| 1 | Bioleaching for resource recovery from low-grade wastes like fly and bottom ashes from |
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| 2 | municipal incinerators: A SWOT analysis |
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| 11 | Highlights |
| 12 | • Circular economy depends on resource recovery from wastes |
| 13 | • Strengths, Weaknesses, Opportunities, and Threats (SWOT) identified for bioleaching |
| 14 | • Bioleaching can be a sustainable technique for metal recovery from low-grade wastes |
| 15 | • Resource recovery demands integrated policy and regulatory framework |
| 16 | |
| 17 | Abstract |
| 18 | Bioleaching (or microbial leaching) is a biohydrometallurgical technology that can be |
| 19 | applied for metal recovery from anthropogenic waste streams. In particular, fly ashes and |
| 20 | bottom ashes of municipal solid waste incineration (MSWI) can be used as a target material |
| 21 | for biomining. Globally, approximately 46 million tonnes of MSWI ashes are produced |
| 22 | annually. Currently landfilled or used as aggregate, these contain large amounts of marketable |
| 23 | metals, equivalent to low-grade ores. There is opportunity to recover critical materials as the |
| 24 | circular economy demands, using mesophile, moderately thermophile, and extremophile |
| 25 | microorganisms for bioleaching. A Strengths, Weaknesses, Opportunities and Threats (SWOT) |
| 26 | analysis was developed to assess the potential of this biotechnology to recover critical metals |

from MSWI wastes. Bioleaching has potential as a sustainable technology for resource
recovery and enhanced waste management. However, stakeholders can only reap the full
benefits of bioleaching by addressing both the technical engineering challenges and regulatory
requirements needed to realise and integrated approach to resource use.

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32 Keywords: Circular economy; critical raw materials; biotechnology; municipal solid waste
33 incineration – MSWI; waste management

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35 **1. Introduction**

36 Our current patterns of consumption are leading to the exhaustion of planetary resources, 37 while simultaneously generating pollution and threatening our survival as a species. There is 38 an urgent need for a change in paradigm in waste management to efficiently recover resources 39 (energy, metals, nutrients) from waste streams, making industrial and urban processes more 40 efficient. According to the recent UN Global Resources Outlook, extractive industries are 41 responsible for half of the global carbon emissions (Oberle et al., 2019). Resource extraction 42 and processing caused 90% of biodiversity loss and water stress, which puts a more dangerous 43 level of pressure on climate and natural life support systems than previously thought. Resources are being extracted three times faster than in 1970, even though the population has only doubled 44 45 in that time, according to the same report. The United Nations Sustainable Development Goals 46 (SDG) set a target to achieve sustainable management and efficient use of natural resources by 2030, by decoupling economic growth from resource use and environmental degradation. This 47 is to achieved by improved resource efficiency, decreased reliance on raw materials, and 48 49 increased recycling to reduce environmental pressure and impact. "Circular economy" and "zero waste" are buzzwords today, but both goals still look unattainable (Velenturf et al., 2019). 50

51 Metals are valuable raw materials for the global economy, and key components of various 52 products such as low-carbon energy technologies, electric vehicles, and electronic and 53 biomedical devices. Large quantities of critical and scarce metals, such as platinum group 54 metals (PGM), rare earth elements (REE), cobalt, vanadium, selenium, and tellurium are required for storage and production of renewable energies, catalytic processes, digital 55 56 communication, and green technologies (Hofmann et al., 2018; Işıldar et al., 2019). Raw 57 materials and metals are considered critical when they have high economic importance 58 combined with an elevated risk of supply, mainly resulting from high production in countries 59 with poor governance (geopolitical instability), limited material replaceability, and low endof-life recycling rates (EC, 2017). 60

61 Municipal solid waste incineration (MSWI) has been globally adopted for the management 62 of vast amounts of waste (a global average of 130 tonnes per year) (Joseph et al., 2018), as it 63 allows energy recovery and reduction in the volume of waste sent to landfill. However, the 64 incineration of municipal solid waste destroys technical value given that once the energy value 65 of waste has been recovered by burning, it is no longer available to the circular economy (Purnell, 2019). Coarse metals (>2mm) remaining in fly ashes and bottom ashes are typically 66 67 recovered, and the residual fraction is recycled as construction material. However, low but significant quantities of high-tech, high-value metals remain in the residual material. Estimated 68 69 annual flows of these high-value metals are in the order of tens of kg and a total content 70 comparable to low-grade active mines (Funari et al., 2015). The potential for urban mining and 71 recovery of secondary resources from MSWI residues is, therefore, promising as a way of 72 closing the loop within a circular economy (Simon and Holm, 2018).

Given the low metal concentrations, economic feasibility is dependent on using low cost,
sustainable technologies, such as bioleaching. Bioleaching is commercially used for the
recovery of metals from low and waste grade ores, in particular for copper (Gomes et al., 2018).

76 These ores would typically be uneconomic to process using conventional comminution-77 concentration-flotation routes. Bioleaching uses microorganisms isolated from natural settings 78 (e.g. extreme environments, acid mine drainage) to generate mineral or organic acid (as 79 metabolites) and improve metal solubility by enzymatic reactions. Bioleaching can be 80 performed by direct contact (primary bioleaching) and by indirect leaching (or secondary 81 bioleaching). The latter only uses the acid produced by bacteria to recover metals without a 82 direct inoculation. This approach may be best suited in some circumstances, e.g. for alkaline wastes where the conditions are not favourable for the survival of the typically acidophilic 83 84 bacteria However, both approaches are acknowledged to lessen environmental and economic 85 drawbacks during treatment of anthropogenic waste, compared to using mineral acids (Funari 86 et al., 2017).

87 This paper aims to assess opportunities and limitations associated with bioleaching of low-88 grade wastes for metal recovery, in the context of the circular economy. It is beyond the scope 89 of this study to provide a comprehensive review of the latest developments of bioleaching. In 90 recent years, there has been an increasing amount of literature on bioleaching, with several extensive reviews covering the topic (Auerbach et al., 2019; Pollmann et al., 2018; Sethurajan 91 92 et al., 2018; Srichandan et al., 2019). Our objectives are to focus on practical aspects and 93 limitations that remain obstacles for the full implementation of bioleaching as a crucial tool for 94 the circular economy.

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96 2. Bioleaching as an alternative for resource recovery from MSWI waste

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98 Current alternatives used at industrial scale for resource recovery from MSWI fly ashes are
99 based on acid leaching (e.g. FLUWA process), and washing processes for salt recovery (e.g.
100 HALOSEP®) (Quina et al., 2018). At lab scale, hydrometallurgical processes for copper and

zinc recovery (Tang et al., 2018), electrodialytic processes (Kirkelund et al., 2015), and
treatment processes combining leaching, selective extraction and adsorption (Tang et al., 2019)
have shown promising results, but are still at low technology readiness levels (TRL).

A recent review of the different removal techniques for metals from fly ash compared bioleaching, carrier in pulp method, chemical extraction with different acids, alkaline leachates and chelating agents, chloride evaporation process, electrodialytic and thermal treatments (Meer and Nazir, 2018). The authors concluded that the selection of the best process depends on the type of fly ash and target metal(s), but also of factors like cost, time and energy (Meer and Nazir, 2018).

110 Bioleaching of bottom ash (BA) and fly ash (FA) with a mixed culture isolated from a 111 natural system showed good yields of metal extraction, with more than 90% Zn, Cu, and 10% 112 Pb removed from FA; while 100% Cu, 80% Zn and 20% Pb were removed from BA samples 113 (Funari et al., 2019). Bioleaching of bottom ashes with pure cultures Acidithiobacillus 114 ferrooxidans or Leptospirillum ferrooxidans, or a mixture of Acidithiobacillus thiooxidans and 115 Acidithiobacillus ferrooxidans in batch tests showed that Al, Cr, Cu, Ni, Mn and the rare earth elements Ce, La, and Er were significantly more extracted with iron-oxidizing bacteria 116 117 compared to abiotic controls (Auerbach et al., 2019). The results are encouraging for industrial application to recover concentrated metals like Al and Cu, simultaneously reducing the cost of 118 119 landfilling the remaining residues. Continuous heap bioleaching at lab scale showed leaching 120 yields for zinc and copper between 18-53% and 6-44% (Mäkinen et al., 2019), but also 121 highlighted the need for further optimisation, in particular regarding acid addition and aeration. Bioleaching using alkaline autochthonous extremophiles isolated from a fly ash landfill site 122 123 showed that Alkalibacterium sp. TRTYP6 could recover 52% of Cu (Ramanathan and Ting, 2016). The use of fungi for fly ash bioleaching showed that fungal morphology of Aspergillus 124

niger was significantly affected during one-step and two-step bioleaching, with precipitation
of calcium oxalate hydrate crystals at the surface of hyphae (Xu et al., 2014).

127 Several factors can influence bioleaching efficacy, such as pH, temperature, pulp density, 128 redox potential, microorganisms or communities involved, particle size, oxygen and iron concentrations, and wastes mineralogy (Sethurajan et al., 2018). Also, metal bioleaching is 129 130 influenced by biomass concentration, metal tolerance of microorganisms, type and amount of metabolic products released into the medium, contact time, and pretreatment (e.g. heating) and 131 132 has to be assessed case by case (Pollmann et al., 2018). Nevertheless, bioleaching can be a 133 flexible and environmentally friendly alternative for conventional processes, as it allows the recovery of valuable resources, but can also reduce the toxicity of the waste for further reuse 134 135 in other applications (e.g. aggregate materials) (Auerbach et al., 2019).

136 **3. SWOT analysis**

Strengths, Weaknesses, Opportunities, and Threats (SWOT) analysis is a framework
technique used in business to facilitate the development of a sustainable market niche by
uncovering new outlooks and identifying problems that would hinder progress (Miller, 2007).
Table 1 summarises a SWOT analysis for the use of bioleaching for metal recovery from MSWI
fly ashes and bottom ashes.

142

144 Table 1. Strengths, Weaknesses, Opportunity and Threats (SWOT) analysis for

| Strengths | Weaknesses |
|--|--|
| Lower environmental footprint (avoids strong mineral acids used in hydrometallurgy methods, lower energy consumption) Not labour intensive Different microorganisms (fungi, isolate/mixed acidophilic/alkaline bacteria) can be used for bioleaching Bioleaching can be achieved in onestep, two-step and spent medium-step Minimal investment and low operating costs compared with hydrometallurgy methods Can be performed <i>in situ</i> Technology readiness level 9 for primary ores Can be used to reduce contamination of wastes/biostabilization | Depends on quantities/concentrations of metals in wastes Presents slow dissolution kinetics and low metal leaching yield Heap bioleaching can be space demanding Adaptation of microorganisms to waste materials is critical Alkaline wastes are not favourable to the growth of acidophile bacteria More data on alkaline bioleaching are needed Not fully reproducible, as it depends on the feedstock material Inhibitory layers hinder cell-mineral interaction (passivation effects) Lack of research dedicated to process development and reactor design No pilot-scale applications for anthropogenic streams such as FA and |
| | BA from MSWI |
| Opportunities | Threats |
| Recovery of metals from low-grade ores and wastes can be a potential offset for remediation and operation costs in both operating and legacy sites Development and use of bioelectrochemical systems for energy production and metal recovery Minimise the environmental impacts of raw materials extraction Fine-tuned bioleaching to enhance selectivity to targeted metals Accelerate carbonation and carbon sequestration using microorganisms | Hight volatility of markets and metal prices Low cost of waste disposal in landfill Low mineral extraction costs ("mineralogical barrier") No alternatives for the downstream processing of excessive biomass produced Lack of satisfactory coverage for substrate materials and inocula Lack of work practices may lead to poo reproducibility Higher bioavailability of potentially |

145 bioleaching for resource recovery from MSWI residues.

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148 **3.1 Strengths**

149 Bioleaching is a technology considered environmentally friendly, for being less aggressive 150 (lower use of concentrated acids/bases) and energy-intensive than traditional 151 hydrometallurgy methods. It can be performed *in situ*, as heap bioleaching, with minimal investment, and low operating costs. The process is not labour-intensive since only a few 152 153 process parameters need essential monitoring and control. Unskilled operators can perform the key maintenance operations with minimal risks (e.g., no use of strong acids like 154 155 hydrofluoric acid), including the manipulation of bacterial matter. MSWI bioleaching 156 bacteria show low biological hazard and risk of contamination, with no identified risks to 157 humans, especially those cultures isolated from natural systems of well-known 158 characteristics. Moreover, bioleaching combines traditional hydrometallurgical methods 159 effortlessly and in a cost-effective manner, because it does not require highly sophisticated 160 monitoring and control instruments, the implementation of which can be expensive. Pure 161 bacterial cultures, mixed (acidophilic or alkaline) bacteria, archaea, fungi, algae and plants 162 metabolites can be used for bioleaching of different metal-bearing wastes. The bioleaching process can be achieved by one-step, two-step and spent medium-step, both in batch or 163 164 continuous modes and showed promising results on the extraction of several metals (Ni, Co, Mo, V, Fe, Zn, Cu, Cr, Cd, W, Pb and Mn) (Srichandan et al., 2019). Metal extraction takes 165 166 place directly by electron supply (e.g. oxidation or reduction reactions) or indirectly by 167 metabolic products of the microorganisms (inorganic or organic acids or excretion of complexing agents) (Auerbach et al., 2019). Bioleaching is a fully developed technology 168 (TRL 9 for primary ores), and has been used at industrial level since 1960 for extraction of 169 170 copper from sulphide ores. Currently, approximately 20% of the worldwide Cu is extracted using bioleaching (Latorre et al., 2016). Bioleaching can reduce contamination of wastes, 171 contributing to their biostabilization and can be used to extract metals from ores or 172

secondary wastes that are too low grade and therefore uneconomic to processing, usinghydrometallurgical or pyrometallurgical methods.

175 **3.2. Weaknesses**

176 For further implementation of bioleaching as a technology for the circular economy, there 177 is a need to improve process dissolution kinetics and metal leaching yields. Currently, 178 bioleaching is slower than traditional extraction methods. Dissolution kinetics can be 179 accelerated by optimisation of process parameters such as pre-treatments, reaction time, pH, 180 temperature, mass transfer rate, nutrient requirements, pulp density, and particle size. Further 181 developments in understanding the biotic factors controlling the inhibitory effects of pulp density on metal extraction are needed (Valix, 2017). Both single-stage, multistage, batch, and 182 183 continuous stirred tank reactors can be developed and tested. To facilitate implementation on 184 waste matrices, there is also a need to assess the process efficiency in larger, commercial 185 relevant scale reactors, whilst focusing on process development and reactor design.

186 Fine-tuned processes assisted with microorganisms do exist, leading to the production of 187 engineered inocula tailored to the target material and capable of high metal tolerance and 188 improved selectivity towards the metals wanted as secondary resource (especially Cu, Co, Mn, 189 V, Zn). The growing use of indigenous bacteria adapted to the environments instead of well-190 known strains from lab collections may overcome some of the limitations in terms of 191 processing times (Pollmann et al., 2018). However, it will be challenging to use bioleaching 192 for recovery of just one element, as further separation and purification techniques will be 193 needed for circularity. The use of mixed culture instead of pure strain shows remarkable 194 synergistic effects, especially against heavy metals that inhibit biomass growth, although pure 195 cultures might demonstrate improved selectivity for the recovery of individual or groups of elements. The use of iron and sulphur (for acidophilic bioleaching) or organic sources (fungal 196 197 and cyanogenic bioleaching) might increase bioleaching costs, as well as the need to add acid 198 to keep the medium pH low (Srichandan et al., 2019). Adaptation of microorganisms to the 199 waste materials can be critical, primarily due to heterogeneous feedstock composition, high 200 buffering, and passivation effects. Alkaline wastes like MSWI ashes are unfavourable for 201 bioleaching using acidophile cultures, so more data on alkaline bioleaching are needed other 202 than fungal bioleaching, which showed limited performances in terms of metal yield, 203 biosorption capacity, and volumes of biomass produced (Luo et al., 2019). Controlling the 204 formation of inhibitory layers may overcome cell growth disruption and decrease metal 205 extraction where a reasonable trade-off between microbial community succession and their 206 energy types metabolisms can be maintained.

Regarding microbial development, there is also a need to better understand partnering of organisms and cell adaptation to the toxic effects of not only metals, but also toxic organic contaminants in the wastes (e.g. polychlorinated dibenzo-p-dioxins) (Valix, 2017). Metal separation from the bio-leachate also demands cost-effective and selective processes to recover metal ions. A major challenge is the recovery of low concentrations of metals from large volumes of dilute leachates (Pollmann et al., 2018).

213 **3.3. Opportunities**

214 Bioleaching has potential to recover metals from low-grade ores and wastes, but also to 215 offset remediation (legacy sites) and operation costs by valorising wastes. This can also 216 contribute to minimising the environmental impacts of raw materials extraction as potentially 217 toxic elements are not discharged to the environment, but recovered for the circular economy. 218 The importance of resource recovery in reducing carbon emissions could receive increased 219 attention and should be leveraged to support the development of bioleaching. It can be expected 220 that the growing demand for hi-tech elements driven by development and uptake of renewable energy technologies, will further promote research on bioleaching for metal recovery from 221 222 wastes (Pollmann et al., 2018). Similarly, resource recovery will play a key role in securing the future availability of critical metals, and further investment in recovery technologies that have
a high potential for implementation in multi-step processes for cost-effective mineral
beneficiation, is likely.

226 Further opportunities reside in fine-tuned bioleaching to enhance selectivity to targeted 227 metals, especially critical raw materials. Extension of bioleaching methods from two steps to 228 three and four steps could mitigate the bacteriostatic effects of waste (Valix, 2017). Another 229 area of development is accelerated carbonation and carbon sequestration in alkaline wastes 230 using microorganisms (Mayes et al., 2018). Recent advances in microbial electrochemical 231 technologies for energy production metal recovery are also promising (Huang et al., 2019; Pollmann et al., 2018). Similarly, reductive bioleaching of oxidised ores and urban biomining 232 233 of electronic wastes (Sethurajan et al., 2018) can promote further research and implementation 234 of bioleaching in municipal solid waste incineration residues.

235 **3.4. Threats**

Bioleaching efficacy can be compromised by the release of potentially toxic metals from wastes, which may affect the microbes used in bioleaching. Adaptation of microorganisms is critical for higher effectiveness of this biotechnology. The current lack of work practices, especially for wastes at pilot scale, may lead to poor reproducibility of metal recoveries in different matrices. Further research is needed to satisfactory cover more substrate materials and inocula. There is also a need to assess the downstream processing in case where excessive biomass is produced in the process.

The major threats for the implementation of bioleaching are statutory and financial factors, such as the volatility of markets and metal prices, currently low mineral extraction costs ("mineralogical barrier"), and the current low cost of landfill disposal; all of which are not favourable to resource recovery. Finally, the possibility of biological hazards needs accurate

assessment at the prototype phase via standardised ecotoxicity tests that still have to be fullydeveloped and should adapt to the proposed technologies.

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250 4. Concluding remarks

251 Bioleaching allows recovery of critical resources from MSWI residues, such as fly ashes 252 and bottom ashes, and in this paper, we have identified the Strengths, Weaknesses, 253 Opportunities, and Threats associated with this biotechnology. Bioleaching is a promising 254 approach for the recovery of metals, in particular critical raw materials, from wastes. 255 However, further developments are still needed to enable sustainable and commercial 256 application to residues from municipal solid waste incineration, to enable scale-up and 257 demonstration of the commercial value. Advances in reactor design and demonstration at 258 commercially relevant scales are critical to the adoption of this biotechnology. Techno-259 economic analysis, life cycle assessment (LCA), life cycle sustainability assessment (LCSA) 260 are tools that can be used to establish and develop the application of bioleaching for resource 261 recovery from wastes.

262 The full implementation of new technologies for resource recovery, such as bioleaching, 263 needs an integrated policy and regulatory framework at all levels – local, regional, national, and international, not just scientific and technical advances. The need for regulatory, 264 265 economic, and fiscal instruments to enforce and incentivise resource recovery is pressing. 266 Some of these instruments might include providing a buffer against price volatility for 267 recovered materials and metals; supporting markets in recyclates; whilst simultaneously 268 investing in research and development and advancing technology readiness levels. Further 269 investment in infrastructure and supply chains to enable resource recovery is also required. 270 Finally, good coverage of testing for suitable microorganisms and heterogeneous substrate 271 materials is essential to fill the existing knowledge gaps and feasibility uncertainties for

alkaline waste. Increasing environmental awareness, as well as limitations of traditional

273 methods for complex materials with low metal content, will expand the development and

application of bioleaching for primary ores. The potential amount of resources, in particular,

critical raw materials such as metals, that can be recovered are relevant and can contribute to

- 276 more sustainable waste management practices, whilst simultaneously avoiding unnecessary
- 277 raw resource extraction and associated environmental impacts.

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