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

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## Abstract

The environmental sustainability of acid mine drainage (AMD) treatment at semi-industrial scale is examined by means of the life cycle assessment (LCA) methodology. An integrated process which includes magnesite, lime, soda ash and $CO_2$ bubbling treatment was employed to effectively treat, at semi-industrial scale, AMD originating from a coal mine in South Africa. Economic aspects are also discussed. AMD is a growing problem of emerging concern that cause detrimental effects to the environment and living organisms, including humans, and impose on development, health, access to clean water, thus also affect economic growth and cause social instability. Therefore, sustainable and cost effective treatment methods are required. A life cycle cost analysis (LCCA) revealed the viability of the system, since the levelized cost of AMD treatment can be as low as R112.78/$m^3$ (€7.60/$m^3$ or \$9.35/$m^3$). Moreover, due to its versatility, the system can be used both at remote locales, at stand-alone mode (e.g. using solar energy), or can treat AMD at industrial scale, thus substantially improving community resilience at local and national level. In terms of environmental sustainability, 1.18Pt or 29.6 kg $CO_{2eq}$ are emitted per treated $m^3$ AMD or its environmental footprint amount to 2.96 Pt/$m^3$. South Africa's fossil-fuel depended energy mix and liquid $CO_2$ consumption were the main environmental hotspots. The total environmental footprint is reduced by 45% and 36% by using solar energy and gaseous $CO_2$, respectively. Finally, AMD sludge valorisation, i.e. mineral recovery, can reduce the total environmental footprint by up to 12%.

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

## 1. Introduction

Access to clean water is a basic human right, and one of the cornerstones of environmental protection in Europe (Eurostat, 2017). Water is critical for sustaining ecosystems, plays a fundamental role in the climate regulation cycle and is also the primary requirement for human survival and socioeconomic development (Eurostat, 2017; Naidoo, 2016). Even though clean water access is taken for granted in the developed world this is not the case for developing countries, which are struggling to keep economic growth but often at the expense of environmental protection and water quality. South Africa is a developing 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, also known as acid rock drainage (ARD), is a common problem at mine sites, primarily at 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 produced from the bio-hydro-geochemical weathering of pyrite and other reactive sulphide bearing minerals, when exposed to oxidising conditions (Masindi et al., 2017). AMD emanating from active or abandoned mines and from mine wastes are often net acidic. These 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) and metalloids (Johnson and Hallberg, 2005). South Africa has a long history in mining and 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 Geoscience, 2010; Naidoo, 2016).

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 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 as magnesite, periclase, brucite, lime, hydrated lime, and limestone, to sustain the process), ii) 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 combination of both) (Johnson and Hallberg, 2005; Masindi, 2017). The most widespread method for AMD neutralization is active treatment, involving addition of an alkaline material (chemical-neutralising agent such as magnesite, lime, calcium carbonate, sodium carbonate, 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 oxidising agent are also required) and cause many of the metals present in solution to 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 (Potgieter-Vermaak et al., 2006).

Even though treatment efficiencies of the available AMD methods are well-established and explored (e.g. (Johnson and Hallberg, 2005; Potgieter-Vermaak et al., 2006), 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., 2014; Tuazon and Corder, 2008). Therefore, herein a full life cycle assessment (LCA) of a 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 environmental sustainability of a typical AMD treatment process, identify environmental hotspots and identify avenues to improve its environmental sustainability, such as resource extraction from AMD sludge. Also, economic and social aspects regarding the sustainability of the treatment system are discussed.

## 2. The case study

Acid mine drainage (AMD) was collected from a coal mine in Mpumalanga Province, South Africa, and was transferred to the premises of the Council for Scientific and Industrial Research (CSIR), Pretoria campus, South Africa, for treatment. The raw mine water was 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 components that contribute to this process. Co-existence of raw mine water, atmospheric oxygen, sulphide minerals (as a source of iron) and waterborne bacteria (to accelerate the reactions) can lead to the production of AMD (Pondja, 2017).

A

B

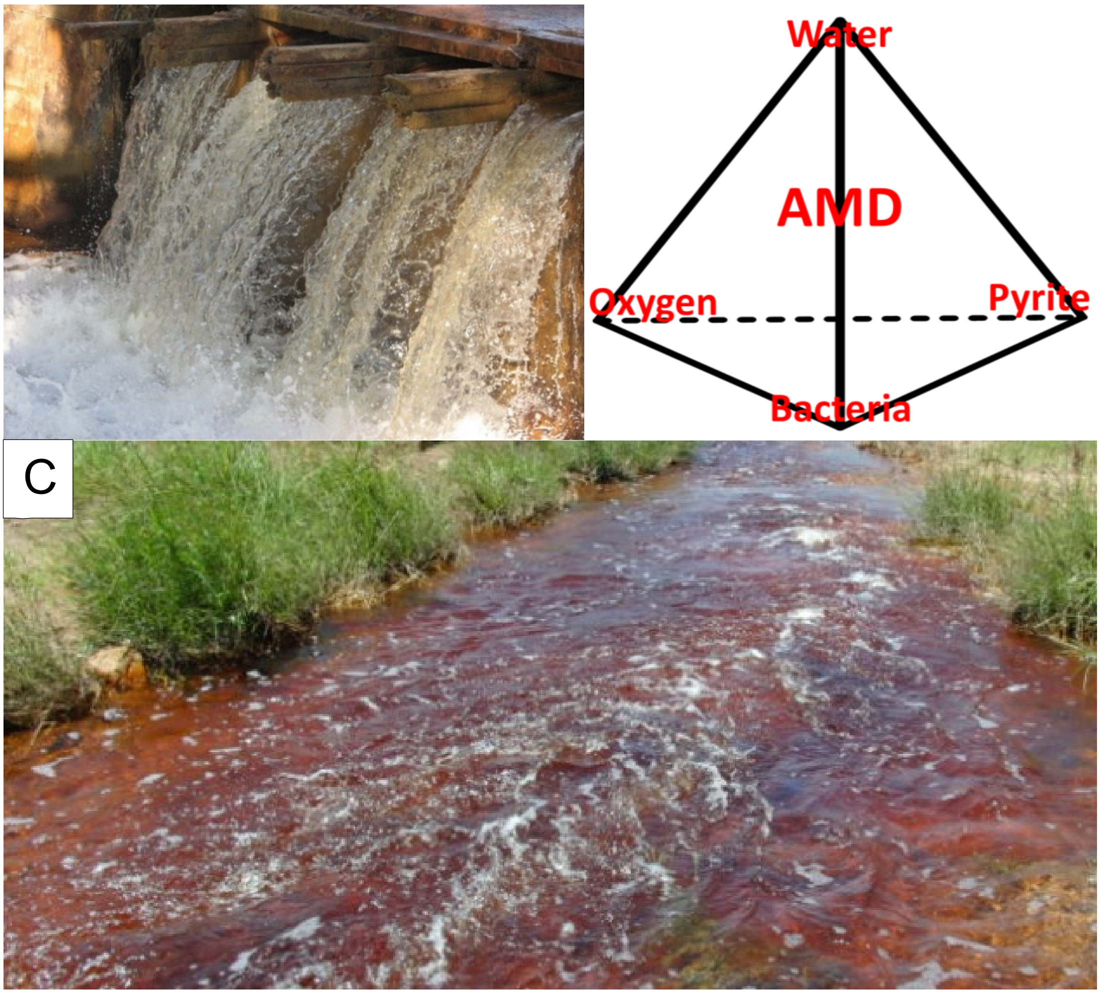


Figure 1. AMD effluent coming out of an underground pit, when it is initially colourless (A) and after being oxidised by atmospheric air in the presence of sulphide minerals and waterborne bacteria (B), gradually turns to red (C).

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).

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 |

As shown in Figure 2 the AMD treatment system comprises the following four discrete process steps: (1) neutralization of AMD and partial removal of sulphates achieved by using calcined cryptocrystalline magnesite (magnesite treatment); (2) addition of 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) $CO_2$ bubbling to correct the pH to 7.5 and recover limestone ($CO_2$ bubbling). The main products of this treatment process comprise the treated AMD effluent and the produced sludge. The latter is typically discarded for landfilling, but it can be also valorized as will be discussed in the sensitivity analyses section. As shown in Table 1, the system is capable of providing a 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).

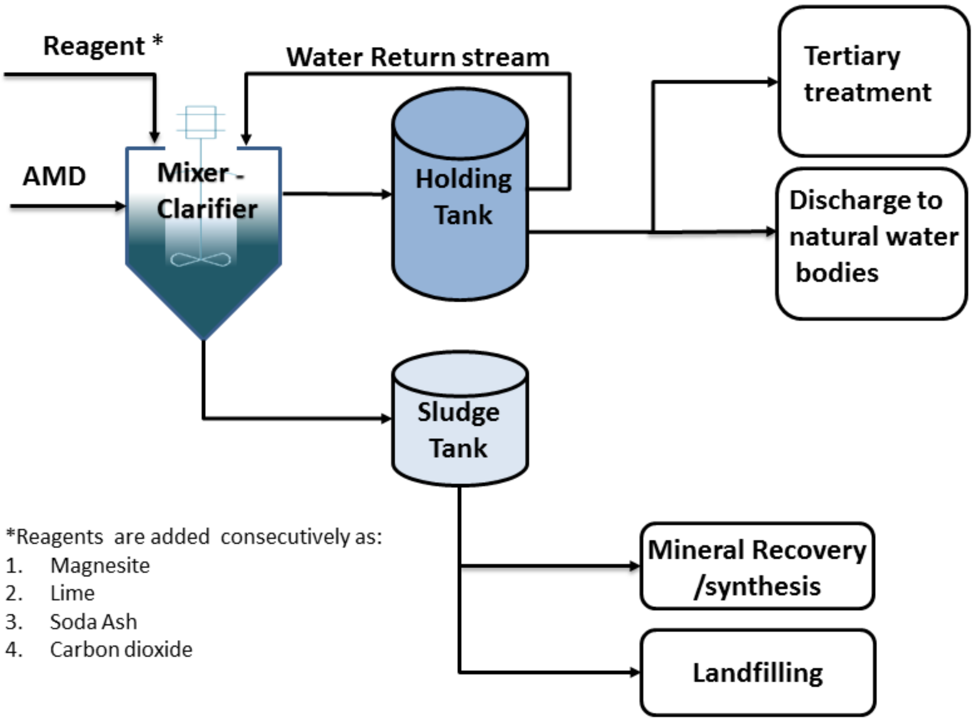


Figure 2: Flow diagram of the semi-industrial AMD treatment unit under study and possible scenarios for the disposal/treatment of the treated effluent and the AMD sludge.

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.

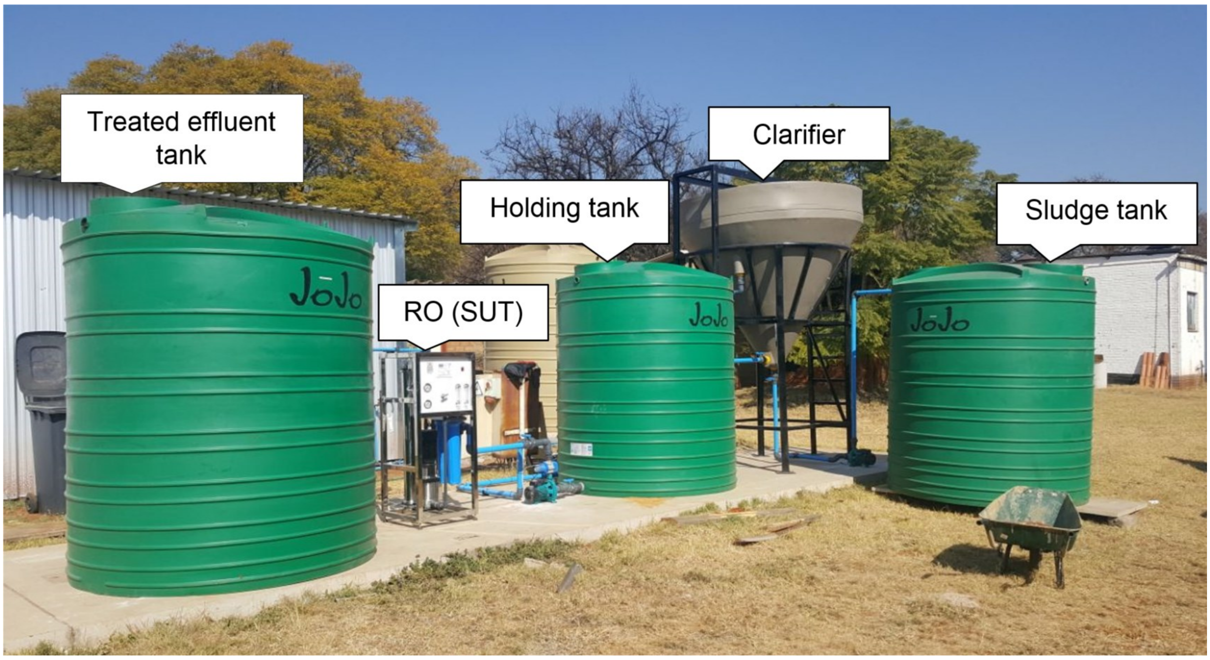


Figure 3: The semi-industrial AMD treatment unit in operation at the premises of CSIR Pretoria campus, South Africa.

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

## 3. Goal and scope and system boundaries

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

In order to quantify the environmental performance of the system 1 $m^3$ of effluent generated by the AMD reactor was set as the functional unit. Therefore, input and output data were normalized per $m^3$ 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 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 perspective. It should be noted that there might be some limitations of applying the ReCiPe method, since this was first developed in and for the European context (Adiansyah et al., 2017a). Nevertheless, the updated version of ReCiPe, i.e. ReCiPe2016, which was used in this work, provides characterisation factors that are representative for the global instead of the European scale, while maintaining the possibility for a number of impact categories to be adapted at a country or continental scale (Huijbregts et al., 2017).

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 LCA are presented (ISO, 2006b). All four AMD treatment steps, along with their main inputs 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. (Foteinis et al., 2018; Ioannou-Ttofa et al., 2016; Ioannou-Ttofa et al., 2017)). AMD transportation from Mpumalanga coalmine to the semi-industrial treatment plant, i.e. CSIR Pretoria campus premises, South Africa, is external to the system boundaries, since future 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 reason is that depending on the final use, e.g. disposal in natural water bodies, irrigation, or 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 required, i.e. reinforced concrete slabs to accommodate the treatment systems, piping, as well as land use were into account separately, i.e. a sub-system for infrastructure was created.

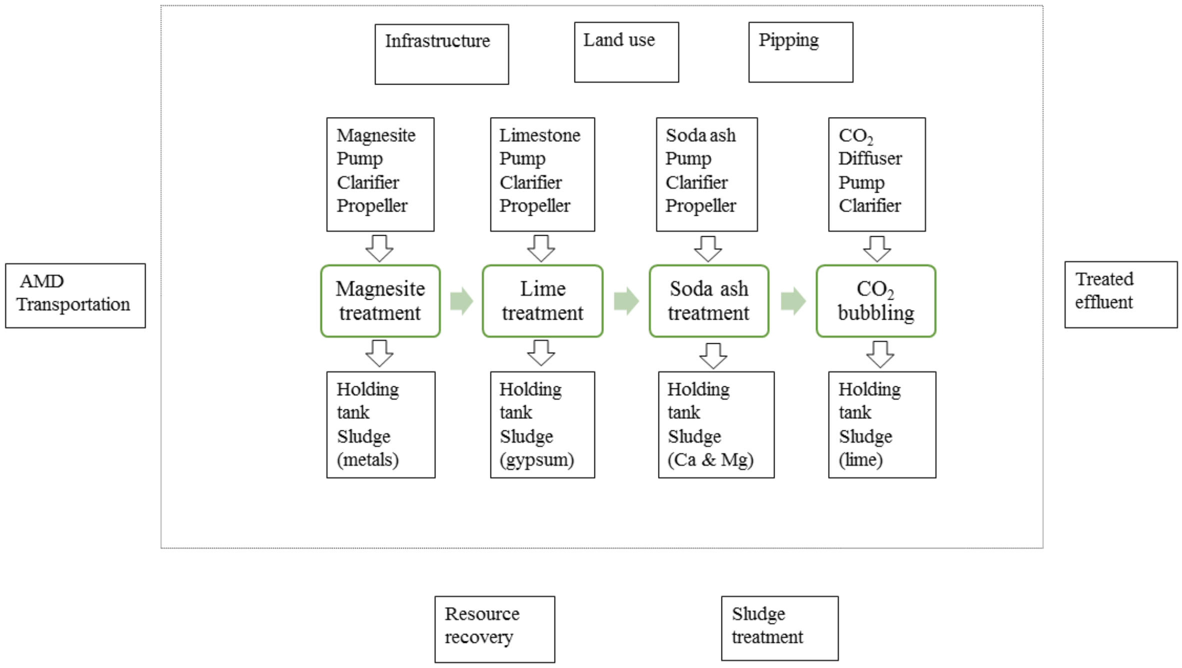


Figure 4: The system boundaries (shown with dashes) of the AMD treatment system.

## 4. Life cycle inventory (LCI)

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 $m^3$ 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 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 taken from (Sulzer Ltd, 2013) and re-scaled to fit the desirable rated output (3 kW). In each of the above treatment step mixing (propeller) lasts for 60 minute. Moreover, according to their manufacturer the clarifier and the tanks under study are made from linear low-density polyethylene (LLDPE), while their life span is at least 10 years (JoJo Tanks Ltd, 2017). Therefore, literature data for LLDPE tanks (Shah et al., 2016) were used, while it was assumed that the tanks and the clarifier will be replaced once during the pilot unit lifespan of 20 years. For the diffuser used for $CO_2$ bundling LCI data were taken from the literature (Ioannou-Ttofa et al., 2016), assuming that its main material is Polyvinyl chloride (PVC). Piping comprised high-density polyethylene (HDPE) pipes (~ 20 m total length) and was 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 (Cherubini et al., 2008). Moreover, a mean transportation distance of 40 km was ascribed to all construction materials and system inputs, except from AMD transportation which is outside of the system boundaries. Finally, it was assumed that 40 $m^2$ of industrial land will be occupied throughout the treatment plant life span.

Table 2: The LCI of the semi-industrial treatment plant for the treatment of 1 $m^3$ 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 |
| **$CO_2$ 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 $m^3/m^3$ | - |
| 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 |

## 5. Life cycle impact assessment (LCIA)

The life cycle impact assessment (LCIA) associates the collected LCI data with specific environmental impacts and damages and also attempts to understand those impacts/damages (ISO, 2006b). Here, a single-issue, i.e. IPCC 2013 for a timeframe of 100 years, and a multi-issue, i.e. ReCiPe, LCIA methods were used. IPCC 2013 compares processes based on $CO_2$ equivalent ($CO_{2eq}$) emissions, i.e. total greenhouse gas (GHG) emissions, used to measure Global Warming Potential (GWP), which is a standard indicator of environmental relevance. This is also included in ReCiPe's midpoint impact category “Climate Change”, but using a single-issue method allows a more direct dissemination of the results to the general public (Foteinis et al., 2018). ReCiPe can express results both at midpoint, where environmental impacts are examined earlier in the cause-effect chain, and endpoint level, where environmental impacts are examined at the end of the cause-effect 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 communicate to the general public. The endpoint or damage-oriented approach, can translate 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 gaps and assumptions stacking up along the cause-effect chain. Nonetheless, endpoint results are easier to communicate to decision- and policy-makers and the general public (Chatzisymeon et al., 2016).

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

(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 following damage categories: i) damage to human health, covering climate change, ozone 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) damage to resource availability, covering mineral resource depletion, and fossil fuel depletion (Adiansyah et al., 2017a; Foteinis et al., 2018). In order to obtain a holistic understanding of the environmental performance of the AMD treatment system under study, both midpoint and 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).

## 6. Economic analysis

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 requirements. Construction costs, maintenance costs, operational costs, occupancy costs, end-of-life costs and non-construction costs are usually included in LCCA (Zhong and Wu, 2015). In this study, an economic evaluation of the prototype AMD treatment system, based 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) 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.

## 7. Results and discussion

### 7.1 Carbon footprint

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

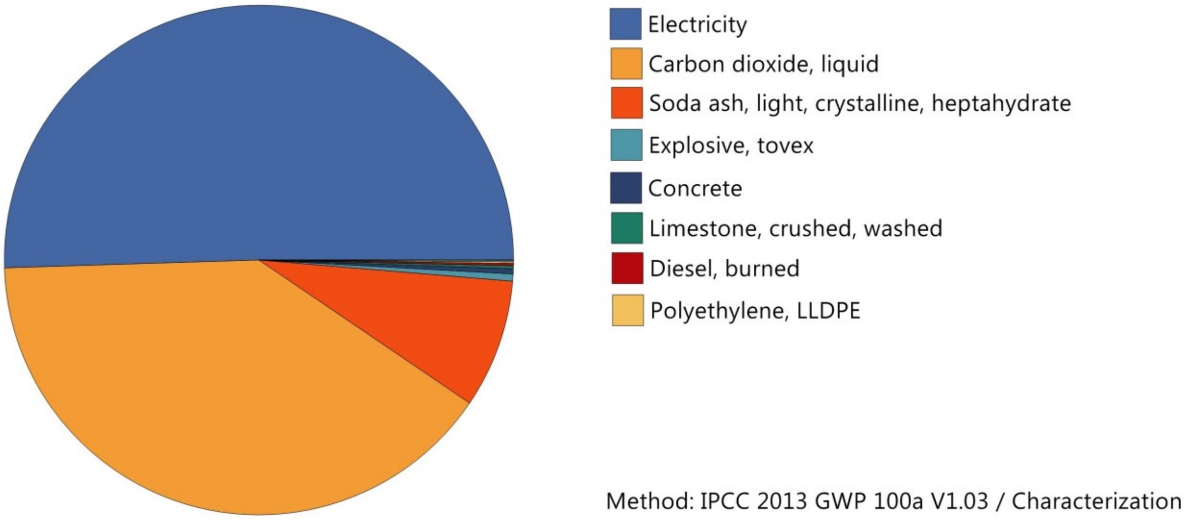


Figure 5: The main contributors to carbon emissions in kg $CO_{2eq}$ per $m^3$ of treated AMD.

## 7.2 ReCiPe LCIA method

### 7.2.1 ReCiPe results at midpoint

LCA findings are first presented at midpoint level, using ReCiPe's LCIA characterisation model and then results are normalized using the world's reference inventories. Figure 6 shows the contribution of all process sub-systems to each of the ReCiPe's 18 midpoint impact categories (characterisation). It is observed that the treatment step that has the highest contribution to all impact categories is the $CO_2$ bubbling stage. Magnesite, limestone, and soda ash treatment yielded comparable scores to most impact categories, while infrastructure (i.e. land use, concrete slab/foundations and piping) has a very low to miniscule contribution to all impact categories (Figure 6).

The high scores (from 25% - 85%) of the $CO_2$ bubbling stage are attributed to the high liquid $CO_2$ amounts required to drive the process, while electricity for water pumping required in this step had a much lower contribution. $CO_2$ 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 $CO_2$ is sourced directly from another production process it could be assumed that it will be free of any environmental burden (Althaus et al., 2007). On the other hand, liquid $CO_2$, which is the most commonly bought and sold form of $CO_2$, is associated with environmental burdens since energy and resources are required for $CO_2$ extraction and purification (Althaus et al., 2007). Here, liquid $CO_2$, originating from ammonia production, was assumed to be used in the bubbling stage. It should be mentioned that if gaseous $CO_2$ could be sourced for future industrial scale AMD treatment plants the total environmental footprint of the process could be further reduced. This scenario is examined in the sensitive analyses section.

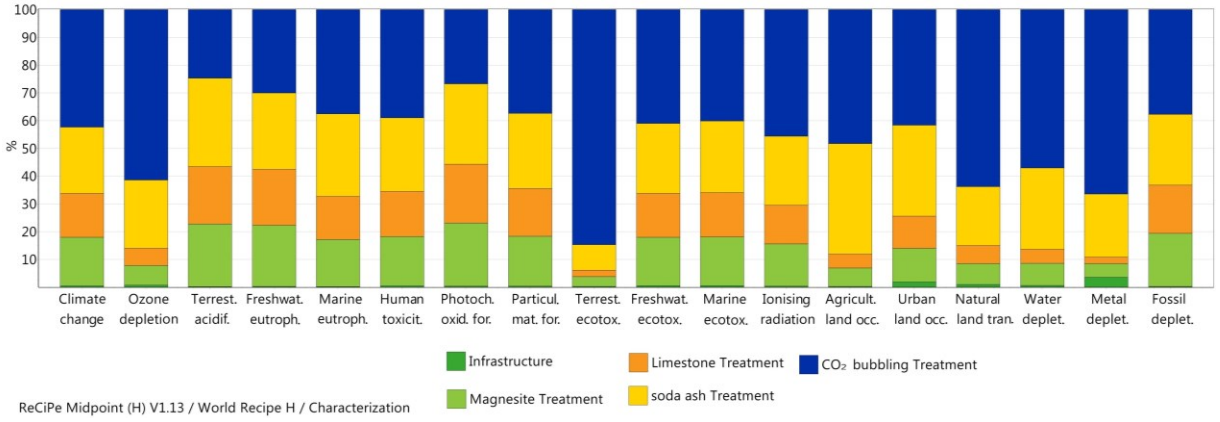


Figure 6: Contribution of the main process steps to ReCiPe's midpoint impact categories for the treatment of 1 $m^3$ of AMD.

Since the electricity inputs during the first three steps are the same, their differences in each midpoint impact categories is attributed to the chemical reagents, i.e. magnesite, limestone, and soda ash as materials, utilized in each step. The higher contribution (ranging between 9% - 32%) of the soda ash step is attributed to the sodium carbonate, i.e. soda ash, input require to drive this process step. Soda ash is mainly produced by the Solvay process, also called ammonia soda process (Mahida Prashantsinh et al., 2015), and hence soda ash (light grade) manufactured from this process was taken into account here. The Solvay process uses salt (NaCl) and limestone as raw materials and involves several treatments to produce 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 one tone of soda ash to be between 2 and 4 tons, depending on the energy source used (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 lower carbon footprints, compared to soda ash (Cherubini et al., 2008; Nazari and Sanjayan, 2016), and this is reflected in Figure 6. Finally, the low contribution of the infrastructure in all impact categories is mainly attributed to: (i) the high life span of the concrete 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 carried out in the same reactor (clarifier).

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 were used) and are shown in Figure 7. Normalisation is an optional step of the LCIA, which transforms the results by dividing each impact category with a corresponding reference value (ISO, 2006b). By doing so, results are compared with reference values and the magnitude of 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 lower (an order of magnitude) normalised scores. The remaining midpoint impact categories yield very low to miniscule normalised scores (Figure 7).

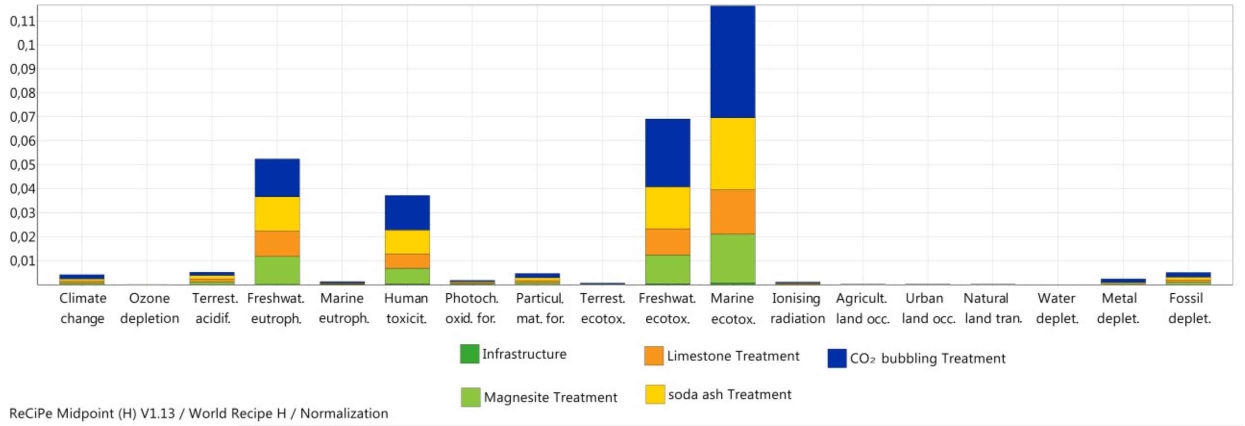


Figure 7: Normalised ReCiPe's midpoint results using the world's reference inventories for the effective treatment of 1 $m^3$ of AMD.

The high scores in the (eco)toxicity (MET, FET and HT) and eutrophication (FE) impact categories are attributed to the mining of the chemical reagents and of the fossil fuels required for electricity generation (South Africa's energy mix is dominated by fossil fuels, 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 fuels. Therefore, the chemical reagents and particularly fossil fuel mining exposes previously buried coal minerals to both oxygen and water, thus releasing, through waterborne emissions, mine-derived sulfate salts. Sulfate emissions could disrupt water balance and ion exchange 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 as soda ash production and fossil fuel refining and combustion release toxic materials, such 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., 2016).

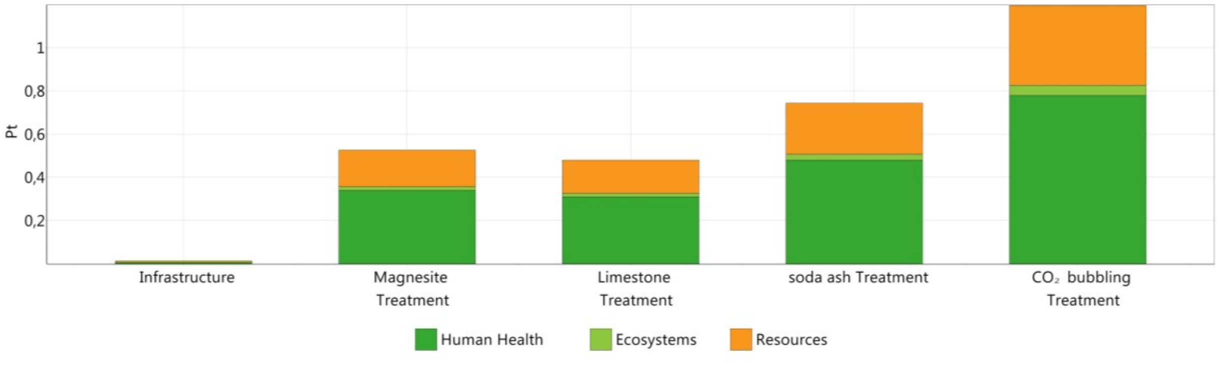
As far as the FE impact category is concerned, mining activity is an increasingly important stressor for freshwater ecosystems, since sulfate can co-vary with other environmental parameters, such as nitrogen and phosphorus, and impact aquatic organisms.

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 eutrophication (Zhao et al., 2017). Also, fossil fuel combustion lead to nitrogen oxides 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 resilient to eutrophication and lower quantities of direct and indirect (e.g. from fossil fuel burning) nitrogen emissions are attributed to the system under study (limestone used in the treatment system is assumed to be recovered and/or properly disposed, thus it does not reach 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).

### 7.2.2 ReCiPe results at endpoint

Figure 8 shows ReCiPe's weighted results at endpoint level (Hierarchist perspective using the normalisation values of the world with the average weighting set). Weighting is an optional step in the LCIA, after normalisation, where results are multiplied by weighting factors corresponding to each impact category. Weighted results can be then aggregated into a single score, in order to access the total environmental footprint of the effective treatment of 1 $m^3$ of AMD. It was found that the weighted damage to human health exhibits the highest score (1.92 Pt), followed by the damage to resources availability (0.932 Pt), while the damage to ecosystem diversity has the lower score (0.109 Pt). Therefore, the aggregated score is found to be 2.96 Pt per $m^3$ of AMD. The high scores of the first two damage categories are attributed to electricity consumption and to the mining/processing of the chemical reagents added in each treatment step. Similarly to IPCC 2013, the treatment step that has the highest aggregated score is $CO_2$ bubbling (1.19 Pt), followed by soda ash (743 mPt), magnesite (526 mPt), and limestone treatment (479 mPt). Moreover, the main environmental hotspot was 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 stirring/propelling and the remaining 15.8% or 0.468 Pt to water pumping. The second environmental hotspot was identified as the liquid $CO_2$ input (1.08 Pt or 36.4%), followed by soda ash, as a material (276 mPt or 9.33%). The remaining chemical reagents had a lower contribution, i.e. magnesite 58.7 mPt or 1.99%, and limestone 11.7 mPt or ~0.4%. Due to their high life span, the $CO_2$ diffuser, the propeller and the pumps had a miniscule contribution (<0.1%), while the tanks, including the clarifier, contribute 3.72 mPt or 0.126% 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 base (i.e. pipping and land use had a miniscule contribution).



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

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

### 7.3 Sensitivity/scenario analyses

#### 7.3.1 Source and form of $CO_2$

The liquid $CO_2$ used in the bubbling stage was found to be the main environmental hotspot of the system. This, is attributed to the large quantities required and to the energy and resources that are required for $CO_2$ extraction, purification, liquefaction, and storage (Althaus et al., 2007). Therefore, a sensitivity analysis concerning the source and form of used $CO_2$ was carried out. To be more specific, the use of processed $CO_2$ in liquid form versus raw gaseous $CO_2$ harvested as a process by-product was studied. This scenario was explored based on the fact that the system can directly use gaseous $CO_2$ produced as a by-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 proximity. In these cases, since the gaseous $CO_2$ is produced as a by-product of another engineering process, it could be assumed that it will be free of any environmental burden (Althaus et al., 2007), thus improving the system's overall environmental sustainability.

A scenario dealing with directly using gaseous $CO_2$ that derives as produced as a by-product, 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 $CO_2$.

#### 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 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 found that when using solar energy the system's environmental sustainability is substantially improved. Specifically, its total environmental footprint is reduced from 2.96 Pt per $m^3$ to 1.64 Pt per $m^3$. Therefore, the introduction of RES, in this case solar energy, can reduce the total environmental footprint by ~45%. Moreover, a third and practically feasible scenario is to combine the use of RES (i.e. solar energy) and gaseous $CO_2$ in the same AMD treatment system. In this case the total environmental footprint of the system is minimized, from 2.96 Pt per $m^3$ in the initial scenario (i.e. current operating conditions) to 0.559 Pt per $m^3$, and is drastically reduced by ~81%, reaching an overall high environmental sustainability.

### 7.3.3 Resource recovery from AMD sludge

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

In order to extract the targeted resources from each sludge stream chemical reagents are required (Bailey et al., 2016), but most importantly large amounts of energy are required, 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 $m^3$ AMD are Al, Ca, Fe, Mg and Mn. Here we examine the effect of one of the main input of the extraction 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 extraction procedure is more complicated and requires further processing, we estimated that the additional environmental burdens of the sludge valorisation are compensated by the fact that here sludge drying was achieved by a laboratory oven (industrial ovens require much less energy input). Future works could deal with the exact environmental performance of the most promising recourses to be recovered.

Regarding this scenario, since the oven is not included in SimaPros's proprietary 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 impact of the drying process and the avoided emissions, resource extractions, etc. attributed to each recovered resource the following were observed: For the magnesite treatment sludge stream, it was found that the environmental gains of Fe (assumed to be recovered as iron 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 reduced by 0.09 Pt or by about 3%). When, recovery of gypsum and brucite (assumed to be 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 9% reduction of the total environmental footprint could be achieved from gypsum but mainly 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 process. Therefore, alternative routes (e.g. disposal) for the sludge generation during the last two treatment stages should be considered. Overall, it was found that sludge valorisation could reduce the total environmental footprint of the AMD treatment process by up to 12%. Hence, results indicate that sludge valorisation could be a promising strategy to offset the environmental impacts of AMD treatment and improve its overall environmental sustainability.

### 7.4 Economic analysis

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

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

**Table 3:** Economic evaluation of the AMD treatment process capital (CAPEX) and operating expenditure (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 m$^3$** | | **R 7.78** |
| **Operating expenditure (OPEX) required for chemical reagents and energy inputs** | | | |
| **AMD, R/m$^3$** | 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/m$^3$** | | **R 105** |

On average, renewable energy technologies are more expensive than the conventional technology on an levelised cost of electricity (LCOE) basis (Jahed et al., 2016). However, 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 using a solar photovoltaic (PV) system in South Africa ranges from R0.915 to R2.07 per 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 the panel (Ross et al., 2016). Overall, this cost is comparable or cheaper (see Table 3) than the cost of purchasing electricity from ESKOM, the state owned enterprise that generates approximately 95% of the electricity used in South Africa (Ross et al., 2016). Therefore, it is inferred that solar energy could be an economically feasible electricity source for AMD in 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. see (Foteinis et al., 2018)), since it is estimated that low to no additional costs would be incurred per treated m$^3$ of AMD. In this case, however, the total electricity cost would have to be paid upfront, i.e. the CAPEX would be higher, but operating costs would be minimized.

## 8. Conclusions

The environmental sustainability of a typical AMD treatment method was examined by means of the life cycle assessment (LCA) methodology. Actual life cycle inventory (LCI) 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 also discussed. AMD is a common problem at mine sites, primarily at abandoned ones, while 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 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 operate off-grid in remote areas) and national level. It can also support other important functions, such as agriculture, thus improving economic sustainability. The systems has an overall low levelized cost per $m^3$ AMD, i.e. R112.78/$m^3$ (€7.60/$m^3$ or \$9.35/$m^3$), which is expected to reduce at industrial level, where economies of scales exist, and if gaseous CO2 can be sourced by a nearby source, e.g. flue gas.

The system was found to have an overall high environmental footprint (29.6 kg $CO_{2e}$ or 2.96 Pt per treated $m^3$ 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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