1 Circular economy landfills for temporary storage and treatment of mineral-

2 rich wastes

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28 Abstract

Many countries face serious strategic challenges with the future supply of both aggregates and critical 29 elements. Yet, at the same time, they must sustainably manage continued multimillion tonne annual 30 31 arisings of mineral-dominated wastes from mining and industry. In an antithesis of Circular Economy principles, these wastes continue to be landfilled despite often comprising valuable components, such 32 33 as critical metals, soil macronutrients and mineral components which sequester atmospheric CO₂. In 34 this conceptual paper, the author's aim is to present a new concept for value recovery from mineral-rich 35 wastes where materials are temporarily stored and cleaned in landfill-like repositories designed to be mined later. The time in storage is utilised for remediating contaminated materials and separating and 36 37 concentrating valuable components. It is proposed that this could achieved through engineering the repository to accelerate "lithomimetic" processes, i.e. those mimicking natural supergene processes 38 responsible for the formation of secondary ores. This paper summarises the concept and justifications 39 40 and outlines fundamental aspects of how this new concept might be applied to the design of future repositories. The proposed concept aims to end the current "linear" landfilling of mineral-rich wastes 41 in favour of reuse as aggregates and ores. 42

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Keywords: Waste management & disposal; Waste valorisation; Sustainability; Remediation; Recycled
material; Leaching; Landfills; Industrial wastes

46 **1. Introduction**

The aim of this paper is to introduce and explain a new technology concept for the management and recovery of resources from high-volume mineral-rich wastes. There are recognised strategic challenges with the future supply of aggregates, critical minerals and elements (Hayes et al., 2018; Gunn et al., 2008; World Bank, 2017; Bazilian, 2018; MPA, 2018; EU, 2020; Sovacool et al, 2020).). At the same time, individual nations must sustainably manage the multimillion tonne annual waste arisings from various economic sectors. Mineral-rich wastes are typically composed of a diverse range of mineral53 dominated materials that include waste rock and tailings materials from mining, residues and slags from metal production, combustion ashes (fossil fuel, biomass, municipal solid waste incineration (MSWI)), 54 construction and demolition wastes, and contaminated dredgings (inland and coastal). Krausmannet al 55 (2017) estimate that the global solid waste flow was 9.7 Pg/yr (\pm 14%) for 2010, and that only 37% of 56 57 non-metallic minerals in end-of-life outflows from stocks were recycled. Recent mineral-rich waste 58 arisings statistics are not readily available but illustrative EU data (https://ec.europa.eu/eurostat/) reveal some 1,537,090,000 t of mineral and solidified waste arisings in 2008 and 620,900,000 t of mining and 59 60 quarrying waste arisings in 2018. Although some of these materials do have accepted markets, these 61 are often limited to specific applications or the demand for the material may be small compared to 62 current (and projected increases in) production. Therefore, most of these materials are currently treated 63 as wastes, which continue to be disposed of in landfills or other engineered impoundments. This is 64 despite often containing valuable resources such as elevated concentrations of critical metals, soil 65 macronutrients and useful mineral components, some of which actively drawdown atmospheric CO₂ (Sapsford et al., 2017, 2019; Spooren et al., 2020; Antonkiewicz et al., 2020; Riley et al., 2020). 66

67 Although for many countries around the world, engineered sanitary landfill remains an aspiration for 68 solid waste management, other countries and supranational bodies such as the European Union are 69 looking towards ending reliance on landfill and are transitioning towards a circular economy (CE). 70 There are many definitions of CE and its precise meaning remains contested (see for example Kirchherr 71 et al, (2017); Korhonen et al (2018); Kalmykova et al (2019)), and critiqued (Corvellec et al., 2022). 72 However, common themes (particularly in the EU framing of CE; McDowall et al, 2017) include (i) 73 closing materials loops through recycling and increasing resource and materials efficiency and (ii) 74 "using cyclical materials flows, renewable energy sources and cascading-type energy flows" (Korhonen 75 et al; 2018). A key part of this (following the waste hierarchy) involves reduction in waste production, 76 reuse, and recycling. Thus, opportunities to recycle, or upcycle wastes are actively sought.

Despite enormous arisings, mineral wastes do not commonly feature prominently in public-facing CE
discourse. For example, despite England's "Our Waste, Our Resources" strategy focusing on the
development of CE and referencing the importance of tackling residual wastes and producer

80 responsibilities, many of the more tangible commitments relate to issues such as packaging, litter, flytipping and residential recycling (HM Government, 2018). Another example is that extractive and 81 82 primary manufacturing industry wastes do not feature in the "butterfly diagram" used extensively in communication of the CE principle (Ellen MacArthur Foundation, 2013). It is of note that mineral-rich 83 84 wastes are overlooked, considering the scale of production and the embodied carbon in such wastes (Gomes et al., 2016; Zhai et al., 2021). This perhaps because in the popular imagination recycling of 85 86 post-consumer goods holds sway, whereas many pre-consumer wastes that result from the production 87 processes are 'out of sight' and therefore 'out of mind'.

88 Pre-consumer wastes split into two broad groups, clean unmixed materials, such as cardboard packaging and metal swarf, which are easy to separate and have very high recycling rates; and, highly mixed 89 materials, often requiring decontamination, which are difficult to process, and therefore, are currently 90 uneconomic to recycle despite containing valuable components. Within many industries there are 91 92 efforts to view wastes as "secondary" or alternative raw materials, in keeping with the industrial symbiosis concept, where one industry's waste (or water/energy) serves as raw materials for the next 93 94 or others in a cluster (Chertow and Ehrenfeld, 2012). Despite this, mineral-rich wastes are typically still 95 landfilled and not used as secondary raw material. This can be attributed to three critical issues: (i) The 96 technical or environmental constraints related to deleterious leachable components (e.g. Luo et al, 2019); (ii) The large volume of arisings and their low economic value as secondary materials; (iii) High-97 98 volume end-uses for the secondary material need to be (but are typically not) contemporaneous with 99 their production. Expounding the latter points, bulk materials, whose unit value is low can be described 100 as having a high "place value" (Shaw et al., 2013). This means that the financial and environmental 101 costs incurred by transportation restrict their use geographically. Thus, if local end-use demand is not 102 contemporaneous with waste production, storage becomes a solution to balance supply and demand. 103 As a result of these constraints, landfill is currently the typical destination for most mineral wastes. 104 Furthermore, there may be some reticence for use of secondary material where there may be (perceived 105 or actual) variation in composition and uncertainty over geotechnical or geochemical performance 106 leading to a preference for virgin materials (See Perkins et al (2021) and references therein).

107 When considering resource recovery from waste and/or recycling it is important to consider that the deportment of value (cf. "deportment" of metals in ore) within the wastes can vary. The valuable target 108 component(s) are either those that can be separated from the bulk material (direct recovery e.g. by 109 leaching), or the residual bulk material, after removal of contaminants that prevent reuse (indirect 110 111 recovery), or a combination of both (see Sapsford et al., 2017). There exists a very large body of research on hydrometallurgical, biohydrometallurgical and pyrometallurgical processes for extraction of 112 valuable resource from mineral-rich industrial and mining wastes (see for example reviews by Jadhav 113 114 and Hocheng, 2012; Sethurajan et al, 2018; Gunarathne et al, 2022). However, there are fundamental problems with the sustainability and economic viability of many of these processes. Here we use the 115 term sustainability to mean maintenance of "genuine wealth" in manufactured, human and natural 116 capital assets (Arrow et al., 2004), without degradation of natural capital through land use change, 117 pollution and carbon emissions. Despite the multimillion tonne arisings of mineral-dominated materials, 118 119 they tend (by definition) to contain sub-economic amounts of useful elements i.e., less than the 120 corresponding ores. To remove these metals, which are present at low concentrations, has consequences 121 for increased energy demand, commensurate carbon footprint and economic cost over and above that 122 for element recovery from primary ores. The energy demand and environmental cost of element 123 recovery for low concentration ores (and by extension of the same logic, wastes) is often high. This can be illustrated by the example of platinum, where extraction from very low concentration primary ores 124 is common and the embodied energy exceeds 100 GJ kg⁻¹ and embodied carbon is 10,000 kg kg⁻¹ 125 (Gutowski et al., 2013). For very low-grade materials physically moving millions of tonnes of waste 126 through a conventional processing system for metals recovery incurs a substantial energy penalty. 127 Furthermore, even biohydrometallurgical processes, often considered as most sustainable, still involve 128 elevated temperature, mixing and reagent addition, each with their own considerable environmental 129 footprint. Thus, when considering direct and indirect value recovery from mineral-rich wastes, the 130 energy required for the separation, be it of resource from residue, or contaminant from secondary raw 131 132 material, needs to be from renewable sources to be sustainable.

2. ASPIRE: A new concept for temporary storage and treatment of waste

The authors propose a step-change in waste repository design for mineral-rich wastes, with a change in 135 focus from solely environmental and health protection to one where environmental protection and 136 137 resource recovery are designed in. The concept involves Accelerated Supergene Processes In Repository Engineering (ASPIRE) and the author's refer to waste repositories following this paradigm 138 as "ASPIRE repositories". Supergene processes are natural lithospheric weathering and pedogenetic 139 140 processes involving downward percolation of meteoric water (i.e., derived from precipitation) and metal 141 leaching from unsaturated metalliferous rock (Lelong et al., 1976) often enhanced by organic derived 142 acids and ligands. Metals are then deposited as an enriched metalliferous secondary ore below the 143 groundwater level during the transition to more chemically reducing conditions (Figure 1(a)).

144 The ASPIRE concept is a "nature-based solution" (Song et al., 2019), which involves mimicking these naturally occurring lithospheric mechanisms, such "lithomimicry" aims to achieve the same effect but 145 146 with waste materials (Figure 1(b)). Interestingly, supergene alteration has been noted to occur in wastes such as mine tailings and smelter residues (Dill, 2015). Furthermore, the authors propose expanding the 147 supergene concept to include other value propositions including phosphate recovery, carbon 148 149 sequestration and the decontamination of the waste matrix. Because natural supergene processes can 150 take millennia, there is a requirement to engineer the processes to accelerate them to anthropogenically 151 relevant timescales. This will require innovative biogeochemical engineering such that processes 152 remove potential contaminants/resources from the bulk material matrix (leaving a cleaned residue) and concentrate them within an anthropogenic ore zone over a prolonged period of waste storage. 153

ASPIRE repositories would be designed to significantly accelerate the ore-formation from the geological timescales of natural supergene process to the order of years in the engineered system. Critically, being a fully lined waste repository system, the external environment remains protected. Inspired by research observations of revegetated industrial wastes (Bray et al., 2018), the ASPIRE concept could also intersect with phytoremediation, phytocapping and phytomining concepts. Here, environmental leaching agents ("lixiviants" in hydrometallurgical parlance) generated from plant roots 160 (which produce low molecular weight compounds that act as ligands for metals) accelerate mobilisation of metals. Solar and self-powered electrokinetic and electrochemical phenomena are proposed for 161 acceleration of solubilisation and transport, followed by biogeochemical trapping of metals/elements, 162 potentially as new "ores". Whilst it is impossible to "short-change" fundamental thermodynamics which 163 164 dictate the minimum energy requirement of separation of species from a parent material, it should be possible to provide that energy in a sustainable way from the solar energy incident on the storage site 165 (insolation flux), directly via photovoltaic power systems or indirectly via photosynthesis (and the 166 167 environmental exergy cascade), albeit sacrificing the process intensity by prolonging the timeframe for the process. 168

The central tenet of the ASPIRE concept is that the "time in storage" is used for material 169 cleaning/resource concentration. This has the dual benefits of facilitating future resource recovery and 170 defines the service-life for the barrier system. The dormant waste undergoes processes to (i) concentrate 171 172 valuable components (e.g. critical metals, phosphate) as an anthropogenic ore to facilitate their future recovery, and (ii) concurrently decontaminate residual mineral material so as to make it available as a 173 bank of material to drawdown for "soft" end-uses in agriculture, forestry, greenspace, landscaping, 174 habitat creation (Song et al., 2019) and/or as a cement/concrete additive or replacement aggregate. As 175 176 such, the ASPIRE concept seeks to reconceptualise waste repositories as "temporary storage systems" or "resource banks". This could involve regional ASPIRE repositories as hubs which import a range of 177 178 mineral-rich wastes arising in the region, in the UK context potentially contributing to the Managed 179 Aggregate Supply System. Alternatively, smaller site-specific repositories could be developed. 180 Importantly, the idea is not necessarily to displace any existing sustainable recycling activities for 181 mineral-rich wastes but to provide a practicable CE solution for materials that would otherwise go to 182 conventional landfill for lack of any other viable means to recover value. We suggest that this concept thus fits into a modified waste hierarchy as shown in Fig 2. 183

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3. The Case for Temporary Storage and *in situ* treatment

187 **3.1 Existing concepts of temporary storage and 'End of Waste'**

Temporary storage for MSW landfill has been proposed such that stored waste undergoes processes to 188 189 accelerate the "stabilisation" of the waste, to facilitate recovery of materials from the landfill mass or 190 the use of the land (Jones et al., 2013). The accelerated stabilisation is achieved either via active aeration 191 to promote the breakdown of organics (Ritzkowski and Stegmann, 2012), or the recirculation of landfill 192 leachate to accelerate anaerobic process and methane production (Reinhart and Al-Yousfi, 1996; Warith, 2002), ultimately allowing a compressed timeline for land restoration or the recovery of 193 194 materials from the landfill mass. Temporary or 'interim' storage of municipal solid waste (MSW) has been undertaken in Germany, in response to lack of a receiving market. Several million tonnes of 195 196 Mechanical Biological Treatment (MBT) sorted wasted was stored in interim landfills (Wagner and Bilitewski, 2009). Temporary storage has also been mooted for e-wastes (Kahhat and Kavazanjian, 197 198 2010). Despite these examples, the practice of temporary storage has not taken hold more widely. Functioning markets for recyclate are important so that there is contemporaneous demand for materials. 199 200 Yet even these relatively high-value recyclates are susceptible to market disruption. In 2017, China 201 banned the import of most plastic waste, this resulted in a sharp reduction in global plastic waste trade 202 flow (Wen et al., 2021). The fact that market volatility is commonplace in markets for relatively high-203 value recycled resources emphasises the challenges of recycling low-value, high-volume and 204 contaminated mineral-wastes and why they are currently landfilled.

There has been some success in England and Wales in utilising secondary materials, this is also encouraged by a levy on virgin aggregates. "End of waste" (EoW) criteria have been developed for some mineral-rich wastes to facilitate reuse: notably, quality protocols developed in England and Wales by the Environment Agency and the Waste and Resources Action Programme (WRAP) for steel slag and pulverised fuel (coal) ash, furnace bottom ash, as well as a Code of Practice developed by MIBAAA (Manufacturers of IBA Aggregates Association in conjunction with the Environment Agency) for incinerator (MSWI) bottom ash. These approaches are framed around Article 6 (1) of the European 212 Waste Framework Directive (2008/98/EC) which sets out end of waste status, and the conditions that if met enable waste which has been recycled or recovered to cease to be classed as waste. The substance 213 or object must meet the following conditions: (i) It is to be used for specific purposes (ii) Market/ 214 demand exists (iii) Achieves the technical requirements for the specific purposes and meets existing 215 216 standards and legislation applicable to products (iv) Its use will not lead to adverse environmental or human health impacts. Thus, the materials referred to above need to demonstrate compliance with the 217 218 quality protocol framework, as well as an appropriate standard such as BS EN 12620, Aggregates for 219 concrete, and associated testing. In addition, markets may well be restricted, for example the Quality 220 Protocol for PFA allows its use in bound or grout applications only.

Despite these examples, many mineral-rich wastes from mining and industry are still largely overlooked 221 and thus poorly integrated into current CE strategies and developing policy (Cisternas et al., 2022). 222 There is a lack of practicable, economically viable and sustainable technologies for returning these 223 224 resources to the CE and the temporal and geographical dislocation between production and end-use (Cisternas et al., 2022). Providing a solution to the temporal dislocation is a key advantage of the 225 226 ASPIRE concept, that once the material is cleaned, it stays in hibernation until end-user market arises. 227 There are historical examples that demonstrate over a decadal scale, materials often thought as valueless 228 waste later become highly valued resources, for example due to advances in recovery technologies, 229 decreases in ore-grade or increase in value of specific components which were previously non-230 economic. For example, the re-mining of Pb/Zn spoil in 1970s / 80s in the northern Pennines (UK) for 231 fluorspar and Ba minerals, driven by demand for F in chemical industries and Ba for oil drilling fluids (Dunham, 1985). 232

It is noteworthy that temporary storage and treatment as a concept is conceptually similar to the "cluster" approach developed in the UK to facilitate the remediation of contaminated soils where a number of sites are in close proximity. The sites share a "hub" for the central decontamination / treatment of contaminated soils (CL:AIRE, 2021). This is similar to the concept proposed here, albeit these remediation hub timescales are shorter, the relative intensification of remediation is economically viable because of the value of land redevelopment.

239 **3.2** Future generations and conventional landfill

240 Landfill remains the destination for wastes that cannot be combusted, composted or separated into recyclates for which there is a current market value. Landfill philosophy has evolved from "dilute and 241 242 disperse" to a "store and contain" containment strategy, with the waste environmentally isolated by use of engineered top and bottom liners (e.g. Cisternas et al., 2022). Allen (2001) critiques containment 243 strategies, raising concerns over the durability of liner systems and problems with clay liners. 244 Landfilling simply postpones the release of contaminants, as for future generations it is not a question 245 of "if" but simply "when" the containment systems for landfilled industrial wastes succumb to natural 246 247 degradation. This then leads to important questions about intergenerational equity for management of wastes. Put simply, should we be burdening future generations with pollution issues from our current 248 waste production? This appears contrary to the sustainability agenda and UN sustainable development 249 goals (United Nations, 2015). 250

Whilst accepting that many of the drivers for diversion of mineral-rich wastes to landfill remain, the authors contend that a sustainable and ethical philosophy would be "store, contain, clean and concentrate" to allow future recovery. It is notable that future recovery of residues from the repositories would likely be in the context of a decarbonised economy, thus future exploitation will likely be of low carbon intensity.

256 It is interesting to consider temporary landfilling of waste in the context of a CE. A critical component of a move to a CE is increased resource efficiency via the provision of a service-based economy. The 257 258 current landfill paradigm is a linear model, often with associated ownership/license surrender by the 259 operator. Where used, facility gate fees cover operational costs but neither the original waste producer 260 nor the landfill operator is paying (post-permit surrender) for the ultimate environmental impact of the 261 waste (i.e. these long-term impacts remain market externalities). Applying the ASPIRE concept changes the landfill operator's function to the provision of a storage and remediation service, and the production 262 of secondary raw materials which will ultimately be removed from site, thus removing the long-term 263 environmental hazard. 264

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266 3.3 Alignment with current waste legislation

To achieve a CE, it is essential to re-use and recycle materials from waste for future use in new 267 buildings, infrastructure, products etc, and to keep these in productive use as long as is feasible. Thus, 268 the treatment of waste materials to facilitate this is vital to deliver a CE. The Waste (England and Wales) 269 270 Regulations 2011 introduced a duty for waste importers, producers, waste carriers and waste 271 management facilities to "take all measures available to it as are reasonable in the circumstances" to 272 apply the waste hierarchy when transferring waste: (i) Prevention (ii) Preparation for re-use (iii) 273 Recycling (iv) Recovery (v) Disposal. The crux of this is the term "reasonable", which ultimately means 274 that aspects such as technical, economic and environmental aspects are considered in order to make a 275 judgement. Despite this legislation, landfill remains a commonly used option for mineral-rich wastes.

Whilst temporary storage and treatment may not be feasible for all landfilled wastes, we suggest that it 276 would be a practicable and sustainable solution for the many mineral-rich materials amenable to 277 278 processing via leaching. Because the ASPIRE concept involves waste materials being reprocessed into products (clean residue and anthropogenic ore) it is in essence a recycling technology. However, the 279 confounding issues of the long-term storage and processing whilst stored in a "landfill-like" repository 280 will likely lead to challenge of its status as recycling technology in the eyes of the waste regulations. 281 282 At minimum, an ASPIRE repository could be viewed as providing a technology option at the base of 283 the waste hierarchy, replacing conventional landfill – and this may be a more regulatory acceptable approach for early adoption of the technology. The Environment Act 2021 provides some potential 284 opportunities for the ASPIRE concept to gain acceptance in the UK. First, it confers power to the 285 286 Government to set regulations on producer responsibilities, including waste prevention and reduction. 287 It also enables regulations related to the re-use and recovery of materials.

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289 4. Candidate Waste Arisings and Legacy Wastes

291 Alkaline combustion ashes, bottom ash and fly ash/Air Pollution Control Residue (APCr) from Biomass, MSWI, sewage sludge, co-firing of biomass/coal will be important wastes in the UK and 292 293 internationally for the conceivable future. By 2050, global arisings of biomass ash will be of the order 294 of 480 Mtpa (Vassilev et al., 2013). Furthermore, the UK's Sixth Carbon Budget (CCC, 2020) highlights the important role of Bioenergy with Carbon Capture and Storage (BECCS) as one of the 295 key technology options for the UK in limiting the contribution to catastrophic global warming. BECCS 296 297 has been estimated to have a CO₂ removal potential of 20-70 MtCO₂ pa (Smith et al., 2016) and further 298 removal through carbonation of the ash arisings e.g., through enhanced terrestrial weathering. Biomass ashes comprise major elements O, Cl, Si, Ca, K, P, Al, Mg, Fe, S, Na, Ti, Mn and are notably enriched 299 compared to coal ash in trace elements in Ag, Au, B, Be, Cd, Cr, Cu, Mn, Ni, Rb, Se, Zn (Vassilev et 300 301 al., 2013; Zhai et al., 2020). Heavy metals and readily soluble metal chlorides are the most recurrent 302 contaminants preventing ash reuse in many of its principal potential applications e.g. soil amendment 303 in silvi/agriculture and admixture in cements and mortars. Statistical analysis of numerous database 304 records shows that compositionally there are four main types of virgin biomass ash: hard wood ash, 305 softwood ash, and grass (straw) and non-grass type agricultural residues. Interestingly, wood ashes 306 have slightly higher trace metals, whereas agricultural residue ashes have notably higher levels of persistent organic pollutants (POPs) (Zhai et al., 2021). Thus, the ASPIRE concept could offer a 307 308 practicable management/decontamination option for ash arisings from projected biomass combustion 309 and future BECCS with potential for further CO₂ mitigation through ash weathering, ash reuse as a 310 cement admixture and nutrient recycling to agri/silviculture whilst also recovering critical elements and 311 minerals.

There is also a clear trajectory for growth in MSWI ash arisings. Bottom ash is readily recyclable, but market limitations mean that its reuse is restricted to prescribed applications, whilst MSWI fly ash/APCr is considered hazardous which prevents recycling. Due to POP content, some 5% of MSWI APCr requires further treatment before it can be disposed of to hazardous waste landfill. UK Energy from Waste capacity was projected to be 15.4 Mtpa by 2020 (Tolvik, 2017), with estimated bottom ash and APCr arisings of 3.1 Mtpa and 0.5 Mtpa respectively. MSWI ash can be enriched in Ca, Cl, K, Mg, N,

Na, P, Ti, trace elements Ag, Cd, Cr, Cu, Ho, I, Mn, Pb, Pr, Re, Sb, Sm, Sn, Zn (Lima et al., 2008; Tang

et al., 2015; Tang and Steenari., 2016; Zhai et al., 2021), the trace element compositions will besusceptible to changes in waste composition.

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322 *4.2 Dredgings*

323 Annually, approximately 40 million wet tonnes of sediments are disposed of in approximately 150 324 licensed disposal sites around the coast of England (Bolam et al., 2006). Most result from dredging of 325 harbours and shipping lanes on (post-)industrial rivers. The EU annually produces around 200 - 250 Mt of marine and 50-60 Mt of freshwater dredgings per year, of which up to 5-10 % and <30%, for marine 326 and fresh water respectively, are sent to "confined disposal" (Mink et al., 2006). Data on total arisings 327 of inland UK contaminated dredgings are sparse but the amounts are likely in the range of hundreds of 328 thousands of tonnes per annum. As with ash, the heavy metal content of dredgings often prevents their 329 330 reuse in applications such as bank protection, habitat creation and conditioning agricultural land (Renalla, 2021). ASPIRE repositories could offer the practicable means to create habitats and green 331 corridors close to damaged waterways while also providing a means to return metals that have escaped 332 into rivers and bound up with sediment to the CE and bioremediating any associated organic 333 334 contaminants.

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336 *4.3 Alkaline Industrial Wastes*

Industrial processes produce a very wide variety of different wastes, many of which are readily recyclable. However, several classes of mineral-dominated residues are produced during metal production that lead to high-volume, low-value materials that are uneconomic to recycle and commonly disposed of in land-based repositories. These include, steelmaking and metallurgical slags, bauxite processing residues, Solvay process wastes from the manufacture of soda ash, and chromite ore processing residues. In terms of global production, steelmaking slags and bauxite residues are particularly important and have rapidly increasing rates of production (170-250 Mtpa and 120 Mtpa
respectively (Power et al., 2011)). These residues are often the product of high temperature processes
that involve additions of alkali materials during extraction. Therefore, the resultant by-products are rich
in alkaline mineral components (e.g. CaO, NaOH, Ca and Na – silicates) that readily hydrate and
dissolve to produce a highly alkaline leachates (Gomes et al, 2016).

After uses for the bulk residues can be limited by a range of factors which include (i) extreme alkalinity and mobility of potentially hazardous metal(loid)s at high pH (e.g. As, Cr, V: Burke et al., 2012; Hobson et al., 2017), (ii) stability issues where weathering of residues can lead to expansion and limit aggregate after uses, (iii) the potential classification of some residues such as bauxite processing residue as Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) which can limit after uses in construction (e.g. Somlai et al., 2008). As such, virgin materials are often preferred over bulk reuse of alkaline residues given the additional costs associated with mitigating potential issues.

There is growing research in value recovery from alkaline industrial residues ranging from bulk material reuse to critical metal recovery and carbon capture (e.g. Gomes et al., 2016; Ujaczki et al., 2017; Pullin et al, 2019; Pan et al, 2020). However, relatively low fluxes of dilute valuable components (e.g. V) occurring in leachates with mixed contaminants makes separation and recovery challenging (e.g. Gomes et al., 2017). As such, there are few examples of large-scale critical raw material recovery from alkaline industrial wastes.

361 The ASPIRE concept brings a range of opportunities to overcome some of these current technical 362 constraints. The extended timescales on which ASPIRE is based provides scope for the gradual leaching and accumulation of critical metals in enrichment zones that would be better candidates for 363 recovery than current technologies (e.g. Gomes et al., 2020). The decontamination of bulk material by 364 365 these leaching processes could produce more stable and less hazardous materials that could open up more avenues for large scale reuse as aggregate (e.g. slags), in land reclamation or in ceramics / building 366 materials (e.g. bauxite residue). Alkaline wastes also have the potential to sequester atmospheric CO₂ -367 See Section 8 below. 368

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370 *4.4 Mine wastes*

Mine wastes include overburden, marginal and sub-ores, tailings and mine water treatment residues. 371 Globally the arisings of these wastes likely run to hundreds of billions of tonnes (Kinnunen and 372 Kaksonen, 2019), for example there are an estimated 39 billion tonnes of mine tailings in Chile alone 373 374 at 0.2 wt.% Cu (Dold, 2020). Globally, mine wastes are deposited in a range of settings from uncontrolled storage in the landscape to highly engineered impoundments, depending on their age and 375 376 the environmental legislation relevant in the geographic jurisdiction. Mine wastes deposited to land in 377 an uncontrolled manner cause a substantial pollution legacy through acid rock drainage and metal-378 leaching (e.g. Wolkersdorfer and Bowell, 2004), and migration of fines (e.g. Fuge et al., 1989). 379 Impoundments can pose risks through dust and failure of dams (Dold, 2020). In the UK, an estimated 73 - 195 Mt of mine wastes were produced from the historical working of lead, copper and tin, and 380 cover an estimated 1960 - 4400 hectares of land (Palumbo-Roe and Colman., 2010). The indicative 381 382 estimated cost, over a 10-year life cycle, to remediate solely the water-related environmental problems due to diffuse pollution caused by non-coal mine wastes in England and Wales was estimated at £35 383 Million (Jarvis et al., 2012). 384

385 There is considerable interest in recovery of resources from mine wastes, and they are one of the more 386 obvious targets for sourcing of metals, particularly as ore grade continue to decrease and/or aggregate 387 resources decrease. Despite this, there has been very limited explicit adoption of the concept of the 388 circular economy amongst large-scale mining firms (Upadhyay et al., 2021). Large bottlenecks for 389 tailings valorisation identified by Kinnunen and Kaksonen (2019) include the low concentration and 390 mass of recoverable elements for economical processing, missing value chains, regulatory barriers, and 391 environmental risks of reprocessing. Applying the ASPIRE repository concept for future mine wastes 392 and tailings could address many of these concerns and at minimum a better option than the "do nothing" 393 option of continued landfilling.

395 4.5 Temporary Storage as a Remediation Strategy for Legacy Wastes

Where "polluter pays" legislation (see Stenis and Hogland, 2002) is in place then there is a compelling 396 case (from the perspective of environmental justice) for legacy mineral-rich wastes to be managed by 397 398 the generator. However, many legacy mineral-rich wastes remain where the waste generator has no liability (due to lack of action-forcing legislation at the time), or no longer exist. In the UK, such legacy 399 400 wastes and orphan sites become the responsibility of local authorities and/or central government. The ASPIRE concept could offer a potentially lower cost alternative to conventional site remediation, which 401 402 often leads to secondary landfilling of heavily contaminated materials in any case. In a sense, this would 403 be a revisitation of the "dig and dump" approach to site remediation that was common in the UK but has become less common due to landfill tax (Hodson, 2010), with the critical difference that the "dump" 404 is into a temporary storage repository where the waste is remediated to allow future recovery. 405

Multi-dimensional value refers to the value as expressed by the range of measurable benefits and 406 407 impacts in the environmental, economic, social and technical domains (Iacovidouet al, 2017). Many 408 legacy sites in the UK, for example mine sites, display multi-dimensional value. For example, important 409 landscape and cultural designations, sites of special scientific interest and/or host rare and valued species of flora and fauna as well as causing environmental pollution (Crane et al, 2017; Sinnett, 2019). 410 It is important to note that an ASPIRE repository could feasibly be constructed at a legacy site, with 411 412 some, or all, of the valuable aspects of the legacy site (which tend to be associated with the upper surface) retained or recreated in the repository cover. 413

414

415 5. Accelerating supergene processes: Lithomimicry and Biogeochemical 416 Engineering

417

418 5.1 Storage time

The requisite duration of the storage of wastes within an ASPIRE repository is an open question but one that dictates the necessary intensity/acceleration of the *in situ* supergene processes. Conceptually, it is possible to envisage (i) shorter-term storge duration and processing of a few years (the system resembling a contaminated land remediation project) (ii) medium term storage and treatment over one to a few decades, through to (iii) long-term intergenerational storage and treatment on a timescale of several decades to a century. It is envisaged that the *in situ* processes could be engineered to correspond to the likely time in storage.

426

427 **5.2** Upper surface and leaching zone

428 The primary hazards to humans and other vertebrates from industrial residues tend to result either from 429 direct contact with the residue, ingestion or inhalation of dusts, or ingestion of rainwater run-off (Nathanail and Earl, 2001; Stange and Langdon, 2016). All these hazards can be significantly reduced 430 if vegetative cover is established on the residue surface, as plant growth will buffer the chemistry of the 431 432 surface layer and minimise dusts and suspended solids in runoff. When revegetation occurs naturally at a site that is initially devoid of life (e.g. after deposition of volcanic ash) the process of initial 433 colonisation by pioneer species, and their subsequent replacement by a wider range of less hardy 434 435 species, is called primary succession (Breeze, 1973; Gemmel, 1972; Bradshaw, 2000; Wong, 2003; Phillips and Courtney, 2022). Without intervention this process, which involves the slow build-up of 436 plant accessible nutrients with successive cycles of plant and microbial growth and decay, typically 437 takes decades and centuries to produce verdant vegetative cover (Ash et al., 1994; Lee and Greenwood, 438 1976; Bradshaw, 2000; Santini and Fey, 2013). However, many industrial residue disposal sites could 439 be transitioned from providing little benefit for people or nature to delivering multiple benefits, or 440 441 ecosystem services, in few years if this natural process is accelerated.

Revegetation and the potential for accelerated ecological succession has been demonstrated on acidic
coal mine (Wali, 1999; Fernandez-Caliani et al., 2021) and metalliferous mine wastes (O'Neill et al.,

1998; Bagatto and Shorthouse, 1999; Wong, 2003; Walker et al, 2004), circumneutral metalliferous 445 mine wastes (Shu et al, 2002; Yang et al, 1997; Sarathchandra et al, 2022; Peñalver-Alcalá et al, 2021), 446 and on alkaline wastes (Breeze, 1973; Courtney et al., 2009; Fellet et al., 2011; Lee and Greenwood, 447 1976; Ash et al., 1994). Interventions at industrial residue disposal sites that can accelerate the 448 449 succession process include conditioning treatments (to provide nutrients, create soil-like structure and buffer extreme pH values) and seeding of plant mixtures (Ash et al., 1994; Courtney et al., 2009; 450 Kumpeine et al., 2008). Grass species appear to be good pioneer species, as species from calcareous 451 452 grassland appear readily established on alkaline wastes and species from acidic heathland on acidic wastes (Ash et al., 1994), but early establishment of nitrogen fixing species (such as legumes) is 453 454 important for accelerated succession (Phillips and Courtney, 2022). Over the last 20 years Courtney and 455 co-workers have investigated the revegetation of bauxite processing residue (red mud); a very alkaline, abiotic waste (Courtney and Timpson., 2004, 2005; Courtney et al., 2009; Courtney and Harrington., 456 457 2012; Courtney et al., 2014). Their approach has been to augment the surface layer of the residue with various combinations of sand-sized material to improve drainage, gypsum to buffer high pH, and 458 459 organic matter to provide a suitable substrate for plant growth, and they have shown that vegetative 460 cover can be established from seed and sustained for more than 20 years. A subsequent investigation 461 has shown that plant roots become established mainly in the amended surface layer, but vegetative cover resulted in a pH reduction >2 pH units and a five-fold reduction in sodicity (compared with an untreated 462 and therefore largely unvegetated control plot) that extended to more than 3x the initial treatment depth 463 (Bray et al., 2017). Such beneficial effects from bio-rehabilitation have also been reported elsewhere 464 (Santini et al., 2015; Zhu et al., 2016; Sarathchandra et al., 2022). 465

Plants can alter the chemistry of any residue upon which they are grown through the chemicals they secrete through their roots. These include CO_2 from respiration in roots; ions transported across the soil-root interface during nutrient uptake, extrusion of H⁺ by plant cells during energy metabolism, and secretion of various organic species (termed "root exudates") (Hinsinger et al., 2003). Plants can secrete a broad array of organic compounds into the rhizosphere, and while the flux is quantitatively quite modest (Guo et al., 2010; Hinsinger et al., 2013), it represents 5% to 21% of the carbon 472 photosynthetically fixed by the plants (Helal and Sauerbeck, 1989; Marschner, 1995). Some root exudates are produced in response to specific plant stresses, but the majority (including sugars, amino 473 acids, and organic acids) are believed to be passively lost from the root (Canarini et al., 2019). Organic 474 acids and phenolics released by plants (including those released to combat excessive aluminium in 475 476 acidic soils (Li et al., 2009; Mora-Marcias et al., 2017) and siderophores to that allow Fe uptake in 477 neutral to alkaline soils (Schenkeveld et al., 2014; Grillet and Schmidt, 2017), can mobilise toxic metal 478 contaminants by chelation of complexation. In healthy soil systems most plant exudates are consumed 479 by rhizosphere-dwelling microbes (resulting in production of CO_2), but in industrial residues, with a 480 thin, poorly established rhizosphere, plant exudates can migrate deeper into the deposit (Bray et al. 481 2018), see Fig 3.

482

483 **5.2** Capture Zone

There are already many existing and developing technologies for the trapping of metals and other 484 485 contaminant species from water into a solid matrix that have variously been developed for treatment of contaminated water (e.g. groundwater, municipal solid waste landfill leachate, mine water, highways 486 and urban runoff) and there are many parallels with the continuing development of such "passive" 487 488 technologies. These include permeable reactive barriers (PRBs), constructed wetlands, sustainable 489 drainage systems (SuDS), swales and bioswales (for examples see Scherer et al., 2000; Pat-Espadas et al., 2018; Woods-Ballard et al., 2007 and Ekka et al., 2021 respectively). The mechanisms of trapping 490 491 variously involve biogeochemical process within the matrix that induce metal sequestration from 492 solution. These typically rely on changes in redox or pH, utilise precipitation by the common ion effect 493 or in situ sulphide production from microbial sulphate reduction, sorption, chelation or coprecipitation 494 (Sapsford et al., 2020). Less well understood are passive systems for removing metal and metalloid oxyanions prevalent in alkaline wastes e.g. V, Al and As in bauxite processing residue (Burke, 2013). 495 For Cr(VI) reductive precipitation is important, for V(V) pH reduction by carbonation and sorption to 496 Fe-(oxy)hydroxides is key, whereas for As oxidation of As(III) to As(V) and then sorption to Fe-497 (oxy)hydroxides is important (Ding et al., 2015; Hobson et al., 2018). In these cases, pH reduction and 498

499 carbonation are key mechanisms for metal capture. Additives such as gypsum and organic matter can 500 reduce the pH value (the former by promoting carbonation, the latter by dissolution of soluble humics) 501 and organic matter can capture metals directly by complexation or indirectly by supporting bioreduction 502 by microorganisms. Such mechanism should be effective in a trapping zone as they have been 503 demonstrated to reverse the mobility of elements including V in bauxite residues (Bray, 2018).

An underlying aim of the design and operation should be a significant concentration of the target metal (or other species) compared to the leached residue. In achieving this, key challenges for capture zone engineering include (i) maintenance of the long-term effectiveness of the biogeochemical removal mechanisms (ii) maintenance and/or control of hydraulic conductivity (See below) (iii) concentration achievable in the capture zone.

509

510 6. Hydraulic and Geotechnical Considerations

511 Many landfills (or in-ground impoundments) of wastes rely to some extent on isolation, preventing or minimising the mobilisation of contaminants and their leakage into the surrounding environment. With 512 the ASPIRE concept of *in situ* processing during temporary storage, however, complete isolation is no 513 longer desirable as external environmental processes are required to be brought to bear on the waste 514 mass to enable no/low input processing. The most important of these is likely to be controlled hydraulic 515 flow into the waste repository. As a result, the repository design moves from a containment facility to 516 something more akin to a funnel-and-gate permeable reactive barrier, where water/lixiviant flow is 517 encouraged (or at least controlled) to pass through the waste mass prior to treatment of the resulting 518 519 liquor. This conceptual change leads to implications for both the hydraulic and geotechnical design of 520 in-ground temporary waste storage.

521

522 6.1 Hydraulic processes in temporary storage

Hydraulic flow is likely to be the major driver of waste processing under natural processes, permitting the mobilisation and transport of both lixiviants and resource. Flow must enter the waste and travel through the entirety of the mass to reach the region where mobilised species are deposited. It is therefore vital to encourage water to flow through the waste mass – optimisation of this flow is dependent on-site conditions, but general issues may be considered here. There are two major environmental sources of water ingress, from rainfall / run-on (more transient) or groundwater (more consistent and predictable), which could be extracted adjacent to, and introduced into the repository.

530 A second major issue is the volume and rate of flow – the optimal volume/rate will be determined by 531 properties of the waste mass including hydraulic conductivity, as well as the rate of resource deposition and thus the minimum residence time in the capture zone. Mineral-rich wastes of interest in temporary 532 storage schemes have a wide range of potential hydraulic conductivities, for example municipal solid 533 waste incineration bottom ash is relatively coarse, with sand-like hydraulic conductivity ($5x10^{-6}$ m/s (de 534 Windt et al., 2011)) whilst finer fly ashes may vary from 10^{-7} to 10^{-10} m/s (Zabielska-Adamska, 2020). 535 Depending on the storage duration, rainwater alone may not provide sufficient water to fully mobilise 536 537 resource even on the long timescales considered here and where it does, the transient nature of rainfall 538 will potentially lead to periods of desaturation which in turn causes preferential flow and thus reduced 539 resource mobilisation. To overcome this, water retention in ponding schemes may be used to attenuate 540 flow transience, buffering water ingress by collection prior to gradual, more continuous release into the 541 waste. Alternatively, fluid exiting the storage facility may be collected and recirculated *via* autonomous 542 systems, for example powered by solar energy.

Flow in porous media, including mineral wastes, is impacted by preferential flow – certain paths through the pore space offer less resistance to flow and so the majority of advective flow takes these paths (Clothier et al., 2008) leaving the majority of the medium (the matrix) untouched. Causes may include heterogeneities at a range of scales such as strata with varying hydraulic conductivity, pore sizes of differing diameter or unsaturated pores where flow cannot occur. Such preferential flow leads to only a proportion of the waste being treated by flushing / leaching, while decontamination of the remaining waste is governed by the rate of contaminant diffusion to preferential flow paths from the matrix. Soil 550 flushing (or "pump and treat") of contaminated land is affected by this process and sometimes results 551 in active pumping continuing for decades to achieve the remediation objectives (Guo et al., 2019). 552 Without some kind of engineered system preferential flow will occur and will limit recovery from the waste mass, so it is appropriate to question whether there exist autonomous natural processes that can 553 554 be engineered to alter fluid flow and/or diffusion. For example, this can, to an extent, be managed by pumping techniques such as pulsed flushing where flow is intermittent to give time for diffusion within 555 the matrix (Cote et al., 2000); this doesn't significantly affect the overall remedial time but it does 556 557 reduce the active flushing time. Similar processes may be helpful in the case of temporary storage where treatment time is not a significant issue, but diffusion alone is unlikely to be wholly effective in moving 558 559 resource, particularly when that resource may be bound tightly in mineral wastes and thus unavailable 560 to the pore fluid. More active technologies to enhance availability and diffusion, such as electrokinetics, 561 have been successfully employed to alter the mobility and availability of resources bound in mineral wastes (Peppicelli et al., 2018), though the challenge to engineer natural processes to autonomously 562 enhance resource availability remains. 563

564 Over time, the hydraulic behaviour of all aspects of a repository may alter, and the repository system needs to be able to adapt to this. The whole waste body will undergo self-weight compaction to a degree 565 566 and pattern determined partly by compaction during placement and the rate at which self-weight develops. With repositories that develop over time with the deposition of new waste, the process of 567 568 compaction and thus alteration of hydraulic flow in the original deposits will be a continuous process. 569 The surface ecosystem will be dynamic with the growth of established plants and potentially 570 successional growth of new plant species, potentially with new root architectures and behaviour which 571 could alter water infiltration, evapotranspiration and so on. Continued leaching may lead to erosional processes, particle breakdown and aggregation, and clogging in the leach zone, whilst calcium-rich 572 573 wastes such as certain fly ashes may be susceptible to cementation with calcium carbonate (Zabielska-574 Adamska, 2020), blocking the pore space and changing flow patterns and causing or preventing any continued waste compaction. The capture zone is by definition an active biogeochemical zone with 575

deposition changing the pore structure and likely reducing the hydraulic conductivity, with challengesthen for hydraulic flow throughout the system.

578

579 **6.2** Geotechnical factors relevant to temporary storage

The geotechnical stability of in- or on-ground waste repositories will be dependent on their site-specific 580 581 design, their profile and the surrounding ground conditions. The desire to have fully saturated wastes (to maximise resource recovery and avoid preferential flow) is problematic for sloped surfaces. It is 582 likely that repository design will require non-sloping or modest-sloping surfaces as steeper slopes would 583 be susceptible to slip failure in the presence of large pore pressures, as well as seepage problems in a 584 585 similar manner to earth dams (Meyer et al., 1994), leading to failure mechanisms such as piping, heave and internal erosion (as noted above). This may be exacerbated with enhanced water flow and pore 586 pressures should measures such as surface ponding and/or recirculation be employed. Settlement and 587 consolidation of the emplaced waste masses will also be impacted by variations in the development of 588 589 pore pressures, alongside factors such as internal erosion and changes on the structure of the waste 590 materials. These processes will require consideration to ensure flow systems and any installed rigid infrastructure remain functional and to avoid adverse features such as differential surface settlement 591 592 occurring. Geotechnical liners employed to encapsulate the waste body will require redesign from 593 typical systems employed in traditional landfill. The inclusion of drainage layers should be avoided to prevent preferential flow of influent water (rain or ground) around rather than through the waste. The 594 saturation of the waste without drainage, however, has the potential to create significant hydraulic 595 596 gradients across the liner which may lead to localised liner failure.

597

598 7. Delivery of ecosystem services

In the proposed ASPIRE repository concept the upper surface can be vegetated to provide root exudatesthat enhance leaching and drive capture zone biogeochemistry. In addition, the opportunity for

601 vegetated upper surface provides opportunities for considerable added value through the delivery of 602 ecosystems services. Restored landfills deliver a range of ecosystem services, or benefits to people, following conversion to 'soft' land uses, including agriculture, forestry, amenity and nature 603 conservation (Li et al., 2019; Zalesny et al., 2020). There is an opportunity to tailor the design and 604 605 species selection to provide the necessary lixiviants as well as delivering ecosystem services. Using the framework provided by Common International Classification of Ecosystem Services (CICES) V5.1 606 (Haines-Young and Potschin, 2018) we explore the opportunity to provide ecosystem services in 607 608 ASPIRE landfills. The CICES focuses on regulation and maintenance (e.g. mediation of wastes, flood 609 risk management, pollination), cultural (e.g. experiential and physical use, education) and provisioning 610 services (e.g. cultivated plants, energy generation, mineral resources), which are underpinned by 611 'supporting' conditions (e.g. primary production; Haines-Young and Potschin, 2018).

612

613 7.1 Regulation and maintenance services

614 Regulation and maintenance services include management of flood risk, temperature, and air, water and soil quality, as well as pollination. It is well known that restored landfills can deliver positive outcomes 615 for nature conservation (MacGregor et al., 2022; Tarrant et al., 2012; Rahman et al., 2013). The sector 616 617 is already accustomed to designing restoration strategies for recreation (see below) and nature 618 conservation end uses, providing a variety of habitat types and amenities. For example, providing a nutrient-poor soil and allowing natural colonisation of plants to facilitate the development of semi-619 620 natural grasslands to provide for pollinators (Rahman et al., 2013). With ASPIRE repositories there 621 would also be a need to ensure that selected species also generated sufficient exudates/lixiviants.

New requirements for biodiversity net gain, introduced in the UK as part of the Environment Act 2021 create an opportunity to deliver biodiversity as an integral part of the system as well as the restoration phase. For example, current landfills, or those planned for habitats of low distinctiveness, such as quarries, or improved grassland, could be restored to provide mosaic habitats of medium or high distinctiveness, including native flower-rich grasslands, to provide pollination services, open grassland, 627 mixed broadleaved woodland and ponds and wetlands (Natural England, 2021). Furthermore, 628 revegetation of landfills can provide many of the services of urban greenspaces, including improved air 629 quality, and reduced temperature and flood risk (Harwell et al., 2021; Li et al., 2019; Pereira et al., 630 2018). As well as providing water storage, including wetlands and ponds will also allow the 631 recirculation of clean water contributing to the regulation of water pollution (Benyamine et al., 2004).

632

633 7.2 Provisioning services

In addition to cleaned residue, and a concentration of potentially valuable elements in the anthropogenic 634 635 ore zone, there are equally important potential applications for the upper surface of the ASPIRE repository (as with other landfills) to provide food and energy. The establishment of agriculture and 636 637 forestry on restored landfills is a common end use and selecting species that can be used in food or timber production whilst providing the necessary lixiviants could ensure ASPIRE repositories are also 638 639 able to contribute provisioning services. Landfills are also increasingly being used for energy 640 generation, through solar farms (Szabó et al., 2017) or biomass crops (Cervelli et al., 2020; Zalesny et al., 2020) and there is also potential here for added value from ASPIRE repositories. This approach can 641 also achieve biodiversity benefits (Cervilli et al., 2020), and in the case of some energy crops, spaces 642 643 for recreation use for example through the creation of community woodlands. For example, the energy 644 crop miscanthus can provide a habitat for farmland birds (Bright et al., 2013) and the brown hare, whose population is declining in areas of intensive agriculture (Petrovan et al., 2017). Creating solar farms on 645 646 landfills, particularly, can overcome some of the tensions with using agricultural land for this purpose 647 (Szabó et al., 2017). As a new concept, an ASPIRE landfill will be well placed to take advantage of the latest developments in other fields, such as the dual use of land for cultivation and solar farms (Toledo 648 649 and Scognamiglio, 2021). These 'agrovoltaic' systems employ a range of technologies to ensure that crop production and energy generation can work in unison, for example, by using vertical photovoltaic 650 651 (PV) panels, or elevating the panels several metres about the ground, allowing for vegetation growth beneath (Toledo and Scognamiglio, 2021). Depending on the configuration and species selection this 652 approach can increase crop yields and improve the efficiency of the PVs (Toledo and Scognamiglio, 653

654 2021). There may also therefore be the potential to combine energy crops, such as miscanthus, with655 solar farms to increase the energy generation from ASPIRE repositories.

656

657 7.3 Cultural services

Many landfills have been restored to high quality greenspaces, often close to where people live, providing space for rest, relaxation, physical activity and contact with nature (Li et al., 2019). These activities provide cultural ecosystem services including health and wellbeing benefits such as improved mental health, physical activity (Li et al., 2019).

662

663 8 Carbon Capture

Any soil surface has the potential for capturing carbon through the biological colonisation and 664 development of the regolith. Thus, an ASPIRE repository, which deliberately includes a vegetated upper 665 surface would achieve carbon capture. Moreover, many mineral-rich wastes contain components which 666 will either directly react with CO₂ or will lead to capture of CO₂ as bicarbonate via mineral weathering. 667 668 The carbonation mechanism is particularly relevant to alkaline wastes, whereby soluble oxide and silicate minerals react with CO₂ and form carbonate minerals (Renforth, 2019). Recent studies have 669 suggested that legacy iron and steel wastes have a carbon capture potential of up to 80 million tonnes 670 in the UK (e.g. Riley et al., 2020, and section 4.3), whilst global estimates suggest between 5-12% of 671 672 global CO₂ emissions could be mitigated through carbonation with alkaline residues (Renforth, 2019). 673 The sequestration process in these wastes is currently limited in disposal settings by low rates of 674 atmospheric CO₂ ingress into heaps and surface armouring of wastes by secondary deposits (e.g. Pullin et al., 2019). An ASPIRE repository could accelerate carbonation rates in a controlled manner through 675 managed weathering of shallow piles (to encourage contact with atmosphere) and accelerated flushing 676 of residues (to minimise surface armouring and accelerate weathering rates). The deliberate engineering 677 of the surface to encourage downward percolation of water and root exudates may transport organic 678

materials into the waste repository where biological mineralisation should further enhance rates of mineral weathering and carbonation. Interestingly, there may well be significant financial incentives associated with this atmospheric CO_2 sequestration (e.g. in operating an auditable negative emissions system: Renforth, 2019) which could assist with long-term management of ASPIRE repositories.

683

684 9 Technology Barriers and Risks

There are foreseeable barriers to the further development of the ASPIRE repository concept, including both the technical, engineering challenges and public perception. There are potentially enhanced risks of contaminant escape due to increased water movement, potential for the development of hydraulic heads on liners and presence of agents that enhance contaminant concentration and mobilisation. However, existing containment engineering for landfills and heap leaching facilities (common in the mining industry) should find application in mitigating this risk. Furthermore, landfill monitoring technology is well established, and environmental monitoring technologies continue to develop apace.

692 There are risks and barriers associated with public perceptions of the technology, similar to those experienced during the remediation of contaminated sites and mining operations. For example, they 693 694 include concerns that there will be adverse impacts on the environment from the release of pollutants and the time taken for resource extraction (Song et al., 2019; Tayebi-Khorami et al., 2019). There may 695 696 also be a lack of trust from local communities that operators are mitigating risks effectivity or acting in 697 their interests (Tayebi-Khorami et al., 2019; Sinnett and Sardo, 2020). Operators may also be resistant 698 to a new technology and any associated liabilities (Tayebi-Khorami et al., 2019; Song et al., 2019), and 699 uncertain that a market exists for the product generated by ASPIRE repositories (Tayebi-Khorami et 700 al., 2019) or the ownership of these products. Currently, there are also regulatory risks to adoption of 701 the ASPIRE concept as with an authentic shift towards a CE model for mining waste management 702 (Tayebi-Khorami et al., 2019; Cisterna et al., 2022), the proposed long timescales may make it difficult 703 to foresee future changes in environmental regulations that might impact on operation or recovery.

Furthermore, there may also be increased cost implications for the operators in terms of long-termmonitoring, which may result in additional running costs compared with a traditional landfill.

706

707 **10. Conclusions**

708 There are many reasons why mineral-rich wastes are currently landfilled. However, it is self-evident that environmental containment offered by a landfill will eventually succumb to environmental 709 processes, potentially allowing pollution to escape to burden future generations. Thus, in a very real 710 sense, landfilled waste becomes "someone else's problem", removed in time, rather than 711 712 geographically, from the producer. Therefore, landfill cannot be considered as an intergenerationally equitable waste management option. Temporary storage is an important concept that has been 713 714 considered but not adopted for MSW but may be more applicable for mineral-dominated wastes from 715 mining and industry. The case for temporary storage is more compelling, due to the disconnect between 716 potential end-users for residues and the waste production. The reuse of mineral-rich wastes is also often 717 limited because of the presence of leachable contaminants. Separating contaminants from clean residue 718 or concentrating valuable components from a waste where they are dispersed requires energy. The 719 ASPIRE concept looks to extend the concept of temporary storage to include in-built biogeochemical 720 engineering and the solar insolation flux to utilise the time in storage for the separation and 721 concentration of contaminants (or resources). Furthermore, there are clear opportunities for adding value through ecosystem services and carbon capture. There are several important engineering and 722 legislative hurdles that remain to be solved. Importantly, recycling materials within ASPIRE 723 repositories is not intended to displace any existing economically viable and sustainable recycling 724 technologies for mineral-rich wastes. However, the concept could provide a potentially practicable 725 Circular Economy solution for materials that would otherwise go to conventional landfill, thus at 726 minimum replacing/displacing landfill with temporary storage and treatment for recycling at the base 727 728 of the waste hierarchy.

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