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Life cycle assessment of management/valorisation practices for metal-sludge from treatment of acid mine drainage

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ABSTRACT

The treatment of acid mine drainage in sulphide mining generates large amounts of metal-rich sludge whose management suppose an environmental quandary worldwide. Although traditional practices have focused on safe disposal in landfills, more environmentally friendly solutions may be adopted. The environmental performance of these solutions can be evaluated using tools like Life Cycle Analysis (LCA) that allows the estimation of the cumulative environmental impacts resulting from all stages in the product life cycle. The main goal of this study is, for the first time, to perform a LCA on different management strategies (i.e., i), encapsulation and disposal in landfill; ii) metal recovery using a chemical scheme; iii) pellets manufacturing for phosphorus removal from wastewaters; and iv) brick fabrication) for the metal-rich sludge generated from an active treatment plant. The manufacturing of pelletized material exhibited the lowest impacts (−35 REQs) due mainly to the environmental benefits of wastewater treatment. Despite the undoubtedly economic interest of recovering metals from the sludge, this route exhibited the highest environmental impacts (38 REQs), especially in the categories of fossil depletion (20,943 kg oil eq), marine (64 kg 1,4-dB eq) and freshwater ecotoxicity (50 kg 1,4-dB eq) due to the use of toluene and trybutylphosphate (TBP). The manufacture of bricks could be also an alternative route by obtaining economic value while exhibiting low environmental impacts (0.32 REQs), mainly on marine and freshwater ecotoxicity (0.7 kg 1,4-dB eq). These impacts can be notably reduced if waste materials are used replacing clays or using renewable energies. The encapsulation of these wastes is also a suitable option with a low environmental impact (0.46 REQs), but slightly greater than the brick manufacturing and with no economic return.

1. Introduction

The exposure of sulphides to oxygen and water in mining sites leads to the generation of acidic waters with high concentrations of sulphate and metal(loid)s, a process known as acid mine drainage (AMD) which is a serious environmental concern worldwide (Younger and Wolkersdorfer, 2004; Akcil and Koldas, 2006). A simplified reaction of sulphide oxidation can be represented as follows:

$$FeS_{2(s)} + 15/4\, O_{2(g)} + 7/2\, H_2O_{(l)} \rightarrow Fe(OH)_{3(s)} + 2SO_{4(aq)}^{2-} + 4H^{+}_{(aq)}$$

The intensity of sulphide oxidation mainly depends on sulphide, oxygen and Fe(II) availability and the absence/presence of alkaline rocks that may neutralize the acidity and slow down these reactions. These processes are particularly intense in the Iberian Pyrite Belt (IPB) which constitutes one of the most important metallogenic provinces in the world, with giant and supergiant polymetallic massive sulphide deposits (Leistel et al., 1997). In order to avoid the deterioration of water quality, mine waters must be treated prior to discharge (Sánchez-España et al., 2005). In this sense, mine waters can be treated by chemical and/or biological mechanisms upon two generic approaches; those considered active, which require continuous inputs of resources to sustain the process, or those passive, that in turn, require relatively little resource input once in operation (e.g., Johnson and Hallberg, 2005; Skousen et al., 2017). Operating mines have traditionally adopted active treatments due to the generation of a huge volume of polluted waters which

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would require large surface areas to treat them by passive technologies (e.g., Wolkersdorfer, 2021). Active treatments remove acidity and metals from AMD by the continuous addition of alkaline materials, leading to the generation of a huge amount of sludge whose management could suppose an environmental quandary worldwide (Macías et al., 2017). A detailed study on the waste production from conventional active and passive treatment plants ($n = 108$) conducted by Zinck and Griffith (2013) mainly in Canada, but also in USA, UK, Australia, Germany, Mexico, Brazil or China, provided average values of around 9500 tons of dry sludge per year, with maximum values of up to 135,000 dry tons per year. The majority of mines considered in this study corresponded to metal mines (46%), precious metal mines (23%), coal (7%), uranium (5%) and others (19%), therefore, the sludge chemical composition varied noticeably.

The sustainability of remediation systems is a factor that is becoming increasingly critical in decision-making (Johnson and Hallberg, 2005) and not only relies on the efficiency of the system but also on the management of the resulting wastes. The suitability of these types of wastes for landfilling disposal has been recently addressed by Macías et al. (2017) who highlighted its hazardousness due to the high potential leaching exhibited upon atmospheric conditions. On the other hand, recycling and reuse of mining and mineral processing wastes can be considered one of the main challenges for future waste management (Lottermoser, 2011) as demand circular economy requirements, particularly due to the large waste amounts (Bhattacharya et al., 2006; Amos et al., 2015; Moreno-Gonzalez et al., 2020). However, wastes generated during AMD remediation have not been perceived as a resource until lately (e.g. Chen et al., 2014; Macías et al., 2017; Orden et al., 2021), and in this sense, potential valorisation schemes should be environmentally evaluated before application.

The cumulative environmental impacts through all stages of mine waste management should be included in the decision-making by the application of unbiased tools. Life cycle assessment (LCA) is an approach ranging from "cradle-to-grave" to "cradle-to-cradle" for assessing products and processes that considers the interdependence of all stages of a product/process, normed according to ISO 14040. LCA enables the estimation of the cumulative environmental impacts resulting from all stages in the product life cycle, often including impacts not considered in more traditional analyses such as raw material extraction, material transportation or ultimate product disposal (EPA, 2006). The concept "life cycle" deals with the main activities in the course of the product's life-span from its manufacture, use and maintenance, to its final disposal, including the raw material acquisition required to manufacture the product, and might be even used to identify circular economy potentials.

The criteria followed by decision-makers to select the best waste management strategy, usually rely on the efficiency performance and cost, rather than on its environmental impact. When deciding between two or more alternatives, LCA can help decision-makers to compare all major environmental impacts associated to different waste management strategies. The implementation of LCA allows identifying the transference of environmental impacts from one media to another or even from one life cycle stage to another. For example, when selecting between two different waste treatments, it may appear that the first would be better for the environment because it causes a higher volume reduction than the second. However, after performing an LCA it might be determined that the first treatment actually causes larger cradle-to-grave environmental impacts considering all three media (i.e. air, water, land). Moreover, if LCA were not implemented, this impact transfer might be unnoticed. This tool has been successfully applied to study the environmental impacts of management strategies of municipal solid wastes (e.g. Yay, 2015; Di Gianfilippo et al., 2016; Quirós et al., 2016) and mineral processing practices (e.g. Northey et al., 2013; Ferreira and Leite, 2015; Burchart-Korol et al., 2016), sewage and industrial sludge (e.g., Rodríguez et al., 2017; Ding et al., 2021; Zhou et al., 2022). However, to our knowledge the LCA methodology has not been still applied to the management of sludge generated by treatment systems of mine waters. Thus, the main goal of this study is to perform a LCA on the management strategies for the resulting metal-rich sludge from the active treatment plant of Almagrera (SW Spain), which has been selected as a representative worldwide example of sludge generated by metal mining (Macías et al., 2017). The environmental impacts associated to different waste management routes were studied including raw material acquisition, manufacturing, transport and handling operations, using the OpenLCA modelling environment. The results of this work may contribute to a more environmentally friendly mining by providing an insight into the cumulative environmental impacts linked to the management of sludge generated by AMD treatment during mining operations.

2. Metal-sludge from Almagrera active treatment plant: potential management strategies

The studied sludge comes from the active treatment of AMD in Almagrera mine (SW Spain), characterized by a semi-arid climate, which was active from 1982 to 2001 to obtain Cu, Pb, Zn and sulphuric acid, from polymetallic sulphide ores. The treatment of AMD in this plant generated about 39,000 ton of metal-rich sludge which were deposited over a pond (Macías et al., 2017). The sludge composition is dominated by Ca (11%) and Mg (7%) from the neutralization reagent, S (9.9%) and metals such as Zn (4.5%), Mn (2.9%), Fe (2.4%), Al (1.3%) or Cu (0.6%) from the AMD (Macías et al., 2017). Traditional management practices for this type of wastes were aimed at to ensure the stability of the sludge and its safe disposal (Rakotonimaro et al., 2017). Both aspects were addressed in Almagrera metal-rich sludge by Macías et al. (2017) who performed leaching tests according to the rules for waste acceptance at European landfills (EC, 2003). Regarding exclusively metals, this sludge could be considered as an inert waste as concentrations in leachates were below the threshold limits established for inert wastes landfills, which evidences a high stability of the waste for potentially toxic metals. However, the sulphate concentrations exceeded the limits established for all type of landfills (i.e. inert, non-hazardous and hazardous) and thus, it requires a further treatment (e.g. additional neutralization, encapsulation, etc.). Therefore, the first route explored in this study consists on the encapsulation of the waste and the final transport to an inert waste landfill to ensure a safe disposal (Fig. 1, Route 1). The material selected to encapsulate the sludge is a Portland cement-based binder, which has been widely used for stabilization/solidification of wastes due to its availability and low cost (Shi and Fernández-Jiménez, 2006).

The potential of sludge from AMD treatments as source of elements of economic interest has been previously reported (e.g. Smith et al., 2013) based on the fact that the sludge may concentrate the metals of interest by several orders of magnitude in relation to the treated AMD. In the case of the Almagrera sludge, there are some base metals at very high concentration, such as Zn or Cu with around 43 g/kg and 6 g/kg respectively, which constitute ore grades clearly mineable in current market conditions (i.e. around 4% for Zn and 0.6% for Cu). The presence of other base metals such as Co (443 mg/kg), industrial metals such as Mn (28 g/kg) or high-tech metals such as rare earth elements and yttrium (200 mg/kg) could increase the potential value of this waste. Macías et al. (2017) reported the presence of these elements in poorly crystalline oxy-hydroxides and/or oxy-hydroxy-sulphates, which may be solubilized under soft acid conditions. Recovery percentages of around 80% for REE and higher than 90% for Zn, Cu, Co, Ni and Cd using dilute acids (HCl and H_2SO_4, 0.5 and 1 M) at a solid:liquid ratio of 1:20 were reported by Macías et al. (2017). This liquor must be subsequently treated in order to obtain a product, which accomplishes the market requirements. The approach studied in the present work is based on a solvent extraction scheme followed by Radzyminska-Lenarcik and Ulewicz (2015) for Zn recovery from metallurgical wastes sludge; the acid liquor comes into contact with an 80% tributyl phosphate (TBP)

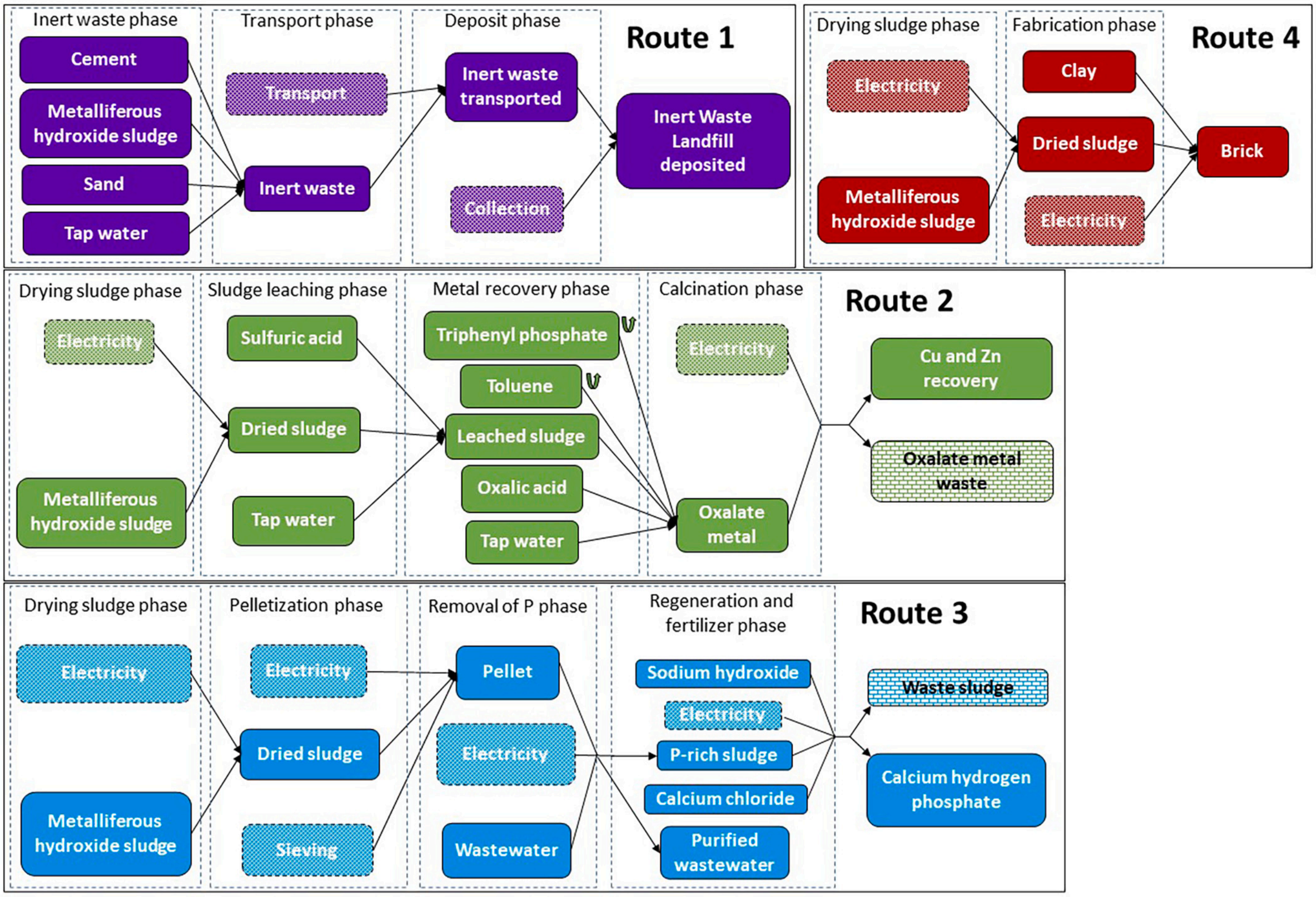

Fig. 1. Flowchart of the four valorisation routes considered in this study. Processes are shown with coloured rectangles and phases with dashed line rectangles. *Route 1:* Encapsulation and disposal in inert landfill; *Route 2:* Metal recovery using a chemical extraction scheme; *Route 3:* Phosphorus removal by pelletized sludge; *Route 4:* Brick fabrication using sludge.

solution in toluene, and shaken at 25 °C for 30 min. A recovery efficiency of 99% and 87% for Zn and Cu, respectively, at pH 2.87 was reached by Radzyminska-Lenarcik and Ulewicz (2015). Then, the backstripping procedure is accomplished by an oxalic rich solution that retain metals as oxalates that could be subsequently transformed in a more marketable product (i.e. oxides) while the solvent is recycled. A flowsheet of the recovery process considered as the second route for the sludge management is shown in Fig. 1 (route 2).

The high content in Al and Fe poorly crystalline oxy-hydroxides and/or oxy-hydroxy-sulphates, considered as highly adsorptive materials, make them also suitable to remove contaminants like metals and nutrients from water. For instance, radionuclides bound to water treatment sludge have been reported by Schneider et al. (2001). Thus, the third management practice explored is the use of the sludge to remove P from wastewater according to the procedure reported by Sibrell et al. (2009). In this way the sludge is dried, sieved and pelletized to produce the sorbent material. The pelletized material is deposited in packed bed contactors through which the wastewater flows. The P retained is desorbed by using a 0.1 M NaOH solution and precipitated as calcium phosphate by the addition of $CaCl_2$. Thus, the calcium phosphate generated as by-product can be sold as fertilizer product while the NaOH strip solution can be recycled (Fig. 1, route 3).

The inorganic components of sludge make them also suitable to be used in a wide range of building materials such as cement and ceramics manufacturing, or bricks production (Rakotonimaro et al., 2017). The main advantage of using sludge as clay substitute in bricks is the immobilization of heavy metals in the matrix during the firing process. Cusidó and Cremades (2012) reported the absence of environmental impacts of bricks manufactured with additions ranging from 5% to 25% wt of sewage sludge from water treatment and paper industries. The process of sludge brick manufacturing explored in this study is similar to that reported by Wang et al. (2003). Sludge is initially dewatered and then dried at 250 °C for 2 h. The proportion of sludge used in the bricks is 20%. The blended material (i.e. dry sludge and clay) spends a 24 h maturation stage, after which the material is exposed to 103 °C during 24 h. Then, the moulded materials are fired at 900 °C for 6 h (Fig. 1, route 4).

3. Methodology

3.1. Goal definition, scoping and system boundaries

The main goal of this study is to analyse the environmental impacts related to different management strategies of hazardous wastes resulting from the treatment of AMD in an active treatment plant. The selected functional unit (FU) is 1 ton of sludge produced during the treatment of AMD, which is representative of the studied process.

The system boundaries included input and output flows of materials and energy resources for the different valorisation routes (Fig. 1), including fertilizer application in route 3. The routes comprised raw material acquisition, manufacturing, transport and handling operations based on disposal in inert landfill (only for route 1). The LCA framework distinguishes the main drivers throughout the material flow, the relationships between unit processes or impact categories, thus describing the life cycle of the different valorisation routes. The LCA was performed using the OpenLCA modelling environment, an open source code developed by GreenDelta for LCA and sustainability assessments (http://www.openlca.org). The Ecoinvent Life Cycle inventory (LCI) database was used (version 3.5) to obtain the environmental information and datasets (https://simapro.com/databases/ecoinvent/). This database, widely recognized as the largest and most reliable LCI database for studies and assessments based on ISO 14040 and 14,044, contains approximately 15,000 LCI datasets within the areas of energy supply, transport, agriculture, chemicals, construction materials, packaging materials, textiles, metals, and metals processing considering unit (UPR) and system processes (LCI). For the present study was used the "cradle-to-grave" approach.

The potential environmental impact was assessed with the ReCiPe method (Goedkoop et al., 2009) at midpoint level Hierarchist (H). The midpoint level provides information on the effects of different impact categories on the environment, their participation in the environmental performance of the system and which inventory item contributes to each impact. The ReCiPe method for LCIA supports a method to calculate life cycle impact category indicators with midpoint and endpoint level results, which is harmonized in terms of modelling principles and choice (Goedkoop et al., 2009). Life cycle impact assessment (LCIA) converts emissions and resource extractions into different environmental impact scores by characterization factors. ReCIPe includes 18 midpoint indicators, however only 11 of them were selected considering the characteristics of the sludge and the valorisation routes: Climate change, fossil depletion, human toxicity, marine ecotoxicity, freshwater ecotoxicity, metal depletion, particulate matter formation, terrestrial acidification, terrestrial ecotoxicity, photochemical oxidant formation and water depletion (RIVM, 2016). The version of ReCiPe used is that from 2008, because indicators included are more appropriate for our study than those of 2016. In the present study, the system expansion approach is applied, e.g. co-products are considered as alternatives to other products and credited. Sensitivity and uncertainty analyses were considered in this study. Background LCA uncertainty is integrated into the Ecoinvent 3.5 database, while foreground uncertainty is established with the data quality of Ecoinvent 3.5 database. Data sources were evaluated corresponding to five characteristics: reliability, temporal correlation, geographic correlation, completeness, and further technological correlation (Weidema et al., 2013).

3.2. Life cycle inventory

Each unit process in the life cycle inventory was built independently of other unit processes, allowing the objective review of individual datasets before the assessment of their contribution to the overall life cycle results. Four different valorisation routes were therefore analysed:

3.2.1. Encapsulation and disposal in inert landfill

The exceeding concentrations of sulphate during leaching tests performed on these wastes demand a further treatment, which in this case consists on the encapsulation of the waste and the final transport to a landfill for inert wastes to ensure a safe disposal. Following the work of Biellen et al. (2014), 300 kg of cement and 390 L of water are added to 1 ton of sludge. As a result, we would obtain 2.59 ton of stabilized sludge (Table 1).

3.2.2. Metal recovery using a chemical extraction scheme

The approach studied in this work is based on a solvent extraction scheme followed by Radzyminska-Lenarcik and Ulewicz (2015) for Zn recovery from metallurgical waste sludge. The sludge is initially dried (50 kW/ton) and then, the sludge is leached with a 0.5 M H_2SO_4 solution (686 kg of H_2SO_4, 13,048 L of H_2O) during 2 h. Then, the acid liquor comes into contact with an 80% TBP solution in toluene (304 kg of TBP, 14434 kg of toluene), and shaken at 25 °C for 30 min. Afterwards, the back-stripping procedure is accomplished by an 0.5 M oxalic solution

Table 1
Inventory results for the valorisation routes referred to the functional unit (1 ton of sludge) used in OpenLCA.

Process	Unit	Value	Source	Unit Process Name
Route 1. Encapsulation and disposal in inert landfill				
Input				
Metalliferous hydroxide sludge	kg	1000	ecoinvent	metalliferous hydroxide sludge
Cement	kg	300	ecoinvent	cement, Portland
Sand	kg	900	ecoinvent	Sand
Tap water	kg	390	ecoinvent	Tap water
transport	kg*km	103,600	ecoinvent	transport, freight, lorry >32 metric ton, EURO6
excavation	m3	0.8	ecoinvent	excavation, skid-steer loader
Output				
Inert waste landfill deposit	kg	2590		
Route 2. Metal recovery using a chemical extraction scheme				
Input				
Metalliferous hydroxide sludge	kg	1000	ecoinvent	metalliferous hydroxide sludge
Electricity for drying	kWh	50	ecoinvent	electricity voltage transformation from high to medium voltage
Sulphuric acid	kg	686	ecoinvent	sulphuric acid
Tap water for leaching process	kg	13,048	ecoinvent	Tap water
Oxalic acid	kg	649	ecoinvent	
Tap water for metal recovery	kg	14,434	ecoinvent	Tap water
Toluene	kg	14,434	ecoinvent	Toluene
Tributyl phosphate	kg	304	ecoinvent	Triphenyl phosphate
Electricity for calcination	kWh	2568	ecoinvent	electricity voltage transformation from high to medium voltage
Output				
Zn	kg	5		
Cu	kg	43		
Oxalate metal waste	kg	20,585		
3. Phosphorus removal by pelletized sludge				
Input				
Metalliferous hydroxide sludge	kg	1000	ecoinvent	metalliferous hydroxide sludge
Electricity for drying	kWh	50	ecoinvent	electricity voltage transformation from high to medium voltage
Electricity for pelletization	kWh	35	ecoinvent	electricity voltage transformation from high to medium voltage
Sieving	kg	700		
Wastewater	m3	8327	ecoinvent	wastewater, average
Electricity for removal P from wastewater	kWh	14,072	ecoinvent	electricity voltage transformation from high to medium voltage
Calcium chloride	kg	2	ecoinvent	calcium chloride
Electricity for drying	kWh	14	ecoinvent	electricity voltage transformation from high to medium voltage
Sodium hydroxide	kg	34	ecoinvent	Sodium hydroxide, without water, in 50% solution state

(continued on next page)

Table 1 (*continued*)

Process	Unit	Value	Source	Unit Process Name
Output				
Calcium hydrogen phosphate	kg	1.4		
Purified wastewater	kg	8327		
Waste sludge	kg	735		
4. Brick fabrication using sludge				
Input				
Metalliferous hydroxide sludge	kg	1000	ecoinvent	metalliferous hydroxide sludge
Electricity for drying	kWh	50	ecoinvent	electricity voltage transformation from high to medium voltage
Clay	kg	2800	ecoinvent	Clay
Electricity for brick fabrication	kWh	37	ecoinvent	electricity voltage transformation from high to medium voltage
Output				
Brick	kg	3500		

(0.9 M) that retain metals as oxalates that could be subsequently transformed into a more marketable product (i.e. oxides) while the solvent is recycled. The energy required for the calcination of these oxalates is 2568 kWh/ton (Table 1). As a result, 43 kg of Cu and 5 kg of Zn are recovered.

3.2.3. *Phosphorus removal by pelletized sludge*

The flow of wastewater treated by the columns was the value reported by Sibrell et al. (2009); 169 L/min/m^2. Considering a column height of 1 m, the density of the material (1.9 ton/m^3) and a working period of 1 year, the total volume of wastewater treated per ton of sludge is around 46,750 m^3. The energy consumption required to treat the wastewater is 79,000 kW/h according to the value reported by Stokes and Horvath (2010) for a wastewater treatment plant in California.

Following the process described by Sibrell et al. (2009), the sludge was stripped with 0.1 M NaOH and the P precipitated was used to produce fertilizer. The elution flow was similar to that reported by these authors (i.e. 43 L/min/m^2) to increase the P concentration in the eluate while the elution time was proportional to the 1-year period considered (i.e. 9 h). As a result, 34 kg of NaOH solution was used as eluent. Then, 2 kg of $CaCl_2$ was added to precipitate the P as calcium hydrogen phosphate. During this step, 1.4 kg of calcium hydrogen phosphate is produced and 735 kg of this material is generated that can be used as fertilizer (Table 1).

3.2.4. *Brick fabrication using sludge*

The substitution of 20% of clay by sludge generated by the active treatment plant during brick manufacturing was explored. The total amount of sludge used was 1 ton while the energy consumption considered during the whole manufacturing process, including both the maturation and firing stages, was 87 kW/h (EC, 1998; Table 1). In total, 2.8 ton of clay were used to produce 3.5 ton of bricks.

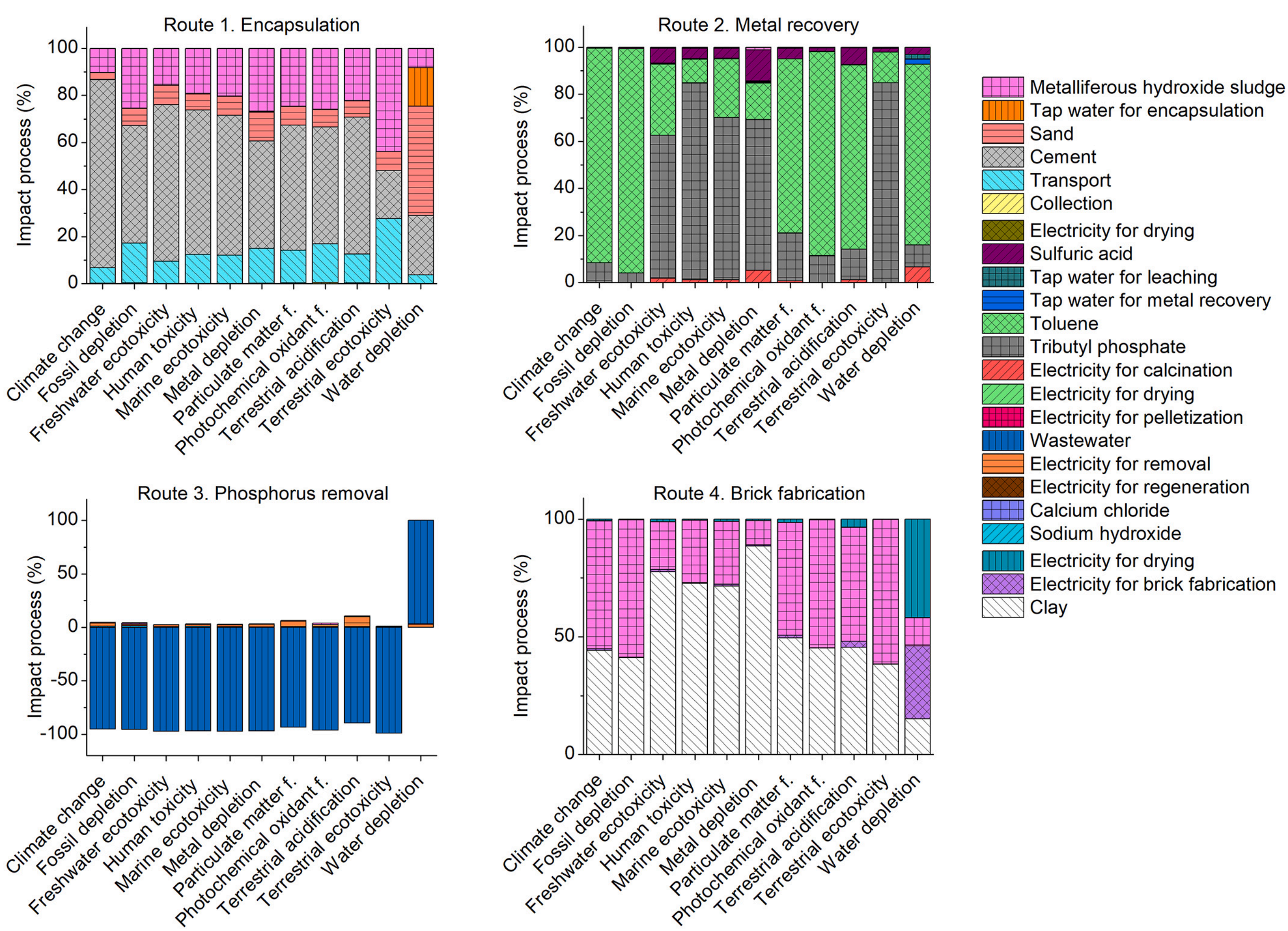

Fig. 2. Impact contribution of each process for the different routes explored.

4. Results and discussion

4.1. Relative impact contributions of the process to the different routes

The relative impact contributions of the different processes for each route were assessed in order to potentially improve the environmental performance of the treatments (Fig. 2). Cement manufacturing turned to be the main impact contributor of route 1, with values between 46 and 80% for the different categories except in terrestrial ecotoxicity (20%) and water depletion (25%). Sand obtaining is the process with higher impact in water depletion (value of 47%). The obtaining of this material, together with the treatment of the metalliferous hydroxide sludge also contributes to the impacts associated to other categories. Despite transport plays a minor role in impacts contribution for most categories, its contribution impacting terrestrial ecotoxicity may be considered as significant (around 28%).

The highest impacts associated to route 2 (metal recovery) are related to the use of toluene and TBP (Fig. 2), chemical reagents commonly used for the recovery of metals, with values higher than 79% in each impact category. Minor impacts were associated to the use of sulphuric and oxalic acids during the metal recovery scheme, especially in the category of metal depletion (values of 10% and 14%, respectively). This fact points out to the need of applying green chemistry procedures to extract metals from these materials. On the contrary, the route 3 (P removal) exhibited negative values in all categories except for water depletion due to the elevated use of wastewater. The use of electricity to prepare the pelletized material also caused positive impact values in this route (Fig. 2).

On the other hand, the use of clay and the treatment of metalliferous hydroxide sludge accounts for >94% of the impacts for route 4 (brick fabrication), except for water depletion, with values dropping to 27%. The electricity necessary for the sludge drying and the brick manufacturing phases is the main contributor to water depletion (value of 42% and 31%, respectively). Thus, this impact distribution highlights the need of using more sustainable materials and renewable energies during the manufacturing of bricks using waste materials.

4.2. Life cycle impact assessment

The environmental impacts generated by the different management/valorisation routes obtained with OpenLCA are shown in Table 2. The main environmental impacts associated to the encapsulation of wastes and disposal in landfills are mainly related to climate change (331 kg CO_2 eq), human toxicity (51 kg 1,4-dB eq) and fossil depletion (47 kg oil eq), metal and water depletion (6 kg Fe eq and 3 m^3, respectively). The route 2, based on metal recovery from the sludge, causes the greatest impact in all categories, with a significant impact on climate change (24,103 kg CO_2 eq), fossil depletion (20,943 kg oil eq), human toxicity (2441 kg 1,4-dB eq) or metal depletion (172 kg Fe eq). In the case of the route 3, based on the use of sludge to remove P, it stands out the impact associated to water depletion (value of 7663 m^3) due to the volume of water consumed in the life cycle. In turn, route 3 shows minimum impacts in other categories, having negative values in all of them due to the use of wastewater within its life cycle. The negative values usually express that the credits/benefits are larger than the burdens to the environment. In this case, recycling wastewater implies credits/benefits from the environment, because freshwater is not taken as a resource, and the phosphorus contained originally in water is removed. Comparing the impacts among different routes, the routes 1 (waste encapsulation and landfilling) and route 4 (brick fabrication) have much lower impact in all categories than route 3 (phosphorus removal) and to a lesser extent to route 2 (metal recovery).

Each impact indicator was normalized using regional Europe release per capita reference values (ReCiPe H Europe 2008 V1.13) to facilitate the comparison across impact categories. The normalized results (REQs) are expressed on a common scale providing information on the relative significance of the valorisation routes for each category indicator (Fig. 3 and Table SM1). In this sense, the highest impact is caused by route 2 in the fossil depletion impact category (value of 13 REQs), followed by marine ecotoxicity and freshwater ecotoxicity (value of 7 and 5 REQs, respectively). In addition, route 2 also generated low impacts in

Table 2
Impact results for the valorisation routes per ton of sludge. Route 1: Encapsulation and disposal in inert landfill; Route 2: Metal recovery using a chemical extraction scheme; Route 3: Phosphorus removal by pelletized sludge; Route 4: Brick fabrication using sludge.

Impact category	Unit	Route 1	Route 2	Route 3	Route 4
Climate change	*kg CO2 eq*	331	24,103	−3885	28
Fossil depletion	*kg oil eq*	47	20,943	−866	8.5
Freshwater ecotoxicity	*kg 1,4-dB eq*	1.1	50	−133	0.7
Human toxicity	*kg 1,4-dB eq*	51	2441	−2609	27
Marine ecotoxicity	*kg 1,4-dB eq*	1.2	64	−124	0.7
Metal depletion	*kg Fe eq*	6	172	−1050	14
Particulate matter formation	*kg PM10 eq*	0.4	24	−14	0.1
Photochemical oxidant formation	*kg NMVOC*	1.0	75	−19	0.2
Terrestrial acidification	*kg SO2 eq*	0.9	66	−34	0.2
Terrestrial ecotoxicity	*kg 1,4-dB eq*	0.03	12.9	−2.4	0.008
Water depletion	*m^3*	3.0	652	7663	1.8

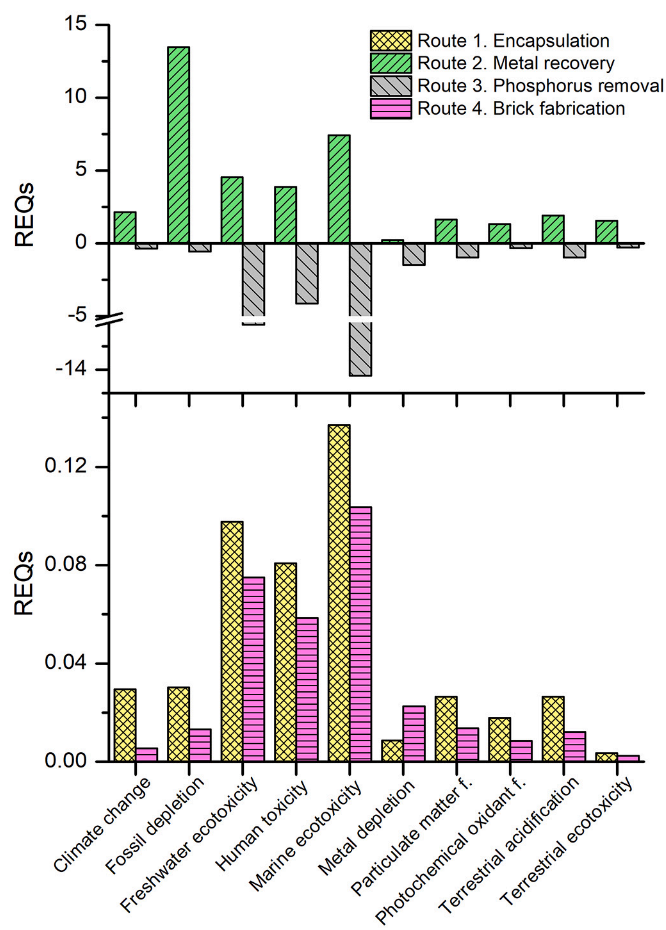

Fig. 3. Resident equivalents (REQs) per capita/yr for the valorisation routes, based on ReCiPe normalization reference values for Europe (Note: for water depletion the ReCiPe H Europe reference value is zero).

terrestrial ecotoxicity and metal depletion (value of 1.6 REQs and 0.2 REQs, respectively). In contrast, the route 3 exhibited the lowest impacts in the categories of marine and freshwater ecotoxicity (values of −14 REQs and − 12 REQs, respectively), due to the treatment of wastewater, as previously commented. Instead, the greatest impacts observed for route 3 are in the categories of terrestrial ecotoxicity, photochemical oxidant formation and climate change (values between −0.29 to −0.34). However, an overall benefit to the environment is observed for this route. Route 1 causes its higher impacts in the marine and freshwater ecotoxicity categories (value of 0.14 and 0.10 REQs, respectively), while the impact in terrestrial ecotoxicity category was low (value of 0.003 REQs). Finally, route 4 exhibited generally lower impact values than route 1, except for metal depletion which exhibited similar values. The greatest impact generated by this route was in the marine ecotoxicity category (value of 0.1 REQs), while the lowest was observed in terrestrial ecotoxicity (value of 0.002 REQs).

4.3. *Potential improvements to the environmental performance of the sludge treatment routes*

The availability of abiotic resources, such as water, minerals, or fossil fuels, is an issue related to their depletion for future generations (Harmsen et al., 2013; Tao et al., 2022). In this sense, more sustainable raw materials have to be used to avoid these effects and cause a lower environmental impact. The previous sections highlighted the impacts associated to several processes and raw materials within the different explored valorisation routes. As previously commented, cement manufacturing and sand obtaining caused the highest impacts during the encapsulation of sludge (Route 1). Cement production causes a high environmental impact and intensive energy consumption. Therefore, the modification of the process or the substitution of cement by a more environmentally friendly material would significantly reduce the impacts caused by this route. In this sense, Mikulčić et al. (2016) propose an improvement of the production system using alternative fuels and a more energy efficient kiln process. These actions made possible to reduce the consumption of fossil fuels and consequently reduce CO_2 emissions. On the other hand, coal ash and dicalcium silicate γ phase (γ-2CaO·SiO_2) could also be used as a substitute for cement, as proposed by Higuchi et al. (2014) for the manufacture of concrete. Thus, CO_2 generated by a power plant can be captured and reacts with γ-2CaO·SiO_2, hardening concrete. A more sustainable solution could be achieved by replacing the cement by other wastes with pozzolanic properties. In this sense, Dandautiya and Singh (2019) tested the joint use of fly ash and copper tailing as replacement of cement. These authors reported a significant environmental improvement, with a notable reduction in the selected midpoint categories (from 25 to 38%). More recently, Liu et al. (2021) used gold mine tailings rich in Si and Al oxides as precursors to produce geopolymers, achieving a marketable product containing up to 60% of tailings. Considering the abundance of this type of wastes within the mines, this latter option would improve notably the environmental (and economic) performance of this route.

The main impacts of route 2 were associated to the use of commercial chemical reagents, such as toluene and TBP. Therefore, the substitution of these products within a greener chemistry framework would help to reduce the impacts associated to the recovery of metals from the sludge. In this respect, the production of toluene from biomass is more environmentally sustainable that its obtaining by traditional chemical processes (Clark et al., 2015; Niziolek et al., 2016). A similar case is observed for TBP which conventionally is synthetized after the reaction of n-butanol with phosphorus oxychloride in batch/semi-batch process. However, more efficient processes would reduce the environmental impacts associated to the synthesis of this reagent. For example, Sen et al. (2020) proposed an improvement of the process using a continuous synthesis method in a micro-reactor. These authors reported a 100% yield for a residence time of 44 min at reaction temperature of 60 °C.

The utilization of the sludge to prepare pelletized materials leads to impacts associated to water removal and electricity. This process, which turned to be the most environmentally friendly of those studied, can be therefore improved by the reutilization of treated wastewater and the use of renewable energies. This latter impact is common to all explored valorisation routes, therefore, the use of renewable energies such as biomass or solar thermal and geothermal technologies in low and medium temperature may reduce the environmental impacts produced (IRENA, 2015).

The valorisation of sludge for brick manufacturing leads to impacts mainly associated to clay obtaining and energy required to treat the sludge. The environmental performance of the process can be notably improved if clay is partially or totally replaced. For that matter, Taha et al. (2016) propose substituting this raw material by treated calamine processing wastes. These authors obtained decreasing costs and waste generation while producing a brick which fulfils the requirements for construction. These facts highlight the need to evaluate individual valorisation schemes of generated sludge during waste water treatment in the mining industry as well as the potential to improve these schemes.

5. Conclusions

Metal-rich sludge generated by AMD treatment may be of environmental concern and requires proper management. In this study, 4 different management/valorisation routes have been proposed for this type of sludge and evaluated their environmental impacts. The evaluation of the routes studied by LCA allows to quantify the environmental impacts associated with each process and valorisation route. In this case, the route with the lowest impacts was route 3, focused on the manufacturing of pelletized material to remove phosphorus from agricultural/urban wastewaters while producing a fertilizer product. This route has mainly net negative impact values due to the environmental benefits of wastewater treatment. However, the highest impacts were associated to water depletion (value of 7663 m^3) and energy consumption. The recovery of valuable metals from sludge has an undoubtedly economic interest, however exhibited the highest environmental impacts in the categories of fossil depletion, marine and freshwater ecotoxicity (values of 6–14 REQs) due to the use of toluene and TBP (impact>79%). An improvement of different processes within this route, such as the production of toluene from biomass or the use of continuous TBP synthesis method in a microreactor would cause a significant reduction of environmental impacts. The other two routes exhibited much lower impacts than the route focused on metal recovery (values <1 REQs). The manufacture of bricks could also be an alternative route by giving economic value to wastes while exhibiting low environmental impacts, mainly on marine and freshwater ecotoxicity (value of 0.7 kg 1,4-dB eq). These impacts can be notably reduced if waste materials are used replacing clays with wastes such as calamine processing wastes or using renewable energies. The encapsulation of these wastes is also a suitable option with a low environmental impact, but slightly greater than the brick manufacturing and with no economic return. The substitution of cement by wastes or more environmentally friendly materials can be a good alternative to reduce these impacts.

The results obtained in this work can be used to support the decision-making process for the sustainability of waste management after the AMD treatment. In this sense, the environmental performance of any proposed action must be assessed using a LCA approach, considering all environmental impacts of materials and processes. An adequate valorisation of the large amount of waste produced would contribute to more environmentally friendly mining. In addition, an important economic profit can be obtained from sludge valorisation, although these processes should be improved to reduce the impacts. This study proposes an interesting approach to improve the environmental performance of sulphide mining worldwide, however some issues remained unsolved, such as a carefully life cost analysis of proposed measures or the inclusion of endpoint indicators.

CRediT authorship contribution statement

Raúl Moreno-González: Writing – original draft, Methodology, Visualization, Writing – review & editing. **Francisco Macías:** Formal analysis, Validation, Writing – review & editing. **Andreas Meyer:** Methodology, Visualization, Writing – review & editing. **Petra Schneider:** Methodology, Visualization, Writing – review & editing. **Jose Miguel Nieto:** Funding acquisition, Investigation, Supervision, Writing – review & editing. **Manuel Olías:** Conceptualization, Funding acquisition, Investigation, Supervision, Writing – review & editing. **Carlos Ruiz Cánovas:** Conceptualization, Funding acquisition, Investigation, Supervision, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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