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Life cycle assessment of a passive remediation system for acid mine drainage: Towards more sustainable mining activity

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A B S T R A C T

Historical mining activity in the Iberian Pyrite Belt has left a huge number of abandoned sites; these cause severe water pollution by acid mine drainage, for which remediation seems to be unaffordable. Although the choice of remediation systems is usually dictated by technical and economic factors, the sustainability of such systems is becoming increasingly important in decision making, and efforts to promote greener remediation measures are being made. Life cycle assessment is a proven methodology that can help in the selection of the best available technology by reducing environmental burdens while ensuring the efficiency of the treatment. The main goal of this study is to perform the first life cycle assessment for dispersed alkaline substrate technology, effective for metal-rich and acid waters, in order to determine the environmental impacts generated throughout its entire life cycle and the factors controlling the environmental performance of this technology. We show that although the construction of the plant initially creates significant environmental impacts, these become negligible within a few years (4.5 years). The results also show that the potential impacts of the plant are closely related to the upstream production chain of the materials employed in this technology. Thus, the replacement of certain material sources and circular usage would lead to a significant decrease in impact values. The replacement of wood chips by forestry waste would reduce emissions by between 50% and 100%. The global warming potential of the plant was 1.86 kg CO_2 eq/m^3, to which limestone dissolution contributes 94% of the total value, and hence the replacement of non-carbonate alkaline materials would significantly decrease the emissions to the atmosphere. This study also finds evidence for the lower carbon footprint of passive treatment in comparison with other wastewater treatment systems analyzed using life cycle analysis. The results of this work may contribute to more environmentally friendly mining by providing an insight into environmental burdens related to the available passive treatment options for acid mine drainage during mining operations and the post-closure phases.

1. Introduction

The exposure of sulfides to oxygen and water at mining sites leads to the generation of acidic waters with high concentrations of sulfate and metal(loid)s in a process known as acid mine drainage (AMD), which is of serious environmental concern worldwide (Akcil and Koldas, 2006). This process is particularly intense in the Iberian Pyrite Belt (IPB), which forms one of the most important metallogenic provinces in the world, with giant and supergiant polymetallic massive sulfide deposits (Sáez et al., 1999). The IPB has been intensively mined from the third millennium BC to the present day (Nocete et al., 2005), and this historical mining activity has left a huge number of abandoned sites, with an estimated total affected area of 4846.72 ha in the Spanish sector of the IPB alone

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(Grande et al., 2014). This has caused severe metallic pollution of the main fluvial systems draining the IPB, the Odiel and Tinto river basins (e.g. Cánovas et al., 2007; Olías et al., 2006). The Odiel river basin has been more intensively affected by mining in terms of surface area; around 54 mines have been exploited, leaving approximately 258 ha of open pits, 1299 ha of waste dumps, 600 ha of tailing dams, and 144 ha of other mining facilities (Grande et al., 2014). Remediation measures are therefore needed in order to avoid the pollution of water bodies. Mine waters can be treated by chemical, biological and/or bio-chemical mechanisms using two generic approaches: active processes, which require continuous inputs of energy and chemicals to sustain them, and passive processes, which require no energy and relatively little resource input once in operation (Johnson and Hallberg, 2005). Mining companies have traditionally implemented active treatments, but are becoming increasingly interested in adopting passive treatments to reduce the high recurrent costs of reagent addition and sludge disposal (Johnson and Hallberg, 2005). Although conventional passive treatments (e.g. anoxic limestone drains, vertical flow wetlands, aerobic wetlands) have been successfully employed in coal mine districts (Younger et al., 2002), their use in metal mining districts such as the IPB is discouraged due to the high acidity and metal content of the waters, as they lead to clogging of the system and loss of the reactivity of the alkalinity-generating material employed (Ayora et al., 2013). A remediation system known as dispersed alkaline substrate (DAS) has been designed and successfully tested for the treatment of highly acidic and metal rich waters (Ayora et al., 2013; Caraballo et al., 2011, 2009; Macías et al., 2012b, 2012c). One recent study considers a possible first step in the restoration of the basin, in which the implementation of 13 DAS treatment plants located at selected acid discharges within the Odiel basin would lead to a noticeable improvement of the water quality (Macías et al., 2017b).

Traditionally, the choice of a remediation system for AMD has been dictated by technical and economic factors, with the environmental performance a secondary consideration. However, the sustainability of remediation systems is a factor that is becoming increasingly important in decision making (Johnson and Hallberg, 2005). Hence, efforts have recently been made to promote greener remediation measures for mine pollution. For example, the US EPA Superfund program requires consideration of all of the environmental effects of remediation measures and the incorporation of options to minimize the environmental footprints of a cleanup (Clu-in.org, 2018).

In this context, life cycle assessment (LCA) is a comprehensive approach that considers the interdependence of all stages of a product/process. The International Organization for Standardization (ISO)-standardized LCA enables the estimation of environmental impacts resulting from all stages in the life cycle of a product, often including impacts that are not considered in more traditional analyses such as raw material extraction, material transportation or ultimate product disposal (ISO 14040, 2006) (US EPA, 2006). The implementation of LCA allows for the identification of the transference of environmental impacts from one medium to another (i.e. from mine waters to hazardous solid wastes) or even from one life cycle stage to another. This tool has been successfully applied to the study of the environmental impacts of management strategies for wastewater and municipal solid wastes (Bisinella de Faria et al., 2015; Di Gianfilippo et al., 2016; Erses Yay, 2015; Pintilie et al., 2016; Quirós et al., 2015). However, LCA has recently been adopted by the mining industry to identify environmental impacts that are associated only with certain mining exploitation processes (e.g. Burchart-Korol et al., 2016; Ferreira and Leite, 2015; Northey et al., 2013). In particular, examples of LCA in AMD treatment are scarce. Tuazon and Corder (2008) compared the use of seawater-neutralized red mud with that of lime to treat AMD in Mount Morgan in Queensland, Australia. Despite the use of red mud generating savings in CO_2 emissions (80%) and electricity use (56%), the fuel usage of this approach was 12 times higher. Hengen et al. (2014) performed a comparative LCA of five different passive and two active AMD treatments in the Stockton Coal Mine in New Zealand, finding evidence for the greater impact of active treatments compared to passive ones for an equivalent level of efficiency. Masindi et al. (2018) employed LCA methodology to assess the environmental sustainability of an active treatment using various materials such as magnesite, limestone, soda ash and CO_2 bubbling to treat AMD originating from a coal mine. These authors reported a high overall environmental footprint due to electricity consumption, although these impacts could be markedly reduced if renewable energy was used.

However, these studies need to be extended to the other technologies and treatments commonly adopted in mine sites. Thus, the main goal of this study is to perform an LCA of a passive treatment (DAS technology) of AMD implemented in the Mina Concepción sulfide mine (in SW Spain) throughout its entire life cycle, and to study the factors controlling the environmental performance of this technology. Mina Concepción has a composition that is representative (in terms of acidity and metal concentration) of the mine waters of the IPB, and therefore can be considered a good example for the application of LCA to this technology. Furthermore, first AMD contribution to the Odiel river is found at this site, and this represents the first attempt to remediate the Odiel river (Macías et al., 2017b). The results of this work could help to achieve more environmentally friendly mining by providing an insight into the environmental burdens arising from AMD treatment that is successfully applied to highly acidic and metal-rich mine waters.

2. The DAS-based AMD treatment plant of Mina Concepción

The AMD passive treatment plant at Mina Concepción (20,000 m^2 total surface area), located in the IPB (SW Spain; 6°40′26″W 37°46′39″N) is a multi-step system for treating AMD based on DAS technology, which is one of the few passive treatments that have been successfully applied to treat high acidity and metal-rich AMD (Macías et al., 2012b, 2012c). DAS-based technology relies on the use of fine-grained alkaline reagents (e.g. limestone sand or caustic magnesia powder) to provide high reactivity and neutralizing capacity, mixed with a coarse inert matrix (i.e. wood shavings) to supply high porosity to the reactive mixture. This technology has been tested in laboratory and pilot experiments, and full-scale implementations in the IPB have shown excellent results regarding metal removal (Ayora et al., 2013).

The full-scale treatment plant (Fig. 1) is fed by two different AMD inputs, from (i) a pit lake overflow (MC1) and (ii) a partially restored waste dump (MC2). The MC1 discharge is characterized by high values of acidity and metal concentrations (pH 2.66, 512 mg/L of Fe, 100 mg/L of Al, 35 mg/L of Zn, and lesser amounts of other trace metal/loids (e.g. Cd, Co and As; Fig. 1). The MC2 AMD is characterized by a lower content of Fe (24 mg/L) and most other metals (e.g. Zn, Cd, As and Co) except for Al (119 mg/L; Fig. 1). The average flow is around 0.8 L/s, although during the dry season this progressively decreases to 0.1 L/s. The treatment plant is initially composed of a natural Fe-oxidizing lagoon (NFOL; Macías et al., 2012b) of around 100 m^2, which enhances iron oxidation and the removal of Fe and As. After this pretreatment, the water runs into the first two alkaline reactive tanks (which are divided into two vessels, RT1 and RT2, of 960 m^3 and 720 m^3 respectively) filled with limestone-DAS, which causes the depletion of trivalent metals.

These tanks are serially connected with two settling ponds (S1 and S2, of 100 m^2 each) that enhance the sedimentation of Fe and Al

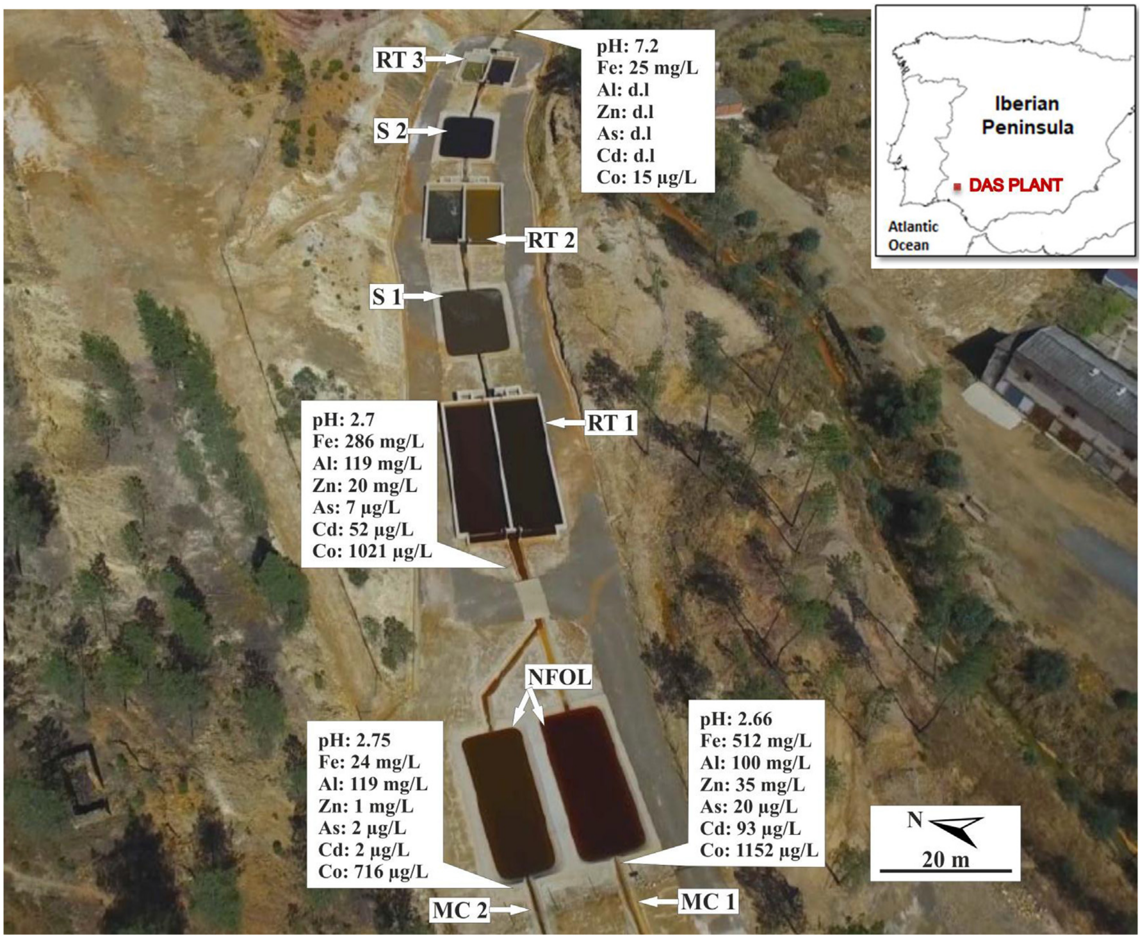

Fig. 1. Aerial view of Mina Concepción DAS passive treatment plant, showing the chemical composition of waters along the plant. The system consists of: 1) pre-treatment (natural Fe-oxidizing lagoon (NFOL); 2) alkaline reactive tanks (RT1 and RT2) filled with $CaCO_3$-DAS, each followed by a decantation pond (S1 and S2), and 3) alkaline reactive tanks (RT3) filled with MgO-DAS.

precipitates. The outflows then reach a MgO-DAS reactive tank (RT3, 400 m^3) where pH values increase to 7.2 and the concentration of divalent metals is reduced to close to the detection limit, and are finally released into the fluvial system (Fig. 1). As a result, an alkaline effluent is obtained with a low metal content, while Fe- and Al-rich sludge accumulates in the reactive tanks (RT1 and RT2) and divalent-rich sludge in RT3. The management of these wastes has been previously addressed by Macías et al. (2012a), who reported the requirement for Fe-rich DAS-type sludge to be disposed in landfills for hazardous wastes, Al-rich sludge to be treated as non-hazardous waste and divalent-rich sludge in landfills for inert waste.

3. Methodology

3.1. Goal definition and scope

The LCA methodology was implemented to quantify the environmental impacts generated by the DAS treatment plant, and to analyze the factors responsible for these impacts. A functional unit (FU) of 1 m^3 of AMD treated water was used. The system boundaries (Fig. 2) comprised the input and output flows of material and energy resources for the construction and use phases; the construction phase encompassed raw material acquisition, manufacturing and construction, while the use phase included the plant's operation, involving the addition and replacement of reagents after material depletion (in the case of the DAS filling), and waste handling operations based on sludge removal for safe disposal.

The passive treatment plant at Concepción has been operating since April 2016, and thus all data concerning the construction and use phases were available. Average values of the flow and chemical composition of water used in the LCA were obtained from monitoring of the plant between April 2016 and April 2017. In total, the plant treated a volume of around 20,000 m^3 during this one-year period. The sludge generated during the treatment was assumed to have been removed for transport to a suitable landfill or for other management options (e.g. metal recovery). These options are currently being investigated, and data quantification for these approaches is not yet available; hence, they were not considered in this study. The present analysis is a “cradle to gate” study in which the end-of-life aspects of the plant (dismantling, demolition and disposal) were not considered. Data were obtained from technical reports on the project's construction (LIFE ETAD Project, 2015; www.life-etad.com), contractor records (e.g. budgets, work plans, technical drawings) and various suppliers of materials. Assumptions regarding the specific properties of a material (e.g. specific weight or density) were obtained from the manufacturer's technical data sheet or, when unavailable, from Spanish commercial manufacturers. Table S1 (Supplementary material) presents detailed information on the primary documentary sources of data and the assumptions made.

The LCA framework, including the material flow and the

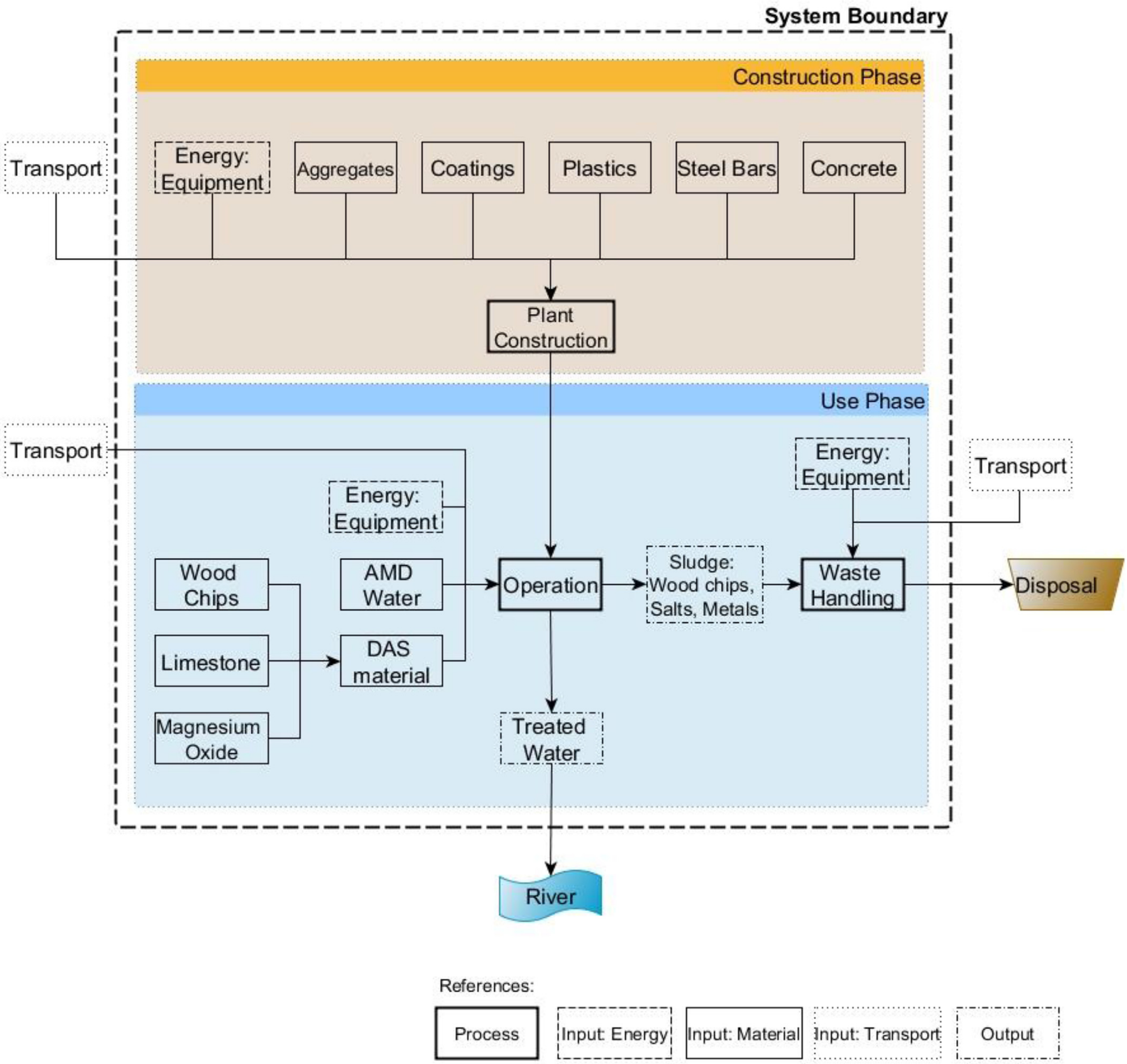

Fig. 2. System boundaries of the DAS treatment plant considered in the LCA. The input and output flows of material and energy resources for the construction and use phases are shown. The construction phase encompasses raw material acquisition, manufacturing and construction, while the use phase includes the plant operation, which involves the addition and replacement of reagents after material depletion (in the case of DAS filling) and waste handling operations based on sludge removal for safe disposal.

relationships between unit processes describing the life cycle of the remediation system, was developed within the OpenLCA modeling environment; this is an open source software application developed by GreenDelta in 2006 for LCA and sustainability assessments (www.openlca.org). Environmental profiles and datasets (for the acquisition of materials, manufacturing and energy use) were obtained from ProBAS+ (www.probas.umweltbundesamt.de) and ELCD 3.2 (http://eplca.jrc.ec.europa.eu/ELCD3/) databases. The final goal of this study was to perform a LCA of a passive AMD treatment (DAS technology) over its entire life cycle and to study the factors controlling the environmental performance of this technology.

3.2. Life cycle inventory

Life cycle inventory (LCI) analysis involves the compilation and quantification of inputs and outputs for a product throughout its life cycle. The inventory data with reference to the functional unit (1 m^3 of treated AMD) are summarized in Table 1, including the energy consumption, material data (types and quantities of materials/products used in each process) and outputs (sludge and direct air emissions).

The environmental impacts related to the construction process are assumed to arise mainly from heavy machinery operations and the materials used. In this case, the construction process involved

Table 1
Inventory results for Mina Concepción passive treatment plant referred to the functional unit (1 m^3 AMD).

Process	Unit	Value	Source	Unit Process Name
Construction*				
Materials				
Concrete	t/m^3	3,116,694.00	ProBas+	Beton
	t/m^3	38.374	ProBas+	Steine-Erden\Porenbetonstein
Metals	t/m^3	113.934	ELCD database 3.2	Steel rebar, production mix, at plant, blast furnace and electric arc furnace route
Plastics				
HDPE Pipes	t/m^3	3.487	ProBas+	hdpe pipes
HDPE liner	t/m^3	3.738	ELCD database 3.2	Polyethylene high density granulate (PE-HD), production mix, at plant
PVC Pipes	t/m^3	2.202	ProBas+	pvc pipe
Geosynthetics	t/m^3	1.316	ProBas+	polypropylene resin
Coatings	t/m^3	5.793	ProBas+	Bitumen
Aggregates	t/m^3	7644.800	ProBas+	Xtra-Abbau\Kies
Energy				
Equipment	kWh/m^3	117,426.600	ProBas+	Raffinerie\Ö-Produkte
Operation				
Materials				
Limestone ($CaCO_3$)	kg/m^3	4.22	ProBas+	Xtra-Abbau\Kalkstein
Wood chips	kg/m^3	3.67	ELCD database 3.2	Dummy_Wood (waste for recovery) Pine wood, production mix, at saw mill, timber, 40% water content
Magnesium Oxide (MgO)	kg/m^3	0.29		
Magnesite ($MgCO_3$)	kg/m^3	0.43	ProBas+	Magnesiumkarbonat
Electricity	kWh/m^3	0.20	ProBas+	El-KW-Park-ES
Energy				
Reagents addition (equipment)	kWh/m^3	0.024	ProBas+	Raffinerie\Ö-Produkte
Waste Handling				
Energy				
Sludge removal	kWh/m^3	0.011	ProBas+	Raffinerie\Ö-Produkte
Outputs				
Sludge	kg/m^3	6.805		Calculation (Table S3)
CO_2 (Direct emission to air)	kg/m^3	1.738		Calculation (Eq. (1))

the use of excavators and dumper trucks for earthworks, motorized graders for ground leveling, and mixer trucks and cranes for concrete preparation and transport. Since the environmental impacts of equipment operation are indirectly determined by energy consumption (Li et al., 2010), the energy use of the equipment was calculated by multiplying the rated engine power and load factor by the running time of each machine (Table S2, Supplementary material). The load factor applied was 60%, since for the majority of the time this type of machinery operates at a load of between 50% and 70% (Janulevičius et al., 2016). The materials used in the construction process can be categorized into materials used for physical building and materials used in supporting items, also known as ancillary materials, including earthworks, formworks and scaffolding (Li et al., 2010). However, ancillary materials (except for the earthworks) are not considered in this study. The construction period was five months in total (from November 2015 to March 2016), and included earthworks (site clearing, leveling and excavation, 40 days), drainage collection systems (minor earthwork arrangements and pipe placement, 20 days), plant infrastructure (completion of concrete structures/tanks and surfaces, 40 days), and auxiliary work (20 days). See Table S2 for further details.

The inputs during the use phase were mainly related to obtaining the DAS material and tank-filling activities, including the equipment used to carry out these tasks. For the equipment, the same assumptions (e.g. engine power, load engine factor, running time) were applied as in the construction phase (Table S2). Calculations based on the numerical model for the plant design estimated the depletion of limestone-DAS material (passivation by ochre was also considered) after treating 90,000 m^3 of water. The emission of CO_2 from limestone dissolution via contact with AMD was considered in the LCA model using Equation (1):

$$CaCO_3\,(s) + H^+\,(AMD) \rightleftharpoons Ca^{2+} + HCO_3^- + H^+\,(AMD) \rightleftharpoons Ca^{2+} + H_2CO_3\,(aq) \rightleftharpoons CO_2(g) + H_2O \quad (1)$$

It is assumed that the CO_2 released to the atmosphere is the difference between the limestone input mass and the output (treated) water alkalinity: $CaCO_3\,(s) - Alk\,(as\ CaCO_3\ eq) = CO_2\,(g)$.

Once the reactive material was depleted, waste handling involved the removal of sludge from the reactive tanks. A mass balance was performed to estimate sludge generation (Table S3). The sludge collection equipment was assumed to be the same as the tank refilling equipment. Table S1 shows the detailed LCI, including calculations and data sources.

3.3. Life cycle impact assessment

The ReCiPe method (Goedkoop et al., 2009) at midpoint level -hierarchist (H) cultural perspective-was used to analyze the potential environmental impacts of each remediation scheme. The hierarchist perspective is based on the most common policy principles concerning the time frame and plausibility of impact mechanisms, and is often considered the default model. The midpoint level provides information on the effects of different impact categories (for example, global warming potential, acidification, water depletion) on the environment, their contribution to the environmental performance of the system and which inventory items contribute (to a higher or lesser degree) to each impact. This allows for an analysis of the factors controlling the environmental performance of the technology, which is one of the goals of this study.

ReCiPe is composed of 18 (midpoint) impact categories, of which only 11 were selected, based on the main environmental issues related to the characteristics of both the technology studied and the region. The iterative approach of the LCA methodology was also applied to impact selection, and impact categories with results equal or close to zero were excluded (e.g. ionizing radiation). The output-related impact categories selected were climate change, terrestrial acidification, photochemical oxidant formation and particulate matter formation, and the (eco)toxicity-related impacts categories were human, terrestrial, freshwater and marine ecotoxicity. The input-related impact categories selected were water depletion, fossil fuel depletion and metal depletion. A brief description of the impact categories is provided in Table S4 (Supplementary material). Further information on the method can be found in the RIVM Report (RIVM, 2016) and the LCIA website (www.lcia-recipe.net [last accessed 30/05/18]). For better comprehension of the relative magnitude of the impact results, the optional step of normalization was carried out based on ISO 14044 (2006). Normalization is a process used to calculate the magnitude of the category indicator results relative to certain reference information. The category indicator results are divided by selected reference values, and this helps in the interpretation of the impact scores by bringing all the results to the same scale and allowing a relative structuring of impacts. In the present study, the annually released mass or resources consumed by an average person living in the European region, based on the geographical system boundary, was selected as a reference. The normalized results are called resident equivalents (REQs), as they provide information about the environmental impact per m^3 of treated AMD compared to the environmental impact of one person over a full year.

3.4. Data quality and limitations

Information related to machine operation (e.g. the age and state of the machines, type of handling of the machine) was not considered in this study. Most data on the unit processes dated back to 2010, and some of them were even taken from 1992. When available, data from Spain were used (for e.g. the Spanish electricity mix); however, information about some of the processes was obtained from German databases (e.g. limestone extraction and processing, aggregate extraction from open-pit quarries, steel bars and pre-cast concrete production). Some of the data related to the European region (e.g. magnesium carbonate mining, bitumen coating including extraction and production in European refinery, plastics, concrete and diesel production). The use of global-scale data was not necessary.

The depletion rate of the limestone-DAS material was determined in the laboratory, since field data are not yet available. However, the calculations were consistent with observations of the Mina Esperanza treatment plant (a similar, smaller-scale DAS plant that has been in operation for two years and six months, with 54,381 m^3 of AMD treated thus far). Incomplete emissions from the MgO production process were used due to the absence of datasets for this material in the databases available for this study. The limited operation time of the Mina Concepción DAS plant may be the source of significant variations in the average annual flow of treated AMD and the quality of its output, considering the changes in annual seasonal flow that are known to occur in the region.

4. Results and discussion

4.1. Life cycle impact assessment

The passive treatment plant commenced operation in April 2016. Since then, 22,164 m^3 of acidic water has been treated, at an average rate of 0.8 L/s. During the operational period, 100% of the As, Cr, Al, Cu, Pb, Zn, Cd and rare earth elements (REE) was removed from the AMD inflows. In addition, more than 95% of the Co and Ni, more than 90% of the Fe and up to 68% of the sulfate was also precipitated. The output waters had pH values of about seven, and the electrical conductivity values were reduced by more than half. No loss of material reactivity or clogging was observed during the monitoring period, thus guaranteeing several additional years of operation without replacing the alkaline materials in the tanks.

The potential environmental impacts of the DAS treatment plant for both phases (construction and use) are shown in Table 2. As can be seen, the construction phase creates the highest environmental life cycle impacts of the DAS plant. The metal depletion impact is entirely associated with the construction of the DAS plant, while the operation of the plant does not contribute to this impact category. For a better understanding of the relative magnitude of each impact indicator, normalization using regional Europe release per capita/year reference values was applied (ReCiPe H Europe 2000 V1.11). The normalized results (REQs) provide information on the relative significance of the product system for each category indicator.

In this context, fossil and metal depletion and climate change are the most significant impacts during the construction stage (Fig. 3). This is consistent with the intensive use of steel for the construction of reinforced concrete tanks and the machinery (which burns fossil fuel) needed for all construction activities, including the upstream chain process related to the extraction of raw materials (sand, gravel and cement). To a lesser degree, human toxicity, terrestrial acidification, particulate matter and photochemical oxidant formation make important contributions. Ecotoxicity-related impacts have the lowest relative relevance. From the treatment of 1 m^3 of AMD, the most relevant impact is climate change (0.00017 inhabitants/yr or 3.4 inhabitants/yr per year of AMD treatment) followed by fossil depletion, terrestrial acidification, particulate matter formation and photochemical oxidant formation. Impacts related to ecotoxicity are, again, the least significant. However, the global warming potential (GWP) (Table 2) after treatment of 1 m^3 of AMD in the DAS treatment plant is 1.86 kg CO_2 eq/m^3, and the dissolution of limestone contributes 94% (1.74 kgCO_2/m^3) to this total value. The remaining 6% of emissions are caused by limestone extraction, MgO manufacturing and machinery use (up to 0.12 kg CO_2 eq/m^3).

In addition, unlike other wastewater treatment industries (Presura and Robescu, 2017), the main source of carbon emissions in passive alkalinity-generating AMD treatments such as DAS technology is not energy consumption but the dissolution of carbonate and subsequent release of CO_2 to the atmosphere. With regard to the water depletion impact, the DAS treatment plant has a water footprint of 0.005 m^3, of which 95% of the total impact is related to upstream processes of indirect water consumption, such as limestone washing.

4.2. Environmental performance of the DAS passive treatment over different timespans

As evidenced by the previous section, the highest environmental impacts of the DAS treatment plant are related to the construction phase. However, time is an important variable in AMD remediation strategies due to the longevity of the AMD, which can last from hundreds to thousands of years (Younger et al., 2002). Table 3 shows the impact values of the DAS treatment plant over three different time periods (4.5, 20 and 100 years). The short-term period (4.5 years) corresponds with the period in which the DAS material is exhausted and replacement is needed. A medium-term period (20 years) was considered since it is the typical design period used for wastewater treatment plants (Yildirim and Topkaya, 2012; Garfí et al., 2017), and long-term operation (100 years) was also examined due to the long-lasting effects of AMD.

Table 2
Impact results for the Mina Concepción passive treatment system per m^3 of AMD treated.

Impact category	Unit	Construction Phase	Use Phase		
			Operation	Waste Handling	Total
Climate Change	kg CO_2 eq/m^3	5.36E+05	1.86E+00	3.41E-04	1.86E+00
Terrestrial acidification	kg SO_2 eq/m^3	1.12E+03	4.69E-04	6.97E-07	4.70E-04
Photochemical oxidant formation	kg NMVOC/m^3	1.69E+03	5.65E-04	1.30E-06	5.66E-04
Particulate matter formation	kg PM10 eq/m^3	4.76E+02	1.77E-04	2.17E-07	1.77E-04
Human toxicity	kg 1,4-DB eq/m^3	2.05E+04	2.60E-04	6.44E-07	2.60E-04
Terrestrial ecotoxicity	kg 1,4-DB eq/m^3	1.11E+01	1.08E-07	8.98E-11	1.08E-07
Freshwater ecotoxicity	kg 1,4-DB eq/m^3	3.96E+01	1.51E-07	8.56E-12	1.51E-07
Marine ecotoxicity	kg 1,4-DB eq/m^3	1.05E+02	1.41E-06	6.52E-10	1.41E-06
Water depletion	m^3/m^3	4.97E+03	4.98E-03	2.00E-07	4.98E-03
Fossil depletion	kg oil eq/m^3	7.98E+04	3.08E-02	1.01E-03	3.18E-02
Metal depletion	kg Fe eq/m^3	3.56E+04	0.00E+00	0.00E+00	0.00E+00

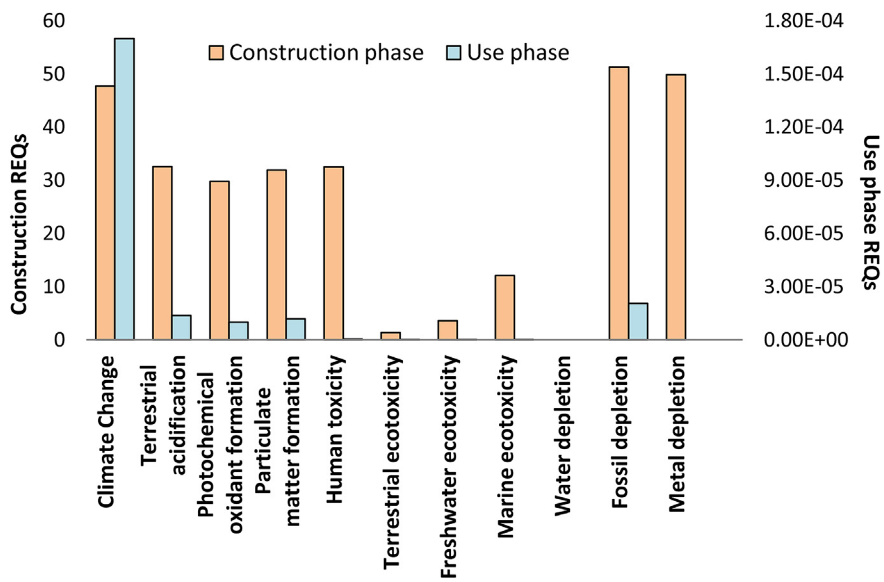

Fig. 3. Resident equivalents (REQs) per fU for the construction and use phases, based on ReCiPe normalization reference values for Europe (Note: the ReCiPe H Europe reference value for water depletion is zero). REQs provide information on the relative significance of the product system for each category indicator, as the environmental impact results per m^3 of AMD treated are compared to the environmental impact of one person over a full year.

Table 3
Impact values of the different processes (construction, C, operation, O, and waste handling, Wh) under different timespan scenarios: 4.5, 20 and 100 years (short, medium and long term). The short-term period corresponds with the DAS material exhaustion time and replacement needs. The 20-year period is the typical design period used for wastewater treatment plants, while the long-term operation corresponds to the long-lasting effects of AMD.

Impact category	Unit	4.5 years; short term			20 years; medium term			100 years; long term		
		C	O	Wh	C	O	Wh	C	O	Wh
Climate Change	kg CO_2 eq/m^3	5.95E+00	1.67E+05	3.07E+01	1.34E+00	7.44E+05	1.36E+02	2.68E-01	3.72E+06	6.82E+02
Terrestrial acidification	kg SO_2 eq/m^3	1.24E-02	4.22E+01	6.27E-02	2.80E-03	1.88E+02	2.79E-01	5.60E-04	9.38E+02	1.39E+00
Photo-chemical oxidant formation	kg NMVOC/m^3	1.88E-02	5.09E+01	1.17E-01	4.23E-03	2.26E+02	5.20E-01	8.46E-04	1.13E+03	2.60E+00
Particulate matter formation	kg PM10 eq/m^3	5.28E-03	1.59E+01	1.95E-02	1.19E-03	7.08E+01	8.68E-02	2.38E-04	3.54E+02	4.34E-01
Human toxicity	kg 1,4-DB eq/m^3	2.27E-01	2.34E+01	5.80E-02	5.11E-02	1.04E+02	2.58E-01	1.02E-02	5.20E+02	1.29E+00
Terrestrial ecotoxicity	kg 1,4-DB eq/m^3	4.40E-04	1.36E-02	7.70E-07	9.89E-05	6.04E-02	3.42E-06	1.98E-05	3.02E-01	1.71E-05
Freshwater ecotoxicity	kg 1,4-DB eq/m^3	1.24E-04	9.72E-03	8.08E-06	2.78E-05	4.32E-02	3.59E-05	5.57E-06	2.16E-01	1.80E-04
Marine ecotoxicity	kg 1,4-DB eq/m^3	1.17E-03	1.27E-01	5.87E-05	2.63E-04	5.64E-01	2.61E-04	5.25E-05	2.82E+00	1.30E-03
Water depletion	m^3/m^3	5.52E-02	4.48E+02	1.80E-02	1.24E-02	1.99E+03	8.00E-02	2.49E-03	9.96E+03	4.00E-01
Fossil depletion	kg oil eq/m^3	8.87E-01	2.77E+03	9.09E+01	2.00E-01	1.23E+04	4.04E+02	3.99E-02	6.16E+04	2.02E+03
Metal depletion	kg Fe eq/m3	3.96E-01	0.00E+00	0.00E+00	8.91E-02	0.00E+00	0.00E+00	1.78E-02	0.00E+00	0.00E+00

The impact values of construction are negligible compared to those associated with the operation phase, in each of these three scenarios. This demonstrates that once the plant has been built, the impacts associated with its construction become insignificant (compared to those of its operation) within only a few years. In comparison to the waste handling process, the impacts associated with construction are higher in terms of the (eco)toxicity and water depletion categories in both the short and medium terms, while higher impacts related to freshwater ecotoxicity were observed only in the long term. The remainder of the impacts associated with construction are only higher in the short-term scenario.

This finding supports previous observations of passive treatments, which are characterized by a relatively high ratio of initial impacts (Higgings and Olson, 2009) in comparison to active systems when only short-term periods are considered (Bayer and Finkel, 2006). However, for AMD treatment, the long term should also be considered due to the longevity of the processes involved.

4.3. Relative contributions of inventory items to the different impact categories

The passive treatment plant was also evaluated in terms of the relative contribution of each input during the construction (Fig. 4A) and operation processes (Fig. 4B). During the construction phase, the environmental impact associated with concrete is dominant (from 47% to 92%) over the impacts related to other input materials (Fig. 4A). An important environmental impact is associated with steel bars, and particularly for the metal depletion impact category, where it accounts for almost 100% of the total contribution. HDPE pipes make a significant contribution to fossil depletion (16.3%), while the rest of the plastic materials are responsible for around 20% of this category. Aggregates contribute between 11% and 14% to the formation of photochemical oxidants, terrestrial acidification, fossil depletion and the formation of particulate matter (Fig. 4A). On the other hand, during operation of the plant (Fig. 4B), the extraction and production of the reactive materials used in DAS technology are responsible for the highest impacts in the different categories. Limestone extraction and processing (grinding, washing and classification) contributes more than 70% to the categories of marine ecotoxicity, photochemical oxidant formation, terrestrial ecotoxicity and water depletion impact. Magnesium carbonate extraction represents 76% of the freshwater ecotoxicity impact, and the remaining contribution is due to the limestone extraction procedure. The electricity input necessary to produce MgO is a main contributor to terrestrial acidification (42%) and fossil depletion (53%). Limestone mining contributes almost 40% to the latter category, while the machinery used to fill the reactive tanks with the materials, and the subsequent removal of sludge when the reactive material is exhausted, accounts for the remaining 10%. The direct emission of CO_2 from limestone dissolution and its consequent release to the atmosphere contribute around 94% of the total greenhouse gas (GHG) emissions of the treatment plant, while the remainder arises from the electricity consumption during MgO production (3.8%), limestone extraction (2.10%), magnesium carbonate extraction and machinery (0.1%). The influence of the waste management process on the environmental impact of the plant is of minor importance, and is related only to the energy consumption necessary for sludge collection for transport to a suitable waste disposal facility.

4.4. Comparison with other LCA studies on AMD water treatment

A straightforward comparison among several water treatment systems cannot be made, since the water sources, chemistry and contaminants targeted for removal differ, and the life cycle stages included in these assessments may vary. The results of various LCA studies can only be directly compared if the assumptions and context of each study are the same (ISO 14040, 2006). For this reason, the results obtained in this study are compared only with other results obtained after applying the LCA methodology to AMD treatments and with the same functional unit (1 m^3 AMD treated).

The ecotoxicity-related impacts generated by the DAS technology plant were of minor importance. These results differ from those obtained for an AMD active treatment plant by Masindi et al. (2018), who found high relative scores for (eco)toxicity impacts that were attributed to the mining of chemical reagents and fossil fuels for electricity generation. This difference can be mainly explained by the electricity use, since the DAS plant is a passive system with almost no energy requirements. In addition, since the geographical system boundaries differ (between South Africa and Spain), the energy mix may vary and the mining processes may be carried out in different ways. Furthermore, the present study accounts for the direct emissions to the air from limestone dissolution, which contribute significantly to the climate change impact. In this context, DAS technology has a lower carbon footprint than other wastewater treatment systems analyzed using the LCA methodology.

In the case of the GWP of the DAS plant, around 1.86 kg CO_2 eq/m^3 of treated AMD was estimated, primarily associated with limestone dissolution (94% of the total value). This value is lower than that of 2.60 kg CO_2 eq/m^3 for an AMD water reclamation plant based on ultrafiltration and reverse osmosis (Goga, 2016) and significantly lower than that of 29.6 kg CO_2 eq/m^3 for an AMD active treatment based on calcined magnesite, limestone, soda ash addition and CO_2 bubbling (Masindi et al., 2018). Although Tuazon and Corder (2008) reported a generation of 4.378 kg CO_2 eq/m^3 for AMD treated in a lime treatment system, the same study reports emissions ranging from 0.528 to 1.550 kg CO_2 eq/m^3 of AMD treated using different mixtures of limestone and recycled wastes such as from a sea-water-neutralized red mud. This finding supports the possibility of using secondary raw materials to reduce the GHG emissions.

The fact that the DAS technology uses carbonate-based materials as acid-neutralizing agents means that carbon emissions have an important role compared to other non-carbonate-based technologies. Similar observations have already been reported by Vince et al. (2008), with regard to the addition of lime in a desalination treatment in order to adjust the alkalinity of the demineralized water to potable water quality standards, and Hengen et al. (2014), who discussed the negative implications of relying on lime as an acidity-mitigating material.

4.5. Potential improvements to the environmental performance of the DAS treatment plant

The previous section highlights the impact of the extraction and processing of raw materials on the environmental performance of the DAS treatment plant. For example, the obtaining and transport of concrete and steel bars made a higher contribution during the construction phase, while the acquisition of carbonate materials and their dissolution generated higher impacts during the use phase. In this context, the adoption of measures based on the circular economy could notably improve the environmental performance of the DAS treatment. Thus, alternative sources of greener raw materials should be explored as substitutes for materials commonly applied in the DAS technology.

For instance, the production of forestry waste in the vicinity of the DAS treatment plant provided a cheap raw material, and the use of wood chips from waste rather than natural pinewood helped to improve the potential emission results in all the impact categories analyzed, reducing almost all impact values by more than 50% (i.e. climate change), or even by almost 100% for some categories (i.e. 95% reduction in marine ecotoxicity) (Fig. 5). The high variability found among the different impact categories relies in the pinewood chips production system, its requirements and emissions. This substitution of greener raw materials could be extended to those that make a higher contribution to environmental impacts. The replacement of the concrete commonly used by building companies by greener versions could help to reduce the environmental impacts of the construction phase. Greener concretes based on the

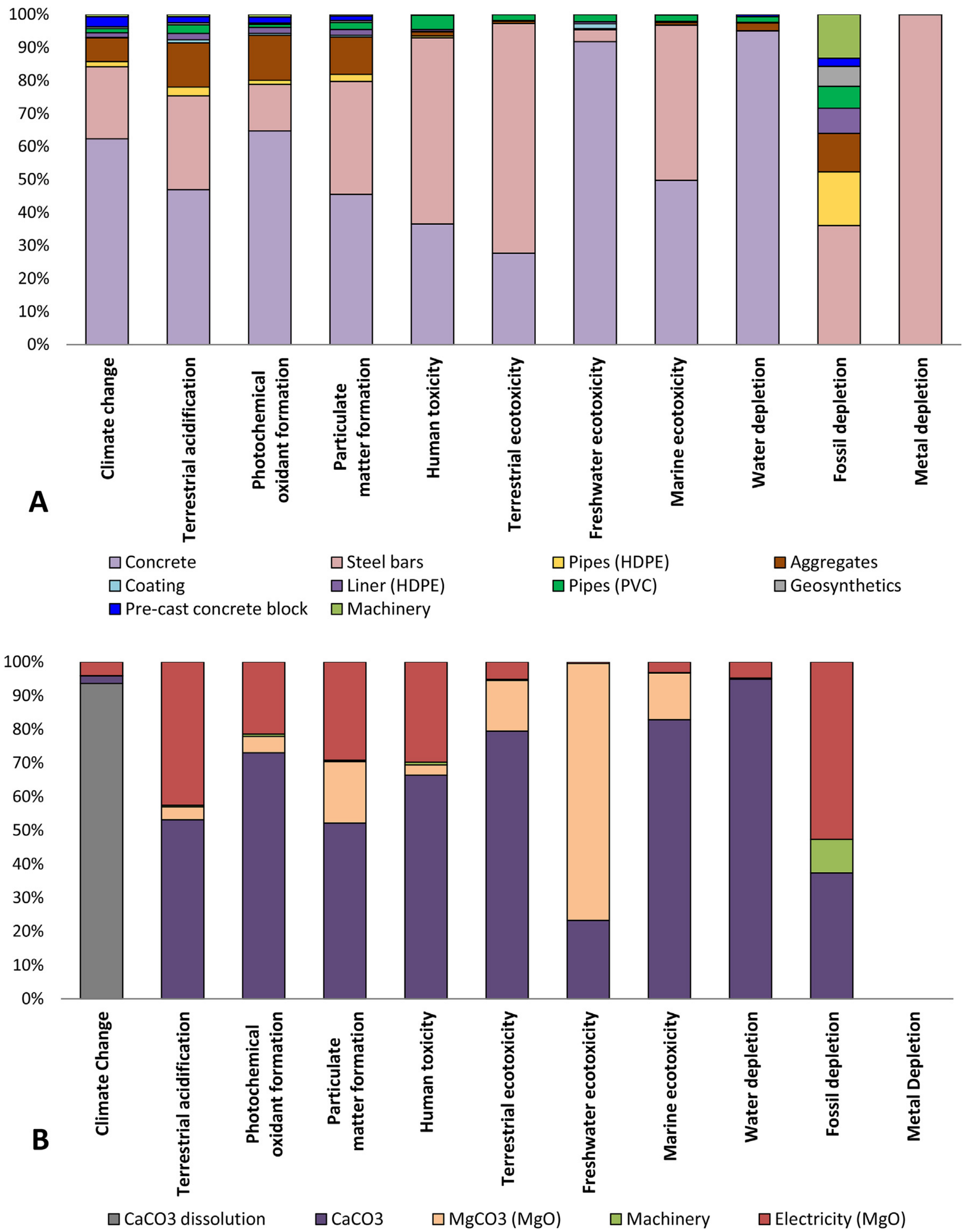

Fig. 4. Impact contribution (%) of each inventory item during the construction (A) and operation (B) processes of the DAS treatment plant.

recycling of construction and demolition waste have been tested, and fulfill the requirements in terms of skid and abrasion resistance (e.g. Yap et al., 2018). However, it remains to be seen whether this greener concrete can support the high acidity found in mine waters for long periods, and further studies are needed before using these materials in the construction of DAS treatment plants. On the other hand, the presence of secondary sources of alkalinity could also reduce the environmental impacts associated with the process of limestone extraction and processing. A promising possibility for obtaining a potential secondary source of calcite would be the route followed by Cárdenas-Escudero et al. (2011), who explored the capacity of phosphogypsum to sequester CO_2, producing calcite among other products. Phosphogypsum is a waste generated at high rates by a fertilizer factory within an industrial complex located close to the treatment plant (Cánovas et al., 2017). The existence of a large number of industries emitting high levels of CO_2 would allow the development of this chemical process at industrial scales. On the other hand, the dissolution of calcite causes a high

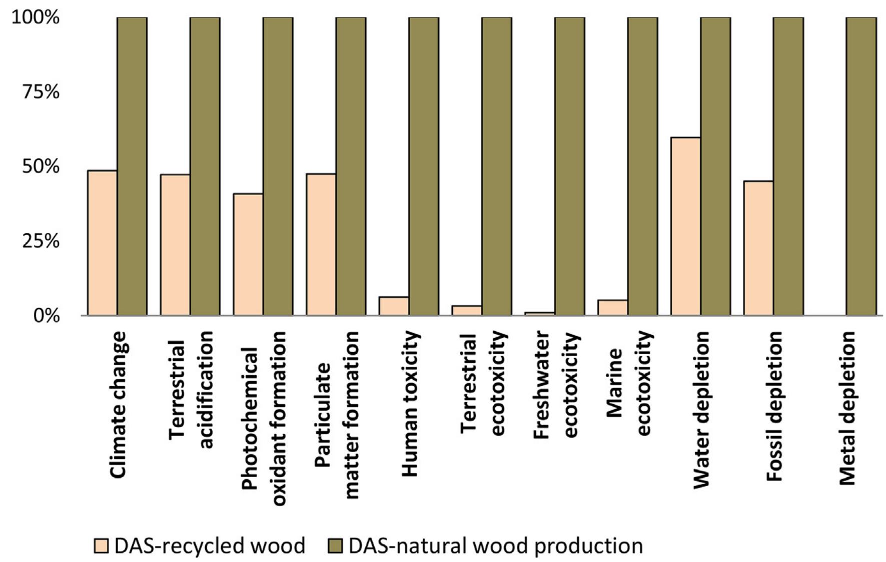

Fig. 5. Differences in impacts related to the use of recycled and natural wood materials to fill the reactive tanks. Results are normalized using the greatest value in each impact category. The use of recycled wood gives a reduction of more than 50% in almost all impact values, and even close to 100% for some categories.

impact on the climate change category, due to the release of CO_2 to the atmosphere, which is caused by the difference in CO_2 pressure between the outflows and the air. The replacement of the limestone commonly used in DAS technology by other non-carbonate alkaline materials could markedly reduce the impacts on climate change. A potential secondary raw material is the fly ashes commonly produced by power plants, which have high contents of CaO and MgO. Bottom and fly ashes from clean biomass fuels have been shown to increase the pH of pore water and to be particularly useful for the recovery of AMD-contaminated mining soils in the IPB (Cruz et al., 2017). The availability of huge amounts of these materials in the vicinity of the treatment plant offers a promising solution, although the technical and economic feasibility needs to be tested at laboratory scale.

Although the impacts caused by waste disposal were outside the scope of the present study, more environmentally friendly solutions can be adopted than disposal in a waste landfill, such as the reuse of these materials for different applications or as secondary raw materials. This could be the case for the Fe-rich sludge generated by the DAS treatment plant, which could be used to synthesize iron oxide pigments. These pigments are characterized by their non-toxicity, chemical stability, durability, variety of colors and their low cost for emerging markets (Legodi and de Waal, 2007). Ayora et al. (2016) also highlighted the potential of the Al-rich wastes generated from limestone-DAS treatment for use in scavenging technological metals of high economic interest, such as rare earth metals. Macías et al. (2017a) proved that these metals (and also the base metals Cu or Zn) can easily be extracted from sludge generated by AMD neutralization by simple leaching with dilute commercial acids. Although the technical performance and the cost-effectiveness are critical points, the environmental impacts related to these waste management strategies can be also assessed. The inclusion of these strategies in the LCA system boundary and the evaluation of the environmental burden using this methodology are of interest, and will be addressed in further investigations.

5. Conclusions

This study represents the first application of LCA to DAS technology, an effective treatment for metal-rich and acid waters. The LCA performed in this study allows quantification of the environmental impacts associated with an AMD passive treatment plant. Although the construction of the plant initially causes significant environmental impacts, these become negligible within a few years (4.5 yr). In comparison with the waste handling process, impacts associated with construction are higher with regard to the categories of (eco)toxicity and water depletion in both the short and medium terms, while higher impacts related to freshwater ecotoxicity were observed only in the long term. However, the remainder of the impacts associated with construction are only higher in the short-term scenario.

The plant GWP value of 1.86 kg CO_2 eq/m^3 is well below other corresponding values reported in LCA studies on AMD treatment. Lower values of GWP have been reported in other studies only with the use of recycled waste. Thus, our results also demonstrated that the choice of materials has a strong effect on the environmental performance of the treatment plant; in particular, it was shown that the main source of carbon emissions is not energy consumption but the type of alkaline material used in the AMD treatment. Since limestone dissolution contributes around 94% to the total GWP, the replacement of non-carbonate alkaline materials would significantly decrease the emissions to the atmosphere.

The LCA applied to the treatment plant allowed us to identify the sources of impacts, and to determine the changes necessary in terms of design, material replacement and the use of greener materials to reduce the environmental impacts and enhance the environmental performance of a treatment plant. The replacement of wood chips by forestry waste can reduce emissions by between 50% and 100%. When data become available, an evaluation of the environmental burdens associated with waste management will be also investigated using this methodology. In addition, the replacement of raw materials during the construction and operation phases to improve the environmental performance of the DAS technology should be addressed in the short term. LCA is shown to be a valuable tool to assess operation and post-closure AMD management options.

Finally, the results of this work can be used to support the decision-making process of a future restoration plan for the Odiel river basin. They may also contribute to more environmentally friendly mining by providing an insight into the cumulative environmental impacts related to the AMD treatment during mining operations. Overall, they contribute to the search for greener remediation solutions. Although the ISO standard attempts to establish a common methodology for the comparison of LCA results, there are still a number of unresolved variability sources (including software, databases, local data and data availability, among others). This limitation is a setback for the applicability of LCA, particularly with regard to decision-making processes and technology selection.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2018.11.224.

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