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

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15 **Abstract**

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

35

36

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

39 **1. Introduction**

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

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

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

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

89

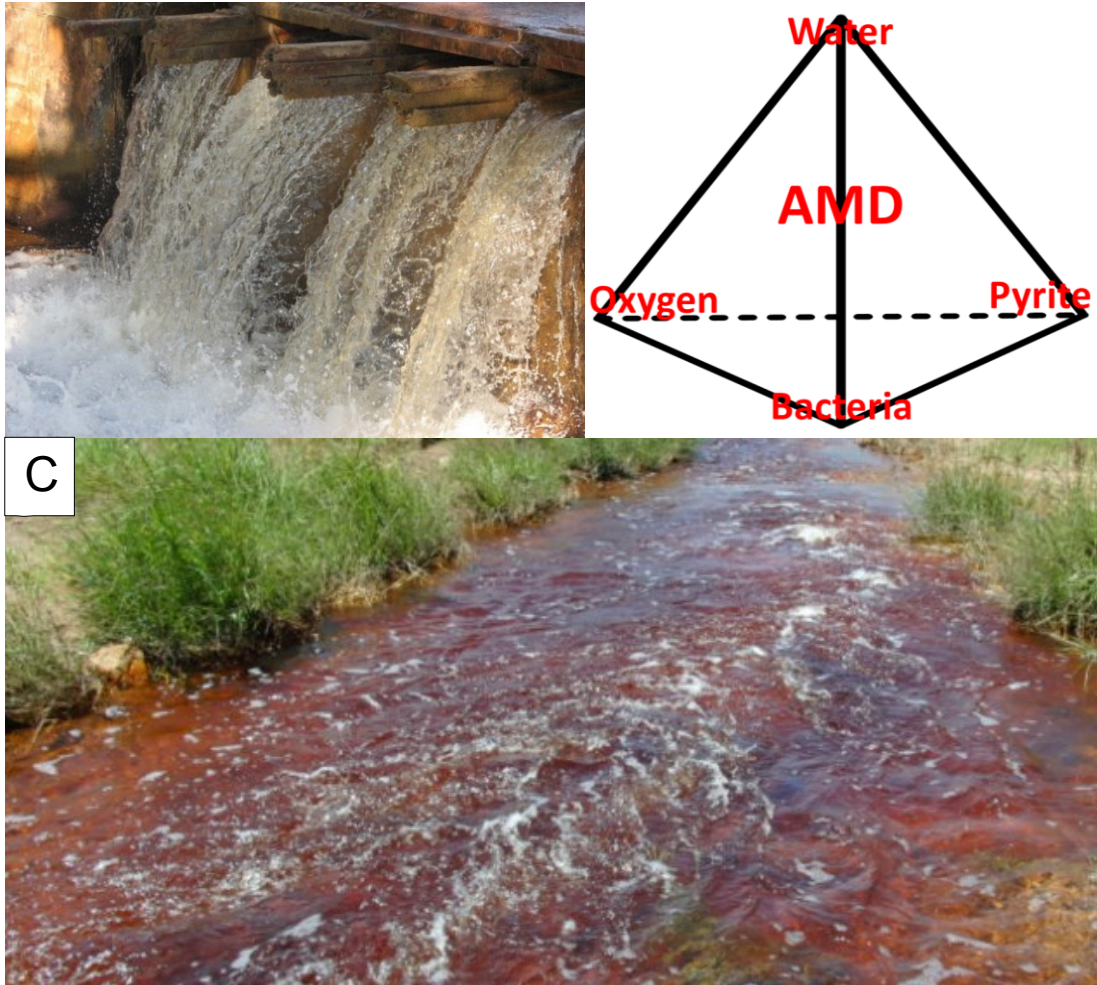
90 **2. The case study**

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

99

A

B



100

C

101

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

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

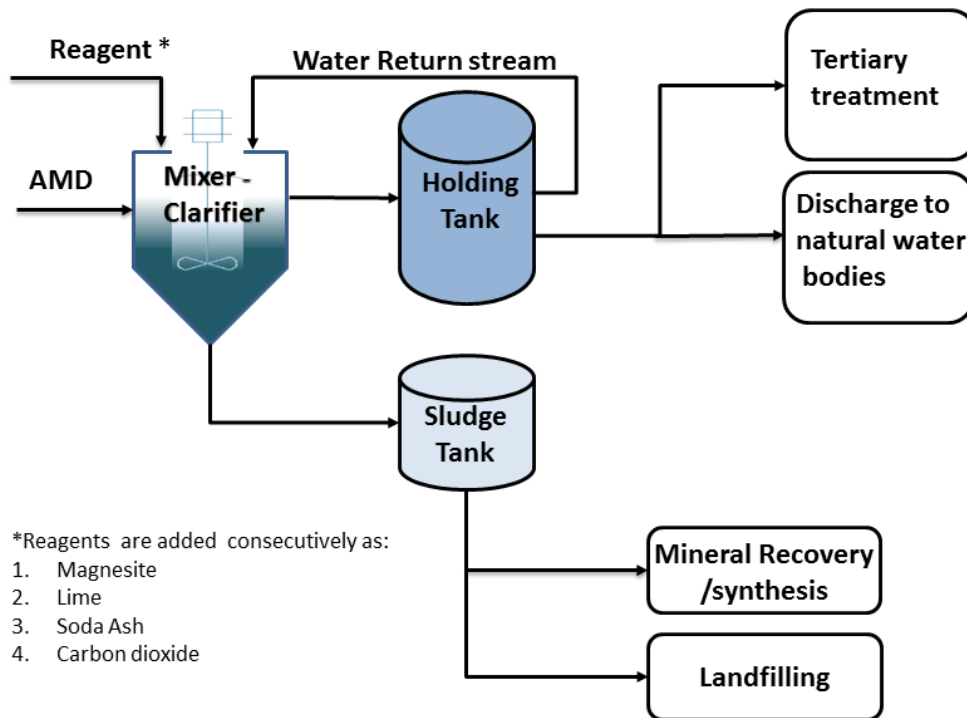
108 Table 1: AMD physicochemical characteristics, before and after treatment (data taken from (Masindi, 2017)).

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	≤ 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

109

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



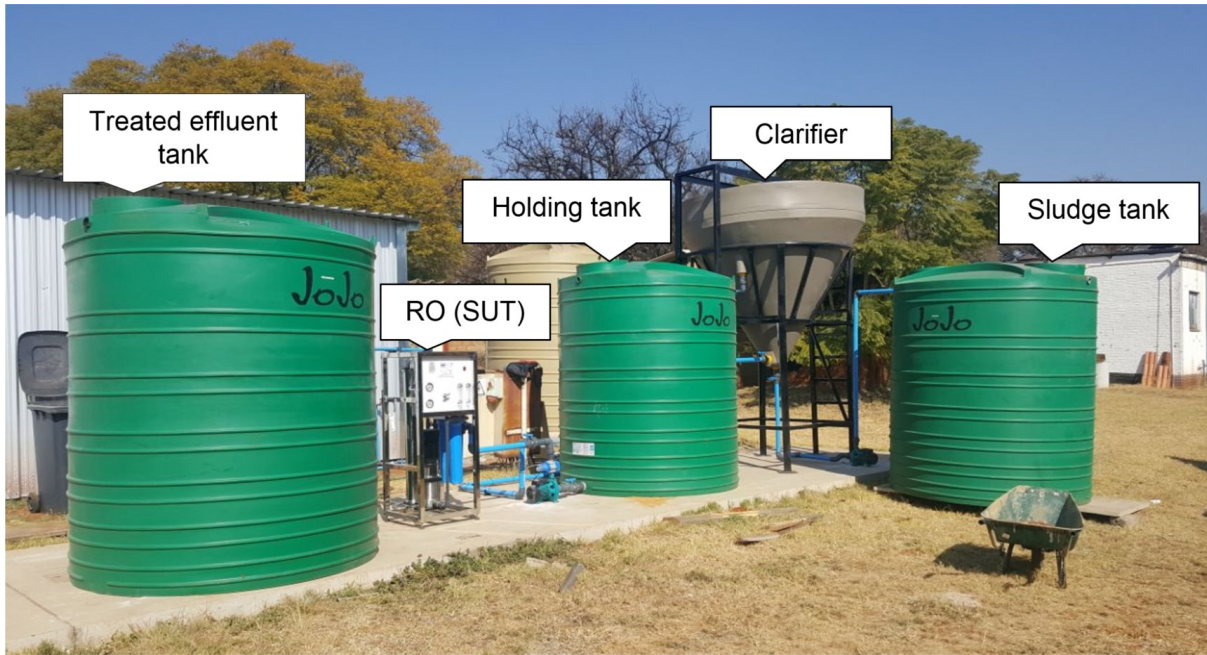
121

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

124

125 The aforementioned system was designed, constructed, and commissioned at the
 126 premises of CSIR Pretoria campus, South Africa, where it operates at semi-industrial scale
 127 and is able to effectively treat 3.5 m³ of AMD daily (Figure 3).

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



134
 135 Figure 3: The semi-industrial AMD treatment unit in operation at the premises of CSIR Pretoria campus, South
 136 Africa.
 137

138 A detailed discussion regarding the four process steps under study can be found in
 139 Masindi (2017). For the magnesite treatment stage 10 kg of magnesite per m^3 of AMD are
 140 added to the clarifier, shown in Figure 2 and Figure 3. The mixture is agitated for 60 minute
 141 and then it is left for another 60 minute to settle, where solid precipitates are gravity settled.
 142 Then, the magnesite treated AMD is transferred to a holding tank and the sludge is
 143 transferred to a separate tank (sludge tank). In this stage Fe-species can be recovered from the
 144 sludge. For the limestone treatment stage, the magnesite-treated effluent is recycled back to
 145 the clarifier and 10 kg/m^3 -AMD of limestone are added into it. The mixture is then agitated
 146 for 60 minute and is left for another 60 minute unstirred, to allow solid precipitates to settle.
 147 The magnesite/limestone treated AMD and the sludge are transferred back to the holding and
 148 the sludge tank, respectively. In this stage, residual Ca (gypsum) and Mg (brucite) can be
 149 recovered from the sludge. For the soda-ash treatment stage, the effluent is recycled back to
 150 the clarifier and 4 kg/m^3 -AMD of soda ash are added, following the same procedure (i.e. 60
 151 minute agitation, 60 minute settling and sludge removal). Finally, in the CO_2 bubbling stage

152 the magnesite/limestone/soda-ash treated AMD is recycled back to the clarifier, where CO₂ is
153 bubbled (45 L/min) until the pH reaches 7.5. Similarly, the effluent is left for 60 minute to
154 settle and then is transferred to the treated effluent tank, while the sludge is collected to the
155 sludge tank (Figure 2 and Figure 3).

156

157 **3. Goal and scope and system boundaries**

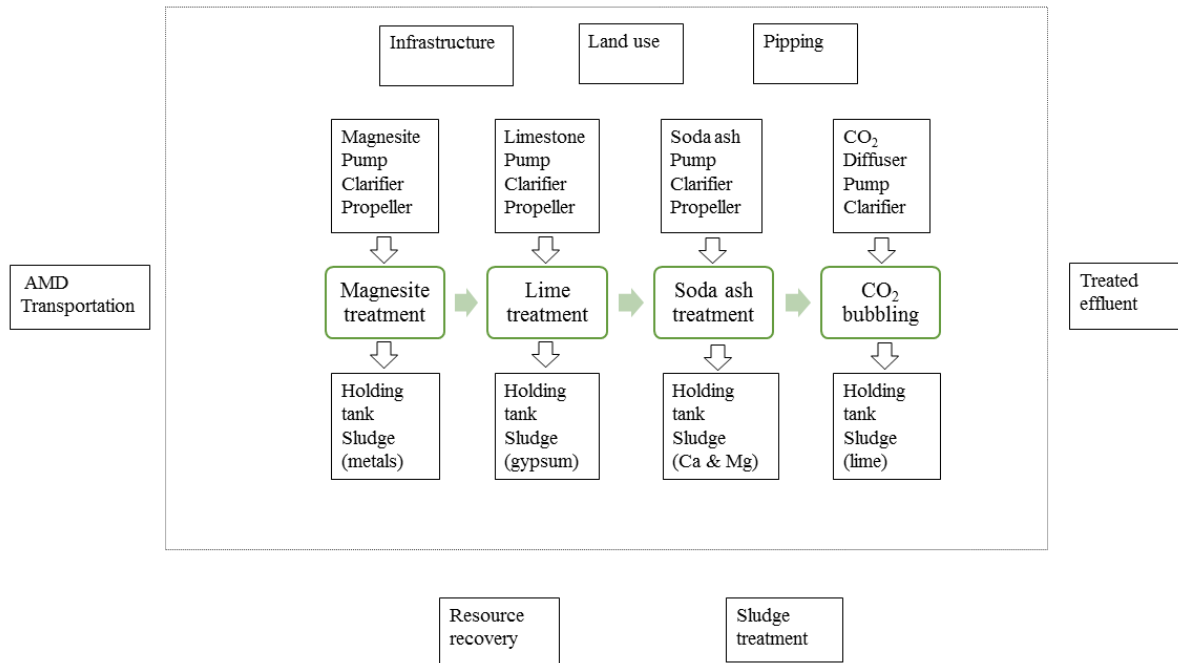
158 The purpose of this assessment is to identify the environmental performance and the
159 main environmental hotspots of a robust AMD treatment method, as well as identify
160 environmental saving avenues through scenario/sensitivity analyses. To this end, primary LCI
161 data were collected from a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

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

172 The environmental modelling was carried out using the SimaPro 8 software package,
173 based on the LCA methodology as set in ISO 14040 and 14044 (ISO, 2006a; ISO, 2006b).
174 The time-related coverage of this work refers to present, i.e. 2018, while its geographical
175 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
177 cycle impact assessment (LCIA) method were used, the latter by employing the Hierarchist
178 perspective. It should be noted that there might be some limitations of applying the ReCiPe
179 method, since this was first developed in and for the European context (Adiansyah et al.,
180 2017a). Nevertheless, the updated version of ReCiPe, i.e. ReCiPe2016, which was used in
181 this work, provides characterisation factors that are representative for the global instead of the
182 European scale, while maintaining the possibility for a number of impact categories to be
183 adapted at a country or continental scale (Huijbregts et al., 2017).

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

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As mentioned above, primary LCI data for the system under study (i.e. semi-industrial AMD treatment plant) were collected from its construction and operation phase in CSIR Pretoria campus, South Africa. Table 2 summarizes the LCI that were used in this work, normalized per functional unit, i.e. the effective treatment of 1 m³ of AMD. It has to be noted that the clarifier and tanks under study, as well as the pumps and the propeller were not identified in SimaPro's proprietary LCI databases, and thus literature data were used as 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 rated power output (0.75 kW). It has to be noted that magnesite, limestone and soda ash are 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 takes 30 minute to move the effluent from the clarifier to the holding tank and another 30

215 minute to move it back to the clarifier. The sludge is much thicker and therefore it takes 10
 216 minute to transfer it from the clarifier to the sludge tank. For the propeller, LCI data were
 217 taken from (Sulzer Ltd, 2013) and re-scaled to fit the desirable rated output (3 kW). In each
 218 of the above treatment step mixing (propeller) lasts for 60 minute. Moreover, according to
 219 their manufacturer the clarifier and the tanks under study are made from linear low-density
 220 polyethylene (LLDPE), while their life span is at least 10 years (JoJo Tanks Ltd, 2017).
 221 Therefore, literature data for LLDPE tanks (Shah et al., 2016) were used, while it was
 222 assumed that the tanks and the clarifier will be replaced once during the pilot unit lifespan of
 223 20 years. For the diffuser used for CO₂ bundling LCI data were taken from the literature
 224 (Ioannou-Ttofa et al., 2016), assuming that its main material is Polyvinyl chloride (PVC).
 225 Piping comprised high-density polyethylene (HDPE) pipes (~ 20 m total length) and was
 226 taken from Ecoinvent database. The chemicals that are used to drive the process were taken
 227 directly from Ecoinvent, apart from magnesite which LCI was taken from the literature
 228 (Cherubini et al., 2008). Moreover, a mean transportation distance of 40 km was ascribed to
 229 all construction materials and system inputs, except from AMD transportation which is
 230 outside of the system boundaries. Finally, it was assumed that 40 m² of industrial land will be
 231 occupied throughout the treatment plant life span.

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

Process	Main parts - chemical reagents	Value	LCI data reference
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)

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
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
CO₂ 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 m ³ /m ³	-
Fe (taken as iron sulfate)		2 kg/m ³	Ecoinvent 3.3
Gypsum		5 kg/m ³	Ecoinvent 3.3
Brucite (taken as magnesium oxide)		1 kg/m ³	Ecoinvent 3.3
Limestone		3 kg/m ³	Ecoinvent 3.3

233 5. Life cycle impact assessment (LCIA)

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

253 At midpoint level ReCiPe comprises the following impact categories: climate change
254 (CC), ozone depletion (OD), terrestrial acidification (TA), freshwater eutrophication (FE),
255 marine eutrophication (ME), human toxicity (HT), photochemical oxidant formation (POF),
256 particulate matter formation (PMF), terrestrial ecotoxicity (TET), freshwater ecotoxicity
257 (FET), marine ecotoxicity (MET), ionising radiation (IR), agricultural land occupation
258 (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
260 endpoint, ReCiPe converts and aggregates most of the midpoint impact categories into the
261 following damage categories: i) damage to human health, covering climate change, ozone
262 depletion, toxicity, and human health associated with PM10 and ozone; ii) damage to
263 ecosystem diversity, covering climate change, acidification, toxicity, and land-use; and iii)
264 damage to resource availability, covering mineral resource depletion, and fossil fuel depletion
265 (Adiansyah et al., 2017a; Foteinis et al., 2018). In order to obtain a holistic understanding of
266 the environmental performance of the AMD treatment system under study, both midpoint and
267 endpoint approaches were used. Moreover, the Hierarchist perspective (H) was employed by
268 using the normalisation values of the world and average weighting (i.e. world ReCiPe H/A).

269 **6. Economic analysis**

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

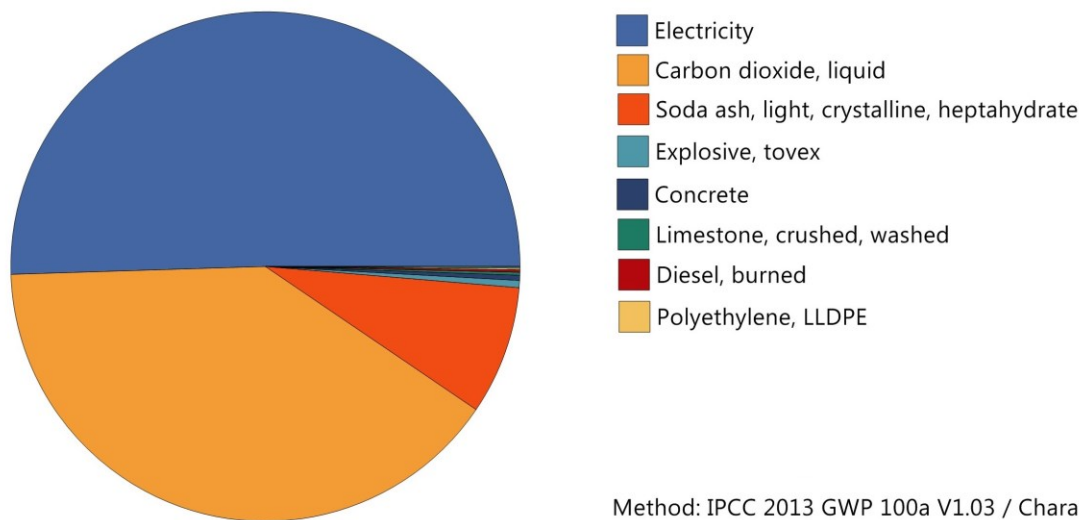
278 It has to be noted that the analysis was focused on accounting for the operating cost,
279 which represent the majority of financial input of the AMD treatment unit, and only under the
280 present conditions, i.e. not accounting for inflation. This analysis intends to act as a screening
281 tool to assess the economic viability of the system under study, rather than specify possible
282 options and provide information about costs and benefits in present monetary value such as in

283 benefit-cost analysis (BCA). For example, treating AMD reduces sulfate waterborne
284 emissions and therefore minimizes the environmental impact on (eco)toxicity and on
285 freshwater and marine eutrophication. In addition, water and land conservation and GHG
286 reduction could be achieved, which lead to monetary benefits (Adiansyah et al., 2017b).
287 Nonetheless, calculating the monetary benefits of AMD treatment is beyond the goals and
288 scope of this work, and could be addressed in future studies.

289 **7. Results and discussion**

290 **7.1 Carbon footprint**

291 Total carbon equivalent ($\text{CO}_{2\text{eq}}$) emissions were estimated using the IPCC 2013 LCIA
292 method for 100 years timeframe and it was found that the effective treatment of 1 m^3 of
293 AMD emits $29.6 \text{ kg CO}_{2\text{eq}}$. Regarding the contribution of each process step, it was found that
294 CO_2 bubbling had the highest score ($13 \text{ kg CO}_{2\text{eq}}$), followed by soda ash treatment (6.9 kg
295 $\text{CO}_{2\text{eq}}$), magnesite treatment ($5.04 \text{ kg CO}_{2\text{eq}}$) and limestone treatment ($4.54 \text{ kg CO}_{2\text{eq}}$). The
296 main environmental hotspot was identified as electricity consumption ($14.5 \text{ kg CO}_{2\text{eq}}$),
297 followed by the liquid CO_2 input for the bubbling process ($11.8 \text{ kg CO}_{2\text{eq}}$). Soda ash and
298 magnesite, as materials, had a lower carbon footprint, 2.41 and $0.558 \text{ kg CO}_{2\text{eq}}$ respectively.
299 The remaining inputs (e.g. Tovex explosive for magnesite and limestone mining and concrete
300 for the system base) had a very low to negligible score. Therefore, the main contributors to
301 the total carbon footprint are electricity consumption from South Africa's fossil fuel-
302 depended energy mix (49.2%), followed by the liquid CO_2 input (40%) and soda ash
303 (8.16%), as shown in Figure 5.



304

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

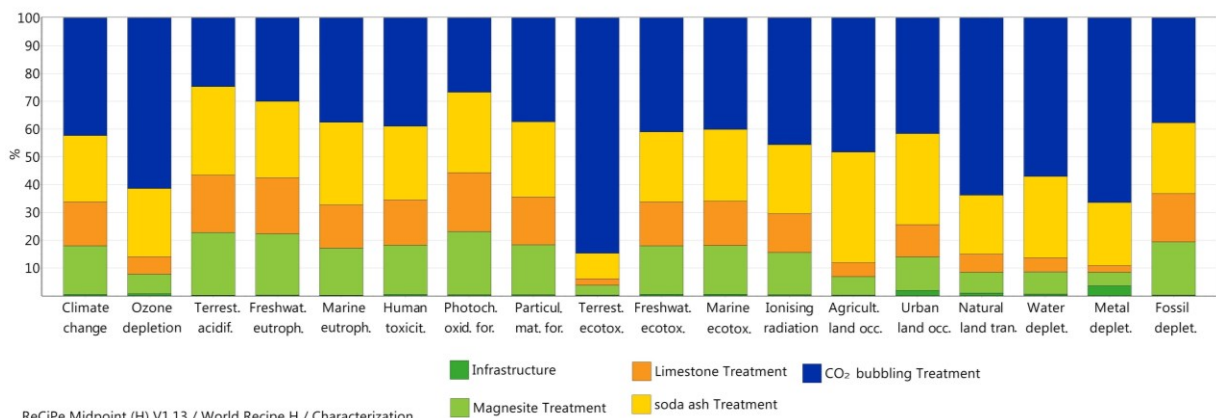
306 7.2 ReCiPe LCIA method

307 7.2.1 ReCiPe results at midpoint

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

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

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

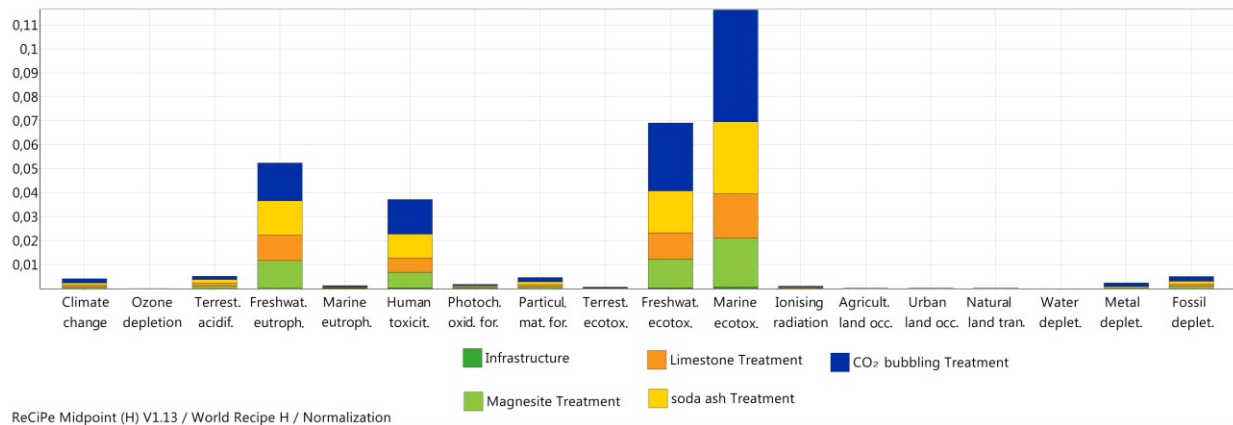


330
 331 Figure 6: Contribution of the main process steps to ReCiPe's midpoint impact categories for the treatment of 1
 332 m³ of AMD.

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

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

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



361

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

364 The high scores in the (eco)toxicity (MET, FET and HT) and eutrophication (FE)
 365 impact categories are attributed to the mining of the chemical reagents and of the fossil fuels
 366 required for electricity generation (South Africa's energy mix is dominated by fossil fuels,
 367 mainly coal (Papadaki et al., 2017)). It has to be noted that CO₂ harvesting and purification
 368 require large amounts of energy (Althaus et al., 2007), in this case electricity from fossil
 369 fuels. Therefore, the chemical reagents and particularly fossil fuel mining exposes previously
 370 buried coal minerals to both oxygen and water, thus releasing, through waterborne emissions,
 371 mine-derived sulfate salts. Sulfate emissions could disrupt water balance and ion exchange
 372 processes, thus causing aquatic organisms to live under stress or even death (Zhao et al.,
 373 2017). Moreover, magnesite, limestone and fossil fuel extraction and transportation, as well
 374 as soda ash production and fossil fuel refining and combustion release toxic materials, such
 375 as heavy metals, sulphurous compounds and polycyclic aromatic hydrocarbons (PAHs) to the
 376 environment, thus also affecting the (eco)toxicity impact categories (Ioannou-Ttofa et al.,
 377 2016).

378 As far as the FE impact category is concerned, mining activity is an increasingly
 379 important stressor for freshwater ecosystems, since sulfate can co-vary with other
 380 environmental parameters, such as nitrogen and phosphorus, and impact aquatic organisms.

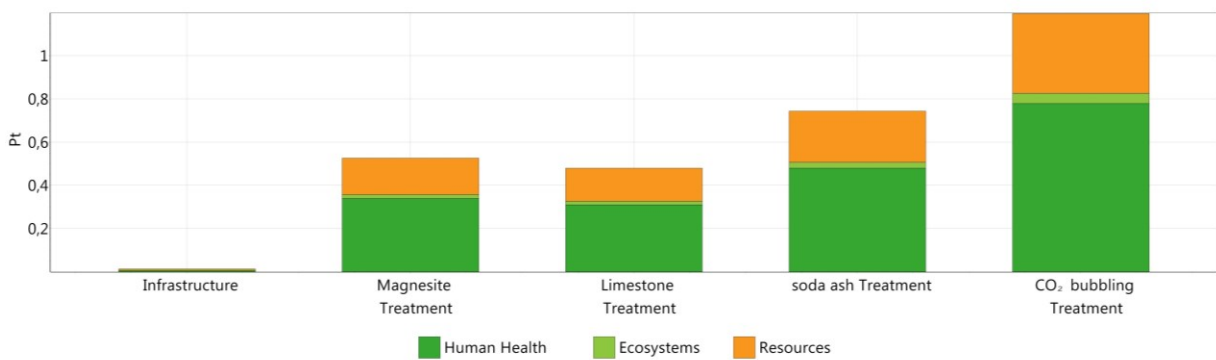
381 The reason is that the introduction of sulfates in natural water matrices, through mining
382 activities, can increase the availability of nitrogen and phosphorus through internal
383 eutrophication (Zhao et al., 2017). Also, fossil fuel combustion lead to nitrogen oxides
384 emissions which impact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

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

394 **7.2.2 ReCiPe results at endpoint**

395 Figure 8 shows ReCiPe's weighted results at endpoint level (Hierarchist perspective
396 using the normalisation values of the world with the average weighting set). Weighting is an
397 optional step in the LCIA, after normalisation, where results are multiplied by weighting
398 factors corresponding to each impact category. Weighted results can be then aggregated into
399 a single score, in order to access the total environmental footprint of the effective treatment of
400 1 m³ of AMD. It was found that the weighted damage to human health exhibits the highest
401 score (1.92 Pt), followed by the damage to resources availability (0.932 Pt), while the damage
402 to ecosystem diversity has the lower score (0.109 Pt). Therefore, the aggregated score is
403 found to be 2.96 Pt per m³ of AMD. The high scores of the first two damage categories are
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
 406 aggregated score is CO₂ bubbling (1.19 Pt), followed by soda ash (743 mPt), magnesite (526
 407 mPt), and limestone treatment (479 mPt). Moreover, the main environmental hotspot was
 408 identified to be electricity from South Africa’s fossil-fuel depended energy mix (1.51 Pt or
 409 51.1% of the total environmental footprint). From it, 35.5% or 1.05 Pt is attributed to
 410 stirring/propelling and the remaining 15.8% or 0.468 Pt to water pumping. The second
 411 environmental hotspot was identified as the liquid CO₂ input (1.08 Pt or 36.4%), followed by
 412 soda ash, as a material (276 mPt or 9.33%). The remaining chemical reagents had a lower
 413 contribution, i.e. magnesite 58.7 mPt or 1.99%, and limestone 11.7 mPt or ~0.4%. Due to
 414 their high life span, the CO₂ diffuser, the propeller and the pumps had a miniscule
 415 contribution (<0.1%), while the tanks, including the clarifier, contribute 3.72 mPt or 0.126%
 416 in total. Finally, the infrastructure contributes 0.44% on the total environmental footprint,
 417 with the majority of those impacts attributed to the reinforced concrete used to construct its
 418 base (i.e. pipping and land use had a miniscule contribution).



419 Method: ReCiPe Endpoint (H) V1.13 / World ReCiPe H/A / Single score

420 Figure 8: Contribution of the main process steps on the aggregated total environmental footprint of the process.

421

422

423

424

425 **7.3 Sensitivity/scenario analyses**

426 **7.3.1 Source and form of CO₂**

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

439 A scenario dealing with directly using gaseous CO₂ that derives as produced as a by-
440 product, e.g. flue gas, was examined. If environmental burden-free gaseous CO₂ is fed into
441 the bubbling stage, then the environmental footprint is reduced by ~36%, i.e. 1.88 Pt instead
442 of 2.96 Pt when using liquid CO₂.

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

444 The second environmental hotspot of the system was identified as the electricity
445 consumption from South Africa's fossil energy mix, which is attributed to the high share of
446 fossil fuels, mainly coal (Papadaki et al., 2017). Therefore, using an electricity mix solely
447 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)
449 systems), an abundant and readily available RES in South Africa, was examined. It was
450 found that when using solar energy the system's environmental sustainability is substantially
451 improved. Specifically, its total environmental footprint is reduced from 2.96 Pt per m³ to
452 1.64 Pt per m³. Therefore, the introduction of RES, in this case solar energy, can reduce the
453 total environmental footprint by ~45%. Moreover, a third and practically feasible scenario is
454 to combine the use of RES (i.e. solar energy) and gaseous CO₂ in the same AMD treatment
455 system. In this case the total environmental footprint of the system is minimized, from 2.96 Pt
456 per m³ in the initial scenario (i.e. current operating conditions) to 0.559 Pt per m³, and is
457 drastically reduced by ~81%, reaching an overall high environmental sustainability.

458 **7.3.3 Resource recovery from AMD sludge**

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

470 In order to extract the targeted resources from each sludge stream chemical reagents
471 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
473 (EcoTherm Economy) was used and the resources that can be extracted per m³ AMD are Al,
474 Ca, Fe, Mg and Mn. Here we examine the effect of one of the main input of the extraction
475 process, i.e. the sludge drying, to identify if sludge valorisation can mitigate the
476 environmental footprint of the examined AMD treatment process. Even though, the
477 extraction procedure is more complicated and requires further processing, we estimated that
478 the additional environmental burdens of the sludge valorisation are compensated by the fact
479 that here sludge drying was achieved by a laboratory oven (industrial ovens require much less
480 energy input). Future works could deal with the exact environmental performance of the most
481 promising recourses to be recovered.

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

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**

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

517 It has to be noted that if AMD is left untreated it could have large economic impacts,
518 since it could cause detrimental effects to the environment and living organisms, including
519 humans, and impose on development, health, access to clean water, thus stressing social
520 sustainability. The proposed treatment system can address, at least partly, the growing
521 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.

524

525 **Table 3:** Economic evaluation of the AMD treatment process capital (CAPEX) and operating expenditure
 526 (OPEX)

Input	Unit cost	Quantity	Total costs (Rand)
Initial capital expenditure (CAPEX) required for infrastructure			
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
Thread 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
Total CAPEX			R 198,721.00
Levelised CAPEX per m³			R 7.78
Operating expenditure (OPEX) required for chemical reagents and energy inputs			
AMD, R/m³	R 0.00	3500	R 0.00
Material, R/ton			
Magnesite	R 1,000	45	R 45.00
Lime	R 2,000	2.5	R 5.00
Soda Ash	R 3,500	15	R 52.50

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
	Total OPEX/m³		R 105

527

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

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
548 collected from a coal mine in Mpumalanga Province, South Africa. Economic aspects were
549 also discussed. AMD is a common problem at mine sites, primarily at abandoned ones, while
550 in water scarce countries, such as South Africa, health and socioeconomic concerns render
551 AMD sustainable treatment imperative. If left untreated AMD can cause detrimental effects
552 to the environment and living organisms, including humans, and impose on development,
553 health, access to clean water, thus stressing social sustainability. The proposed treatment
554 system can address, at least partly, the growing problem of AMD pollution and improve
555 community resilience at local (the system can operate off-grid in remote areas) and national
556 level. It can also support other important functions, such as agriculture, thus improving
557 economic sustainability. The systems has an overall low levelized cost per m^3 AMD, i.e.
558 $\text{R}112.78/\text{m}^3$ ($\text{€}7.60/\text{m}^3$ or $\text{\$}9.35/\text{m}^3$), which is expected to reduce at industrial level, where
559 economies of scales exist, and if gaseous CO_2 can be sourced by a nearby source, e.g. flue
560 gas.

561 The system was found to have an overall high environmental footprint ($29.6 \text{ kg CO}_{2\text{e}}$
562 or 2.96 Pt per treated m^3 AMD), which is mainly attributed to electricity consumption from
563 South Africa's fossil-fuel depended energy mix and liquid CO_2 consumption. The
564 introduction of renewable energy, i.e. solar energy, and directly sourcing gaseous CO_2 from
565 other production process, e.g. flue gas, can axe the total environmental footprint by up to
566 81%. AMD sludge valorisation, i.e. mineral recovery, can also be used as a strategy to
567 mitigate AMD's environmental footprint, but more research is needed.

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