LCA of Greywater Management within a Water Circular Economy Restorative Thinking Framework

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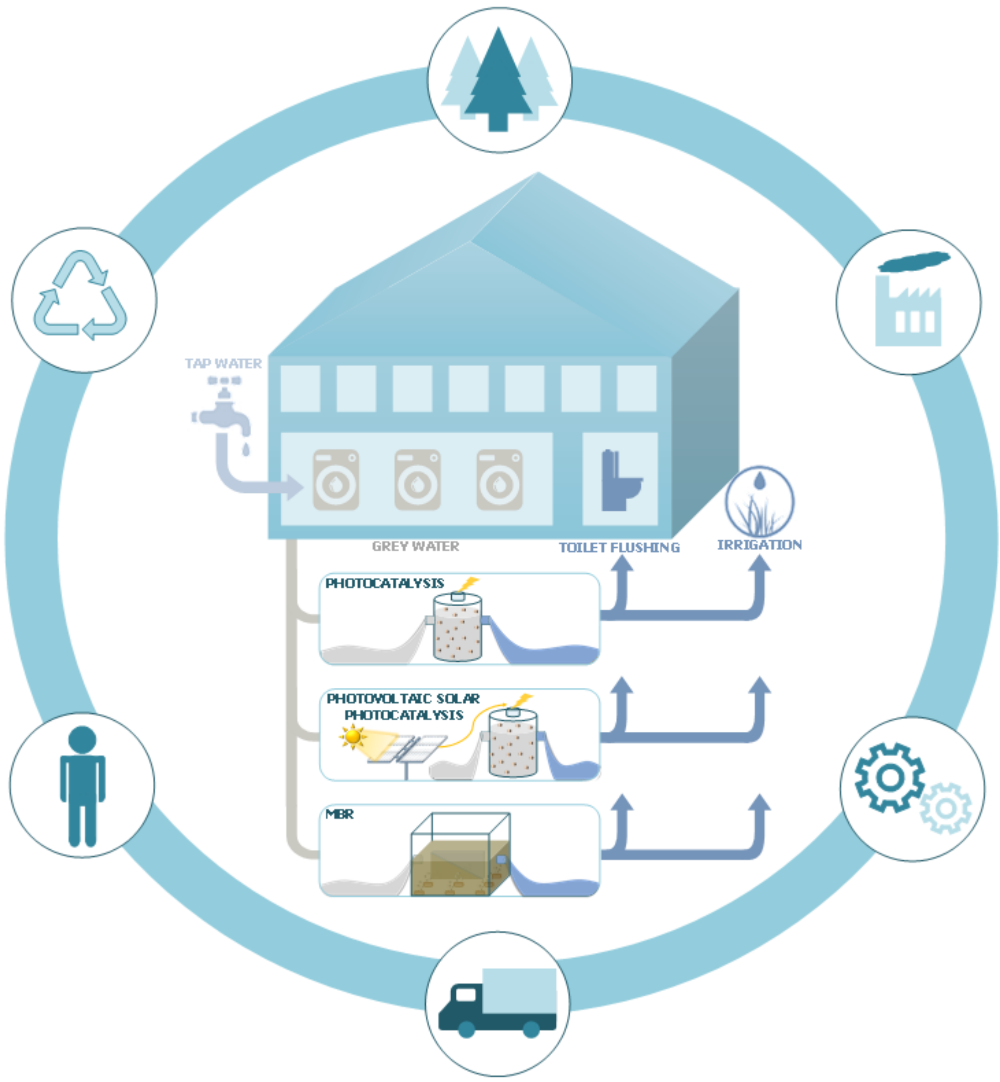
GRAPHICAL ABSTRACT



ABSTRACT

Greywater reuse is an attractive option for the sustainable management of water under water scarcity circumstances, within a water circular economy restorative thinking framework. Its successful deployment relies on the availability of low cost and environmentally friendly technologies. The life cycle assessment (LCA) approach provides the appropriate methodological tool for the evaluation of alternative treatments based on environmental decision criteria and, therefore, it is highly useful during the process conceptual design. This methodology should be employed in the early design phase to select those technologies with lower environmental impact. This work reports the comparative LCA of three scenarios for greywater reuse: photocatalysis, photovoltaic solar-driven photocatalysis and membrane biological reactor, in order to help the selection of the most environmentally friendly technology. The study has been focused on the removal of the surfactant sodium dodecylbenzenesulfonate (SDBS), which is used in the formulation of detergents and personal care products and, thus, widely present in greywater. LCA was applied using the Environmental Sustainability Assessment (ESA) methodology to obtain two main environmental indicators in order to simplify the decision making process: natural resources (NRs) and environmental burdens (EBs). Energy consumption is the main contributor to both indicators owing to the high energy consumption of the light source for the photocatalytic greywater treatment. In order to reduce its environmental burdens, the most desirable scenario would be the use of solar light for the photocatalytic transformation. However, while the technological challenge of direct use of solar light is approached, the environmental suitability of the photovoltaic solar energy driven photocatalysis technology to greywater reuse has been demonstrated, as it involves the smallest environmental impact among the three studied alternatives.

Keywords

Life cycle assessment (LCA); Light emitting diodes (LEDs); Membrane biological reactor (MBR); Photocatalysis; Photovoltaic solar energy.

1. Introduction

The economic, environmental, and social impact of past water resources development and the present water scarcity lead to a new paradigm in water resource management~~,~~. Therefore, the application of sustainable water supply solutions is essential (Ortiz et al., 2015; Wilcox et al., 2016). In this scenario, the implementation of a circular economy strategy results in a promising approach. This concept~~,~~ has been already introduced in several environmental policy initiatives of the European Commission (EC) (European Commission, 2017a). The circular economy restorative thinking demands that wastewater should be considered a valuable non-conventional resource used to sustain scarce life-essential resources (Abu-Ghunmi et al., 2016). Therefore, the development of wastewater recycling systems has gained attention over the last years (Guo et al., 2014; Holloway et al., 2016; Wilcox et al., 2016). However, limited awareness of potential benefits among stakeholders and the general public, and lack of a supportive and coherent framework for water reuse are the major barriers currently preventing a wider spreading of this practice in the EU. For these reasons the EC is working on legislative or other instruments to boost water reuse when it is cost-efficient and safe for health and the environment (European Commission, 2017b).

One of the most interesting alternatives is the on-site treatment and reuse of greywater in households, hotels, and sport centers (Fountoulakis et al., 2016; Gabarró et al., 2013; March et al., 2004; Merz et al., 2007; Sánchez et al., 2010). Greywater is domestic wastewater originated in washing machines, kitchen sinks, baths, and hand basins. Spanish law allows its recycling under several circumstances (Real Decreto 1620/2007, 2007). Hence, it is adequate for toilet flushing, irrigation, laundry, fire extinguishing, groundwater discharge or car and window washing (Ghunmi et al., 2011; Liberman et al., 2016; Santasmasas et al., 2013). This kind of water contains surfactants, which are compounds commonly used in the formulation of detergents and personal care products that represent an environmental hazard due to their low biodegradability and their ability to provoke foams (Suárez-Ojeda et al., 2007). One of the most representative surfactants is the sodium dodecylbenzenesulfonate (SDBS) (Dominguez et al., 2016; Sanchez et al., 2010; Sanchez et al., 2011). Several methods have been considered for greywater treatment in literature including biological, chemical, and physico-chemical processes (Ghunmi et al., 2011). However, most of these techniques are ineffective for the total removal of surfactants or they can only transport these contaminants to a different phase resulting in a concentrated waste volume (Dhouib et al., 2005). One of the most environmentally friendly options is the use of constructed wetlands, however, their use is limited by the requirement of large land spaces (Ghunmi et al., 2011).

Advanced oxidation processes (AOPs) have been presented as environmentally friendly treatments for wastewater remediation; they achieve the successful degradation of different contaminants of emerging concern (CECs) (Dominguez et al., 2016; Rodríguez et al., 2016; Serra et al., 2011; Wankhade et al., 2013). AOPs are based on the in situ generation of reactive oxidizing species (ROS), mainly hydroxyl radicals ($^{\bullet}OH$) (Fernández-Castro et al., 2014; Muñoz et al., 2006). Among them, heterogeneous photocatalysis appears as an attractive emerging technology to treat greywater because it avoids secondary pollution and works at ambient temperature and pressure (Dominguez et al., 2016). As seen in Eq. 1, in this process a source of appropriate light (hʋ) and a solid semiconductor material, the photocatalyst, are necessary to promote the mineralization of the organic pollutant (Kumar and Bansal, 2013).

$$\text{Organic pollutant} \xrightarrow{\text{Photocatalyst / h}\upsilon} \text{Intermediate compounds} \longrightarrow CO_2 + H_2O \quad (1)$$

Solar light is the most environmentally friendly light source (Fig. 1) and solar-assisted photocatalysis has shown positive results over the last years in the removal of emerging contaminants (Malato et al., 2016). However, several barriers still need to be overcome for its full implementation worldwide (Spasiano et al., 2015). First, the solar UV spectral irradiance reaching the Earth's surface is not homogeneous. Another disadvantage already reported is that large areas might be required for the treatment (Muñoz et al., 2006). Furthermore, the most commonly employed photocatalyst, TiO_2, is only excited for radiations in the ultraviolet region (UV), which only represents about 3.00–4.00% of the solar spectrum (Spasiano et al., 2015). Thus, the effective application of TiO_2 photocatalysts to the removal of recalcitrant compounds requires artificial illumination (Ibhadon and Fitzpatrick, 2013). Hg lamps have known ample use in laboratory studies; however, they have low efficiency in the transformation of energy into light and short useful life, thus, making photocatalysis energy intensive. The use of light emitting diodes (LEDs) provides a more energy efficient alternative with longer useful life and lower price than the traditional photocatalytic mercury lamps (Song et al., 2016). Besides, the use of solar photovoltaic panels as primary energy source (Dominguez-Ramos et al., 2010) appears as the ultimate goal to convert photocatalysis into a sustainable treatment.

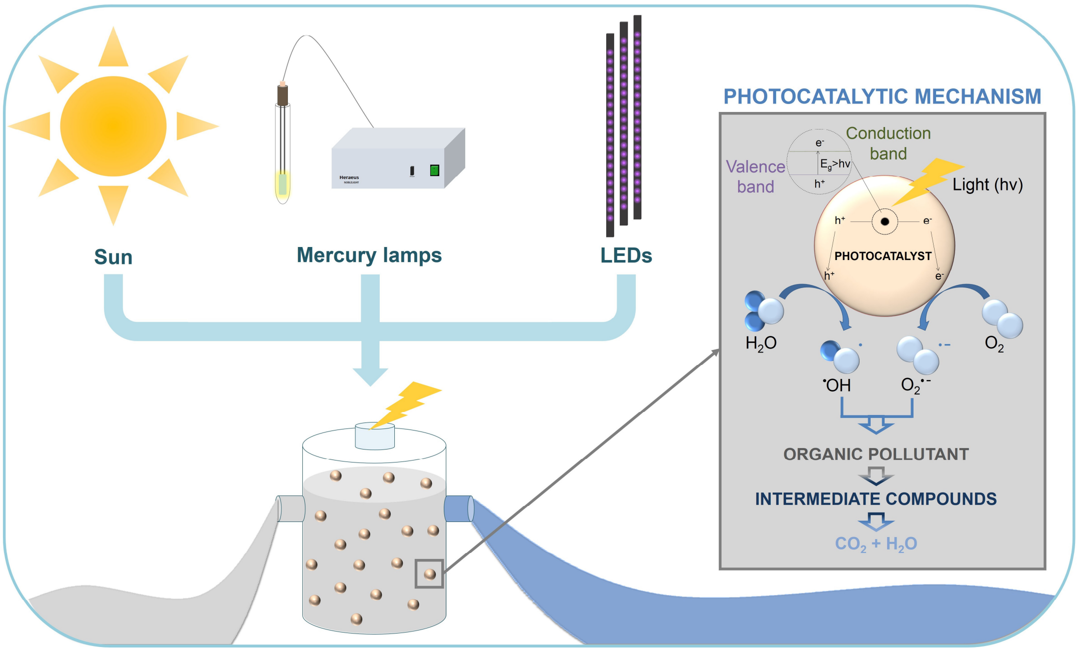


Fig. 1. Light source alternatives in photocatalysis.

Another promising technical alternative to treat greywater consists in the use of membrane biological reactors (MBR), which combine traditional activated sludge biological treatment with membrane filtration (Atanasova et al., 2017; Chai et al., 2013; Fountoulakis et al., 2016; Gander et al., 2000; Merz et al., 2007). This technology provides high efficiencies in the removal of surfactants, good effluent quality, high mixed liquor suspended solids concentrations, small space requirements, and reduced sludge production (Chai et al., 2013; De Gisi et al., 2016; Dhouib et al., 2005; Gander et al., 2000; Merz et al., 2007). Both, photocatalysis and MBR, have shown their suitability for the treatment of greywater (Sánchez et al., 2010; Santasmasas et al., 2013). Nevertheless, their deployment generates an environmental impact associated with an intensive use of resources (chemicals and energy) and the construction of the required infrastructures (Giménez et al., 2015; Rodríguez et al., 2016; Ortiz et al., 2007). Thus, the application of the above-mentioned technologies should be preceded not only by evaluation of the degradation and mineralization yield, but also by the complete environmental assessment (Chatzisymeon et al., 2013; Giménez et al., 2015; Rodríguez et al., 2016). In this sense, Life cycle assessment (LCA) appears as a reliable methodology to define, evaluate, quantify and reduce the potential impacts of the lifecycle stages (from 'cradle' to 'grave') of a product, activity or process (Corominas et al., 2013; Margallo et al., 2014a; Serra et al., 2011), supporting the environmental decision-making process (García-Herreo et al. 2017a). The inputs and outputs of the system, such as energy, reagents, materials, emissions, waste, and environmental impacts are quantified in LCA (Chong et al., 2010, Serra et al., 2011). The implementation of the LCA tool in green chemistry processes supports the development of more sustainable concepts based on the relationship between the selection of compounds and process parameters and the resulting environmental impacts (Kralisch et al., 2015). While LCA has been widely applied to MBR treatments (Ortiz et al., 2007; Pretel et al., 2016; Zang et al., 2015), only scarce studies evaluating the environmental performance of photocatalysis can be found in literature (Chatzisymeon et al., 2013; Giménez et al., 2015; Muñoz et al., 2005). Furthermore, it is worth remarking that most LCA studies applied to photocatalytic treatments are performed in lab scale, which unquestionably limits the usefulness of the results regarding the real large-scale application (Chatzisymeon et al., 2013; Giménez et al., 2015; Muñoz et al., 2005).

Within these premises, this work provides an LCA study to assess and compare the environmental impacts generated in the treatment of greywater by photocatalysis, photovoltaic solar-driven photocatalysis, and MBR. It will also identify the environmental bottlenecks in order to address the main technological challenges for greywater reuse.

2. Methodology

LCA is carried out according to the requirements of the ISO 14040 and ISO 14044 international standards (ISO, 2006a; ISO, 2006b). Therefore, LCA is applied in the following stages: definition of the goal and scope of the study, development of the life cycle inventory (LCI), life cycle impact assessment (LCIA) and results interpretation.

2.1. Goal and scope

This research aims to assess the environmental sustainability of three alternatives for greywater treatment, photocatalysis, photovoltaic solar-driven photocatalysis, and MBR. It provides an appropriate framework to evaluate the opportunities for process success leading also to the identification of hot-spots, which are the stages with the highest environmental impact. The purpose of the system is to treat greywater with high degree of removal of SDBS, allowing its reuse for toilet flushing and garden irrigation. SDBS has been selected as target pollutant due to its environmental persistence and because the treatment is applied to hotel laundry greywater, where SDBS is a key component. Thus, the functional unit is defined on the basis of the same treated volume of greywater and the same amount of SDBS removed. In order to establish the amount of SDBS removed, a minimum threshold accomplished by the three scenarios within a given treatment time has to be selected (Muñoz et al., 2005). Therefore, 1.00 m^3 of treated greywater with 90.0% reduction of the SDBS initial concentration is designated as functional unit. All the mass and energy inputs and outputs will be referred to this unit. The use of a similar functional unit that considers the same treated water volume and a fixed reduction level of the contaminant has been previously reported in literature. For instance, Muñoz et al. (2005) defined as functional unit the removal of 15.0% DOC from 1.00 m^3 kraft pulp mill wastewater, and Serra et al. (2011) selected as functional unit the removal of 93.0% total organic carbon in 250 mL of wastewater with 500 mg L^{-1} of α-methyl-phenylglycine.

The study is carried out from a 'cradle to gate' pathway, considering the extraction, production, and transportation of raw materials, the greywater treatment and the management of generated waste. This approach is developed for three scenarios based on photocatalytic, photovoltaic solar photocatalytic, and MBR technologies.

Scenario 1 (Sc. 1), photocatalytic technology: photocatalytic studies were performed in laboratory to obtain kinetic data, and after modeling the process, scale-up was carried out. The commercial photocatalyst used is TiO_2 Aeroxide® P25 (Evonik Industries). One g L^{-1} of TiO_2 was added to the effluent and kept for 0.50 h premixing in the dark to reach adsorption equilibrium before the photocatalytic treatment started. The photocatalyst loading was selected after the results attained in preceding works (Dominguez et al., 2016). The photocatalytic reactor (APRIA Systems S.L. Photolab LED/160) is constituted of 1.00 L jacketed annular reactor, 5.00 L mixing tank and 40 LEDs LZ1-00U600 (LED Engin). LEDs emit in a wavelength between 365 nm and 370 nm, being the total electrical power between 1.00 W and 100 W. A fan (San Ace 80, Sanyo Denki) is used to keep LEDs temperature in the suitable range (20.0 ºC - 30.0 ºC) to keep constant radiation over time and high lamp lifetime. SDBS concentration is quantified by means of an UV-1800 spectrophotometer (Shimadzu) at 223 nm. The waste TiO_2 obtained after the photocatalytic treatment is sent to a municipal landfill.

Scenario 2 (Sc. 2), photovoltaic solar-driven photocatalysis: the photocatalytic studies detailed in Sc. 1 were used for process scale-up as well. Since the existing photovoltaic panels have different materials and processing requirements that lead~~s~~ to diverse emission profiles, a global average share of different photovoltaic panels is considered;

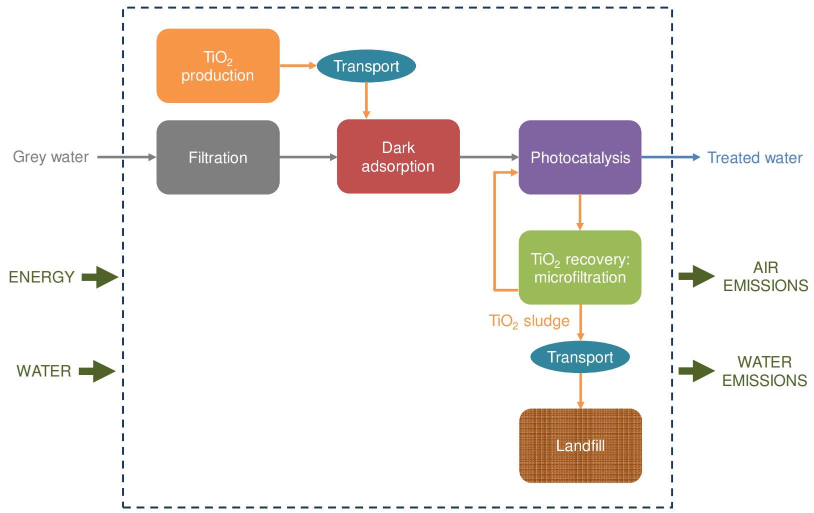
these include mono-silicon 47.7%, multi-silicon 38.3%, cadmium-telluride 6.4%, amorphous-silicon 5.10%, ribbon-silicon 1.50%, and copper-indium-gallium-diselenide 1.00%.

Scenario 3 (Sc. 3), MBR technology: all the data have been collected from literature. The selected MBR has a submerged configuration in order to reduce energy consumption (Khan et al., 2016). The membrane is a flat sheet ultrafiltration polyethersulfone membrane with 50 nm and a permeate flux of 19.2 L m^{-2} h^{-1} (Santasmasas et al., 2013). The hydraulic retention time (HRT) is estimated as 25.3 h (Santasmasas et al., 2013). It is assumed that the sludge retention time (SRT) is 35 days (Gori et al., 2010) because high SRTs cause endogenous respiration in the biomass reducing the sludge production (Gander et al., 2000). For the biomass conditions an average mixed liquor total suspended solids (MLTSS) of 8.00 g L^{-1} is taken as reference (Gori et al., 2010). The sludge is supposed to be treated by incineration and then deposited in a municipal landfill, which is one of the most common processes in the wastewater area. However, an alternative option consisting on sludge compost-stabilization for its land application has been also analyzed (Sc. 3b).

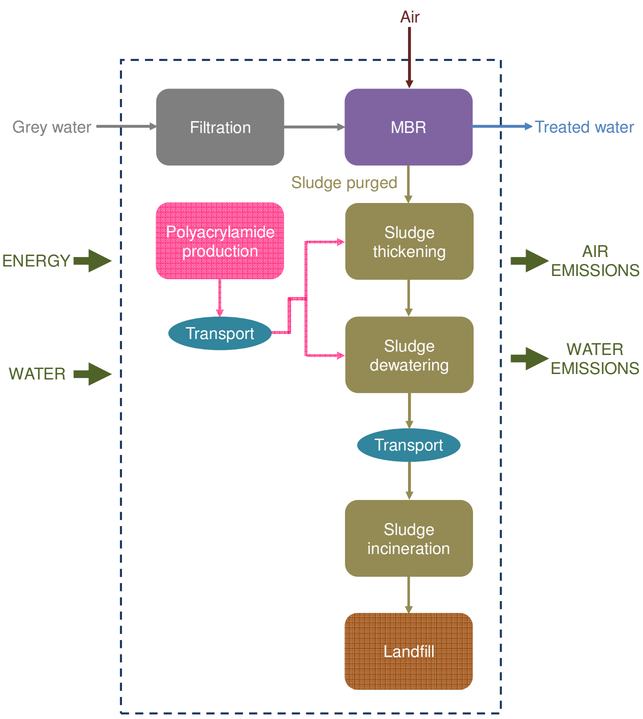
Fig. 2 shows the flow diagram and the system boundaries considered for each treatment. The main system flows are the energy inputs, water, manufacture of the reagents used in each treatment (extraction of resources, manufacture, and transport) and their outputs to the environment. The systems boundaries for Sc. 1 and Sc. 2 are the same since the only difference between both scenarios is the method to obtain the required energy, being the electricity grid in Sc. 1 and renewable energy in Sc. 2.

It is to be highlighted that in order to simplify the LCA application, the infrastructure related to the three greywater treatments has not been considered (Giménez et al., 2015). Moreover, the contribution of the infrastructure to the impacts of these processes is typically negligible owing to the long lifetimes of the considered industrial installations and because its impact is insignificant compared to the impact produced by the operation phase (Garcia-Herrero et al., 2017a; Hospido et al., 2012).

a).



b).



c).

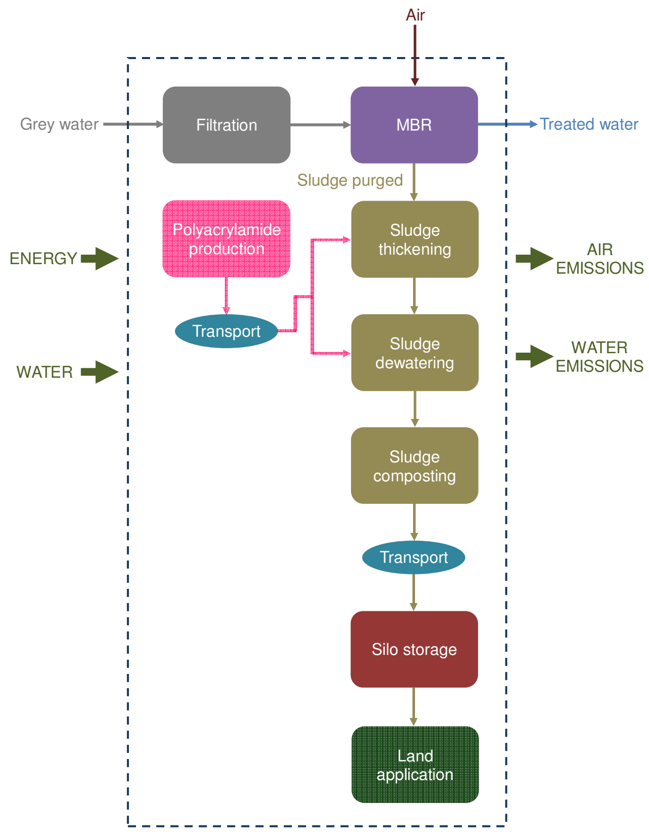


Fig. 2. System boundaries for greywater treatment in a). Sc. 1 (photocatalysis) and Sc 2. (photovoltaic solar-driven photocatalysis), b). Sc. 3 (MBR with sludge incineration) and c). Sc. 3b (MBR with sludge composting).

The three scenarios are multi-functional processes, in which greywater treatment is the main function, and the recovery of energy in the landfill site and in the incinerator are additional functions. Furthermore, a modification of Sc. 3 has been set out in the MBR variation assessment. In this case (Sc. 3b), after composting, the sludge is used as fertilizer, adding a new function to the system. In these systems, the environmental burdens associated with a particular process must be partitioned over the various functional flows of that process (Margallo et al., 2014b).

According to the ISO recommendation, this work solved the existence of additional functions gaining credit by the reduction of the emissions related to the co-products. That is to say, the impact of the co-product manufacturing is subtracted from the original systems. In this case, for energy and material valorization, the 'avoided' emissions of conventional production of electricity and fertilizer were subtracted from those produced during waste treatment.

This procedure requires identifying the type of material substituted or displaced. In Sc. 3, the energy mix is the substituted process, whereas in Sc. 3b the displaced fertilizer is ammonium sulfate.

2.2. Life cycle inventory

The mass and energy flows considered within the scope of the work are recorded in the life cycle inventory (LCI), which collects the most relevant input and output data for the scenarios under study in separate unit processes. In this work the data are taken either from fieldwork (Dominguez et al., 2016) or from literature; the sources and quality of the LCI per functional unit are depicted in Table 1, and detailed in Table 2. The natural resources consumption and the environmental burdens associated to the systems can be estimated from these values.

The main hypothesis assumed in the inventory phase of the LCA can be summarized as follows:

- For the process scale-up and estimation of energy consumption, reagents, and waste, both scenarios are assumed to be implemented in a hotel laundry to treat greywater with 50.0 mg L^{-1} of SDBS.
- The treatment is assumed to be carried out in Santander, Cantabria, Spain, in a hotel of 75 guests.
- It is assumed that each guest produces 1 kg of laundry per day, including 2 bed sheets, 1 pillow slip and 1 towel (Filimonau et al., 2011), and that 13 L of fresh water are required to wash 1 kg of laundry (Máša et al., 2013).

- The photocatalytic treatment works in batch mode, 20.6 h day^{-1}, all year round. This time has been extrapolated from the results previously obtained at laboratory scale by the authors (Dominguez et al., 2016).
- The MBR works in continuous mode, with a hydraulic retention time (HRT) of 25.6 h. This value has been estimated taking into account data taken from literature (Santasmasas et al., 2013).
- To improve data quality and consider the local idiosyncrasy in Sc. 1 and Sc. 3, the electricity mixed provided by the PE database is adapted to the characteristics of the Spanish mix of 2016, which contains 40.5% of renewable sources and 35.3% of fossil fuel based sources.
- The energy employed in Sc. 2 is taken from photovoltaic solar panels.
- The electricity consumption corresponds to a treatment time required to remove 90.0% of the initial SDBS concentration, being 19.5 h for photocatalysis and 25.6 h for MBR. Moreover, in the case of the photocatalytic treatment an additional time of 0.50 h has been considered for dark adsorption of the photocatalyst, 0.14 h for pumping the greywater to the system and 1.00 h for pumping the treated water during the TiO_2 separation step.
- A photocatalyst recovery stage by means of microfiltration membranes has been taken into account in the case of the photocatalytic treatment (Rivero et al., 2006). It is assumed that the TiO_2 is fully recovered and it can be reused 10 times in a closed cycle (Muñoz et al., 2006). Then it is disposed of in landfill; it should be transported along 32.8 km by a 28 tones Euro 4 truck.
- TiO_2 is delivered to the consumer after transport by a Euro 4 truck with a maximum total capacity of 28 tones along 1596 km from the production plant of

Evonik Industries in Frankfurt, Germany (Evonik Industries, 2017; Muñoz et al., 2005).

- The manufacturing of the membranes is considered as part of the infrastructure and, therefore, it is not considered in this work.
- In the MBR treatment the membrane-cleaning step is based on air scouring avoiding backwashing cycles or the use of chemicals (Liberman et al., 2016).
- The data used for the sludge treatment are recompiled from literature (Hospido et al., 2005; Suh and Rousseaux, 2002). The sludge is thickened and dewatered on-site; the addition of polyacrylamide is required in both stages. Then, it is transported by a 28 tones Euro 4 truck along 32.8 km to an incineration plant located in a landfill site placed in Meruelo, Cantabria, Spain, where it is treated and disposed of (Suh and Rousseaux, 2002).
- The polyacrylamide is transported by a 28 tones Euro 4 truck along 722 km after its manufacture in a plant of Derypol, S.A. in Les Franqueses del Vallés, Spain (Derypol, 2017).

2.3. Life cycle impact assessment

The life cycle impact assessment (LCIA) calculates environmental indicators from the LCI data. It implies further classification and characterization of these indicators, including their additional and non-mandatory normalization and weighting (Garcia-Herrero et al., 2017b). In this work, the software selected for the modeling of the two treatments under study is the LCA software GaBi 6.0 and the database of PE International (PE International, 2016).

The Environmental Sustainability Assessment (ESA) method followed in this work was initially developed by Irabien et al. (2009). Accordingly, a first classification stage is performed in which the inventory data are organized in different impact categories. Then, the possible impact of each resource consumption or emission is estimated using a characterization factor (CF) (Garcia-Herrero et al., 2017b).

To conduct the environmental assessment, the two main indicators considered are the natural resources consumption (NRs) and the environmental burdens (EBs). The consumption of energy ($X_{1,1}$), materials ($X_{1,2}$), and water ($X_{1,3}$) are considered within the NRs and the primary burdens to air ($X_{2,1}$), water ($X_{2,2}$), and land ($X_{2,3}$) are included in the EBs. These indicators are based on the environmental sustainability metrics established by the Institution of Chemical Engineers (IChemE, 2002). Specifically, the EBs are classified in 12 impact categories. The atmospheric burdens are atmospheric acidification (AA), global warming (GW), human health effects (HHE), photochemical ozone formation (POF), and stratospheric ozone depletion (SOD). The impact categories for the water burdens are aquatic acidification (AqA), aquatic oxygen demand (AOD), ecotoxicity to aquatic life (metals to seawater) (MEco), ecotoxicity to aquatic life (other substances) (NMEco), and eutrophication (EU) (García et al., 2013). For the land burdens the categories are given by the amount of hazardous and non-hazardous waste produced and its management (Margallo et al., 2014a).

Since the environmental sustainability indicators employed in this study are expressed in different units depending on the environmental impact category considered, their normalization is recommended. Therefore, with the purpose of conducting a comparison in a common basis, dimensionless impacts indicators are required (Garcia-Herrero et al., 2017a). The NRs are normalized regarding the natural resource with the highest impact and the EBs regarding the threshold values specified in the European Pollutant Release and Transfer Register (E-PRTR, 2006).

Eq. (2) and Eq. (3) show the calculations used to normalize the NRs and EBs:

$$X^*_{1,i} = \frac{X_{1,i}}{X^{ref}_{1,i}} \quad (2)$$

$$X^*_{2,j,k} = \frac{X_{2,j,k}}{X^{ref}_{2,j,k}} \quad (3)$$

where "i" represents the NRs indicators (energy, materials, and water), "j" symbolizes the environmental compartments (air, water, and land) and "k" designates the environmental impacts to the corresponding compartment.

Then, $X_{1,i}$ represents the consumption of each NRs, $X^*_{1,i}$ is the normalized $X_{1,i}$, $X^{ref}_{1,i}$ is the reference natural resource, $X_{2,j,k}$ designates the environmental burdens to the corresponding compartment, $X^*_{2,j,k}$ is the normalized $X_{2,j,k}$, and $X^{ref}_{2,j,k}$ is the reference environmental burden.

After normalization, a weighting stage is developed. This procedure ranks the different impact categories taking into account their relative importance (EC JCR, 2010). Thus, the normalized NRs and EBs variables are aggregated as shown in Eq. (4) and Eq. (5):

$$X_1 = \sum_{i=1}^{i=n} \alpha_{1,i} \cdot X^*_{1,i} \quad n \in [1, 3] \quad (4)$$

$$X_{2,j} = \sum_{k=1}^{k=m} \beta_{2,j,k} \cdot X^*_{2,j,k} \quad m \in [1, 5] \text{ if } 1 \leq j \leq 2 \wedge m \in [1, 2] \text{ if } j = 3 \quad (5)$$

where $\alpha_{1,i}$ is the weighting factor for the NRs and $\beta_{2,j,k}$ is the weighting factor for the EBs.

In this work it is considered that the three natural resources are equally important, then $\alpha_{1,i}$ is 1/3 for each i. This assumption is taken as it is the best way to obtain a single indicator that allows comparison of the three proposed greywater treatments (Margallo et al., 2014a).

3. Results and discussion

3.1. Natural resources

The consumption of NRs, including energy ($X_{1,1}$), materials ($X_{1,2}$), and water ($X_{1,3}$), is analyzed for all the scenarios. The results are normalized regarding the natural resource with the highest impact, which is water for the three scenarios (Table 3).

The energy ($X_{1,1}$) embraces the consumption of electricity, steam, diesel, and natural gas. Sc. 2 is the most energy intensive, bringing the total energy demand close to 1304 MJ, while in Sc. 1 and Sc. 3 the energy demand is 450 MJ and 162 MJ, respectively. As it can be observed in Table 4, 99.5% of the energy consumed in Sc. 1 and 99.84% of the one required by Sc. 2 is demanded by the photocatalytic process. This is mainly due to the intensive energy demand of the light source, which represents the main hot-spot of the system. Therefore, the influence to $X_{1,1}$ of cleaning water and transport, production, consumption, and end of life of TiO_2 are below 0.50% in the three scenarios and, thus, it can be considered negligible. It has to be highlighted that in Sc. 3 the $X_{1,1}$ takes negative values in the sludge treatment stage due to the fact that during incineration thermal energy is produced.

Within the materials ($X_{1,2}$), TiO_2 is considered for Sc. 1 and Sc. 2 while air and polyacrylamide are taken into account for Sc. 3. Nevertheless, it is necessary to assess not only the amount of materials but also the toxicity and environmental impacts of their production and consumption. This point will be analyzed in the next section by means of the study of the environmental burdens. The results show that the demand of materials associated to the primary energy transformation is the major contributing factor to this indicator. The consumption of material resources is significantly higher in Sc. 3, 2481 kg, than in Sc. 1, 77.1 kg, and Sc. 2, 24.1 kg. The main reason behind this result lies on the high demand of air required by the MBR, implying high consume of materials for the energy required in the aeration process. This behavior was also previously reported in literature, where the energy consumption required for the aeration is the parameter that has the most significant influence in the environmental performance of biological reactors (De Feo and Ferrara, 2017). In Sc. 1, although the consumption of materials in the photocatalysis represents 92.4% of the indicator, cleaning water and TiO_2 production have contributions of 6.66% and 0.79%, respectively. In the case of Sc. 2, the intake of materials in the photocatalysis diminishes to 69.2%, while the TiO_2 production increases to 11.0% and the cleaning water to 19.5%. In Sc. 3, the production of polyacrylamide, the sludge treatment and the transport have a contribution below 0.01% to $X_{1,2}$, because the aeration required by the MBR has a contribution near 100%.

Despite the fact that water consumptions for the reagents production and for cleaning are included within the indicator $X_{1,3}$, the hot-spot is the water required for the primary energy transformation. According to Table 4, the value ranges from 96.8~~1~~% for Sc. 2 to 100% for Sc. 3, being the contribution of other stages to the indicator $X_{1,3}$ minimal.

Consistent with the results, Sc. 3 has the greatest global consumption of NRs (X_1 = 0.57), displaying a value 1.62 times higher than Sc. 1 (X_1 = 0.35) and 1.46 times higher than Sc. 2. This behavior is mainly due to the high energy consumption in the aeration (2072 m^3 m^{-3} greywater).

3.2. Environmental burdens

The environmental burdens to air and water are estimated following the methodology explained above. Before the normalization process, global warming represents the highest impact in all the scenarios. The main reason is the emission of greenhouse gases during energy production (CO_2, CO, etc.), the consumption of coal and energy in the manufacture of reagents (CH_4, CO, CO_2, NO_X, N_2O), diesel consumption and production and landfill emissions (NO_X, N_2O), and the transport of reagents and wastes (NO_X, N_2O). It is worth noticing that in the energy consumption for the Sc. 1 and Sc. 3, the electricity grid mix selected might have an important impact on the quantity of greenhouse gas emissions and in derived results (De Feo and Ferrara, 2017). Therefore, as it was previously specified, the Spanish mix of 2016 is selected for both scenarios. The smallest score for this environmental burden is obtained in Sc. 2 (2.14 kg CO_2 eq.) being almost 6-fold smaller than in Sc. 1 (12.7 kg CO_2 eq.) and 2-fold smaller than in Sc. 3 (4.42 kg CO_2 eq.). Regarding the aquatic indicators, the EU has the highest impact on the three scenarios before the normalization owing to the emissions of nitrogen, ammonia, phosphate, and chemical oxygen demand during energy production.

Table 5 shows the EBs to air and water normalized using the European threshold values (E-PRTR, 2006). After normalization, the HHE and POF become the most important categories among air metrics for the three scenarios. The principal reason is that, although GW has the highest air impact, when it is referenced to its threshold value ($1.00 \cdot 10^8$ kg CO_2 eq.) the normalized results are reduced by 8 orders of magnitude. Nonetheless, lower thresholds for HHE and POF are used as reference (1000 kg benzene eq. and 1000 kg. ethylene eq., respectively). In the case of water impacts, there are no significant differences after the normalization process because the threshold values are lower than those in the air categories.

The EBs to air in Sc. 2 are smaller than in the other two scenarios for all the indicators with the exception of the HHE. This high contribution to human toxicity in Sc. 2 is due mainly to the extraction of raw materials and the manufacturing of components for the photovoltaic solar panels fabrication. For instance, regarding the copper part of the cables, electric components, and electronic devices, the toxicity is frequently related to the mining and processing of the raw metal, particularly to the disposal of sulfidic ore tailings (Corona et al., 2017). Nevertheless, the development of photovoltaic panels that do not require toxic elements such as cadmium or rare elements like tellurium is under study (Tsang et al., 2016), which will diminish the influence of the HHE in the photovoltaic solar-driven photocatalysis in the future.

Regarding EBs to water, all the indicators are slightly smaller in Sc. 2 than in the other scenarios. Sc. 3 shows the highest total aquatic EBs due to its high NMeCo value, behavior mostly associated with the disposal of sludge incineration wastes (Pretel et al., 2016). Additionally, the total EBs of Sc. 1 are slightly higher due to the high energy demand of the light source in photocatalysis. However, LEDs have been evolving rapidly over the last few years (Song et al., 2016), the development of energy efficient LEDs with the same intensity of radiation but less electricity demand seems feasible. Thus, an extraordinary environmental progress of the photocatalytic treatment seems feasible within the upcoming years.

3.3. MBR variation assessment

Since the EBs to water in Sc. 3 are slightly higher than in Sc. 1 and Sc. 2~~,~~ due to the landfilling of the sludge, a variation in the MBR has been performed in order to assess the environmental impact of an alternative sludge. Therefore, a new scenario, Sc. 3b, is considered. In this process, the sludge is thickened and dewatered on-site following the same procedure as in Sc. 3. Nevertheless, after the dewatering process, the sludge is stabilized by composting and then transported and stored for several days before its use in land stabilization. All the data used for the analysis are taken from literature (Hospido et al., 2005; Suh and Rousseaux, 2002). The results obtained are shown in Fig. 3. Although a reduction in the NRs and EBs is expected in Sc. 3b, both alternatives have similar environmental performance. This is because in the MBR what causes greater consumption of resources and generation of impacts is the energy used in the aeration of the reactor and, thus, the loads avoided, both by incineration and by composting, are minimal compared to aeration.

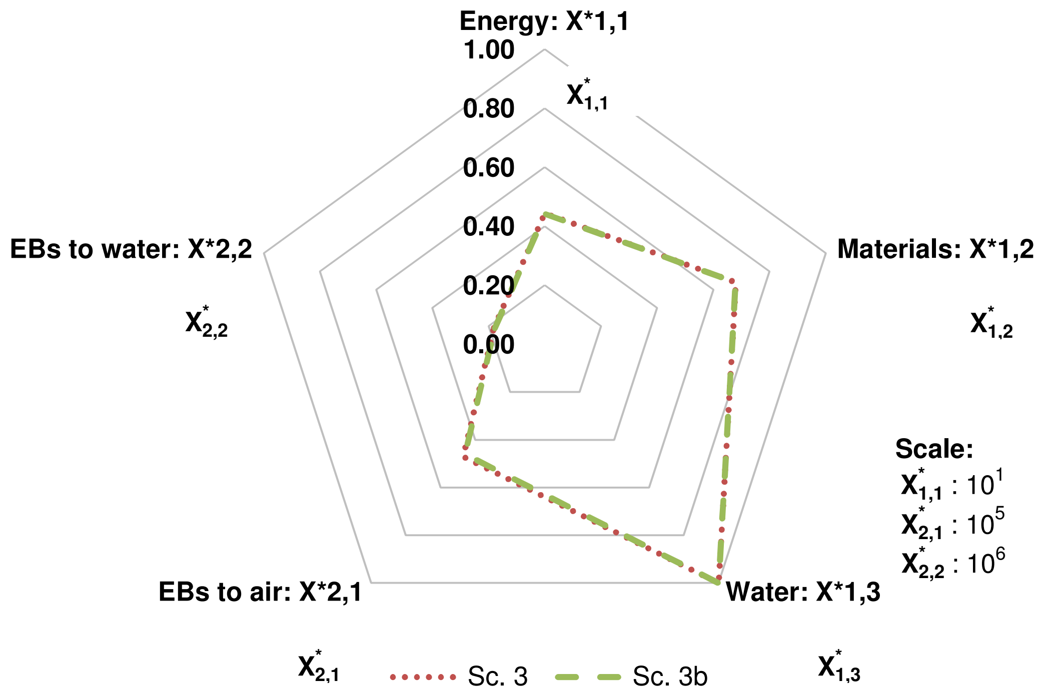


Fig. 3. NRs and EBs dimensionless variables for Sc. 3 (MBR with sludge incineration) and Sc. 3b (MBR with sludge composting).

The EBs for both scenarios are detailed in Table 6. Sc. 3 shows slightly higher EBs than Sc. 3b. This trend is observed for all the indicators but for the water aquatic ecotoxicity, due to the presence of heavy metals in the sludge applied to agricultural fields in Sc. 3b. However, it has to be remarked that the presence of heavy metals in air is more important than in the aquatic medium because they have more possibilities to directly contact human beings. Regarding the air burdens, Sc. 3 shows a higher global warming indicator as a result of the greenhouse gases emissions from the incineration step. It has to be highlighted that the human toxicity is the indicator with the highest contribution to the EBs in Sc. 3, owing to the heavy metals present in the gaseous effluent generated during the incineration of the sludge (Suh and Rousseaux, 2002). Furthermore, in Sc. 3 the stratospheric ozone depletion also shows a high value due to the landfill gas emissions originated when the incinerated sludge is landfilled. Thus, taking all this into account, Sc. 3b can be considered the best alternative for the MBR treatment of greywater.

4. Conclusions

This work provides technological and environmental decision criteria to use clean, safe, and renewable solar energy for the treatment of greywater under a circular economy of water. The LCA methodology is applied to evaluate the environmental impacts of three greywater treatment alternatives, photocatalysis, photovoltaic solar-driven photocatalysis, and MBR. The analysis shows that photovoltaic photocatalysis driven by solar energy is the most sustainable scenario from the environmental point of view. The variable that contributes mostly to the use of natural resources and the generation of environmental burdens is energy consumption. This is due to the high energy requirements of the light source, which is the main bottleneck of photovoltaic solar-driven photocatalysis and photocatalysis scenarios. Therefore, this study determines the main hot-spot of an emerging technology such as photocatalysis. The analysis and the results allow to promote the deployment of the technology through its combination with photovoltaic solar energy. This can be considered as the first step in establishing the best available techniques for greywater reuse.

Despite the higher consumption of natural resources observed in the MBR~~,~~ due to the high air consumption, their EBs are lower than in the photocatalysis scenario. However, due to the landfill of the sludge, the EBs to water in the MBR scenario are slightly higher than in the photocatalysis and photovoltaic solar photocatalysis scenarios.

Taking into account the environmental assessment of the greywater reuse process through the scenarios considered, future technological challenges have to be addressed under an environmentally friendly framework. Energy consumption could be optimized to a large extent to avoid the excess of energy applied and, therefore, to allow the process to operate in a sustainable manner.

In this context, despite the potential of photocatalysis for greywater treatment, there are still some key technological issues related to its application that have to be solved, with the high energy demand being the main one. Thus, the development of more energy efficient light sources is being studied. In order to reduce their environmental burdens, the most desirable scenario would be the use of solar light. Nonetheless, further research is needed to overcome some important issues like the development of photocatalysts that are active under visible light, which could help to implement solar photocatalysis for the treatment of greywater.

Thus, to achieve a sustainable greywater treatment, future discussion including technical and economic evaluations should be performed in order to complement the LCA study.

Acknowledgements

Financial support from the Spanish Ministry of Economy and Competitiveness and from FEDER funds for projects CTM2013-43539-R, CTM2015-69845-R and CTQ2015-66078-R (MINECO/FEDER, UE) is gratefully acknowledged. Sara Dominguez and Jara Laso would also like to express their gratitude for the FPI postgraduate research grants (BES-2013-064055 and BES-2014-069368).

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