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Environmental life cycle assessment of polyhydroxyalkanoates production by purple phototrophic bacteria mixed cultures

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ABSTRACT

Bioplastics offer a promising sustainable alternative to petroleum-based plastics due to their biodegradability as well as favourable thermal and mechanical properties. Among different types of biobased polymers, the production of polyhydroxyalkanoates (PHA) using purple phototrophic bacteria (PPB) and low-value substrates has gained increasing interest. Despite the momentum, challenges regarding the scalability and environmental feasibility of this biopolymer production pathway remain. In response, this study employs an exploratory LCA approach to quantitatively assesses the potential environmental implications of PHA production in powder form and the joint management of the organic fraction of municipal solid waste (OFMSW) through a novel photobiorefinery system that uses PPB mixed cultures. Environmental impacts were tested under multiple improvement scenarios and benchmarked against the production of conventional fossil-based granulate or unprocessed plastics, including low density polyethylene (LDPE), polyethylene terephthalate (PET) and polyurethane (PU). The photobiorefinery stage was found to have the greatest contribution to the impact categories, particularly due to direct emissions, consumption of electricity and production of extractive chemical agents used. These factors accounted for over 70% of the photobiorefinery impact in all cases. Avoided impacts provided net favourable outcomes in terms of carbon footprint and fossil resources when comparing PHA production to conventional plastics, especially PET and PU, with impact reductions ranging from 30% to 60%, respectively. However, when considering other impact categories like eutrophication, this situation was less favourable. The exploration of alternative scenarios offered significant impact reductions, especially when renewable electricity or an environmentally friendly extraction agent is used. Moreover, minimizing methane losses or co-producing hydrogen in the photobiorefinery had a notably positive effect on the carbon footprint, reducing the impact by more than 2 t of CO_2 eq per t of PHA powder compared to the base case. Therefore, the implementation of feasible improvement measures in the short term can position PHA produced by mixed cultures as a sustainable alternative to petroleum-based plastics.

1. Introduction

Approximately 57 million of the nearly 390 million metric tonnes (Mt) of plastic manufactured worldwide in 2021 were produced in Europe (Plastics Europe, 2022). Only 9% of all plastic ever made has been recycled, 12% has been burned, and the remainder builds up in landfills or the environment, according to the UNEP (UNEP, 2022). Furthermore, approximately 80% of the plastic waste in the oceans comes from land, usually from poorly managed landfills and curbsides plundered by sea tides and wind (Rosenboom et al., 2022). This plastic enters the food chain as microplastics, endangering biodiversity (Wang et al., 2021). In this context, bioplastics have emerged as a real, sustainable alternative, as they can be produced from renewable resources, are biodegradable and have similar thermal and mechanical properties to petroleum-based plastics.

Biobased polymers used as bioplastics can be subdivided into three types, plant-based (i.e., thermoplastic starch, TPS), polymerised biomonomers (i.e., polylactic acid, PLA), and extracted biopolymers (polyhydroxyalkanoates, PHA) (Meereboer et al., 2020). PLA and PHA are biodegradable (de Castro et al., 2021), specifically, PLA is compostable

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but not marine biodegradable like PHA (Meereboer et al., 2020). Biodegradability is essential if we want to move towards circular economy practices. Moving towards bio-based and biodegradable polymers allows for a more sustainable option, especially if the output of biodegradation becomes the production input for the same polymer in a reasonable time frame within the biological cycle (Braungart et al., 2007). Therefore, PHA are becoming important due to their many advantages such as their biodegradability, biocompatibility, controllable thermal and mechanical properties (Laycock et al., 2014). In the industrial setting, PHA is produced by fermentation using engineered, pure aerobic heterotrophic microorganisms that are fed relatively pure plant-derived substrates like sugars and starch. However, for the time being, the price of commercial production of PHA is as high as €2.2–5.0 per kg, while the price of fossil-based plastics, such as low-density polyethylene (LDPE) or polyethylene terephthalate (PET) is around €1.0 per kg (Liu et al., 2021). The most commonly used strategies studied to reduce these costs are using mixed microbial communities as biocatalysts for PHA generation under non-sterile conditions and applying a variety of low-value substrates, such as industrial and municipal waste and by-products (Tsang et al., 2019). One strategy that is gaining momentum in recent years is the use of purple phototrophic bacteria (PPB) (Fradinho et al., 2021; Sali and Mackey, 2021).

The use of PPB for the production of PHA has several key decisive factors: a) PPB can deliver up to 90% PHA yields on substrates, almost three times higher than aerobic yields (Fradinho et al., 2019), b) PPB does not require aeration, as they obtain their energy from light (Hülsen et al., 2014) and c) PPB can accumulate PHA while growing, and nutrient availability drives the accumulation process (Fradinho et al., 2016). However, there are still uncertainties regarding the scalability and the environmental suitability of this technology (Fradinho et al., 2021). In this sense, there is a clear need to develop biorefineries capable of producing several products at the same time, eliminating uncertainties of seasonal variability and feedstock heterogeneity (Alibardi et al., 2020). For example, the technical feasibility of the production of PHA together with hydrogen and biogas in a photobiorefinery with PPB from food waste (Allegue et al., 2020), lignocellulosic waste (Allegue et al., 2021) or the organic fraction of municipal solid waste (OFMSW) (Allegue et al., 2022) have recently been studied. Regarding environmental performance, it is necessary to ensure that novel bioplastics avoid larger environmental pressure than that produced by fossil plastics. This can be achieved through standardised methods such as Life Cycle Assessment (LCA), a widely used and recognized tool for assessing the environmental impacts of product systems (ISO, 2006a; 2006b), or the utilization of Product Category Rules (PCR), which provide instructions to perform LCA on specific topics (EDP, 2022).

To date, several studies have been conducted to evaluate the environmental performance of PHAs using LCA. These studies have shown that PHA production usually has a lower global warming impact. However, it involves high energy requirements that penalise other impact categories, such as eutrophication or depletion of abiotic resources (Bassi et al., 2021; Saavedra del Oso et al., 2021). In general terms, studies conducted so far proved that the environmental impacts of PHA production depend on several factors, including the feedstock used, the energy sources supplied, and technical features of the production process, especially the effects of upscaling (Baioli et al., 2019). Furthermore, although many studies focus on using waste as a substrate for PHA generation, only a few consider the simultaneous environmental effects of treating the OFMSW (Bassi et al., 2021). Additionally, even though the use of PPB in PHA production is a novel approach that has the potential to improve the environmental performance of PHA production (Allegue et al., 2022), as far as we are aware, there are no studies in the literature that thoroughly evaluate the environmental feasibility of this biopolymer production pathway and identify any potential sustainability challenges associated with scaling up this technology. Filling this research gap is particularly important given the current need to transition to more sustainable and environmentally friendly biorefinery products.

This study aims to quantitatively assess the potential environmental implications of the joint production of PHA and the management of OFMSW through a novel photobiorefinery system involving PPB. To that purpose, the environmental performance of the produced PHA is benchmarked against conventional plastics (viz., LDPE, PET, and polyurethane (PU)) to verify the potential environmental savings of this biopolymer. It should be noted that in addition to environmentally evaluating PHA production by PPB for the first time through LCA, the scientific significance of the present article also lies in the upscaling of this novel process derived from experimental work and the exploration of different scenarios related to the photobiorefinery concept. The structure of this article is organized as follows. Section 2 details the materials and methods employed in this study, encompassing the definition of the case study, the definition of goals and scope, the elaboration of life cycle inventory, the impact assessment methodology selected, and the sensitivity analysis approach followed. The subsequent Section 3 delves into the results and discussion, wherein the environmental implications of PHA production and the joint management of OFMSW are examined. This section analyzes the base case scenario and explores multiple improvement scenarios to highlight the potential impact reductions and trade-offs under different conditions. Finally, Section 4 encapsulates the conclusions drawn from this study, outlining the key findings, implications, and avenues for future research.

2. Material and methods

2.1. Definition of the case study

The foreground data of the system have been estimated based on bibliography research and own modelling derived from laboratory and pilot scale experiments, mainly those published in (Allegue et al., 2022). All data used in the modelling are shown in Tables S1–S7 from the Supplementary Material. Energy efficiencies and consumptions were scaled by designing a hypothetical full-scale installation in Spain and deriving consumptions/costs from process engineering calculations. 290 days·year^{-1} of work at 24 h·day^{-1} were assumed with a daily demand of 95 t OFMSW (ww)·day^{-1} and an annual maximum PHA production of 605.5 t PHA. The standard macroscopic characteristics of the OFMSW are as follows: a content in total solids (TS) of 20% (Tyagi et al., 2018), volatile solids (VS) of 90% of TS (Ahmed et al., 2021), and a chemical oxygen demand (COD) fraction of 1.2 kg COD·kg TS^{-1} (Tyagi et al., 2018). In the present study, 45.5 t of OFMSW are required to produce 1 t of PHA according to own modelling.

A simplified process flow diagram is shown in Fig. 1. In summary, the first stage of the photobiorefinery system involves the waste collection and pretreatment of the OFMSW, where the residues are processing to promote particle size reduction. After the physical pretreatment, steam explosion is used to further process and assist in the hydrolyzation and solubilization of organic matter. The next step is an acidogenic fermentation process for the purpose of maximizing the volatile fatty acid (VFA) production. The acidogenic fermentation outlet is split into two streams, the liquid one directed towards the photofermentation process and the solid (sludge) one towards anaerobic digestion treatment. The photoheterotrophic process is conducted using raceway reactors, with the optimal operating conditions for maximizing PHA production by means of a mixed PPB culture. Finally, in the last stage, PHA extraction in powder form is performed from the obtained biomass. In the anaerobic digestion process, sludge is mixed with the biomass obtained after PHA extraction, and the biogas produced is fed into a combined heat and power (CHP) plant. Further details and different strategies used in the photobiorefinery system are provided below.

2.1.1. Waste collection and pretreatment

The photobiorefinery process starts with citizens' collection of selectively separated OFMSW. Then, diesel trucks drive 50 km to the

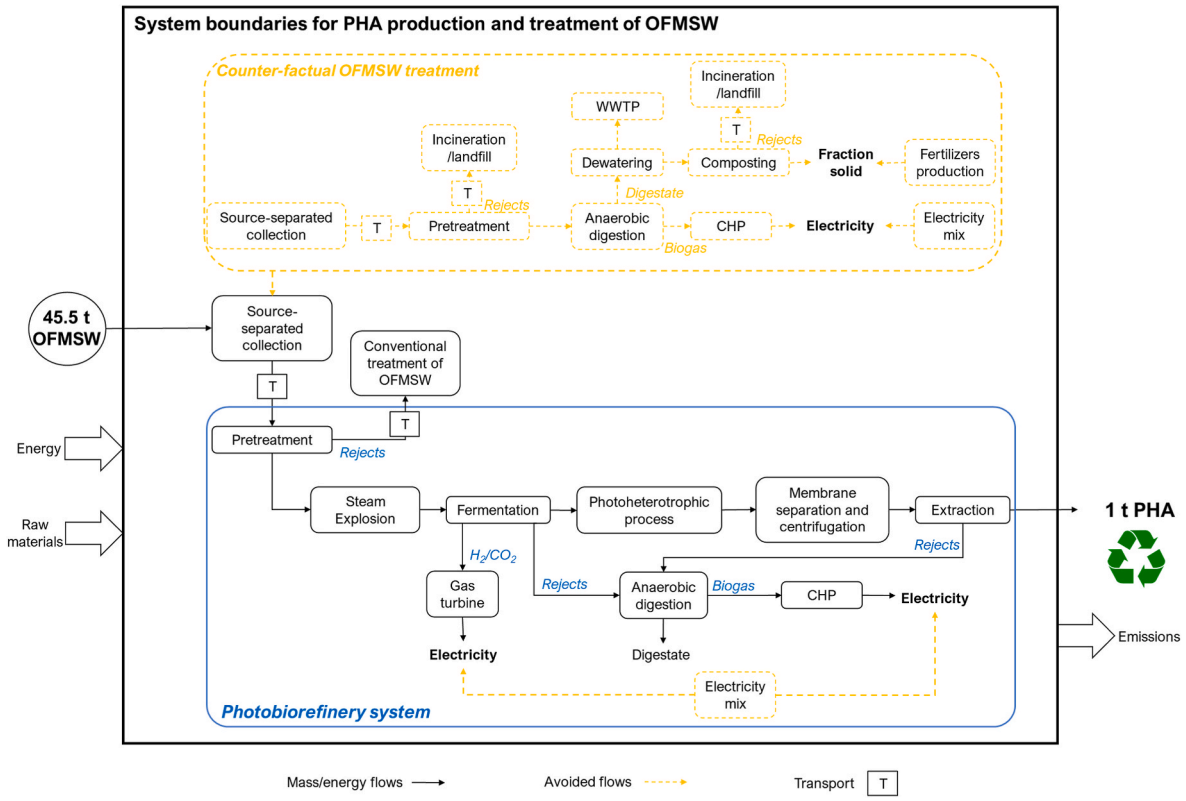

Fig. 1. System boundaries to produce polyhydroxyalkanoates (PHA) in powder form and treat the OFMSW.

photobiorefinery where an industrial shredder is used to homogenise and reduce particle size with an electricity usage of 2.43 kWh·(t OFMSW)$^{-1}$ (Perkoulidis et al., 2010). At this point, a rejection of 10% occurs (Bassi et al., 2021), due mainly to non-organic components. The remaining OFMSW is stored in a pre-heating chamber until use, where it is directly heated up to 60 °C by steam recovered from the steam explosion pretreatment.

2.1.2. Steam explosion pretreatment

This system consists of a hydrolysis reactor, where the steam enters, and a flash tank to relieve the pressure. The operating parameters chosen were 150 °C and 38 min following Allegue et al. (2022). The calculated amount of steam needed to heat the entire volume during the reaction time is 1.5 t of steam. A recovery of heat from the excess flash vapours (saturated steam at 105 °C) produced during the previous reaction to the pre-heating stage of the substrate leads to considerable saving in the energy consumption (Cano et al., 2014). The solids solubilization obtained is 44%, with a soluble to total COD ratio of 0.4 (Allegue et al., 2022).

2.1.3. Acidogenic fermentation

An upflow anaerobic sludge bed (UASB) reactor was used to model the acid fermenter, with a pH control of 5.5, enough to inhibit methanogens, and a temperature of 55 °C. The advantage of using a UASB is the easy collection of a liquid effluent that is largely free of solids that can be used directly in the PPB-based photobioreactor. Any undigested waste or excess sludge is sent to an anaerobic digester. The stream is fed at an organic loading rate (OLR) of 4 g COD·(L·d)$^{-1}$, and a hydraulic retention time (HRT) of 5 days. A solids (VSS) solubilization of 53%, a yield of 0.66 g COD equivalent short chain carboxylic acids (SCCAs)·(g COD added)$^{-1}$ and a hydrogen productivity of 345 mL H_2·(L·d)$^{-1}$ (Allegue et al., 2022) was used to perform the modelling calculations. A gas stream, composed of 80% H_2 and 20% CO_2, is obtained in this stage and subsequently used for electricity production in a gas turbine.

2.1.4. Photobioreactor and PHA production

The effluent from the acid fermenter, consisting of a stream rich in SCCAs, was further diluted before being fed into a membrane photobioreactor (MPBr). The extent of dilution was calculated from a mass balance at an OLR of 1 g COD·(L·d)$^{-1}$, a 2-day HRT, and a 4-day solids retention time (SRT). It was assumed that artificial lights are used 12 h a day, with natural lighting during the day. The energy consumption for the photobioreactor light is 2.2 W L^{-1}. The yields used for modelling are a consumption of 60% of the COD, a biomass yield ($Y_{X/s}$) of 0.97 g COD·(g COD)$^{-1}$, and a PHA productivity (q_{PHA}) of 0.61 g PHA·(L·d)$^{-1}$, with a percentage of 55% of PHA on dry mass, and hydrogen productivity (q_{H2}) of 83 mL H_2·(L·d)$^{-1}$. However, hydrogen accumulation in this stage is not feasible due to the industrial design applied for the scalability of the system (raceway reactor). These values were sourced from Allegue et al. (2022).

PHA is extracted from the biomass produced according to literature yields, i.e., 90% efficiency (Bassi et al., 2021). Finally, the PHA is purified and further processed to obtain a powder form. The rejected biomass is recirculated to the anaerobic digester. The methanogenic potential of the PPB biomass was estimated as 210 mL CH_4·(g VS)$^{-1}$ as determined by Hülsen et al. (2020). The water extracted from this process is recirculated into the inlet of the photobioreactor to reduce the water footprint of the photobiorefinery.

2.1.5. Anaerobic digestion and combined heat and power (CHP) plant

The results have been modelled based on typical anaerobic digester parameters. The methanogenic capacity of the waste was determined as 336 L CH_4·(kg VS)$^{-1}$, with an HRT of 20 days and a solids conversion into biogas of 65%. The digestate can be used as an organic soil amendment, while the biogas is directed to a CHP plant to produce steam and electricity. The energy content of biogas was estimated to be 28 MJ·Nm^{-3} (IEA, 2022a,b). The total energy generated in the CHP would be divided into 15% losses, 30% electricity, 30% hot water (directly used to heat the anaerobic digester) and 25% exhaust gas (used

for the production of steam on a boiler) as described in Cano et al. (2014).

2.2. Goal and scope

2.2.1. Goal and functional unit

The aim of this LCA is to evaluate the environmental implications of producing PHA in powder form and the joint management of OFMSW. LCA is a well-established and widely recognized methodology for comprehensively assessing the environmental impacts of products and processes throughout their life cycle. In this study, LCA is chosen as the analytical framework due to its capability to account for the various stages of PHA production and OFMSW management, from raw material extraction to PHA powder production. While the approach is attributional since the PHA production of the system will not result in a globally significant demand change, a system expansion approach is adopted to account for the avoided impact of OFMSW treatment. In this sense, the function of the system was expanded to include not only the production of PHA in powder form but also the treatment of the organic waste as an additional function of the system under evaluation. The functional unit (FU) of the proposed system is "production of 1 t of PHA powder and treatment of 45.5 t of OFMSW". Furthermore, to address the multifunctionality of the proposed system, the environmental benefits of the electricity co-produced in the photobiorefinery were obtained by substituting the avoided impacts of the conventional benchmarked production (i.e., electricity from the national electricity mix). Notably, we analyse and discuss the impact of different photobiorefinery co-productions through several proposed scenarios in Section 2.5.

2.2.2. System boundaries

Within the novel proposed system, the following life-cycle stages were included: i) avoided management of OFMSW through conventional treatment, ii) production of PHA powder in a photobiorefinery, and iii) management of waste from the photobiorefinery. Fig. 1 illustrates the system diagram, including the stages mentioned above. This study used a "cradle-to-factory gate" approach, given the uncertainty associated with the use and end-of-life of PHA as well as to emphasize the innovation and potential significance of the photobiorefinery pathway in sustainable waste management and bioplastic production. Regarding the counterfactual OFMSW treatment, the current management of this waste closely depends on the geographical context. According to the location of the hypothetical plant (Spain), this conventional treatment is based on anaerobic digestion and composting (Bassi et al., 2021). In the case of the photobiorefinery stage, its industrial modelling was based on experimental work (Section 2.1) and the literature. Within the literature addressing the application of LCA for the environmental evaluation of PHA, the studies focused on the production from organic waste are very limited. Vea et al. (2021) evaluated the environmental burdens of producing PHA from molasses, a by-product of the sugar industry. Asunis et al. (2021) analysed the impacts of producing PHA but, in this case, from a by-product of dairy industries, such as cheese whey. Currently, only one study bases its analysis on obtaining PHA through OFMSW (Bassi et al., 2021). This set of articles was taken as crucial references to support modelling the photobiorefinery stage. Infrastructure and treatment of digestate within this stage were considered out of the system boundaries. Finally, the treatment of rejects from the photobiorefinery was assumed to be the same as the counterfactual OFMSW.

2.3. Life cycle inventory

This section provides a summary of the main data collected for the life cycle inventory (LCI) (Table 1) and describes the main assumptions and data sources used. The foreground processes involved in the PHA generation were estimated using own modelling and literature. Regarding background processes, i.e., energy and raw materials delivered to the foreground system, data was obtained from the ecoinvent database v3.8 using the cut-off system model (Wernet et al., 2016). The list of ecoinvent datasets used is presented in the Supplementary Information (Table S8). For the foreground processes, the electricity required was regionalised based on the Spanish electricity mix for the year 2021, which includes 25% natural gas, 24% wind, 22% nuclear, 12% hydropower, 8% photovoltaic, 4% other renewables, 2% coal, and 3% other sources (REE, 2022). We conducted a sensitivity analysis to evaluate the impact of using 100% renewable electricity on the environmental performance of PHA, as described in Section 2.5.

Waste collection is carried out using a EURO6 truck with a capacity >32 t. According to literature (Vea et al., 2021), distances around 50 km are usually estimated. The first stage of the photobiorefinery plant involves all the steps necessary to favour the particle size reduction and the processing of the residues (e.g., separation, grinding) as well as their associated inputs (mainly energy). Following the physical pretreatment stage is the steam explosion, which utilises the complete residue after particle reduction and consumes electricity and steam (self-supplied by the photobiorefinery). The output of this stage is a hydrolysate in reduced solid form. The next stage is fermentation, where the inputs are the hydrolysate from the steam explosion pretreatment, chemicals (KOH to maintain a pH of 5.5), and electricity for agitation. The outputs of this stage are a hydrolysate in reduced solid form and a gas stream composed of 80% H_2 and 20% CO_2, which is used for electricity production in a gas turbine (supplied to the national grid).

In the photoheterotrophic process using raceways, the inputs include the liquid fraction of the hydrolysate, a water stream for diluting the liquid fraction, electricity for agitation and lighting, solar energy, and the area of occupied marginal land. Outputs include the liquid exiting through the membrane (to water treatment and recirculation), biomass with PHA, and direct emissions (i.e., biogenic CO_2). Membrane separation, centrifugation and PHA extraction and processing stages are highly intensive regarding electricity and chemical consumption, especially the latter one. In the anaerobic digestion process, the inputs are the wet solid fraction obtained after fermentation and PHA extraction, the heat from the CHP waste steam in a heat exchanger to maintain a process temperature of 37 °C (self-supplied) and the electricity for agitation and compression of biogas to the CHP plant. The outputs are biogas and digestate. Finally, in the CHP plant, the inputs are the biogas produced and water. Thermal energy in form of waste steam, electricity and direct emissions are the outputs of this process. Steam production from CHP, along with the thermal energy recovered in the steam explosion (1.4 t of steam per t of PHA in total), are directly used in the photobiorefinery, specifically, in the anaerobic digestion and the OFMSW pretreatment stages, respectively. The electricity co-production in the proposed system, particularly in the CHP plant and in the gas turbine after UASB fermentation (2972 kWh·t PHA^{-1} and 5066.67 kWh·t PHA^{-1}, respectively), is supplied directly to the national grid. These values are exceeded by the total electricity consumption in the photobiorefinery, which amounts to 9382 kWh per t of PHA powder produced. In this sense, the highest energy consumption corresponds to the lighting of the raceway reactor; even if they are used only 12 h per day, it consumes 4590 kWh·t PHA^{-1}. It is important to highlight that the influence of using only natural light during the photofermentation stage is analysed and discussed in Section 3.2.

Further information of the mass and energy balances can be found in the Supplementary Material (Section S1). Additionally, the modelling of the counterfactual OFSMW treatment is based on Bassi et al. (2021), and the LCI can be found in the Supplementary Information (Table S9). It is worth noting that this process is also assumed to treat the rejects of the photobiorefinery (Table S10).

2.4. Impact assessment

Brightway2 and its graphical user interface, Activity Browser (Steubing et al., 2020), were used as the LCA software to model the product system and generate the impact results. Brightway2 is an

Table 1
Life cycle inventory to produce 1 t of polyhydroxyalkanoates (PHA) in powder form and treatment of 45.5 t OFMSW.

1. Collection and transport of OFMSW					
Inputs					
Material/Energy flow	Units	Amount	Min	Max	Reference
Transport by truck	t·km	2275.00	1820.00	2730.00	Vea et al. (2021)

2. Pretreatment of OFMSW					
Inputs					
Material/Energy flow	Units	Amount	Min	Max	Reference
Electricity	kWh	110.60	88.48	132.72	Perkoulidis et al. (2010)
Outputs					
Emissions/waste	**Units**	**Amount**	**Min**	**Max**	**Reference**
Transport to treatment (OFMSW reject)	t·km	40.50	20.25	60.74	Bassi et al. (2021)
Waste treatment (OFMSW reject)	t	4.05	4.05	4.05	Cano et al. (2014)

3. Steam explosion					
Inputs					
Material/Energy flow	Units	Amount	Min	Max	Reference
Electricity	kWh	14.11	11.28	16.93	Zimbardi et al. (2002)
Outputs					
Emissions/waste	**Units**	**Amount**	**Min**	**Max**	**Reference**
H_2O (gas)	kg	318.50	318.50	318.50	Based on Allegue et al. (2022)

4. Fermentation in UASB					
Inputs					
Material/Energy flow	Units	Amount	Min	Max	Reference
Electricity	kWh	173.81	139.05	208.57	Bassi et al. (2021)
Calcium oxide	kg	12.29	12.29	12.29	Based on Allegue et al. (2022)
Outputs					
Coproducts	**Units**	**Amount**	**Min**	**Max**	**Reference**
Electricity, from gas turbine (avoided impact of the Spanish electricity mix production by substitution)	kWh	5066.67	4053.33	6080.00	Allegue et al. (2022) & Cano et al. (2014)
Emissions/waste	**Units**	**Amount**	**Min**	**Max**	**Reference**
NOx	kg	456.17	456.17	456.17	Based on NETL (2022)

5. Photoheterotrophic process in raceway					
Inputs					
Flows from nature	Units	Amount	Min	Max	Reference
Land use, marginal land	m^2	0.23	0.23	0.23	Onen Cinar et al. (2020)
Material/Energy flow	**Units**	**Amount**	**Min**	**Max**	**Reference**
Water	m^3	111.14	111.14	111.14	Based on Allegue et al. (2022)
Electricity (pumping and mixing)	kWh	9.22	7.38	11.06	Davis et al. (2016)
Electricity (lighting)	kWh	4590.00	3672.00	5508.00	Based on Allegue et al. (2022)
Outputs					
Emissions/waste	**Units**	**Amount**	**Min**	**Max**	**Reference**
Biogenic CO_2	t	1.10	1.10	1.10	Based on Allegue et al. (2022)
Wastewater	m^3	102.00	102.00	102.00	Based on Allegue et al. (2022)

6. Membrane separation and centrifugation					
Inputs					
Material/Energy flow	Units	Amount	Min	Max	Reference
Electricity (membrane)	kWh	10.88	8.70	13.06	Davis et al. (2016)
Citric acid (cleaning and recovery of the membrane)	g	104.96	104.96	104.96	LaTurner et al. (2020)
NaOCl (cleaning and recovery of the membrane)	g	40.05	40.05	40.05	LaTurner et al. (2020)
Electricity (centrifugation)	kWh	275.00	220.00	330.00	Davis et al. (2016)

7. PHA extraction and processing					
Inputs					
Material/Energy flow	Units	Amount	Min	Max	Reference
Electricity (chemical extraction)	kWh	20.00	16.00	24.00	Bassi et al. (2021)
Electricity (filter press)	kWh	13.00	10.40	15.60	Bassi et al. (2021)
Electricity (dehydrator)	kWh	1954.00	1563.20	2344.80	Bassi et al. (2021)
Sodium hydroxide (NaOH) 50%	t	0.40	0.28	0.40	Bassi et al. (2021)

(*continued on next page*)

Table 1 (*continued*)

7. PHA extraction and processing					
Inputs					
Material/Energy flow	Units	Amount	Min	Max	Reference
Sodium hypochlorite (NaOCl)	t	0.30	0.30	2.00	Bassi et al. (2021)
Water	m^3	40.00	12.00	40.00	Bassi et al. (2021)
Outputs					
Products	**Units**	**Amount**	**Min**	**Max**	**Reference**
PHA powder	t	1.00	1.00	1.00	Based on Allegue et al. (2022)
8. Anaerobic digestion					
Inputs					
Material/Energy flow	Units	Amount	Min	Max	Reference
Electricity (agitation)	KWh	129.90	103.92	155.88	Bassi et al. (2021)
Electricity (compression biogas to CHP)	kWh	2081.48	1665.18	2497.78	Bassi et al. (2021)
Water	m^3	3.20	3.20	3.20	Bassi et al. (2021)
Output					
Coproducts	**Units**	**Amount**	**Min**	**Max**	**Reference**
Digestate	t	10.90	10.90	10.90	Based on Allegue et al. (2022)
Emissions/waste	**Units**	**Amount**	**Min**	**Max**	**Reference**
Biogenic CO_2	t	2.30	2.30	2.30	Based on Allegue et al. (2022)
Biogenic CH_4 (anaerobic digester)	kg	19.11	0.00	72.62	Bassi et al. (2021)
Biogenic CH_4 (storage of digestate)	kg	24.21	2.80	44.59	Bassi et al. (2021)
9. CHP plant					
Outputs					
Coproducts	Units	Amount	Min	Max	Reference
Electricity (avoided impact of the Spanish electricity mix production by substitution)	kWh	2972.00	2377.60	3566.40	Based on Allegue et al. (2022)
Emissions/waste	**Units**	**Amount**	**Min**	**Max**	**Reference**
Direct emissions	See Supplementary Information (Table S10)				

open-source LCA software platform that enables the user to build and analyse LCA models (Mutel, 2017). Regarding the life cycle impact assessment (LCIA) method, this study uses the Environmental Footprint a method for assessing the environmental impact of products and services. It was developed by the European Commission as a common and standardised methodological approach that enables to robustly assess and compare a wide range of environmental impacts (Fazio et al., 2018).

2.5. Sensitivity analysis

The method used to address the uncertainty associated with the LCA results was the error propagation via Monte Carlo simulation with 1000 iterations. This method involves randomly sampling parameters based on their probability distribution and computing the results for each iteration. The probability distribution assigned to each parameter is provided in Table 1. The uncertainty related to the background processes was directly considered from the ecoinvent database v3.8. This method was used to provide a more accurate estimate of the range of possible outcomes and to identify the most significant sources of uncertainty in the LCA model. Regarding the sensitivity analysis, we focused on the main parameters addressed in the literature: efficiency of the PHA production stages (Asunis et al., 2021; Bassi et al., 2021; Vea et al., 2021), the composition of the national electricity mix used (Asunis et al., 2021; Bassi et al., 2021), % methane losses in anaerobic digestion systems and in combustion engines (Bassi et al., 2021), recirculation of chemical extraction agents to avoid its use or use of alternative agents with less environmental impact (e.g., dimethyl carbonate) (Bassi et al., 2021). We also explored the effects of multifunctionality, given the relevance of biorefineries for generating co-products with high-added value (Puyol et al., 2017; Sekoai et al., 2021; Vea et al., 2021). The influence of all these parameters and others on the environmental impact results was explored in a preliminary LCA. As a result, we modelled five additional scenarios to test the robustness of the results in different conditions: a) minimisation of methane leakages, b) use of renewable electricity, c) alternative use of environmentally friendly extraction agents, d) co-production of hydrogen and e) PHA powder production solely using natural light. For the third scenario, the alternative extraction agent chosen was dimethyl carbonate, a versatile reagent with relatively low toxicity for human health and the environment and a high recyclability rate. We modelled this alternative process according to the information found in (Asunis et al., 2021) (see Supplementary Information). For the fourth scenario, Fig. 2 presents the new system boundaries of the photobiorefinery system, including two new steps for H_2 production, namely PSA purification, and compression (250 bar). Alternative inventory data for all the proposed scenarios can be found in Supplementary Information (Tables S11-S13).

3. Results and discussion

3.1. Life cycle impact assessment

This section presents the LCIA results for the base case scenario of PHA powder production and OFMSW treatment. Fig. 3 provides a breakdown of these results by life cycle stages, namely avoided management of OFMSW through conventional treatment, PHA powder production in the photobiorefinery, waste management from the photobiorefinery, and avoided production of the Spanish electricity mix. This figure also includes the probability distribution of impacts based on Monte Carlo simulation. For benchmarking purposes, the results presented herein are compared against three fossil-based plastics in granulates or unprocessed form, namely low-density polyethylene (LDPE), polyethylene terephthalate (PET), and polyurethane (PU), which were selected based on their technical features and ability to perform a function similar to PHA (Baioli et al., 2019; Bassi et al., 2021). Given

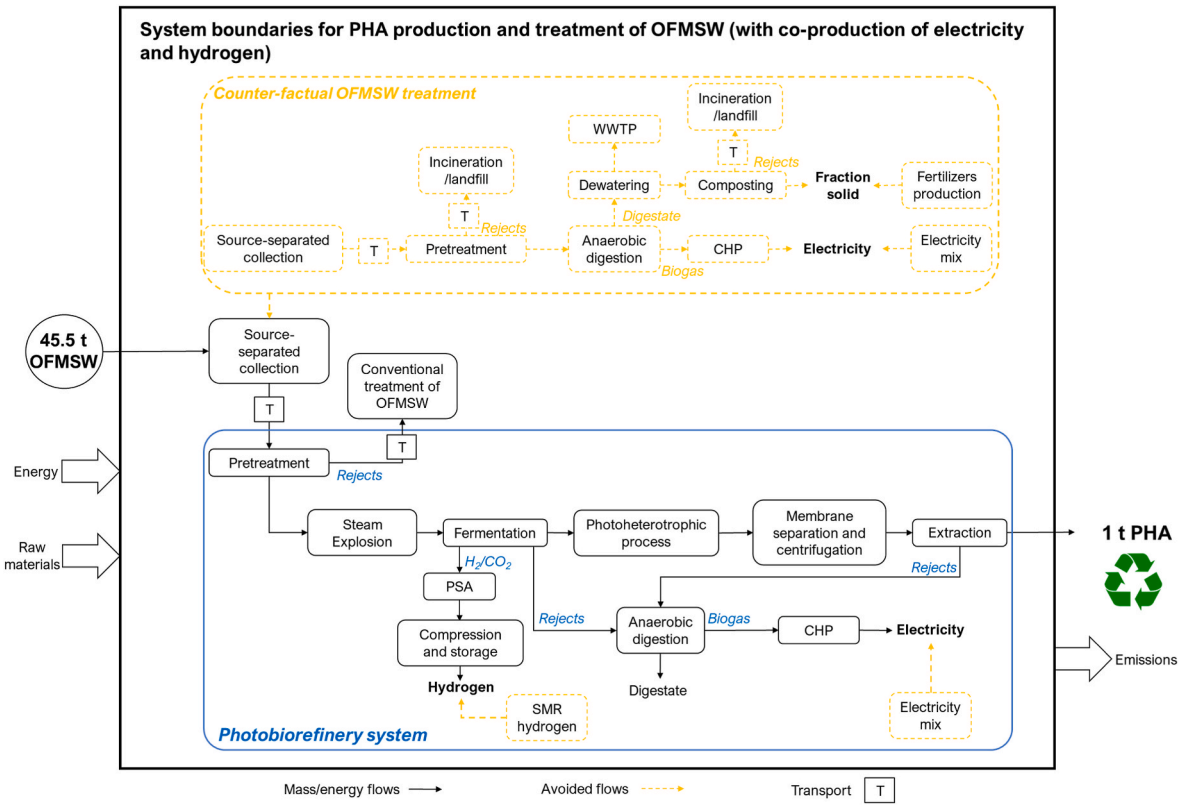

Fig. 2. System boundaries for the production of polyhydroxyalkanoates (PHA) in powder form and treatment of OFMSW. Scenario of the co-production of electricity and hydrogen.

that this study does not specify a particular function for the PHA produced and the focus on the photobiorefinery pathway, 1:1 replacement ratio was selected as a reasonable choice (Bassi et al., 2021). The impact results for these plastics are represented as red lines in Fig. 3. To focus our analysis and discussion, we highlight six impact categories, namely climate change, ozone depletion, acidification, eutrophication (freshwater), resource use (fossils) and resource use (minerals and metals). Results for the complete range of impact categories of the Environmental Footprint method are available in the Supplementary Information (Table S14).

The LCIA results for the base case scenario show a mean value of 1.88E t CO_2 eq·t PHA^{-1} for climate change. This value falls in the lower part of the range reported in the literature, which usually are between 1 and 6 t CO_2 eq·t PHA^{-1} (Baioli et al., 2019). The photobiorefinery stage was found to have the most significant contribution to the climate impact category, mainly due to direct emissions from the anaerobic digester and cogeneration (mainly biogenic methane and dinitrogen monoxide), which accounted for 2.26 t CO_2 eq·t PHA^{-1}. The PHA accumulation and extraction stages also significantly contributed to the climate impact category, with more than 2 t CO_2 eq·FU^{-1} resulting from the consumption of electricity and the production of extractive chemical agents used. It is worth noting that the avoided processes resulted in a significant environmental saving of around 4 t CO_2 eq·t PHA^{-1}, which provided a lower net result than those found in the literature. When the carbon footprint results are compared to conventional plastics in granulate or unprocessed form, which vary between the 1.8–5.4 t CO_2 eq·t plastic evaluated^{-1}, PHA powder production provides impact reductions of more than 30%–60% compared to PET and PU, respectively. However, PHA production is not competitive with LDPE. The uncertainties associated with the Monte Carlo simulation are significant, but they confirm the study's conclusions when benchmarked against PET and PU.

There is a close relationship between the categories of impacts of climate change and consumption of fossil resources, which means that the conclusions drawn regarding the former are mainly applicable to the latter. Thus, the main impacts of fossil resource use are concentrated in the PHA accumulation and extraction stages, which are energy- and material-intensive. However, significant environmental benefits are also found due to the avoided production of the average electricity mix production for the year 2021, and, in particular, the avoided production from fossil-based power technologies (which account for around 25% of the electricity mix). This leads to a net impact value of around 70 GJ, which also falls within the lower range of results reported in the scientific literature (Baioli et al., 2019). Considering the uncertainties associated with the Monte Carlo simulation, it can be concluded with a high degree of confidence that PHA powder production results in lower use of fossil resources throughout the life cycle compared to the three types of conventional plastics in granulate form evaluated, with impact reductions ranging from 38% to 51%.

The hotspot analysis found similar results for the remaining impact categories, but the benchmarking results against conventional plastics were markedly different. The photobiorefinery plant was the primary contributor to the impact in all four categories. Electricity demand and the use of extractive chemical agents were the main drivers of ozone depletion, eutrophication (freshwater), and resource use (minerals and metals), with contributions exceeding 70% in all cases. Meanwhile, direct emissions from the anaerobic digester and cogeneration were the main factor in acidification, contributing over 65% to the impact. The avoided processes associated with PHA powder production also had significant environmental benefits, particularly in the avoided production of the average electricity mix. When the environmental profile of these categories was benchmarked against fossil-based plastics in granulate or unprocessed form, the average performance was always worse than that of LDPE. The results from 1000 iterations of the Monte Carlo simulation provided confident conclusions for the impact categories of ozone depletion, acidification, and eutrophication (freshwater). Comparisons with PET and PU also revealed that the impacts of

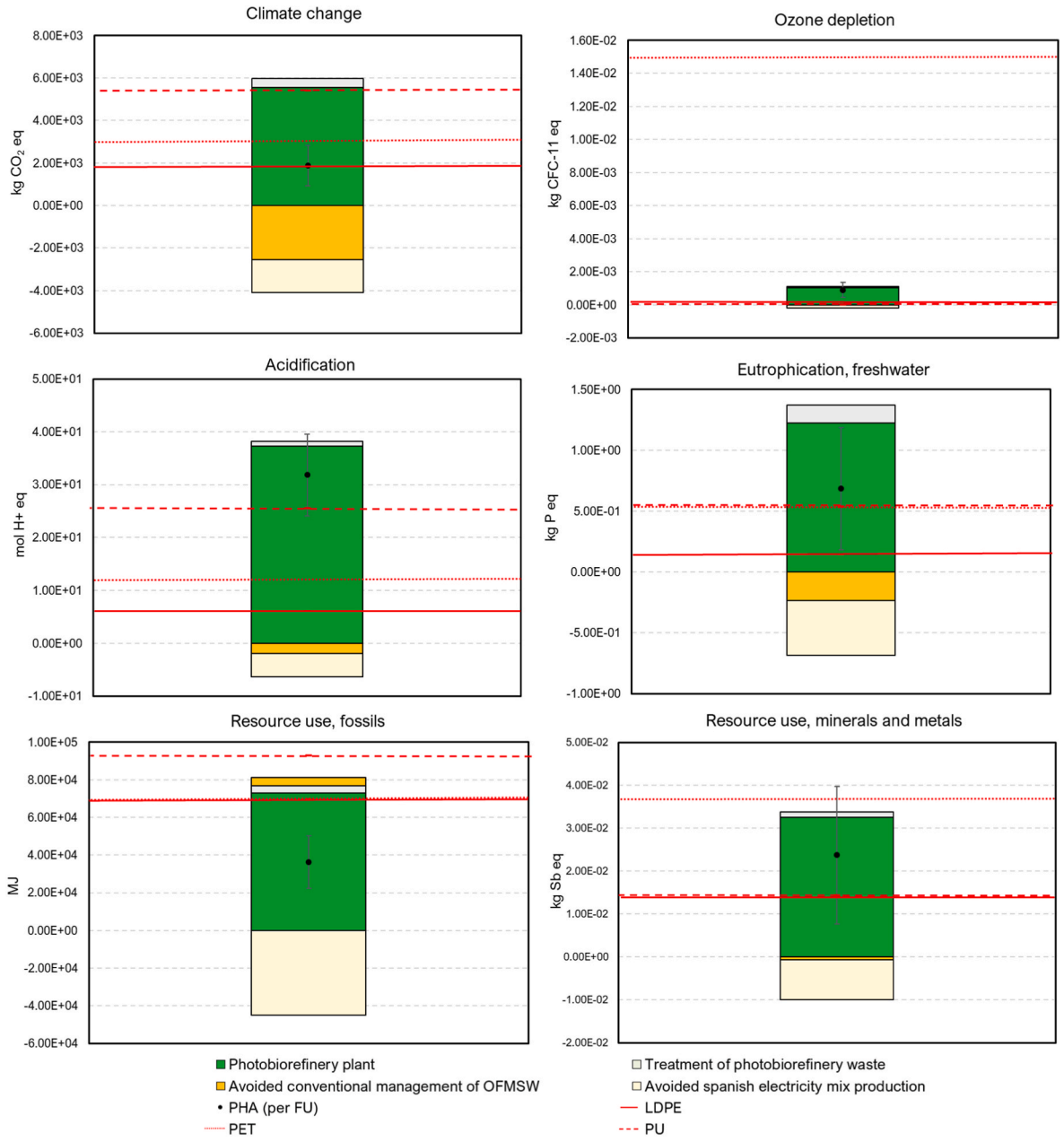

Fig. 3. Life cycle impact assessment results for the production of 1 t of polyhydroxyalkanoates (PHA) in powder form and treatment of 45.5 t OFMSW.

PHA powder production were consistently higher in the 1000 estimated iterations for the categories of acidification and ozone depletion, respectively. However, in the other cases, the associated levels of uncertainty did not allow for a clear preference to be determined between the biopolymer studied and conventional plastics.

3.2. Sensitivity analysis

Fig. 4 presents the sensitivity of the LCIA results to the six additional scenarios presented in Section 2.5: a) minimisation of methane leakages, b1) use of solar PV electricity, b2) use of wind electricity, c) avoiding sodium hypochlorite and alternative use of environmentally friendly extraction agents, d) co-production of hydrogen, and e) PHA production solely using natural light. Results for the complete range of impact categories of the Environmental Footprint method are available in the Supplementary Information (Table S15).

Fig. 4 (Sc A) depicts how the life cycle impact results are affected by reducing methane losses in the PHA powder production system, primarily in the anaerobic digestion, storage of digestate and cogeneration stages. Details on the modelling of the methane leakages minimisation are included in the Supplementary Material (Table S11). The differences between scenarios are only observed for the climate change impact category, which displayed a remarkable 110% reduction in carbon footprint compared to the base case (-196.68 kg CO_2 eq·FU^{-1}). With this new scenario, PHA production can compete favourably in carbon footprint compared to fossil-based plastics. These results highlight the significance of reducing methane losses to mitigate climate change. Methane losses can occur due to several factors, including incomplete digestion, leaks in the system, and venting during maintenance. Proper management and monitoring of the anaerobic digestion and cogeneration processes can help to minimise these losses and ensure optimal biogas production (IEA, 2022a,b). Additionally, if these measures are combined with actions on certain CHP parameters, e.g., increase in CHP efficiency, greater avoided impacts per functional unit can be achieved.

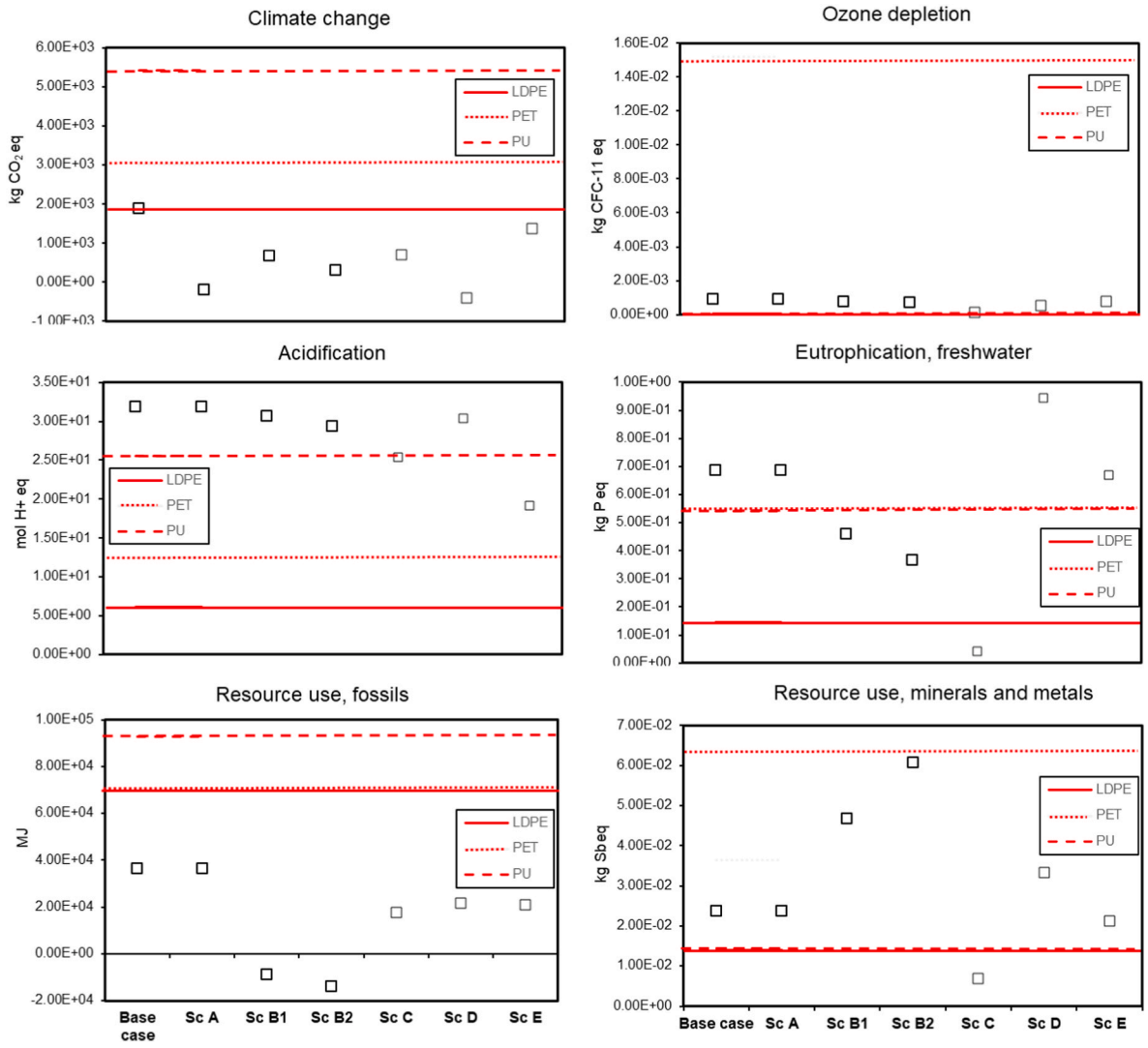

Fig. 4. Life cycle impact assessment results for the production of 1 t of polyhydroxyalkanoates (PHA) in powder form and treatment of 45.5 t OFMS. Scenario A: minimisation of methane leakages; Scenario B1: solar PV electricity; Scenario B2: wind electricity; Scenario C: alternative PHA extraction; Scenario D: co-production of hydrogen; Scenario E: 100% natural light (this scenario involves the treatment of 22.8 t OFMSW per t of PHA powder).

A 10% efficiency enhancement results in a significant 10% reduction in the carbon footprint indicator, ranging the rest of impact categories from reductions of 2% in acidification to as much as 15% in resource use (fossil fuels). However, variability in the biogas heating value, an important factor in cogeneration efficiency, also plays a role in environmental performance. Upon evaluating the influence of using a lower heating range of data, it was found that such a modification would lead to increased impacts across the selected indicators. This effect is particularly pronounced, with some impacts showing an upward shift exceeding 8%, as observed in the carbon footprint indicator. Thus, optimizing methane management and enhancing CHP efficiency contribute significantly to the environmental feasibility of PHA production within a circular biorefinery framework.

Fig. 4 (Sc B1 and B2) shows the influence of the type of electricity used on the environmental impact results. Specifically, the analysis examined the influence of using electricity generated from renewable sources, namely solar photovoltaic and wind, instead of the Spanish electrical mix in the foreground processes. The LCIs of these systems were directly modelled from available datasets in ecoinvent 3.8 (Table S8 from the Supplementary Material). The results show that using renewable electricity can significantly reduce the environmental impact of PHA powder production, with the most substantial improvements observed in climate change, eutrophication, and the use of fossil resources. For instance, solar photovoltaic or wind electricity can decrease the carbon footprint by more than 65% and 85%, respectively. However, using renewable electricity may also penalise specific impact categories, as in the case of resource use (metals and minerals). Despite this, adopting renewable electricity for PHA powder production can significantly improve its environmental performance and provide a more sustainable alternative to conventional plastics. Therefore, implementing this measure in photobiorefineries should be considered a crucial step towards transitioning to a more sustainable and environmentally friendly bioeconomy in the short term.

Fig. 4 (Sc C) examines the impact of environmentally friendly extraction agents, specifically dimethyl carbonate (DMC), on the LCIA results. The LCI of these unit processes was directly modelled from available datasets in ecoinvent 3.8 (see Table S8). DMC has a significant positive impact on all impact categories, especially eutrophication (freshwater) and resource use (metals and minerals), resulting in a more favourable environmental performance of PHA powder compared to the three conventional unprocessed plastics evaluated in four of the six impact categories studied. This aligns with the literature as DMC is less toxic than traditional solvents (Andreasi Bassi et al., 2021; Asunis et al., 2021; Righi et al., 2017). Furthermore, DMC can effectively extract PHA

from the biomass and is easily recovered and reused, reducing waste and improving process efficiency. Therefore, using DMC as an extraction agent for PHA production can provide a more sustainable and environmentally friendly alternative to traditional extraction methods, ultimately reducing the overall environmental impact of the biopolymer production process.

Fig. 4 (Sc D) illustrates the sensitivity of the results to co-producing hydrogen in a photobiorefinery plant. Following the rationale presented in Section 2.2.1, the environmental benefits of the hydrogen co-produced in the photobiorefinery were obtained by substituting the avoided impacts of the conventional benchmarked production, namely, steam methane reforming (SMR). As a result, the co-production of hydrogen has a notably positive effect on the climate change category, reducing the impact by more than 2 t of CO_2 eq compared to the base case. However, some impact categories, such as eutrophication and resource use (minerals and metals), are negatively affected due to the increased impact associated with additional electricity consumption to purify and compress the hydrogen, which is not fully offset by the avoided production of SMR hydrogen. In this regard, the total energy consumption of the plant for this scenario amounts to 12,591.9 $kWh{\cdot}FU^{-1}$, while the electricity co-production corresponds to 2972 $kWh{\cdot}FU^{-1}$. Consequently, there is a net increase in impact for these categories. Overall, the co-production of hydrogen can help to improve the environmental profile of PHA powder production in photobiorefineries as long as it combines with additional measures aimed at sustainability, e.g., the use of renewable electricity or additional co-production. Specifically, considering the potential inclusion of digestate management and biofertilizer production within the boundaries of the system, along with co-production of hydrogen, could yield significant reductions in environmental impacts. While digestate management is not typically identified as a critical hotspot in this type of systems, its environmental benefits, such as the avoidance of commercial fertilizers and potential nutrient recovery, are noteworthy. In summary, the co-production of hydrogen in photobiorefineries represents a potential opportunity to increase biobased polymers' efficiency, sustainability, and profitability. Additionally, increased co-functions of the system, alongside PHA production, could further enhance the sustainability potential of the photobiorefinery, aligning with circular economy principles and reducing overall environmental burdens. Future research may explore the feasibility and quantifiable benefits of integrating digestate management into the system boundaries, ultimately contributing to a more comprehensive sustainability picture of emerging biopolymer pathways.

Fig. 4 (Sc E) addresses the sensitivity of the results to the energy consumption associated with the lighting of the photofermentation process. In this regard, only natural lighting was considered for the raceway reactor. Although this reduces the system's productivity by half (the photobiorefinery jointly manages 22.75 t of OFMSW per t of PHA in powder form, and the maximum annual production is around 300 t of PHA), the energy balance is favourably improved, resulting in a reduction of approximately 60% in total electricity consumption. This fact translates into improvements in all impact categories, with key environmental indicators, namely climate change, fossil resource use, and acidification, positively influenced by the decreased energy demand. Although the productivity of the photofermentation process is lower due to limited hours of optimal lighting, the environmental benefits outweigh this drawback. The reduction in electricity consumption contributes to a more sustainable and resource-efficient production process. Furthermore, the use of natural light aligns with the principles of renewable energy and supports the transition to low-carbon biorefineries. Overall, this strategic choice of relying exclusively on natural light for the photofermentation process in PHA powder production represents a viable approach to minimizing the environmental impacts, especially if is combined with additional measures (e.g., alternative PHA extraction), and demonstrates how the integration of sustainable design into the scalability process can strike a balance between productivity and energy efficiency.

In conclusion, the comprehensive examination of the photobiorefinery system presented in this study sheds light on its pivotal role in advancing sustainable resource management, circular economy principles, and the attainment of key Sustainable Development Goals (SDGs), particularly SDG 7 (Affordable and Clean Energy) and SDG 11 (Sustainable Cities and Communities). The sensitivity analysis unveiled the potential of improvement measures, such as co-production of hydrogen, integration of renewable electricity sources, and utilization of environmentally friendly extraction agents, to significantly mitigate the environmental impact of PHA powder production while fostering a more resource-efficient process. The photobiorefinery's inherent ability to harmonize waste-energy nexus and capitalise on natural light underscores its potential to strike a balance between productivity and energy efficiency (Alao et al., 2022). Such advancements exemplify the promise of photobiorefinery systems in propelling the transition towards a more sustainable bioeconomy, aligning closely with global sustainability agendas, and facilitating the responsible stewardship of resources. Furthermore, the relevance and applicability of these cleaner production strategies extend beyond the scope of this study, offering valuable insights for broader regional and global contexts (Kisser et al., 2020; Koley, 2023).

4. Conclusions

This study provides a comprehensive assessment of the potential environmental implications of PHA powder production and the joint management of OFMSW through a novel photobiorefinery system that uses PPB. While the base case results obtained from the LCIA and the Monte Carlo simulation, reveal a favourable performance of PHA powder production in terms of carbon footprint and fossil resources use, particularly when benchmarked against granulates or unprocessed fossil-based PET and PU, they also underscore the need for improvement measures, especially when the comparison is carried out against LDPE in granulates form or under other impact categories, such as eutrophication or acidification. The sensitivity analysis highlights the potential benefits of mitigation strategies in the photobiorefinery like minimisation of methane losses or the co-production of hydrogen, as well as the utilization of sustainable practices such as the use of environmentally friendly extraction agents such as DMC, renewable electricity or only natural light for the photofermentation process. Thus, implementing feasible improvement measures in the short term can position PHA produced by PPB as a sustainable alternative to petroleum-based plastics. Moreover, this study underscores the importance of adopting a sustainable-by-design approach in the early stages of biorefinery development significantly enhancing the environmental performance of emerging products, such as PHA. Based on exploratory LCA, this pre-market approach not only enables decision-makers to identify and address potential environmental impacts throughout the product life cycle but also helps to build consumer trust and confidence in emerging biopolymers, ultimately supporting their successful adoption and commercialisation in the market. Future lines of work will focus on further quantitative analysis, such as Social Life Cycle Assessment, completing the sustainability picture of the PHA production presented in this paper, more comprehensive environmental evaluation of PHA from photobiorefineries across its entire life cycle, including its use and end-of-life phases, and direct comparisons with fossil plastics in specific applications.

CRediT authorship contribution statement

Mario Martin-Gamboa: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Visualization. **Luis D. Allegue:** Conceptualization, Formal analysis, Investigation, Writing – original draft, Visualization. **Daniel Puyol:** Conceptualization, Writing – review & editing, Supervision. **Juan Antonio Melero:**

Conceptualization, Writing – review & editing, Supervision. **Javier Dufour:** Methodology, Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare no conflict of interest.

Data availability

All the research data is shared in the Supplementary Material

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2023.139421.

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