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Life cycle assessment of a coupled solar photocatalytic–biological process for wastewater treatment

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ABSTRACT

A comparative life cycle assessment (LCA) of two solar-driven advanced oxidation processes, namely heterogeneous semiconductor photocatalysis and homogeneous photo-Fenton, both coupled to biological treatment, is carried out in order to identify the environmentally preferable alternative to treat industrial wastewaters containing non-biodegradable priority hazardous substances. The study is based on solar pilot plant tests using α-methyl-phenylglycine as a target substance. The LCA study is based on the experimental results obtained, along with data from an industrial-scale plant. The system under study includes production of the plant infrastructure, chemicals, electricity, transport of all these materials to the plant site, management of the spent catalyst by transport and landfilling, as well as treatment of the biodegradable effluent obtained in a conventional municipal wastewater treatment plant, and excess sludge treatment by incineration. Nine environmental impact categories are included in the LCA: global warming, ozone depletion, human toxicity, freshwater aquatic toxicity, photochemical ozone formation, acidification, eutrophication, energy consumption, and land use. The experimental results obtained in the pilot plant show that solar photo-Fenton is able to obtain a biodegradable effluent much faster than solar heterogeneous photocatalysis, implying that the latter would require a much larger solar collector area in an industrial application. The results of the LCA show that, an industrial wastewater treatment plant based on heterogeneous photocatalysis involves a higher environmental impact than the photo-Fenton alternative, which displays impact scores 80–90% lower in most impact categories assessed. These results are mainly due to the larger size of the solar collector field needed by the plant.

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1. Introduction

Compared to other regions, water supply and sanitation in the EU are fairly well developed, but still 20% of all surface water in the Union is seriously threatened with pollution (EU, 2002). In recent years, considerable attention has been paid to the so-called priority hazardous substances, listed in annex X of the Water Framework Directive (EU, 2000). This list includes 33 substances and groups of substances presenting a significant risk to or via the aquatic environment, including such risks to waters used for drinking water production. Heavy metals, pesticides, polyaromatic hydrocarbons, and

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chlorinated solvents, are some representative examples of the kind of compounds included. Once this list has been adopted, the commission will propose community-wide water quality standards and emission controls for these substances (EU, 2001).

In the same context, the IPPC Directive (EU, 1996) requires the development of technologies and management practices for pollution prevention and water recycling in several industrial sectors. Biological treatment is nowadays considered to be among the best available technologies (BAT) for wastewater treatment (Sarrià et al., 2002; Gernjak et al., 2006), but the Water framework Directive addresses many pollutants which are toxic, chemically stable or difficult to mineralise. For this reason, the destruction of these pollutants must be put into practice by other non-biological technologies, such as advanced oxidation processes (AOPs), which are those methods based on the in situ production of transient radical species of great oxidant power, specially hydroxyl radicals ($OH^{\bullet}$) (Gogate and Pandit, 2004a, b; Pera-Titus et al., 2004). The potential of AOPs for destroying almost any organic contaminant is widely recognised, but it is also known that they entail higher costs as compared to biological treatment, due to an intensive use of energy and expensive reactants (Andreozzi et al., 1999). In order to overcome this problem, substitution of electricity by solar energy, as well as coupling AOPs to biological treatment have been proposed as suitable strategies (Gernjak et al., 2006). On the one hand, solar energy use is a well-known capability of some AOPs, namely heterogeneous photocatalysis and homogeneous photo-Fenton, which can use solar photons from the VIS and/or UV region to produce $OH^{\bullet}$ radicals (Bauer and Fallmann, 1997). On the other hand, the combination of an AOP as a preliminary treatment to obtain a biodegradable wastewater, followed by an inexpensive biotreatment, seems to be an economically attractive option.

Another important feature of the IPPC Directive is the fact that it considers the environment as a whole. As a consequence, the concept of BAT must consider, in addition to technical feasibility and costs, potential trade-offs between environmental issues, in order to avoid creating a new and more serious environmental problem when solving another (European Integrated Pollution Prevention and Control Bureau (EIPPCB), 2005). Therefore, a complete environmental assessment of any emergent technology should be carried out before considering it as a BAT for wastewater treatment. In this context, life cycle assessment (LCA) appears as one of the most accepted tools for this purpose (Guinée et al., 2002). LCA is a method to define and reduce the environmental burdens from a product, process or activity by identifying and quantifying energy and materials usage, as well as waste discharges, assessing the impacts of these wastes on the environment and evaluating opportunities for environmental improvements over the whole life cycle (Consoli et al., 1993). Because of its holistic approach, LCA is becoming an increasingly important decision-making tool in environmental management. Its main advantage lies in broadening the system boundaries to include all burdens and impacts in the life cycle of a product or a process, and not focusing on the emissions and wastes generated by the plant or manufacturing site only (Azapagic and Clift, 1999).

In a recent paper (Muñoz et al., 2005), a simplified LCA based on laboratory experiments was carried out on several solar AOPs, including heterogeneous photocatalysis and photo-Fenton. However, a more detailed study based on pilot- and full-scale plants is needed. The aim of this work is to perform an environmental assessment, by means of LCA, of coupling solar-driven AOPs to conventional biological treatment, comparing two possibilities with regard to the advanced oxidation step: heterogeneous semiconductor photocatalysis and homogeneous photo-Fenton. The model compound used is α-methyl-phenylglycine (MPG, a precursor in pharmaceuticals), selected because of its non-biodegradability and high water solubility.

2. Materials and methods

2.1. Pilot plants

Heterogeneous photocatalysis and photo-Fenton tests were conducted in pilot plants located in INETI (Instituto Nacional de Engenharia, Tecnologia Industrial e Inovaçao, Lisbon-Portugal) and PSA (Plataforma Solar de Almería, Almeria-Spain), respectively. Both plants consist of compound parabolic collectors (4.16 m^2 aperture area) exposed to sunlight, a reservoir tank, a recirculation pump and connecting tubing, being operated in batch mode. The collector is tilted according to the local latitude (40° in Lisbon, 37° in Almeria) including 20 Pyrex absorber tubes and aluminium reflectors with a concentration factor of one. The illuminated volume when the collector is completely exposed to sunlight is 44.6 L while the total volume is 82 L. The plants are equipped with pH, temperature, dissolved oxygen, and UV radiation intensity monitoring. More details on these plants can be found elsewhere (Gernjak et al., 2006; Lapertot et al., 2006).

2.2. Chemicals

The target compound used in this study is technical-grade MPG (α-methyl-phenylglycine, $C_9H_{11}NO_2$), dissolved in distilled water at a concentration of 500 mg/L. Iron sulphate ($FeSO_4 \cdot 7H_2O$), hydrogen peroxide (reagent grade, 30% w/v) and sulphuric acid for pH adjustment were provided by Panreac, while Titanium dioxide P25 was supplied by Degussa Ibérica.

2.3. Analytical determinations

Mineralisation was monitored by measuring the dissolved organic carbon (DOC) with a Shimadzu 5050-A TOC analyzer. MPG was analysed using reverse-phase liquid chromatography with UV detector in a HPLC-UV (Agilent series 1100). Chemical oxygen demand (COD) was determined with analytical kits from Merck in the ranges of 100–1500 mg COD/L, or 10–150 mg COD/L. Hydrogen peroxide concentrations were analysed by iodometric titration, while ammonia and anions by ionic chromatography (LC-IC). Iron determination was done by colorimetry with 1,10-phenantroline. Biodegradability of the treated wastewater was determined by means of the Zahn–Wellens test (Lapertot et al., 2006).

2.4. Experimental set-up

In the photo-Fenton experiments, the synthetic wastewater was prepared in the reactor by dissolving MPG. Then pH was adjusted to 2.7–2.9, ferrous sulphate (20 mg/L) and hydrogen peroxide were added, and the collectors uncovered. Hydrogen peroxide was repeatedly determined, and small amounts were added as consumed in order to avoid disappearance. In the heterogeneous photocatalysis experiments, the synthetic wastewater was prepared as a 3 L concentrated solution and adjusted to the batch volume afterwards. Titanium dioxide was added (200 mg/L) and after 15 min, the collectors were uncovered.

3. Application of LCA

The experimental data obtained in the above described advanced oxidation tests were used as the starting point for applying the LCA methodology, according to the ISO standards (International Standardisation Organisation (ISO), 1997).

3.1. Goal and scope

The ultimate goal of the LCA study is to compare MPG wastewater treatment by the two solar-driven AOPs tested in the pilot plants: homogeneous photo-Fenton and heterogeneous photocatalysis.

Fig. 1 shows a flow diagram for the system under study. As can be seen, it includes, for both alternatives, the production and installation of the CPC field, production of all chemicals consumed and transport of all these materials to the plant site. Also included is electricity production, management of spent catalysts through transport, inertisation and landfilling, as well as further treatment of the plant effluent in a municipal wastewater treatment plant (MWWTP).

One of the key elements of LCA to be set in the scope of the study is the functional unit, which is defined as the unit of service used as the basis for comparison of alternatives. In this case study the functional unit chosen is "to treat 1 m^3 of synthetic MPG solution (500 mg/L) in order to destroy non-biodegradable and/or toxic compounds, achieving an effluent quality that allows discharge to the aquatic ecosystem".

Although the assessment is based on the experimental data obtained in the pilot plants, the system has been modelled in the LCA considering an industrial-scale plant. The main hypotheses taken into account for such a plant are the following:

- The assumed location is Almeria (Spain), which receives sunrise to sunset 18.6 W/m^2 of yearly average global UV radiation (Malato et al., 2001).
- The treatment capacity is set to 2500 m^3/year of MPG effluent.
- The plant works in batch mode, 12 h/day, all year round.
- A useful life of 15 years is assumed.
- Titanium dioxide is assumed to be reused 10 times in a closed cycle (Malato et al., 2000). This chemical would be separated from suspension by attaining the point of zero charge at pH value around 7 and allowing about 95–97% of

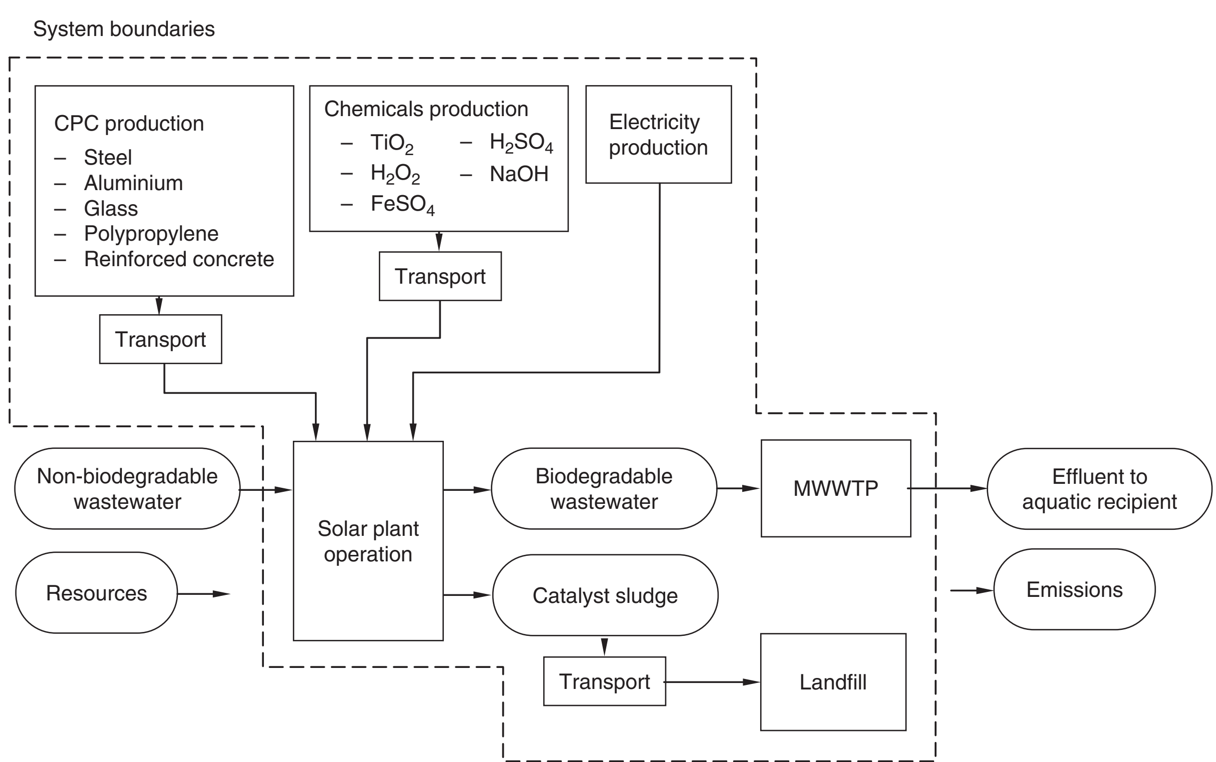

Fig. 1 – Flow diagram and system boundaries in the LCA study.

the catalyst to settle (Blanco et al., 2001). The supernatant would be passed through a microfiltration membrane in order to remove the remaining catalyst in suspension (Fernández-Ibáñez et al., 2003). Iron is assumed not to be reused, but precipitated as hydroxides at the end of each batch by rising the pH to 7, and removed by conventional settling. Both catalysts are assumed to be thickened and transported by truck as a 5% dry mass slurry.

- Biological treatment is assumed to take place off-site in a MWWTP, as the AOPs produce a biodegradable effluent the characteristics of which allow discharge to a public sewer.
- Since the effluent is transported to the MWWTP by a public sewer, the excess (if any) hydrogen peroxide is consumed by oxidation of organic matter present in the urban wastewater stream, preventing further damage of the biological reactor's biomass.

3.2. *Inventory analysis*

Table 1 shows a summary of the data sources employed in the inventory analysis. As can be seen, the material and energy balance of the whole system is obtained with a mixture of data from pilot plant, industrial plant, literature, and models. In order to convert inputs from technosphere (electricity, chemicals, etc.) to their associated consumption of resources and emissions to air, water and soil, the Swiss Ecoinvent LCA database has been used (Frischknecht et al., 2004). The inventory for treating the solar plant effluent in a MWWTP as well as for landfilling of spent catalysts has also been obtained from specific calculation tools included in this database (Doka, 2003).

3.3. *Impact assessment*

The mandatory elements of life cycle impact assessment (LCIA) according to ISO 14042 (International Standardisation Organisation (ISO), 1999) have been carried out, namely: (1) selection of impact categories and models, (2) assignment of inventory results to impact categories (classification), and (3) calculation of category indicator results (characterisation). The impact categories included are global warming potential, ozone depletion potential, human toxicity potential, freshwater aquatic toxicity potential, photochemical oxidant formation potential, acidification potential, eutrophication potential (EP), non-renewable energy consumption (EC), and land use. The characterisation models used are those proposed by the Institute of Environmental Sciences of Leiden (Guinée et al., 2002).

4. Results and discussion

4.1. *Experimental results*

Table 2 shows the composition of the MPG solution, before and after applying the solar advanced oxidation, that is, until biodegradability is achieved. The Zahn–Wellens test showed that, in order to obtain a biodegradable wastewater, MPG must completely disappear. The photo-Fenton and heterogeneous photocatalysis treatments allow to obtain a biodegradable wastewater when DOC is around 120 and 40 mg/L, respectively (Oller et al., 2006). In both experiments, the nitrogen content of MPG is mainly found as ammonia in the final effluent, while nitrate is almost absent (Table 2).

Table 1 – Data collection and data sources

Process	Sources
CPC infrastructure	Data from the first commercial plant applying solar photocatalysis, owned by Albaida Recursos Naturales y Medioambiente S.A. in Almeria. Inventory of materials per m^2 of CPC in this plant, supplied by the plant designer, Ecosystem (Vincent, 2005). CPC area is calculated for from the experimental results obtained in the pilot plants.
Chemicals consumed (H_2O_2, $FeSO_4$, TiO_2, H_2SO_4, NaOH)	Data derived from the pilot plant experiments. All consumptions obtained from the dose applied, except in the case of TiO_2, since it is assumed to be used 10 times (Malato et al., 2000).
Transports	Distances and transport models suggested by the Ecoinvent database for western Europe (Frischknecht et al., 2004).
Pumping requirements	For solar AOPs the pump power is extrapolated from the Albaida plant.
TiO_2 microfiltration	Energy consumed by this process is derived from a pilot plant in PSA (Fernández-Ibáñez et al., 2003).
Stirring	Mechanical stirrer power has been determined by Ecosystem (Vincent 2005).
Effluent to MWWTP	The composition of the effluent from the advanced oxidation step is obtained from the pilot plant experiments.
MWWTP	Inventory for sewage treatment and management of sludge, obtained from the Ecoinvent tool for wastewater treatment (Doka, 2003), by specifying the composition of the input wastewater.
Process emissions	The only process emissions that have been calculated are CO_2 from DOC degradation by AOPs which are calculated stoichiometrically. All other emissions to air, water and soil are included in the Ecoinvent datasets used.
Landfill	Inventory for residual material landfill obtained from the Ecoinvent tool for landfills (Doka 2003), by specifying the composition of the input waste. Stabilisation of the waste with cement is taken into account.
Land use	Land occupied by the solar plant is extrapolated from the Albaida Plant in Almeria (Vincent, 2005).

Table 2 – Experimental data obtained in the solar advanced oxidation tests

Treatment	Solar photo-Fenton	Solar heterogeneous photocatalysis
Initial effluent (non-biodegradable)		
MPG (mg/L)	500	500
DOC (mg/L)	330	330
COD (mg/L)	1270	1270
N-total (mg/L)	42	42
N-ammonia (mg/L)	0	0
N-nitrate (mg/L)	0	0
Final effluent (biodegradable)		
MPG (mg/L)	0	0
DOC (mg/L)	120	40
COD (mg/L)	504	214
N-ammonia (mg/L)	27	36
N-nitrate (mg/L)	<1	<1
Accumulated UV radiation and time required		
Q_{uv} (kJ/L)	12	252
t_{30W} (min)	70	1500

However, N-ammonia and N-nitrate do not amount the expected total of 42 mg/L, meaning that a small nitrogen fraction remains in organic intermediates resulting from MPG degradation. On the other hand, Table 2 also shows the accumulated values of solar UV radiation needed per litre wastewater to achieve the biodegradability point (Q_{uv}), as well as the corresponding "normalised illumination time" (t_{30W}), which refers to a constant solar UV power of 30 W/m^2. From these two parameters, it can be seen that solar photo-Fenton is able to treat MPG much faster than solar heterogeneous photocatalysis. This has clear implications in plant design, since the CPC aperture area required for a full-scale plant treating this wastewater is calculated using (Blanco and Malato, 2003)

$$S_{cpc} = \frac{Q_{uv} \cdot V}{T_{sun} \cdot 3600 \cdot UV_g}, \qquad (1)$$

where S_{cpc} is the collector aperture area needed by the plant, V is the wastewater volume to be treated (2500 m^3/year), T_{sun} is the overall operation time (4380 h/year) and UV_g is the global average UV irradiation, sunrise to sunset, at the plant site (18.6 W/m^2). A solar photo-Fenton plant would require 100 m^2 of solar collectors, while the corresponding plant based on heterogeneous photocatalysis would require 2150 m^2, a figure 21 times higher.

4.2. *LCA results*

Table 3 summarises the inventory for the alternatives under study, highlighting the amount of materials needed to build the CPC field, as well as the electricity and chemicals consumed. With regard to landfilling of the catalysts, only the amount landfilled per functional unit is shown; the environmental burdens of this process have been calculated using the Ecoinvent model for landfills (Doka, 2003), which allocates the corresponding burdens (EC, emissions to air and water, etc.) as a function of the waste composition, which in this case is a 5% dry mass TiO_2 slurry, and a 5% dry mass $Fe(OH^{\bullet})_3$ slurry. Concerning the biological treatment, which has been assumed to be carried out in a conventional MWWTP, Table 3 does not show the process inventory either. The Ecoinvent tool for MWWTPs (Doka, 2003) has been used to quantify the inventory of this process, including transport through the sewer, primary settling, aerobic biological treatment with partial nitrification, phosphate removal by means of precipitation, and sludge management by anaerobic digestion and incineration. This model allocates the MWWTP infrastructure, consumption of energy, chemicals, excess sludge production and treatment, etc., from the specific composition of the wastewater to be treated, which in this case study is that of the biodegradable effluent in Table 2.

All the data obtained through this inventory analysis have been introduced in the software Simapro 6.0 (Pré Consultants, 2006) and LCIA has been applied as described in Section 3.3, in order to obtain the impact category scores shown in Fig. 2. This figure allows, at the same time, to compare both alternatives, as well as to identify the most significant subsystems. With regard to the latter, the figure shows the contribution of four sub-systems: solar field infrastructure, hydrogen peroxide, electricity, MWWTP, and "others", which refers to all the remaining processes included in the LCA with a low individual contribution to the overall impacts. On the other hand, impact categories cannot be compared with each other, since they are measured in different units. As a consequence, this figure cannot be used to tell, for example, whether the analysed AOPs have a more serious effect on human toxicity than, let us say, on global warming.

The main aspect to highlight from Fig. 2 is that, in all impact categories, the environmental performance of solar photo-Fenton coupled to biological treatment is much better than that of coupling heterogeneous photocatalysis to biological treatment. The environmental impact of photo-Fenton is 80–90% lower, depending on the impact category, with the exception of EP, for which it is 30% lower. It can be

Table 3 – Disaggregated inventory table for 1 m^2 of compound parabolic collector and for both alternatives under study

Inputs (per functional unit)	1 m^2 compound parabolic collector	Solar photo-Fenton	Solar heterogeneous photocatalysis
From nature			
Occupation, industrial site (m^2 year)	2.76	0.14	2.4
From technosphere			
CPC infrastructure:			
Stainless steel (kg)	7.81	0.021	0.45
Galvanized steel (kg)	0.17	4.4E-04	9.5E-03
Aluminium extruded and anodised × 2 (kg)[a]	9.68	0.026	0.55
Borosilicate glass tube × 2 (kg)[a]	6.72	0.018	0.38
Extruded polypropylene (kg)	0.2	5.3E-04	1.1E-02
Injection moulded polypropylene (kg)	1.2	3.2E-03	0.069
Concrete (m^3)	0.32	8.5E-04	0.018
Reinforcing steel (kg)	31	0.083	1.8
Concrete blocks (m^3)	0.02	5.3E-05	1.1E-03
Materials transport by rail (kg km)	14,089	38	807
Materials transport by lorry 32 t (kg km)	45,579	15	327
Auxiliary materials and energy:			
Electricity, medium voltage, UCTE profile (kWh)[b]		0.72	18
Hydrogen peroxide (pure) (kg)		1.7	
Iron sulphate (kg)		0.10	
Titanium dioxide (kg)			0.02
Sulphuric acid (pure) (kg)		0.13	
Sodium hydroxide (pure) (kg)		0.08	
Chemicals transport by rail (kg km)		4012	12
Chemicals transport by lorry 32 t (kg km)		669	2
Outputs (per functional unit)			
To nature			
Carbon dioxide (kg)		0.77	1.06
To technosphere			
Spent catalyst management:			
Transport by lorry 16 t (kg km)		38	20
Catalyst landfilling (fresh weight) (kg)[c]		0.76	0.40
Effluent treatment in MWWTP (m^3)[d]		1	1

[a] The amount of aluminium and glass is doubled, since optical elements are changed during the plant lifetime.
[b] European Union for the co-ordination of transmission of electricity.
[c] Inventory for the landfilling process calculated with the Ecoinvent tool for landfills (Doka, 2003).
[d] Inventory for sewage treatment calculated with the Ecoinvent tool for MWWTP (Doka, 2003).

seen that in most impact categories, the worse performance of heterogeneous photocatalysis is originated by two sub-systems, namely the solar field and electricity consumption. As it has been shown, a solar plant applying this AOP would require a large CPC field (2150 m^2), involving a considerable consumption of materials to build this infrastructure. In addition, the larger the solar field, the higher the power needed to pump the wastewater through the system. It must be also considered that microfiltration of the TiO_2 catalyst supernatant is an energy-intensive process, explaining the high contributions of the electricity sub-system. On the other hand, the main impacts of the photo-Fenton alternative are caused by hydrogen peroxide, which is the main input material to the process, and also by production of the solar field (100 m^2). For both alternatives, treatment of the biodegradable effluent in a MWWTP is only a critical sub-system in the EP category; this is mainly due to the nitrogen compounds discharged in the MWWTP effluent to the aquatic environment, mostly ammonium and nitrate derived from the parent compound, MPG.

5. Conclusions

LCA has been used to compare, from an environmental point of view, two strategies for treating MPG wastewaters, based on solar-driven AOPs coupled to biological treatment, namely heterogeneous semiconductor photocatalysis and homogeneous photo-Fenton.

The results of the LCA show that, mainly due to the larger size of the solar CPC field, an industrial wastewater treatment plant based on heterogeneous photocatalysis involves a higher environmental impact than the photo-Fenton

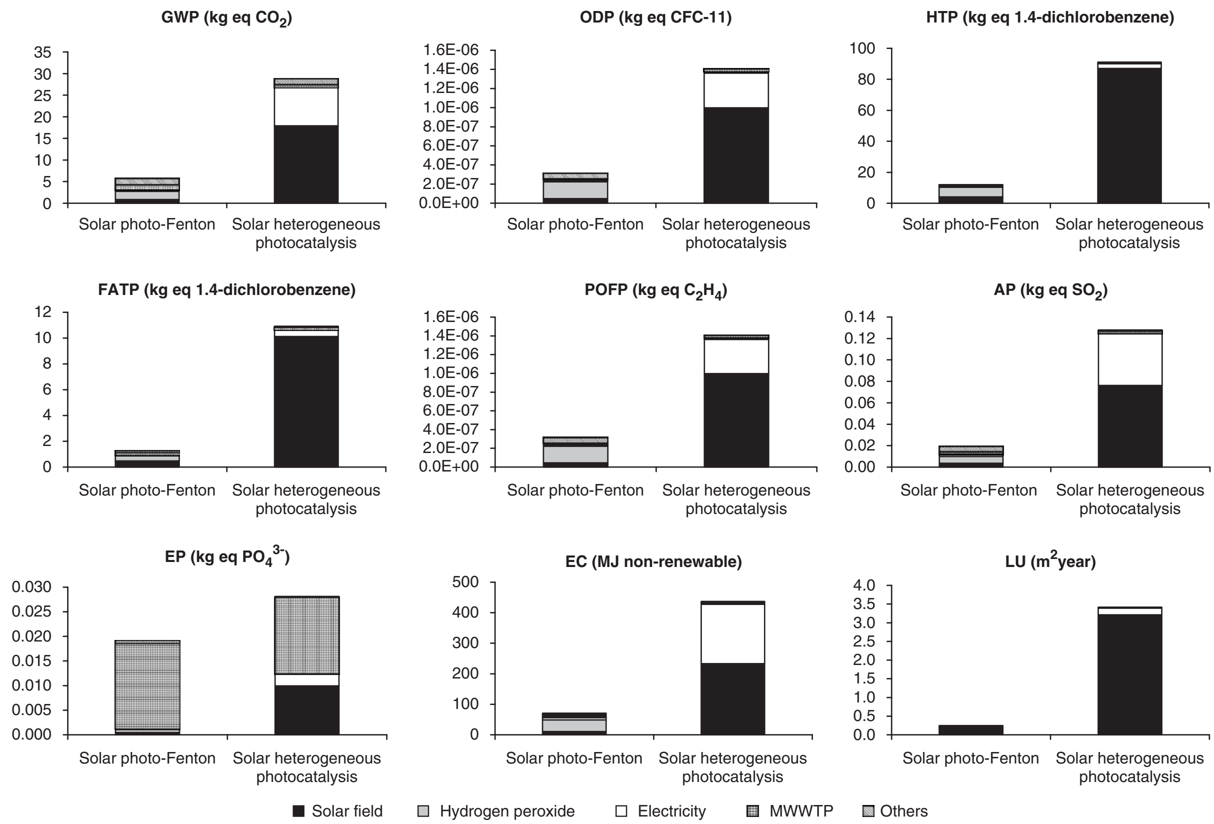

Fig. 2 – Life cycle impact assessment results for the alternatives under study.

alternative, which displays impact scores between 80% and 90% lower in most impact categories assessed.

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