challenges await us ahead as they can hardly be regarded as economical. Therefore, the
435 chemistry mechanism of landfill environments should be well understood and fundamental and
436 practical barriers of the recovering process in landfill leachate, which will lead us one step closer
437 to resource recovery paradigm for a circular economy in closed landfill.

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