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Assessing the sustainability of acid mine drainage (AMD) treatment in
 South Africa

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15 Abstract

The environmental sustainability of acid mine drainage (AMD) treatment at semi-industrial 16 scale is examined by means of the life cycle assessment (LCA) methodology. An integrated 17 process which includes magnesite, lime, soda ash and CO₂ bubbling treatment was employed 18 to effectively treat, at semi-industrial scale, AMD originating from a coal mine in South 19 Africa. Economic aspects are also discussed. AMD is a growing problem of emerging 20 concern that cause detrimental effects to the environment and living organisms, including 21 humans, and impose on development, health, access to clean water, thus also affect economic 22 growth and cause social instability. Therefore, sustainable and cost effective treatment 23 methods are required. A life cycle cost analysis (LCCA) revealed the viability of the system, 24 since the levelized cost of AMD treatment can be as low as R112.78/m³ (€7.60/m³ or 25 $9.35/m^3$). Moreover, due to its versatility, the system can be used both at remote locales, at 26 stand-alone mode (e.g. using solar energy), or can treat AMD at industrial scale, thus 27 substantially improving community resilience at local and national level. In terms of 28 environmental sustainability, 1.18Pt or 29.6 kg CO_{2eq} are emitted per treated m³ AMD or its 29 environmental footprint amount to 2.96 Pt/m³. South Africa's fossil-fuel depended energy 30 mix and liquid CO₂ consumption were the main environmental hotspots. The total 31 environmental footprint is reduced by 45% and 36% by using solar energy and gaseous CO₂, 32 respectively. Finally, AMD sludge valorisation, i.e. mineral recovery, can reduce the total 33 34 environmental footprint by up to 12%.

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37 Keywords: wastewater treatment; water management; scenario analysis; Acid rock drainage
38 (ARD); hazardous wastes; SimaPro

39 **1. Introduction**

Access to clean water is a basic human right, and one of the cornerstones of 40 environmental protection in Europe (Eurostat, 2017). Water is critical for sustaining 41 ecosystems, plays a fundamental role in the climate regulation cycle and is also the primary 42 requirement for human survival and socioeconomic development (Eurostat, 2017; Naidoo, 43 2016). Even though clean water access is taken for granted in the developed world this is not 44 the case for developing countries, which are struggling to keep economic growth but often at 45 the expense of environmental protection and water quality. South Africa is a developing 46 47 country that faces water scarcity issues. On top of this, its water systems are severely harmed by different forms of pollution, including acid mine drainage (AMD) (Naidoo, 2016). AMD, 48 also known as acid rock drainage (ARD), is a common problem at mine sites, primarily at 49 50 abandoned ones, and one of the main environmental challenges facing the mining industry worldwide (Council for Geoscience, 2010; Johnson and Hallberg, 2005). It is mainly 51 produced from the bio-hydro-geochemical weathering of pyrite and other reactive sulphide 52 bearing minerals, when exposed to oxidising conditions (Masindi et al., 2017). AMD 53 emanating from active or abandoned mines and from mine wastes are often net acidic. These 54 55 effluents pose an additional risk to the environment, since they often contain elevated concentrations of metals (iron, aluminium and manganese, and possibly other heavy metals) 56 57 and metalloids (Johnson and Hallberg, 2005). South Africa has a long history in mining and 58 its economy is still largely driven by a strong mining industry, nonetheless growing evidence suggest that its water resources have been grossly impacted by AMD (Council for 59 Geoscience, 2010; Naidoo, 2016). 60

A wide array of treatment methods, such as ion-exchange, adsorption, bio-sorption, chemical-neutralising agents, coagulation and precipitation, have been proposed for AMD treatment (Johnson and Hallberg, 2005; Masindi et al., 2017). In general, treatment methods

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64 can be divided into those that use either chemical or biological mechanisms and they can be further classified as i) active (they require continuous inputs of neutralisation materials, such 65 as magnesite, periclase, brucite, lime, hydrated lime, and limestone, to sustain the process), ii) 66 67 passive (they require relatively little resource input once in operation and could involve the use of wetland, reactive barriers and lime drains), or iii) integrated (i.e. they entail the 68 combination of both) (Johnson and Hallberg, 2005; Masindi, 2017). The most widespread 69 method for AMD neutralization is active treatment, involving addition of an alkaline material 70 (chemical-neutralising agent such as magnesite, lime, calcium carbonate, sodium carbonate, 71 72 sodium hydroxide, and magnesium oxide and hydroxide) that will raise the pH, accelerate ferrous iron rate of chemical oxidation (to this end active aeration or additional chemical 73 74 oxidising agent are also required) and cause many of the metals present in solution to 75 precipitate as hydroxides and carbonates (Johnson and Hallberg, 2005). Lime treatment is the most commonly used active treatment method, due to its high efficiency and low cost 76 (Potgieter-Vermaak et al., 2006). 77

Even though treatment efficiencies of the available AMD methods are well-78 established and explored (e.g. (Johnson and Hallberg, 2005; Potgieter-Vermaak et al., 2006), 79 80 this is not the case for their environmental sustainability, where only a few cases dealing with the environmental sustainability of AMD treatment systems are available (Hengen et al., 81 82 2014; Tuazon and Corder, 2008). Therefore, herein a full life cycle assessment (LCA) of a 83 typical AMD treatment method is carried out, using primary life cycle inventory (LCI) data collected from a semi-industrial AMD treatment plant. The goal is to assess the 84 environmental sustainability of a typical AMD treatment process, identify environmental 85 86 hotspots and identify avenues to improve its environmental sustainability, such as resource extraction from AMD sludge. Also, economic and social aspects regarding the sustainability 87 of the treatment system are discussed. 88

90 2. The case study

Acid mine drainage (AMD) was collected from a coal mine in Mpumalanga Province, 91 South Africa, and was transferred to the premises of the Council for Scientific and Industrial 92 Research (CSIR), Pretoria campus, South Africa, for treatment. The raw mine water was 93 94 initially colourless, but after reacting with atmospheric air it turned red (Figure 1), due to the oxidation of ferrous to ferric ions. The AMD tetrahedron in Figure 1b shows all relevant 95 components that contribute to this process. Co-existence of raw mine water, atmospheric 96 97 oxygen, sulphide minerals (as a source of iron) and waterborne bacteria (to accelerate the reactions) can lead to the production of AMD (Pondja, 2017). 98

99

A

В



Figure 1. AMD effluent coming out of an underground pit, when it is initially colourless (A) and after being
oxidised by atmospheric air in the presence of sulphide minerals and waterborne bacteria (B), gradually turns to
red (C).

As far as its physical and chemical characteristics are concerned, the AMD under study is very acidic with pH 2, and contains high amounts of sulphate, Fe, Al and Mn, Mg and Ca (Table 1).

108	Table 1: AMD physicochemical	characteristics,	before and after	treatment (data taken	from	(Masindi,	2017)
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Parameters	Initial concentration	Treated effluent	
pH	2	7.5	
Acidity (mg/L CaCO ₃)	800	≤ 0.01	
Alkalinity (mg/L CaCO ₃)	<5	80	
Aluminium (mg/L Al)	300	\leq 0.01	

Calcium (mg/L Ca)	300	≤ 0.01
Electrical Conductivity (mS/m [25°C])	600	200
Iron (mg/L Fe)	8,000	≤ 0.01
Magnesium (mg/L Mg)	300	0.5
Manganese (mg/L Mn)	75	≤ 0.01
Sodium (mg/L Na)	≤ 0.01	5
Sulphate (mg/L SO ₄)	30,000	50
Total Dissolved Solids	3,500	1,000
Total Hardness (mg/L)	2,000	200

As shown in Figure 2 the AMD treatment system comprises the following four 110 discrete process steps: (1) neutralization of AMD and partial removal of sulphates achieved 111 by using calcined cryptocrystalline magnesite (magnesite treatment); (2) addition of 112 113 limestone to reduce water hardness and residual sulphate as gypsum (limestone treatment); (3) soda ash addition to reduce residual Ca and hardness (soda ash treatment); (4) CO₂ 114 bubbling to correct the pH to 7.5 and recover limestone (CO₂ bubbling). The main products 115 of this treatment process comprise the treated AMD effluent and the produced sludge. The 116 latter is typically discarded for landfilling, but it can be also valorized as will be discussed in 117 the sensitivity analyses section. As shown in Table 1, the system is capable of providing a 118 119 high quality treated water output, which meets South Africa's water quality standards to be safely returned to nature, or used for industrial and agricultural purposes (DWAF, 1996). 120





Figure 2: Flow diagram of the semi-industrial AMD treatment unit under study and possible scenarios for thedisposal/treatment of the treated effluent and the AMD sludge.

The aforementioned system was designed, constructed, and commissioned at the premises of CSIR Pretoria campus, South Africa, where it operates at semi-industrial scale and is able to effectively treat 3.5 m^3 of AMD daily (Figure 3).

At the time of writing, a reverse osmosis (RO) followed by chlorination tertiary treatment system is under testing in order to explore the possibility to produce drinking water; a viable product for South African rural communities. All process steps take place in the same reactor (i.e. clarifier), since each process step has to be completed before moving on to the next step. This reduces the system's initial capital expenditure and less space is occupied, i.e. land use is minimized.



Figure 3: The semi-industrial AMD treatment unit in operation at the premises of CSIR Pretoria campus, SouthAfrica.

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A detailed discussion regarding the four process steps under study can be found in 138 Masindi (2017. For the magnesite treatment stage 10 kg of magnesite per m^3 of AMD are 139 added to the clarifier, shown in Figure 2 and Figure 3. The mixture is agitated for 60 minute 140 and then it is left for another 60 minute to settle, where solid precipitates are gravity settled. 141 Then, the magnesite treated AMD is transferred to a holding tank and the sludge is 142 143 transferred to a separate tank (sludge tank). In this stage Fe-species can be recovered from the sludge. For the limestone treatment stage, the magnesite-treated effluent is recycled back to 144 the clarifier and 10 kg/m³-AMD of limestone are added into it. The mixture is then agitated 145 for 60 minute and is left for another 60 minute unstirred, to allow solid precipitates to settle. 146 The magnesite/limestone treated AMD and the sludge are transferred back to the holding and 147 the sludge tank, respectively. In this stage, residual Ca (gypsum) and Mg (brucite) can be 148 149 recovered from the sludge. For the soda-ash treatment stage, the effluent is recycled back to the clarifier and 4 kg/m³-AMD of soda ash are added, following the same procedure (i.e. 60) 150 minute agitation, 60 minute settling and sludge removal). Finally, in the CO₂ bubbling stage 151

the magnesite/limestone/soda-ash treated AMD is recycled back to the clarifier, where CO_2 is bubbled (45 L/min) until the pH reaches 7.5. Similarly, the effluent is left for 60 minute to settle and then is transferred to the treated effluent tank, while the sludge is collected to the sludge tank (Figure 2 and Figure 3).

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3. Goal and scope and system boundaries

The purpose of this assessment is to identify the environmental performance and the 158 main environmental hotspots of a robust AMD treatment method, as well as identify 159 environmental saving avenues through scenario/sensitivity analyses. To this end, primary LCI 160 data were collected from a semi-industrial AMD treatment plant (Figure 3) and simulated 161 using the LCA methodology. Since, AMD constitute an environmental problem of emerging 162 concern in South Africa, and beyond, the results of this work are of interest to researchers, 163 decision and policy makers, as well as the mining and water/wastewater industry, which all 164 constitute the intended audience of this work. 165

In order to quantify the environmental performance of the system 1 m³ of effluent generated by the AMD reactor was set as the functional unit. Therefore, input and output data were normalized per m³ of effluent water treated by AMD reactor. Furthermore, the attributional (ALCA) approach was used, since it estimates the environmental impacts of a product/system attributed to the delivery of a specified amount of the functional unit (Chatzisymeon et al., 2016), which is the case here.

The environmental modelling was carried out using the SimaPro 8 software package, based on the LCA methodology as set in ISO 14040 and 14044 (ISO, 2006a; ISO, 2006b). The time-related coverage of this work refers to present, i.e. 2018, while its geographical coverage is South Africa and areas that are affected by AMD pollution. Moreover, average 176 technology was assumed and a single-issue (IPCC 2013) and a multi-issue (ReCiPe2016) life cycle impact assessment (LCIA) method were used, the latter by employing the Hierarchist 177 perspective. It should be noted that there might be some limitations of applying the ReCiPe 178 method, since this was first developed in and for the European context (Adiansyah et al., 179 2017a). Nevertheless, the updated version of ReCiPe, i.e. ReCiPe2016, which was used in 180 this work, provides characterisation factors that are representative for the global instead of the 181 European scale, while maintaining the possibility for a number of impact categories to be 182 adapted at a country or continental scale (Huijbregts et al., 2017). 183

184 In Figure 4 the system boundaries, which define the smallest elements (i.e. unit processes) for which input and output data are quantified in the LCI and are included in the 185 LCA are presented (ISO, 2006b). All four AMD treatment steps, along with their main inputs 186 187 and outputs are included in the system boundaries. For the AMD treatment plant, a useful lifetime of 20 years was taken into account, which is in line with relevant literature (e.g. 188 (Foteinis et al., 2018; Ioannou-Ttofa et al., 2016; Ioannou-Ttofa et al., 2017)). AMD 189 transportation from Mpumalanga coalmine to the semi-industrial treatment plant, i.e. CSIR 190 Pretoria campus premises, South Africa, is external to the system boundaries, since future 191 192 treatment systems are expected to be built near the AMD sources. Furthermore, since this is a cradle to gate LCA the final use of treated water is external to the system boundaries. The 193 194 reason is that depending on the final use, e.g. disposal in natural water bodies, irrigation, or 195 drinking water production, a different environmental burden/benefit would be ascribed to each route, thus making the LCA specific for the chosen route. Moreover, the infrastructure 196 required, i.e. reinforced concrete slabs to accommodate the treatment systems, piping, as well 197 198 as land use were into account separately, i.e. a sub-system for infrastructure was created.



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Figure 4: The system boundaries (shown with dashes) of the AMD treatment system.

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202 4. Life cycle inventory (LCI)

As mentioned above, primary LCI data for the system under study (i.e. semi-industrial 203 AMD treatment plant) were collected from its construction and operation phase in CSIR 204 Pretoria campus, South Africa. Table 2 summarizes the LCI that were used in this work, 205 normalized per functional unit, i.e. the effective treatment of 1 m³ of AMD. It has to be noted 206 that the clarifier and tanks under study, as well as the pumps and the propeller were not 207 identified in SimaPro's proprietary LCI databases, and thus literature data were used as 208 209 proxies. Specifically, for the pumps that are required to transfer the effluent between tanks and the clarifier LCI data from (Xylem Inc, 2011) were used and re-scaled according to their 210 rated power output (0.75 kW). It has to be noted that magnesite, limestone and soda ash are 211 212 inserted in the system in a slurry form, i.e. semi-liquid mixture of the chemical with water, and each require 10 minute pumping from their storing tank to the clarifier. Furthermore, it 213 takes 30 minute to move the effluent from the clarifier to the holding tank and another 30 214

215 minute to move it back to the clarifier. The sludge is much thicker and therefore it takes 10 minute to transfer it from the clarifier to the sludge tank. For the propeller, LCI data were 216 taken from (Sulzer Ltd, 2013) and re-scaled to fit the desirable rated output (3 kW). In each 217 of the above treatment step mixing (propeller) lasts for 60 minute. Moreover, according to 218 their manufacturer the clarifier and the tanks under study are made from linear low-density 219 polyethylene (LLDPE), while their life span is at least 10 years (JoJo Tanks Ltd, 2017). 220 Therefore, literature data for LLDPE tanks (Shah et al., 2016) were used, while it was 221 assumed that the tanks and the clarifier will be replaced once during the pilot unit lifespan of 222 20 years. For the diffuser used for CO₂ bundling LCI data were taken from the literature 223 (Ioannou-Ttofa et al., 2016), assuming that its main material is Polyvinyl chloride (PVC). 224 Piping comprised high-density polyethylene (HDPE) pipes (~ 20 m total length) and was 225 226 taken from Ecoinvent database. The chemicals that are used to drive the process were taken directly from Ecoinvent, apart from magnesite which LCI was taken from the literature 227 (Cherubini et al., 2008). Moreover, a mean transportation distance of 40 km was ascribed to 228 all construction materials and system inputs, except from AMD transportation which is 229 outside of the system boundaries. Finally, it was assumed that 40 m^2 of industrial land will be 230 occupied throughout the treatment plant life span. 231

Process	Main parts -	Value	LCI data reference	
chemical reagents				
		Infrastructure		
Land use	Industrial land	40 m ²	CORINE 121a	
Transportation	Euro 4 lorry	40 km	Ecoinvent 3.3	
Piping	HDPE	20 years	Industry data 2.0	
Clarifier	LLDPE	10 years	(Shah et al., 2016)	
Holding tank	LLDPE	10 years	(Shah et al., 2016)	

Table 2: The LCI of the semi-industrial treatment plant for the treatment of 1 m³ AMD

Magnesite treatment				
Propeller	3 kW	60 min	(Sulzer Ltd, 2013)	
Pumping	0.75 kW	80 min	(Xylem Inc, 2011)	
Magnesite	MgCO ₃	10 kg/m ³	(Cherubini et al., 2008)	
Electricity	South African mix	4 kWh/m ³	Ecoinvent 3.3	
	I	Limestone treatment		
Propeller	3 kW	60 min	(Sulzer Ltd, 2013)	
Pumping	0.75 kW	80 min	(Xylem Inc, 2011)	
Limestone		10 kg/m ³	Ecoinvent 3.3	
Electricity	South African mix	4 kWh/m ³	Ecoinvent 3.3	
		Soda ash treatment		
Propeller	3 kW	60 min	(Sulzer Ltd, 2013)	
Pumping	0.75 kW	80 min	(Xylem Inc, 2011)	
Soda ash	MgCO ₃	4 kg/m^3	Ecoinvent 3.3	
Electricity	South African mix	4 kWh/m ³	Ecoinvent 3.3	
		CO ₂ bubbling		
Pumping	0.75 kW	70 min	(Xylem Inc, 2011)	
CO ₂ Diffuser	EPDM	10 years	(Ioannou-Ttofa et al., 2016)	
Carbon dioxide	CO ₂	45 L/min	Ecoinvent 3.3	
Electricity	South African mix	0.875 kWh/m ³	Ecoinvent 3.3	
Treated effluent tank	LLDPE	10 years	(Shah et al., 2016)	
		Outputs		
Treated AMD effluent (water)		$0.97 \text{ m}^3/\text{m}^3$	-	
Fe (taken as iron sulfate)		2 kg/m^3	Ecoinvent 3.3	
Gypsum	Gypsum		Ecoinvent 3.3	
Brucite (taken as magnesium oxide)		1 kg/m^3	Ecoinvent 3.3	
Limestone		3 kg/m^3	Ecoinvent 3.3	

5. Life cycle impact assessment (LCIA)

The life cycle impact assessment (LCIA) associates the collected LCI data with 234 specific environmental impacts and damages and also attempts to understand those 235 impacts/damages (ISO, 2006b). Here, a single-issue, i.e. IPCC 2013 for a timeframe of 100 236 237 years, and a multi-issue, i.e. ReCiPe, LCIA methods were used. IPCC 2013 compares processes based on CO₂ equivalent (CO_{2eq}) emissions, i.e. total greenhouse gas (GHG) 238 emissions, used to measure Global Warming Potential (GWP), which is a standard indicator 239 of environmental relevance. This is also included in ReCiPe's midpoint impact category 240 "Climate Change", but using a single-issue method allows a more direct dissemination of the 241 results to the general public (Foteinis et al., 2018). ReCiPe can express results both at 242 midpoint, where environmental impacts are examined earlier in the cause-effect chain, and 243 endpoint level, where environmental impacts are examined at the end of the cause-effect 244 245 chain (Ioannou-Ttofa et al., 2016). The midpoint approach provides a robust understanding of the environmental performance of the AMD treatment pilot-unit, but results are hard to 246 communicate to the general public. The endpoint or damage-oriented approach, can translate 247 248 environmental impacts into issues of concern, such as human health, natural environment and natural resources, but it is associated with higher levels of statistical uncertainty due to data 249 gaps and assumptions stacking up along the cause-effect chain. Nonetheless, endpoint results 250 are easier to communicate to decision- and policy-makers and the general public 251 (Chatzisymeon et al., 2016). 252

At midpoint level ReCiPe comprises the following impact categories: climate change (CC), ozone depletion (OD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), human toxicity (HT), photochemical oxidant formation (POF), particulate matter formation (PMF), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine ecotoxicity (MET), ionising radiation (IR), agricultural land occupation (ALO), urban land occupation (ULO), natural land transformation (NLT),water depletion 259 (WD), mineral resource (metal) depletion (MRD), fossil fuel depletion (FD). In order to reach endpoint, ReCiPe converts and aggregates most of the midpoint impact categories into the 260 following damage categories: i) damage to human health, covering climate change, ozone 261 262 depletion, toxicity, and human health associated with PM10 and ozone; ii) damage to ecosystem diversity, covering climate change, acidification, toxicity, and land-use; and iii) 263 damage to resource availability, covering mineral resource depletion, and fossil fuel depletion 264 (Adiansyah et al., 2017a; Foteinis et al., 2018). In order to obtain a holistic understanding of 265 the environmental performance of the AMD treatment system under study, both midpoint and 266 267 endpoint approaches were used. Moreover, the Hierarchist perspective (H) was employed by using the normalisation values of the world and average weighting (i.e. world ReCiPe H/A). 268

269 6. Economic analysis

270 A useful tool to assess economic sustainability the life cycle cost analysis (LCCA), i.e. the cost of an asset, or its parts throughout its life cycle, while fulfilling the performance 271 requirements. Construction costs, maintenance costs, operational costs, occupancy costs, end-272 of-life costs and non-construction costs are usually included in LCCA (Zhong and Wu, 273 2015). In this study, an economic evaluation of the prototype AMD treatment system, based 274 275 on the LCCA methodology, was carried out. In the analysis, the initial capital expenditure (CAPEX) for setting up the system, as well as maintenance and operating expenses (OPEX) 276 277 were taken into account.

It has to be noted that the analysis was focused on accounting for the operating cost, which represent the majority of financial input of the AMD treatment unit, and only under the present conditions, i.e. not accounting for inflation. This analysis intends to act as a screening tool to assess the economic viability of the system under study, rather than specify possible options and provide information about costs and benefits in present monetary value such as in benefit-cost analysis (BCA). For example, treating AMD reduces sulfate waterborne
emissions and therefore minimizes the environmental impact on (eco)toxicity and on
freshwater and marine eutrophication. In addition, water and land conservation and GHG
reduction could be achieved, which lead to monetary benefits (Adiansyah et al., 2017b).
Nonetheless, calculating the monetary benefits of AMD treatment is beyond the goals and
scope of this work, and could be addressed in future studies.

289 7. Results and discussion

290 7.1 Carbon footprint

Total carbon equivalent (CO_{2eq}) emissions were estimated using the IPPC 2013 LCIA 291 method for 100 years timeframe and it was found that the effective treatment of 1 m³ of 292 AMD emits 29.6 kg CO_{2eq}. Regarding the contribution of each process step, it was found that 293 CO₂ bubbling had the highest score (13 kg CO_{2eq}), followed by soda ash treatment (6.9 kg 294 CO_{2eq}), magnesite treatment (5.04 kg CO_{2eq}) and limestone treatment (4.54 kg CO_{2eq}). The 295 main environmental hotspot was identified as electricity consumption (14.5 kg CO_{2ea}), 296 followed by the liquid CO₂ input for the bubbling process (11.8 kg CO_{2eq}). Soda ash and 297 magnesite, as materials, had a lower carbon footprint, 2.41 and 0.558 kg CO_{2ea} respectively. 298 The remaining inputs (e.g. Tovex explosive for magnesite and limestone mining and concrete 299 for the system base) had a very low to negligible score. Therefore, the main contributors to 300 the total carbon footprint are electricity consumption from South Africa's fossil fuel-301 depended energy mix (49.2 %), followed by the liquid CO₂ input (40%) and soda ash 302 303 (8.16%), as shown in Figure 5.



Figure 5: The main contributors to carbon emissions in kg CO_{2eq} per m³ of treated AMD.

306 7.2 ReCiPe LCIA method

307 7.2.1 ReCiPe results at midpoint

308 LCA findings are first presented at midpoint level, using ReCiPe's LCIA characterisation model and then results are normalized using the world's reference 309 inventories. Figure 6 shows the contribution of all process sub-systems to each of the 310 ReCiPe's 18 midpoint impact categories (characterisation). It is observed that the treatment 311 step that has the highest contribution to all impact categories is the CO₂ bubbling stage. 312 Magnesite, limestone, and soda ash treatment yielded comparable scores to most impact 313 categories, while infrastructure (i.e. land use, concrete slab/foundations and piping) has a 314 315 very low to miniscule contribution to all impact categories (Figure 6).

The high scores (from 25% - 85%) of the CO₂ bubbling stage are attributed to the high liquid CO₂ amounts required to drive the process, while electricity for water pumping required in this step had a much lower contribution. CO₂ is mainly generated as a by-product from various industrial production processes, primarily from ammonia or hydrogen production, and almost half of the amount produced is used directly in its gaseous form in the

close neighbourhood, mainly to produce urea or methanol. If gaseous CO₂ is sourced directly 321 from another production process it could be assumed that it will be free of any environmental 322 burden (Althaus et al., 2007). On the other hand, liquid CO_2 , which is the most commonly 323 bought and sold form of CO₂, is associated with environmental burdens since energy and 324 resources are required for CO₂ extraction and purification (Althaus et al., 2007). Here, liquid 325 CO_2 , originating from ammonia production, was assumed to be used in the bubbling stage. It 326 should be mentioned that if gaseous CO₂ could be sourced for future industrial scale AMD 327 treatment plants the total environmental footprint of the process could be further reduced. 328 329 This scenario is examined in the sensitive analyses section.





Since the electricity inputs during the first three steps are the same, their differences 333 in each midpoint impact categories is attributed to the chemical reagents, i.e. magnesite, 334 limestone, and soda ash as materials, utilized in each step. The higher contribution (ranging 335 between 9% - 32%) of the soda ash step is attributed to the sodium carbonate, i.e. soda ash, 336 input require to drive this process step. Soda ash is mainly produced by the Solvay process, 337 also called ammonia soda process (Mahida Prashantsinh et al., 2015), and hence soda ash 338 (light grade) manufactured from this process was taken into account here. The Solvay process 339 uses salt (NaCl) and limestone as raw materials and involves several treatments to produce 340

341 soda ash and therefore various emissions and environmental impacts are generated (Mahida Prashantsinh et al., 2015). As a result, the estimated CO_{2eq} emissions for the production of 342 one tone of soda ash to be between 2 and 4 tons, depending on the energy source used 343 344 (Nazari and Sanjayan, 2016). On the other hand, magnesite (4% - 23%) and limestone (2% -21%) can be obtained directly through mining and refining and therefore are associated with 345 lower carbon footprints, compared to soda ash (Cherubini et al., 2008; Nazari and Sanjayan, 346 2016), and this is reflected in Figure 6. Finally, the low contribution of the infrastructure in 347 all impact categories is mainly attributed to: (i) the high life span of the concrete 348 349 slab/foundations and piping (20 years) (ii), its main inputs are not associated with hazardous or carcinogenic emissions and (iii) land use is not extensive since all main treatment steps are 350 carried out in the same reactor (clarifier). 351

352 In order to get a better idea of the relative magnitude of each treatment step, results were normalised using the world's reference inventories (i.e. the world normalisation factors 353 were used) and are shown in Figure 7. Normalisation is an optional step of the LCIA, which 354 transforms the results by dividing each impact category with a corresponding reference value 355 (ISO, 2006b). By doing so, results are compared with reference values and the magnitude of 356 357 each impact is identifyed. The most affected impact categories are, from higher to lower scores, MET, FET, FE and HT, while the impact categories TA, FD, PMF, CC yielded much 358 lower (an order of magnitude) normalised scores. The remaining midpoint impact categories 359 360 yield very low to miniscule normalised scores (Figure 7).



Figure 7: Normalised ReCiPe's midpoint results using the world's reference inventories for the effective
 treatment of 1 m³ of AMD.

The high scores in the (eco)toxicity (MET, FET and HT) and eutrophication (FE) 364 impact categories are attributed to the mining of the chemical reagents and of the fossil fuels 365 required for electricity generation (South Africa's energy mix is dominated by fossil fuels, 366 367 mainly coal (Papadaki et al., 2017)). It has to be noted that CO_2 harvesting and purification require large amounts of energy (Althaus et al., 2007), in this case electricity from fossil 368 fuels. Therefore, the chemical reagents and particularly fossil fuel mining exposes previously 369 buried coal minerals to both oxygen and water, thus releasing, through waterborne emissions, 370 mine-derived sulfate salts. Sulfate emissions could disrupt water balance and ion exchange 371 372 processes, thus causing aquatic organisms to live under stress or even death (Zhao et al., 2017). Moreover, magnesite, limestone and fossil fuel extraction and transportation, as well 373 as soda ash production and fossil fuel refining and combustion release toxic materials, such 374 375 as heavy metals, sulphurous compounds and polycyclic aromatic hydrocarbons (PAHs) to the environment, thus also affecting the (eco)toxicity impact categories (Ioannou-Ttofa et al., 376 2016). 377

As far as the FE impact category is concerned, mining activity is an increasingly important stressor for freshwater ecosystems, since sulfate can co-vary with other environmental parameters, such as nitrogen and phosphorus, and impact aquatic organisms. 381 The reason is that the introduction of sulfates in natural water matrices, through mining activities, can increase the availability of nitrogen and phosphorus through internal 382 eutrophication (Zhao et al., 2017). Also, fossil fuel combustion lead to nitrogen oxides 383 384 emissions which impact ME (Ioannou-Ttofa et al., 2017). Marine ecosystems are affected to a much lower degree, compared to freshwater ecosystems, since they are in general more 385 resilient to eutrophication and lower quantities of direct and indirect (e.g. from fossil fuel 386 burning) nitrogen emissions are attributed to the system under study (limestone used in the 387 treatment system is assumed to be recovered and/or properly disposed, thus it does not reach 388 389 the sea).

Lastly, the less affected midpoint impact categories, i.e. TA, FD, PMF and CC, are mainly affected by mining activities and fossil fuel extraction and burning. The latter directly affect CC and TA, while also releases particulate matter and leads to their depletion, thus affecting PMF and FD impact categories, respectively (Foteinis et al., 2018).

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7.2.2 ReCiPe results at endpoint

Figure 8 shows ReCiPe's weighted results at endpoint level (Hierarchist perspective 395 using the normalisation values of the world with the average weighting set). Weighting is an 396 optional step in the LCIA, after normalisation, where results are multiplied by weighting 397 factors corresponding to each impact category. Weighted results can be then aggregated into 398 a single score, in order to access the total environmental footprint of the effective treatment of 399 1 m³ of AMD. It was found that the weighted damage to human health exhibits the highest 400 score (1.92 Pt), followed by the damage to resources availability (0.932 Pt), while the damage 401 402 to ecosystem diversity has the lower score (0.109 Pt). Therefore, the aggregated score is found to be 2.96 Pt per m³ of AMD. The high scores of the first two damage categories are 403 404 attributed to electricity consumption and to the mining/processing of the chemical reagents

405 added in each treatment step. Similarly to IPCC 2013, the treatment step that has the highest aggregated score is CO₂ bubbling (1.19 Pt), followed by soda ash (743 mPt), magnesite (526 406 mPt), and limestone treatment (479 mPt). Moreover, the main environmental hotspot was 407 408 identified to be electricity from South Africa's fossil-fuel depended energy mix (1.51 Pt or 51.1% of the total environmental footprint). From it, 35.5% or 1.05 Pt is attributed to 409 stirring/propelling and the remaining 15.8% or 0.468 Pt to water pumping. The second 410 environmental hotspot was identified as the liquid CO₂ input (1.08 Pt or 36.4%), followed by 411 soda ash, as a material (276 mPt or 9.33%). The remaining chemical reagents had a lower 412 contribution, i.e. magnesite 58.7 mPt or 1.99%, and limestone 11.7 mPt or ~0.4%. Due to 413 their high life span, the CO₂ diffuser, the propeller and the pumps had a miniscule 414 contribution (<0.1%), while the tanks, including the clarifier, contribute 3.72 mPt or 0.126% 415 416 in total. Finally, the infrastructure contributes 0.44% on the total environmental footprint, with the majority of those impacts attributed to the reinforced concrete used to construct its 417 base (i.e. pipping and land use had a miniscule contribution). 418







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425 7.3 Sensitivity/scenario analyses

426 7.3.1 Source and form of CO₂

The liquid CO₂ used in the bubbling stage was found to be the main environmental 427 hotspot of the system. This, is attributed to the large quantities required and to the energy and 428 resources that are required for CO₂ extraction, purification, liquefaction, and storage 429 (Althaus et al., 2007). Therefore, a sensitivity analysis concerning the source and form of 430 used CO₂ was carried out. To be more specific, the use of processed CO₂ in liquid form 431 versus raw gaseous CO₂ harvested as a process by-product was studied. This scenario was 432 explored based on the fact that the system can directly use gaseous CO₂ produced as a by-433 434 product from various industrial production processes, such as ammonia and hydrogen production, or even sourcing it from a power plant's flue gas, if such facilities are in the close 435 proximity. In these cases, since the gaseous CO_2 is produced as a by-product of another 436 engineering process, it could be assumed that it will be free of any environmental burden 437 (Althaus et al., 2007), thus improving the system's overall environmental sustainability. 438

A scenario dealing with directly using gaseous CO_2 that derives as produced as a byproduct, e.g. flue gas, was examined. If environmental burden-free gaseous CO_2 is fed into the bubbling stage, then the environmental footprint is reduced by ~36%, i.e. 1.88 Pt instead of 2.96 Pt when using liquid CO_2 .

443 **7.3.2** Renewable energy to power the treatment system

The second environmental hotspot of the system was identified as the electricity consumption from South Africa's fossil energy mix, which is attributed to the high share of fossil fuels, mainly coal (Papadaki et al., 2017). Therefore, using an electricity mix solely comprising renewable energy sources (RES) could reduce the system's overall environmental 448 footprint. In this work the use of solar energy (3 KWp single-Si panels photovoltaic (PV) systems), an abundant and readily available RES in South Africa, was examined. It was 449 found that when using solar energy the system's environmental sustainability is substantially 450 improved. Specifically, its total environmental footprint is reduced from 2.96 Pt per m³ to 451 1.64 Pt per m³. Therefore, the introduction of RES, in this case solar energy, can reduce the 452 total environmental footprint by ~45%. Moreover, a third and practically feasible scenario is 453 to combine the use of RES (i.e. solar energy) and gaseous CO₂ in the same AMD treatment 454 system. In this case the total environmental footprint of the system is minimized, from 2.96 Pt 455 per m³ in the initial scenario (i.e. current operating conditions) to 0.559 Pt per m³, and is 456 drastically reduced by ~81%, reaching an overall high environmental sustainability. 457

458 7.3.3 Resource recovery from AMD sludge

459 Finally, the last scenario examined is AMD sludge valorisation, i.e. the recovery of resources that are contained in the generated AMD sludge. Resource recovery from AMD 460 sludge has recently attracted attention, as this could be a promising strategy to reduce AMD 461 treatment overall cost (e.g. see (Masindi, 2017)). Nonetheless, this entails further AMD 462 sludge processing and energy inputs and therefore it could be associated with high 463 environmental impacts that could render sludge valorisation unattractive, from the 464 environmental perspective. For this reason, a screening, in terms of environmental relevance, 465 for each recovered resource was carried out, by using the substitution approach. The concept 466 behind substitution is that the production of a co-product by the system under study causes 467 another production process in another system to be avoided, which results to avoided 468 emissions, resource extractions, etc. (Wardenaar et al., 2012). 469

470 In order to extract the targeted resources from each sludge stream chemical reagents471 are required (Bailey et al., 2016), but most importantly large amounts of energy are required,

472 since the sludge needs to be dried at 105°C for 24 hrs. In this work a typical laboratory oven (EcoTherm Economy) was used and the resources that can be extracted per m³ AMD are Al, 473 Ca, Fe, Mg and Mn. Here we examine the effect of one of the main input of the extraction 474 475 process, i.e. the sludge drying, to identify if sludge valorisation can mitigate the environmental footprint of the examined AMD treatment process. Even though, the 476 extraction procedure is more complicated and requires further processing, we estimated that 477 the additional environmental burdens of the sludge valorisation are compensated by the fact 478 that here sludge drying was achieved by a laboratory oven (industrial ovens require much less 479 480 energy input). Future works could deal with the exact environmental performance of the most promising recourses to be recovered. 481

Regarding this scenario, since the oven is not included in SimaPros's proprietary 482 483 databases, LCI from literature were used as proxies (Jungbluth, 1997). Electricity was assumed to originate from South Africa's energy mix. When simulating the environmental 484 impact of the drying process and the avoided emissions, resource extractions, etc. attributed 485 to each recovered resource the following were observed: For the magnesite treatment sludge 486 stream, it was found that the environmental gains of Fe (assumed to be recovered as iron 487 488 sulfate, instead of iron oxide/hydroxide) can slightly exceed the environmental impacts of electricity consumption of the drying process (i.e. the total environmental footprint can be 489 reduced by 0.09 Pt or by about 3%). When, recovery of gypsum and brucite (assumed to be 490 491 recovered as magnesium oxide) from the limestone treatment step was examined, a higher reduction on the total environmental footprint was observed. Specifically, a 0.27 Pt or about 492 9% reduction of the total environmental footprint could be achieved from gypsum but mainly 493 494 from brucite recovery. Finally, it was found that the environmental gains of limestone recovery did not exceed the environmental impacts of electricity consumption of the drying 495 process. Therefore, alternative routes (e.g. disposal) for the sludge generation during the last 496

497 two treatment stages should be considered. Overall, it was found that sludge valorisation 498 could reduce the total environmental footprint of the AMD treatment process by up to 12%. 499 Hence, results indicate that sludge valorisation could be a promising strategy to offset the 500 environmental impacts of AMD treatment and improve its overall environmental 501 sustainability.

502 7.4 Economic analysis

Regarding the life cycle cost analysis (LCCA), results were promising. Table 3 503 summarizes all inputs, i.e. chemical reagents and electricity, that contributed to the process 504 operating costs. Specifically, it was found that the initial capital expenditure for setting up the 505 system was very low, since it only comprises two linear low-density polyethylene (LLDPE) 506 tanks, one LLDPE clarifier, the piping, the pumps and the stirrer. Moreover, the capital cost 507 (20 years life span) was estimated at ~R200,000 (€13,500 or \$16,500, exchange rate taken at 508 January 2018), which when normalized per treated m³ AMD is miniscule, i.e. R7.78 (€0.52 or 509 0.65) per m³ AMD. Therefore, the economic evaluation was focused on accounting for the 510 operating cost, i.e. chemical reagents and electricity, of the AMD treatment unit. Table 3 511 summarizes the capital and operating costs, which contributed towards the system's total 512 513 cost. The normalized operating cost of the AMD treatment unit was found to be R105 (€7.08 or \$8.71) per m³ AMD. Therefore, the levelized cost for the treatment of 1 m³ AMD is 514 R112.78 (\notin 7.60 or \$9.35), with operating costs being the main contributors (i.e 6.90%), 515 compared to the capital cost (i.e. 93.10%). 516

It has to be noted that if AMD is left untreated it could have large economic impacts, since it could cause detrimental effects to the environment and living organisms, including humans, and impose on development, health, access to clean water, thus stressing social sustainability. The proposed treatment system can address, at least partly, the growing problem of AMD pollution and improve community resilience at local (the system can

- 522 operate off-grid in remote areas) and national level. It can also support other important
- 523 functions, such as agriculture, thus improving economic sustainability.

Lime

Soda Ash

- 524
- 525 Table 3: Economic evaluation of the AMD treatment process capital (CAPEX) and operating expenditure526 (OPEX)

Input	Unit cost	Quantity	Total costs (Rand)
Initial capita	l expenditure (CAP)	EX) required for infras	tructure
50 mm PVC Ball Valve	R 403.00	10	R 4,030.00
154 ml Oatey PVC Cement glue	R 155.00	6	R 930.00
PVC Adapter 50×63 mm 1/2"	R 38.00	15	R 570.00
PVC Adapter 50 mm 1/2"	R 63.00	6	R 378.00
PVC Union Plain 50 mm	R 75.40	10	R 754.00
50 mm PVC Elbow 16 bar	R 67.00	30	R 2,010.00
50 mm PVC T-piece 16 bar	R 72.50	15	R 1,087.50
Fhread Sealing Tape 19mm×30m	R 25.80	3	R 77.40
Tank Connector 50 mm	R 95.90	5	R 479.50
Tap 25 mm	R 45.50	6	R 273.00
PVC Pipe 50 mm	R 37.50	30	R 1 125.00
Tank (replaced once)	R 10,000.00	6	R 60,000.00
Clarifier, mixer and stand	R 90,000.00	1	R 90,000.00
Concrete slab	R 15,000.00	1	R 15,000.00
Contingency cost			R 22,007.00
Το	otal CAPEX		R 198,721.00
Levelise	d CAPEX per m ³		R 7.78
Operating expenditur	e (OPEX) required	for chemical reagents a	nd energy inputs
AMD, R/m ³	R 0.00	3500	R 0.00
Material, R/ton			
Magnesite	R 1,000	45	R 45.00

CO ₂	R 8,000	20	R 160.00
Electricity, R/kWh			
Pump (0.75 kW)	R 1.41-2.21	16.5	R 24.26
Agitator (3 kW)	R 1.41-2.21	3	R 80.85
	R 105		

On average, renewable energy technologies are more expensive than the conventional 528 technology on an levelised cost of electricity (LCOE) basis (Jahed et al., 2016). However, 529 530 these costs depend on the specific technology, power rating and various others parameters. For example, (Ross et al., 2016) estimated that the present LCOE of producing electricity 531 using a solar photovoltaic (PV) system in South Africa ranges from R0.915 to R2.07 per 532 533 kWh. This cost is sensitive to changes in the discount rate, the level of insolation at the location where the panels will be placed, the initial cost of the system and the efficiency of 534 the panel (Ross et al., 2016). Overall, this cost is comparable or cheaper (see Table 3) than 535 the cost of purchasing electricity from ESKOM, the state owned enterprise that generates 536 approximately 95% of the electricity used in South Africa (Ross et al., 2016). Therefore, it is 537 538 inferred that solar energy could be an economically feasible electricity source for AMD in 539 South Africa. Also, the current cost of electricity from solar PV systems suggest that the AMD treatment system could viably operate off-grid, with the addition of a power bank (e.g. 540 541 see (Foteinis et al., 2018)), since it is estimated that low to no additional costs would be incurred per treated m³ of AMD. In this case, however, the total electricity cost would have to 542 be paid upfront, i.e. the CAPEX would be higher, but operating costs would be minimized. 543

544 8. Conclusions

545 The environmental sustainability of a typical AMD treatment method was examined 546 by means of the life cycle assessment (LCA) methodology. Actual life cycle inventory (LCI)

547 data were directly sourced from a semi-industrial AMD system, treating real effluent collected from a coal mine in Mpumalanga Province, South Africa. Economic aspects were 548 also discussed. AMD is a common problem at mine sites, primarily at abandoned ones, while 549 550 in water scarce countries, such as South Africa, health and socioeconomic concerns render AMD sustainable treatment imperative. If left untreated AMD can cause detrimental effects 551 to the environment and living organisms, including humans, and impose on development, 552 health, access to clean water, thus stressing social sustainability. The proposed treatment 553 system can address, at least partly, the growing problem of AMD pollution and improve 554 community resilience at local (the system can operate off-grid in remote areas) and national 555 level. It can also support other important functions, such as agriculture, thus improving 556 economic sustainability. The systems has an overall low levelized cost per m³ AMD, i.e. 557 R112.78/m³ (ϵ 7.60/m³ or \$9.35/m³), which is expected to reduce at industrial level, where 558 economies of scales exist, and if gaseous CO2 can be sourced by a nearby source, e.g. flue 559 gas. 560

The system was found to have an overall high environmental footprint (29.6 kg CO_{2e} or 2.96 Pt per treated m³ AMD), which is mainly attributed to electricity consumption from South Africa's fossil-fuel depended energy mix and liquid CO_2 consumption. The introduction of renewable energy, i.e. solar energy, and directly sourcing gaseous CO_2 from other production process, e.g. flue gas, can axe the total environmental footprint by up to 81%. AMD sludge valorisation, i.e. mineral recovery, can also be used as a strategy to mitigate AMD's environmental footprint, but more research is needed.

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