# Polyhydroxyalkanoates (PHA) production from biogas in waste treatment facilities: Assessing the potential impacts on economy, environment and society

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**Abbreviations:** Combined Heat and Power (CHP)**;** Elimination Capacity (EC)**;** Empty Bed Residence Time (EBRT)**;** Height-to-diameter ratio (H/D)**;** Internal Rate of Return (IRR)**;** Municipal Solid Waste (MSW)**;** Net Present Value (NPV)**;** Pressure (P)**;** Purchased Equipment Cost (PEC)**;** Polyhydroxyalkanoate (PHA)**;** Removal Efficiency (RE)**;** Temperature (T)**;** Total Investment Cost (TIC)**;** Volume (V)**;** Volatile Organic Compounds (VOCs)**;** Volatile Sulfur Compounds (VSCs)

**Abstract**

Using the biogas generated from organic waste anaerobic treatment to produce polyhydroxyalkanoates (PHAs) has emerged as an attractive alternative to heat and power generation (CHP) in waste treatment plants. The sustainability of biogas combustion for CHP, biogas bioconversion into PHA and a combination of both scenarios was compared in terms of environmental impact, process economics and social responsibility according to the IChemE Sustainability Metrics. Although PHA production presented higher investment and operational costs, a comparable economic performance was observed in all biogas valorization scenarios regarding net present value (0.77 M€) and internal rate of return (6.4±0.2%) due to the higher market value of biopolymers. The PHA production entailed a significant reduction of atmospheric acidification and odor emissions compared to CHP despite showing higher land, water, chemicals and energy requirements. Job creation associated to biopolymer industry and the increasing public demand for bioproducts were identified as fundamental aspects for enhancing social and local acceptance of waste processing facilities. This study demonstrated that PHA production from biogas constitutes nowadays a realistic alternative to CHP in waste treatment plants and that PHA can be produced at a competitive market price when biogas is used for internal energy provision (4.2 €·$kg^{-1}$ PHA).

**Keywords:** Biogas valorization; Biopolymer; Circular economy; IChemE Sustainability Metrics; Methanotrophic bacteria; Polyhydroxyalkanoates.

## 1. Introduction

Biogas is a renewable resource produced during the anaerobic digestion of organic substrates and it is composed of variable concentrations of $CH_4$ (40-75%), $CO_2$ (25-60%), $N_2$ (0-2%), $O_2$ (0-1%), $H_2S$ (0.005-2%) and other minor compounds [1]. The global biogas production was estimated at 58.7 billion $Nm^3$ in 2014, with an associated energy production potential of $3.5 \cdot 10^5$ GWh [2]. Government support schemes and fiscal incentives introduced in the past decades, including feed-in tariffs and tax exemptions, have encouraged its utilization to produce renewable energy (62,704 GWh produced in 2017 in Europe) [3]. Methane ($CH_4$) present in biogas can be transformed into electric and thermal energy in combined heat and power production (CHP) gas engines. Despite latest CHP systems are able to recover up to 85% of the total energy present in biogas, the high capital investment, the excessive operation and maintenance costs and the reduced lifespan of CHP engines limit their economic viability [4]. Consequently, massive amounts of biogas are often flared or vented to the atmosphere (92 Mt $CH_4 \cdot y^{-1}$ according to the World Bank), significantly contributing to the global emission of greenhouse gases [5].

Recent changes in regulatory frameworks, such as the reduction of fiscal incentives and the emergence of new strategies for waste and plastic management in a circular economy, have shifted the attention towards the use of the major biogas components ($CH_4$ and $CO_2$) as building blocks for the chemical industry [6, 7]. Particularly, the use of biogas to produce polyhydroxyalkanoates (PHAs) has emerged as an attractive alternative to conventional heat and power generation. This is especially relevant for municipal and agroindustrial waste treatment facilities, which account for 80% of the total biogas plants installed in Europe [3]. These polyesters are currently being used to produce biodegradable plastics with mechanical characteristics similar to those of traditional oil-based plastics, with a significant minimization of the associated environmental impacts [6, 7]. However, the high cost of raw materials

(especially carbon substrates), accounting for 40-50% of the total PHA production costs, increases the current PHA selling price (4-20 €·$kg^{-1}$ PHA), making it difficult to compete with conventional plastics [8]. In this context, the biogas produced in bulk quantities in waste treatment plants and landfills constitutes a globally available and low-cost source of $CH_4$ for PHA production that could potentially replace 20-30% of the global plastic demand [9]. Overall, integrating the production of high added-value products in waste processing facilities could potentially enhance their economic viability but also respond to the growing consumer demand for renewable bioproducts and anticipate the increasingly restrictive environmental policies.

In this paper, the use of biogas for heat and power generation and as feedstock for biopolymer production was compared in terms of environmental impact, process economics and social responsibility according to the IChemE Sustainability Metrics [10]. A third scenario involving the use of a fraction of biogas to cover the energy demand of PHA production from biogas was also considered. To the best of the authors' knowledge, this is the first study evaluating the sustainability aspects of PHA production from biogas integrated in a waste treatment facility.

## 2. Materials and methods

### 2.1. Methodology, goal and scope definition

This comparative assessment is based upon the triple-bottom-line of sustainability, which combines the evaluation of environmental responsibility, economic performance and social development. A medium-size municipal solid waste treatment plant (750,000 person-equivalent) treating 300 t·$d^{-1}$ of organic urban waste with a biogas production of 24,000 $Nm^3 \cdot d^{-1}$ at an average composition of 60, 35, 2.5, 0.5, 0.4 and 1.6% in $CH_4$, $CO_2$, $N_2$, $O_2$, $H_2S$ and minor compounds, respectively, was considered as a base case scenario. The IChemE Sustainability Metrics provide a set of ratio indicators for measuring process impact [10]. In

this particular study, most ratio indicators are referred to 1,000 $Nm^3$ of biogas, which corresponds to the hourly biogas production of the plant. Three different scenarios were considered for evaluation: (I) combustion of all the biogas in CHP units for electricity and heat production, (II) bioconversion of the $CH_4$ contained in biogas into PHA followed by extraction and purification, and (III) a combination of scenarios I and II, where the power and heat necessary for PHA production, extraction and purification are provided by biogas-fueled CHP units (Fig.1).

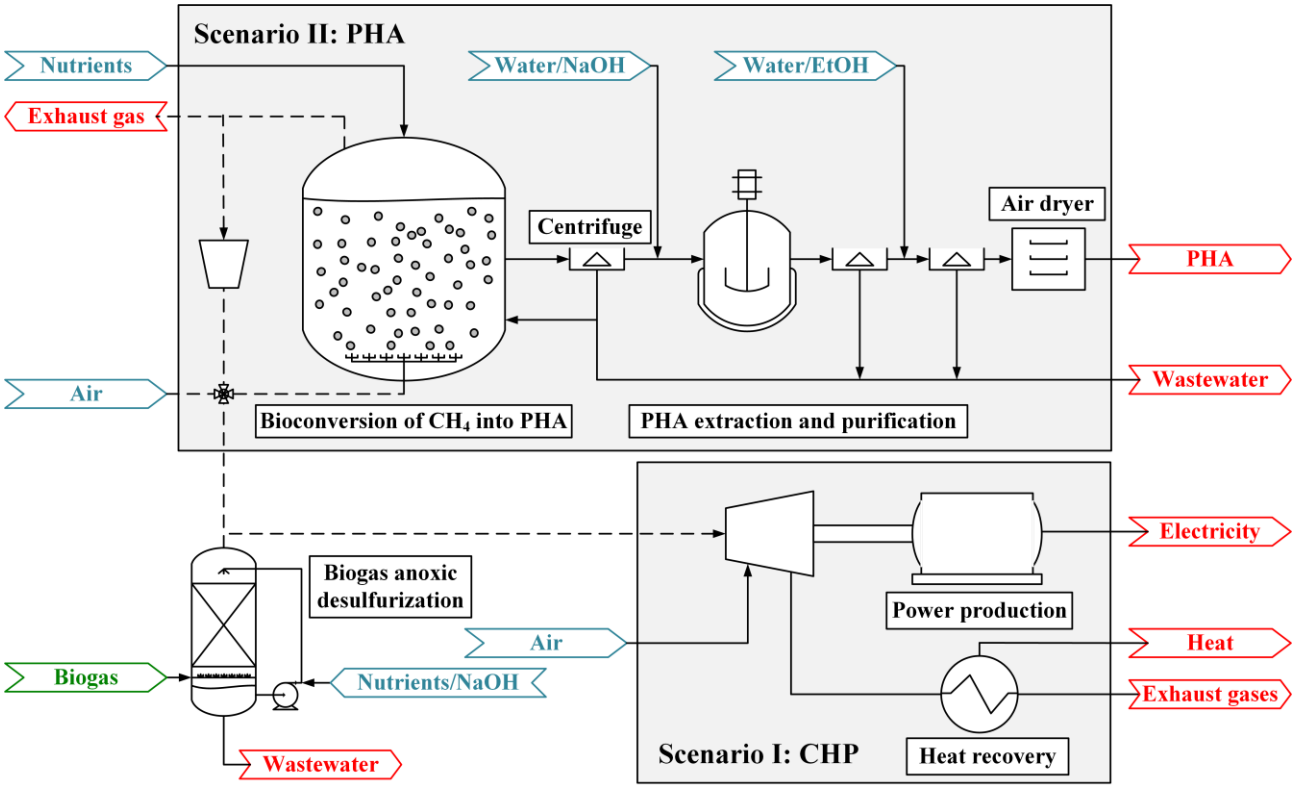



**Figure 1**. Simplified process flow diagram for desulfurized biogas combustion in CHP units (Scenario I) and for the bioconversion of $CH_4$ into PHA (Scenario II).

## 2.2. Process design

Biogas thermodynamic properties such as density, calorific power and Wobbe index were obtained from the Swedish Gas Technology Center report [11]. Relevant design parameters for desulfurization (e.g. $H_2S$ removal efficiency (RE), nitrate requirements), CHP (e.g. air excess, power efficiency) and PHA extraction (e.g. pH, product recovery and purity) were obtained from the literature [4, 12, 13, 14, 15]. $CH_4$-RE, $CH_4$ elimination capacity (EC), PHA productivity and other operational parameters for the production of PHA from biogas were obtained from previous experimental studies conducted in our laboratory [16, 17]. Detailed information on the design parameters and sizing of the equipment can be consulted in the Supporting information (Table S1).

### 2.2.1. Biogas desulfurization

A biogas desulfurization stage for reducing $H_2S$ content below 100 $ppm_v$ was designed before biogas valorization. Although this step is common to all biogas valorization alternatives, its economic and environmental performances were included in the present study for a better estimation of the overall impact of the whole biogas valorization processes. Biological anoxic desulfurization was selected among all commercial physicochemical and biological desulfurization technologies based on its cost-effectiveness, low demand for chemical reagents and high $H_2S$-EC (>120 g $S \cdot m^{-3} \cdot h^{-1}$) [18]. Anoxic desulfurization of biogas is based on the oxidation of $H_2S$ by sulfur oxidizing bacteria able to use nitrate instead of $O_2$ as electron acceptor for the partial or complete oxidation of $H_2S$ to elemental sulfur or sulfate, respectively [19]. A biotrickling filter packed with a mixture of inert plastic material and activated carbon was designed with an empty bed residence time (EBRT) of 3 min for the desulfurization of biogas. The anoxic removal of volatile organic compounds (VOCs) and volatile sulfur compounds (VSCs) was considered negligible during this stage, given the low operating EBRT [20, 21]. Mineral medium (50 $g \cdot L^{-1}$ of $NaNO_3$ and 1 $mL \cdot L^{-1}$ of micronutrients solution) was

considered to be continuously sprayed over the packed bed with a trickling liquid velocity of 10 $m \cdot h^{-1}$. Complete $H_2S$ oxidation is ensured by addition of nitrate in excess (2.5 mol N added per mol S removed), increasing selectivity towards the formation of sulfate and avoiding the precipitation of elemental sulfur in the packing media. Liquid in the biotrickling filter was renewed when nitrate concentration fell below 0.01 g $N\text{-}NO_3^-$ $L^{-1}$ [14].

### 2.2.2. Combined heat and power production

In CHP systems, electricity is generated by the combustion of desulfurized biogas and subsequently, heat is recovered from the combustion exhaust gases in the form of steam or hot water. A gas engine was selected as model technology for the co-production of electricity and heat in scenarios I and III as they are considered the most cost-effective (400-1,100 €·$kW_{el}^{-1}$) and efficient (70-85% of energy recovery) alternative for CHP in small and medium size plants (1,100-3,000 $kW_{el}$ installed) [4]. The amount of air supplied for combustion constitutes a key operational parameter of CHP plants and it is measured by the λ factor, which correlates the real air molar flow supplied with the stoichiometric amount necessary for complete combustion. λ factors lower than the stoichiometric value (< 1) result in high electricity and heat recovery performance, but also in an increased pollutant emission due to incomplete combustion of the fuel. A λ factor of 1.3 is usually recommended for biogas mixtures. A total efficiency (η) of 85% was considered in this study, 40% ($\eta_{el}$) as electricity and 45% ($\eta_{th}$) as thermal energy. Complete oxidation of $CH_4$ and the residual $H_2S$ to $CO_2$ and $SO_2$, respectively, was assumed for calculation purposes. CO, $SO_2$ and $NO_X$ emissions of 8.46, 1.25 and 6.96 g·$Nm^{-3}$ biogas, respectively, were selected according to Paolini et al. [22]. Finally, an average annual operating time of 8,000 h was set for the CHP gas engines.

### 2.2.3. PHA biosynthesis, extraction and purification

A bubble column bioreactor equipped with internal gas recycling was chosen for methanotrophic culture growth and subsequent PHA accumulation in a single stage. The $O_2$ required for biological $CH_4$ oxidation was provided by the addition of air at a molar ratio $O_2$:$CH_4$ = 1.5 [17]. An internal gas recirculation rate ($Q_r/Q_0$, where $Q_r$ and $Q_0$ stand for the recirculation and fresh gas inlet molar flows, respectively) of 10 was selected in order to increase turbulence and ensure a $CH_4$-RE of 90%. $CH_4$-EC was estimated at 60 g $CH_4 \cdot m^{-3} \cdot h^{-1}$ during both the growth and the PHA accumulation phases [18]. Almost complete solubilization and biodegradation of the VOCs and VSCs present in biogas was considered at this stage given the high gas-liquid contact time in the bubble column bioreactor (EBRT ~ 1.2 h ) [23].Biomass (0.67 g biomass$\cdot g^{-1}$ $CH_4$) and PHA (0.55 g PHA$\cdot g^{-1}$ $CH_4$) yields on $CH_4$ were selected according to stoichiometry (Supplementary material: S2). A fed-batch strategy was implemented, consisting of an initial growth phase in which fresh mineral medium (30 $g \cdot L^{-1}$ of $NaNO_3$ and 1 $mL \cdot L^{-1}$ of Whittenbury micronutrients solution) is supplied to the culture broth at a dilution rate of 0.03 $d^{-1}$, to attain a final biomass concentration in the cultivation broth of 30 g biomass$\cdot L^{-1}$, followed by a nutrient-limiting stage in which PHA is accumulated up to 40% $w \cdot w^{-1}$ [24]. It was assumed that 7.5% of the accumulated PHA is consumed during the growth phase as energy source by methanotrophic bacteria [17]. At the end of the accumulation phase, 25% of the liquid medium is extracted for PHA downstream processing. This value was selected as the optimum dilution rate in order to maintain a constant biomass concentration, avoid metabolites accumulation and allow continuous process operation [17].

A NaOH digestion method, described by Lopez-Abelairas et al. (2015), was selected for PHA extraction and purification. This method was selected due to its low-cost, reduced equipment requirements, low environmental impact, and high product recovery (80%) and purity (92%) [16]. According to the described method, culture broth from the bioreactor is centrifuged to increase solid content up to 8% $w \cdot w^{-1}$. After centrifugation, 90% of the liquid fraction is

recycled to the PHA bioreactor to recover residual biomass and nutrients. NaOH is then added for the digestion process (0.8 g NaOH·g biomass$^{-1}$) which takes place in a continuous stirred tank reactor at 37 ºC for 5 h. Complete biomass solubilization and negligible PHA losses were assumed during this process. The product stream is then concentrated to 10% w·w$^{-1}$, and 60% of the liquid fraction is reused, given its high NaOH content. The heavy fraction is then double-washed with water and ethanol and subsequently dewatered to a concentration of 40% w·w$^{-1}$. Finally, solid PHA is dried with air (60 ºC) in a desiccator tray to a final PHA purity of 92%.

### 2.3. Capital and operational costs

The net present value (NPV), internal rate of return (IRR) and payback period were used as economic performance indicators. Net present value was calculated according to Equation (1) [25].

$$NPV = \sum_{t=0}^{t=20} \mathrm{FCF}^{\mathrm{t}}/(1+\mathrm{i})^{\mathrm{t}} - \mathrm{TIC} \qquad (1)$$

Where *FCF* stands for the free cash flow at time *t*, *i* represents the interest rate and *TIC* accounts for the total capital investment at year 0. A time of 20 years, a tax rate of 30% and an interest rate of 5% were considered for NPV calculations. For the calculation of *FCF*, capital investment was assigned to year 0, linear depreciation over the first 10 years of the project was assumed and a circulating capital of 2% over the initial capital investment was set. IRR was calculated as the value of the interest rate at NPV equal to zero.

Costing for CHP was calculated according to Wellinger et al. (2013), assuming an investment cost of 1,000 €·kW$_{el}$$^{-1}$ and operation and maintenance costs of 0.015 €·kWh$^{-1}$ [4]. Since CHP units should be replaced after 80,000 working hours (as per manufacturers' instructions), gas engines cost was considered in duplicate in this analysis for a 20-year horizon. The TIC of anoxic desulfurization, PHA production and extraction for the different scenarios was estimated based on the individual purchased equipment cost (PEC) according to Lang's method. A Lang

196 factor of 4.1 was selected as recommended by Ulrich et al. (2004) for solid-liquid processes
197 [26, 27]. Individual equipment costs were determined by comparing literature, online
198 equipment quoting tools and queries to international manufacturers [28, 29]. All prices were
199 updated to 2019 € considering an annual inflation rate of 1.94% (2017 to 2022 expected EU-28
200 inflation rate). PEC and TIC for the different scenarios are shown in Table 1.

201 **Table 1**. Summary of purchased equipment cost and total investment cost.

| Item | Cost (€) | | |
|---|---|---|---|
| | **I CHP** | **II PHA** | **III CHP+PHA** |
| Biotrickling filter | 114,000 | 114,000 | 114,000 |
| Pumps | 25,000 | 86,000 | 86,000 |
| Compressors | 90,000 | 920,000 | 590,000 |
| Centrifuge | - | 164,000 | 115,000 |
| Dryer | - | 15,000 | 15,000 |
| PHA Synthesis bioreactor* | - | 1,216,000 | 380,000 |
| PHA Extraction reactor | - | 113,500 | 80,000 |
| Auxiliary equipment | 30,000 | 94,000 | 40,000 |
| CHP system* | 4,600,000 | - | 3,200,000 |
| **PEC** | 259,000 | 1,506,500 | 1,040,807 |
| **TIC** | 5,661,900 | 7,392,650 | 7,844,000 |

*Costs including equipment and installation

202

203 Operational costs included water, steam, electricity and chemical reagents (Table 2).
204 Wastewater treatment costs were considered negligible given the low organic load of water
205 effluents in biogas upgrading and valorization units compared to those typically produced
206 during anaerobic digestion. Water and chemical reagent requirements were calculated
207 according to mass balances. Power requirements for pumps and compressors were estimated
208 with Equation 2 according to Estrada et al. [25].

209 $$P = (Q \cdot \Delta P) / \eta \quad (2)$$

Where P stands for the power requirements (kW), Q represents the fluid volumetric flow ($m^3 \cdot s^{-1}$), ΔP is the pressure drop (kPa) and η is the efficiency of pumps and compressors (70%). Biogas was considered as a waste stream of the anaerobic digestion process and therefore, its acquisition cost was set equal to zero. Average purchase and selling energy prices for industrial applications in Spain were selected (Table 2). 40% of the total electricity produced in scenario I was considered for the pretreatment, sorting and anaerobic digestion of the municipal solid waste (MSW) in the three scenarios (personal communication from industrial waste operators). The heat produced in the CHP units was only considered for internal provision of steam (0.08 kg steam · $kg^{-1}$ waste), as district heating implementation is not a common practice in medium size waste treatment facilities.

**Table 2**. Summary of costs for utilities and raw materials.

| Item | Cost | Unit |
|---|---|---|
| Biogas | 0 | €·$Nm^{-3}$ |
| Steam | 0.014 | €·$kg^{-1}$ |
| Water | 0.85 | €·$m^{-3}$ |
| Electricity selling price | 0.054 | €·$kWh^{-1}$ |
| Electricity purchase price | 0.095 | €·$kWh^{-1}$ |
| $NaNO_3$ | 0.30 | €·$kg^{-1}$ |
| NaOH | 0.29 | €·$kg^{-1}$ |
| Micronutrients | 0.10 | €·$kg^{-1}$ |
| Ethanol | 0.71 | €·$L^{-1}$ |

## 3. Results and discussion

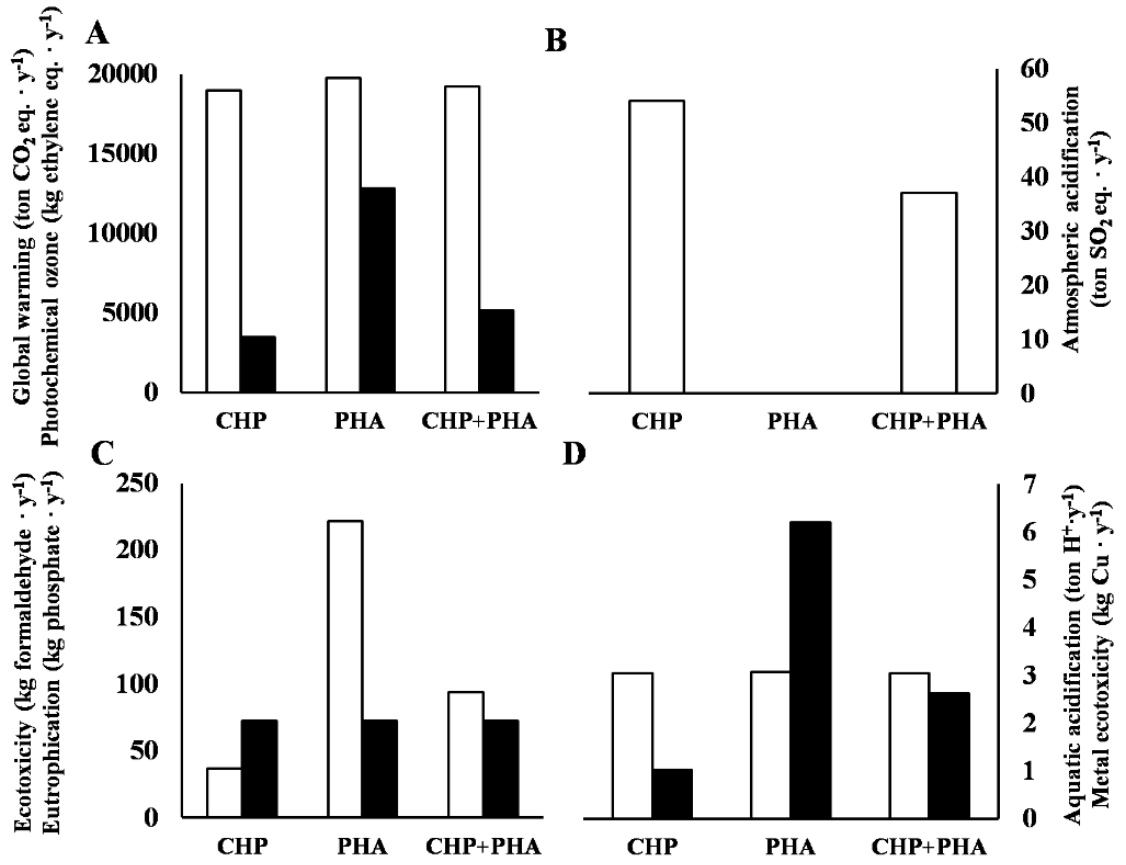
### 3.1. Environmental indicators

#### 3.1.1. Emissions, effluents and waste generation

226 The land, atmospheric and aquatic impacts of the selected biogas valorization scenarios were
227 evaluated according to the IChemE Sustainability Metrics. The impacts on land were limited to
228 the disposal of packing material used in the desulfurization biotrickling filter. Although inert
229 packing material is typically disposed of as non-hazardous waste, the addition of activated
230 carbon to improve biomass adherence turns the spent packing material into a potentially
231 hazardous waste (500 $kg \cdot y^{-1}$ in all scenarios). Notwithstanding, this amount of hazardous solid
232 waste disposal is negligible compared to the high treatment capacity of the model waste facility
233 (300 $t \cdot d^{-1}$) or the biodegradable polymer production (690.5 and 213.5 t $PHA \cdot y^{-1}$ in scenarios II
234 and III, respectively) which would potentially replace the same amount of oil-based plastics
235 largely sent to landfill (75.5% of the total plastic generated, according to EPA) [30].
236 Gas pollutants released during biogas valorization ($CH_4$, $CO_2$, $H_2S$ and VOCs) are major
237 contributors to global warming, ozone depletion and atmospheric acidification. The three
238 evaluated scenarios showed a comparable global warming potential, measured as $CO_2$
239 equivalents (Figure 2.A). The $CO_2$ produced during biogas combustion represented the main
240 contribution in scenario I, while non-biodegraded-$CH_4$ and $CO_2$ produced in $CH_4$
241 biodegradation equally contributed to the global warming burden in PHA production scenario
242 II. At this point, it must be stressed that the IChemE methodology does not take into
243 consideration the potential positive impacts of the evaluated technologies, such as $CH_4$
244 mitigation or waste nitrate depletion. In this regard, Rostkwoski et al. (2012) reported that the
245 use of $CH_4$ from biogas for PHA production entailed a negative global warming potential
246 ranging between -1.94 and -6.06 kg $CO_2$ equivalent$\cdot kg^{-1}$ PHA [9].
247 The highest photochemical-ozone potential burden was associated to the non-degraded $CH_4$
248 released to the atmosphere during the biological PHA production stage (12.8 t ethylene
249 equivalent$\cdot y^{-1}$) (Figure 2.A). Conversely, the PHA production scenario (II) showed negligible
250 atmospheric acidification and odor emission (Figure 2.B). This reduced acidification potential

was attributed to the transfer of VOCs and volatile sulfur compounds (VSCs) present in the desulfurized biogas to the liquid phase during methanotrophic cultivation. In contrast, the combustion of biogas in gas engines resulted in high amounts of $SO_2$ and $NO_2$ released to the atmosphere (54.1 t $SO_2 \cdot y^{-1}$ in scenario I).

Eutrophication, ecotoxicity to aquatic life and water acidification were identified as the main aquatic impacts and were associated to the partial consumption of nutrients and micronutrients supplied during the desulfurization and PHA production stages. Considering that nitrate is completely consumed by methanotrophic bacteria during PHA production, the major contributors to aquatic eutrophication were the nitrate and nitrite present in the desulfurization effluent (0.01 g $N\text{-}NO_3^- \cdot L^{-1}$ and 0.01 g $N\text{-}NO_2^- \cdot L^{-1}$) (Figure 2.C). However, this impact is marginal in comparison with typical digestate production and composition in waste treatment plants (1-5 g $N\text{-}NH_4^+ \cdot L^{-1}$). Moreover, if a secondary nitrogen loaded effluent (i.e nitrified digestate) is used as trickling liquid for electron acceptor supplementation, a positive effect on water eutrophication would be expected [19, 31]. PHA production showed the highest impact on aquatic life due to the presence of heavy metals and chloride in the micronutrient solution required for methanotrophic biomass growth (Figure 2.C and 2.D). Nevertheless, it must be highlighted that the mineral salt medium composition herein used was optimal for laboratory conditions, which in general includes excess metal concentration. In the prospective full-scale PHA production scenario, micronutrients supply would be reduced to minimize production costs, concomitantly reducing the associated impacts. Finally, the aquatic acidification burden estimates the amount of protons released to the aquatic medium. In our particular study, $H^+$ ions were released after the solubilization of $H_2S$ into the liquid medium during biogas desulfurization (Figure 2.D). However, as this process operated at neutral pH, water acidification was not significant.

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276 **Figure 2.** Environmental impacts evaluated according to the IChemE Sustainability Metrics.
277 Atmospheric impacts: (A) Global warming potential (white bars) and photochemical ozone
278 depletion potential (black bars), (B) Atmospheric acidification (white bars). Aquatic impacts:
279 (C) Ecotoxicity to aquatic life (white bars) and eutrophication potential (black bars), (D)
280 Aquatic acidification (white bars) and metal ecotoxicity (black bars).

281 ### 3.1.2. Resource usage

282 A comparative resource usage assessment including land, water, materials and energy was
283 performed for the three biogas valorization alternatives. The compact nature of gas engines and
284 heat exchangers resulted in a footprint 5× lower than that required for PHA production, where
285 high reactor volumes are required for an effective $CH_4$ gas-liquid mass transfer (Figure 3.A).

The area devoted to the cultivation of methanotrophic bacteria was 10× larger than that required for PHA extraction and purification. However, the relatively large area necessary for PHA production from biogas is rather marginal (0.03 $m^2 \cdot (Nm^3\ biogas \cdot d^{-1})$) compared to the extensive areas typically required for pre-processing of MSW (0.26-0.65 $m^2 \cdot (Nm^3\ biogas \cdot d^{-1})$), wastewater treatment (~0.1 $m^2 \cdot (Nm^3\ biogas \cdot d^{-1})$) or for farming and harvesting in agro-industrial facilities (>500 $m^2 \cdot (Nm^3\ biogas \cdot d^{-1})$) [32, 33, 34]. Consequently, the compactness of CHP units would only be advantageous in small-scale facilities. It should also be noted that the land required for PHA production, which is the technology showing the highest footprint, is unlikely to suffer significant depreciation since no recalcitrant chemical contamination of the soil is expected from these activities.

PHA biosynthesis and extraction also exhibited the highest water consumption rates due to the high amount of nutrients and NaOH required by the methanotrophic community during growth and by the extraction process (Figure 3.B). In scenarios II and III, 50-60% of the water consumption was employed in PHA biosynthesis, 30% in PHA extraction, and only 10% was devoted to biogas desulfurization. Internal water reuse strategies were already considered in the PHA production mass balances. Nonetheless, further optimization of the processes should be carried out in order to minimize the total water demand. In CHP, the water demand was associated to the addition of nutrients and NaOH during the biological desulfurization step (0.77 $m^3$ water·1000 $Nm^{-3}$ biogas treated regardless of the scenario). The substitution of synthetic mineral medium for *in-situ* available water streams such as digestate or treated wastewater would significantly minimize the water and chemical demand and boost the cost-efficiency of all biogas valorization alternatives evaluated in this paper. Accordingly, Lebrero et al. (2016) demonstrated that substituting a synthetic mineral medium by a $NO_3^-$ -supplemented liquid effluent from anaerobic digestion did not influence biogas anoxic desulfurization performance, achieving $H_2S$ removal efficiencies higher than 98% [19]. Similarly, Zeng et al. (2018)

311 investigated the use of aerated biogas slurry as recirculating liquid in anoxic biotrickling filters.
312 In this case, an aeration stage was necessary to support the nitrification of $NH_4^+$-rich effluents
313 (>5,000 $mg \cdot L^{-1}$ in some waste streams of MSW treatment plants) into $NO_3^-$ [31].
314 The material depletion indicator included the use of chemical reagents and packing material,
315 but excluded other materials such as spare parts, oils and lubricants necessary for pumps,
316 compressors, gas engines or heat exchangers maintenance. Hence, the only material depletion
317 considered in the CHP scenario was associated with nutrient supply ($NaNO_3$ and
318 micronutrients), pH control (NaOH) and packing material replacement in the desulfurization
319 step (39.3 $kg \cdot 1000\ Nm^{-3}$ biogas, Figure 3.C). PHA production showed the highest material
320 utilization (249.35 and 106.06 $kg \cdot Nm^{-3}$ biogas for scenarios II and III, respectively), the main
321 contributors being nitrate consumption and NaOH and EtOH requirements for extraction. PHA
322 production and purification stages exhibited a similar impact on material consumption (~ 40%
323 of the total material utilization).
324 The scenario exclusively devoted to PHA production showed the highest energy consumption
325 (3,526 kWh consumed·1000 $Nm^{-3}$ biogas treated, Figure 3.D). In this case, the high internal
326 gas recirculation rates and air supply requirements for complete $CH_4$ oxidation accounted for
327 74% of the total energy consumption. In contrast, CHP showed a positive balance on energy
328 production with 1,047·kWh produced·1000 $Nm^{-3}$ biogas treated (Figure 3.D). The hybrid
329 scenario was energetically self-sufficient when allocating 69% of the biogas flow rate to heat
330 and energy co-generation and the remaining 31% to PHA production. Interestingly, a standard
331 MSW treatment plant already allocates about 40% of the energy generated for internal use
332 (MSW pre-processing and anaerobic digestion and digestate treatment), while the remaining
333 60% of the biogas energy is sold, a practice that might not be economically sustainable in the
334 absence of fiscal incentives. Increasing the internal energy usage to satisfy also the energy
335 demand for PHA production, extraction and purification would reduce substantially the price

of the PHA being produced as well as boost the economic viability of anaerobic digestion plants by reducing its dependence from the electricity market.

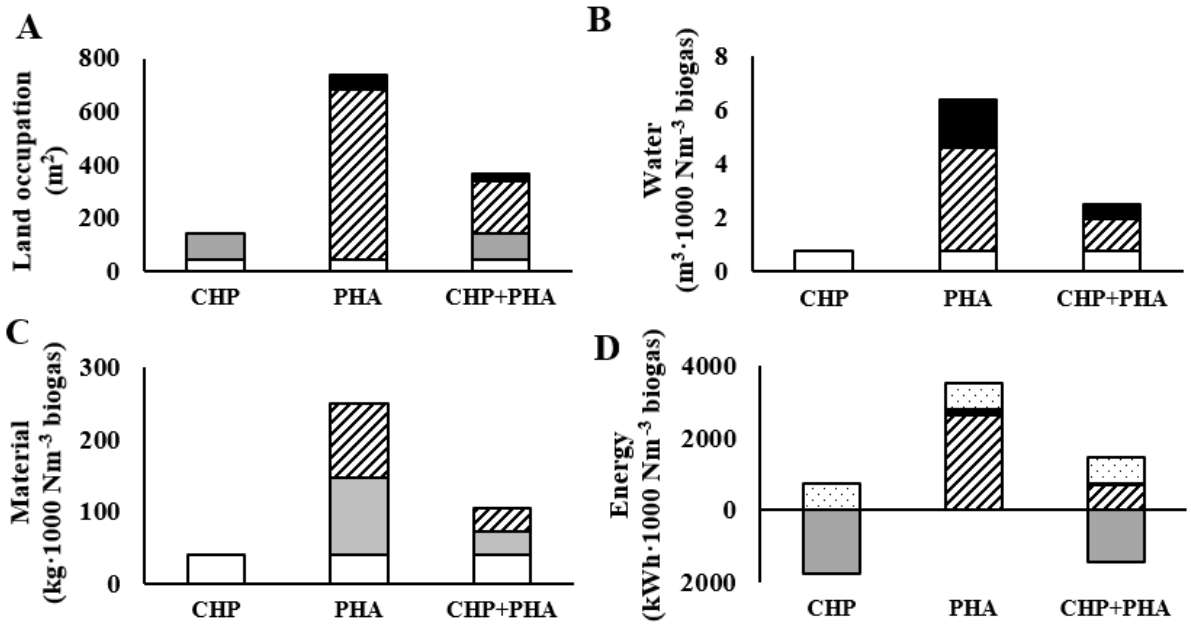


**Figure 3**. Environmental performance evaluated according to the IChemE Sustainability Metrics: (A) Land occupation, (B) Water demand, (C) Materials usage and (D) Net energy consumption (or production). The contribution of each stage to the global impact is represented as white (biogas desulfurization), light grey (CHP), striped (PHA production), black (PHA extraction and purification) and dotted (waste sorting and anaerobic digestion) bars.

### 3.2. Economic indicators

TIC values of 5.7, 7.4 and 7.8 M€ were estimated for scenarios I, II and III, respectively, with a TIC of 1,1 M€ corresponding to the anoxic biogas desulfurization unit (Table 1). For CHP, the heat and power cogeneration system represented the highest contribution to the total TIC (4.60 M€). The civil work and construction of the bubble column bioreactors accounted for only 16 and 6% of the TIC in scenarios II and III, respectively, assuming concrete as the construction material (190 €·m$^{-3}$ of reactor). The use of more expensive materials such as stainless steel would result in economically unsustainable prices of up to 2,500 €·m$^{-3}$. In this

352 regard, the use of concrete for the construction of high volume aerated bioreactors is a widely
353 used practice, i.e. activated sludge bioreactors in wastewater treatment plants. The air and
354 biogas compressors required for the continuous operation of the fermenter equipped with
355 internal gas recirculation were the most expensive equipment in PHA production, representing
356 65 and 57% of the total PEC in scenarios II and III, respectively.
357 Total operational costs of CHP were estimated at 367,417 €·$y^{-1}$ with 30% mainly attributed to
358 the use of raw materials, water and energy during the desulfurization step and the remaining
359 70% corresponding to the high operation and maintenance cost of gas engines. PHA production
360 showed the highest operational costs with 5,1 M €·$y^{-1}$, mainly due to the intensive biogas-air
361 compression, which accounted for 58% of the total operational costs. Additionally, the cost of
362 the raw materials was significant (15% of the total operational costs) due to the large quantities
363 of nitrate and NaOH required for biomass growth and PHA extraction, respectively (Fig.4).
364 CHP presented the lowest sales revenue with 1.1 M €·$y^{-1}$, including savings from steam and
365 electricity consumption and excess electricity sale at market price. Conventional CHP from
366 biogas exhibited a positive NPV of 765,730 €, an IRR of 6.7% and a payback period of 16
367 years. These results demonstrated that the investment in CHP systems in the absence of feed-
368 in tariffs is not highly profitable and it would be feasible only in medium and large biogas
369 production facilities with a high internal energy consumption. Sales revenues in scenarios II
370 and III were highly dependent on PHA selling price, currently ranging from 4 to 20 €·$kg^{-1}$
371 according to literature [8]. Considering the purity (92 %) and the waste-based production of the
372 biopolymer, the selling price of biogas-based PHAs is expected to be in the low end of the
373 selling range, which will make it suitable for low-cost applications. Then, PHA selling prices
374 were calculated for a NPV equal to that of the CHP scenario (765,730 €) which would turn
375 PHA production from biogas competitive against CHP. Prices were estimated at 8.8 and 4.6
376 €·$kg^{-1}$ in scenarios II and III, respectively, with corresponding sales revenues of 6.0 and 1.6 M

€·y$^{-1}$. IRR, which gives an insight of the profitability of the investment, were comparable to the CHP scenario (6.3% for scenarios II and III, respectively), with payback periods of 17 years.

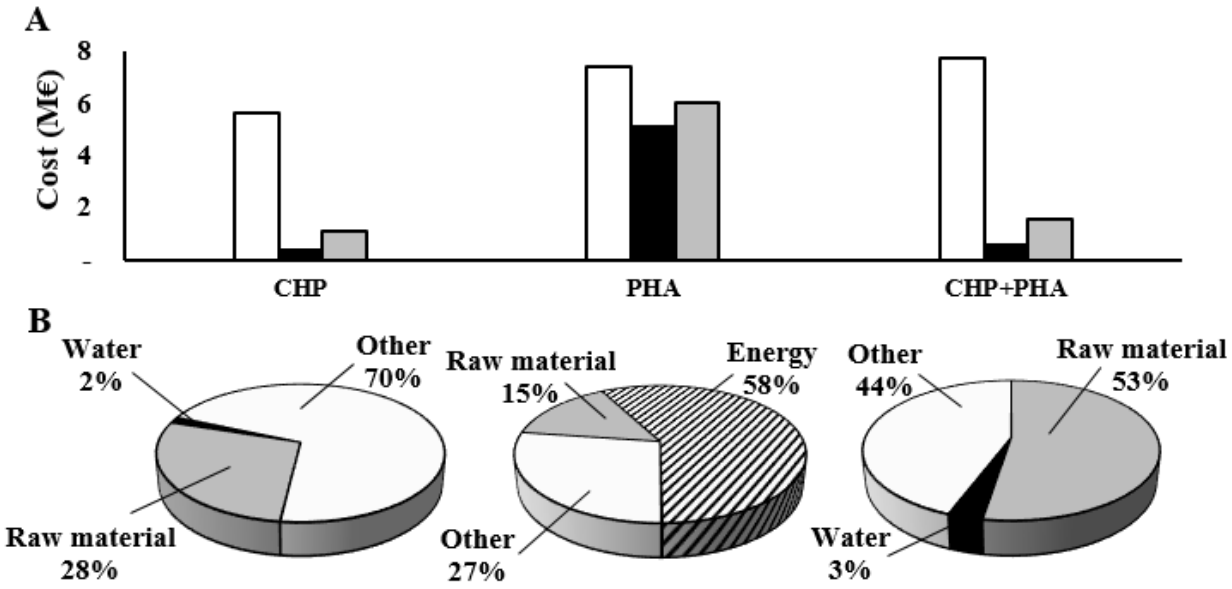


**Figure 4.** Economic performance of the biogas valorization scenarios. (A) Total cost share: capital investment cost (TIC) (white bars), operational cost (black bars), and sales revenue (light grey bars). (B) Operational cost share: raw material cost (light grey), energy cost (striped), water cost (black) and additional costs (white).

The break-even price of PHA for scenarios II and III was calculated as the price at which the NPV becomes positive (NPV>0) and corresponds to the PHA production costs. Interestingly, the hybrid alternative presented the lowest PHA production cost (4.2 €·kg$^{-1}$), while 8.6 €·kg$^{-1}$ was estimated for PHA production in scenario II, likely due to the increased energy costs. These values are in agreement with the current PHB market selling price (4.3 €·kg$^{-1}$ PHA) and with those reported by Levett et al. (2016) in a similar techno-economic study using pure $CH_4$ under pressurized and thermophilic conditions (3.9 €·kg$^{-1}$ PHA) [27, 35]. Likewise, the values are significantly lower than those first reported by Listewnik et al. (2007) using natural gas at a

392 similar production scale (13.6-16.5 €·kg$^{-1}$) and below the median price found in literature for
393 other kind of carbon substrates (6.8 €·kg-1 PHA) [27, 36].
394 These results showed that implementing PHA production in waste treatment plants constitutes
395 already a realistic alternative to biogas utilization as energy vector, showing no additional
396 financial risks. In this sense, PHA production from biogas could potentially provide a value
397 added product at a competitive market price, in spite of the early stages of development of the
398 technology and the low maturity of the biopolymer market.
399

400 **3.3. Social indicators**

401 IChemE Metrics social indicators aim at assessing attitude towards employees, suppliers,
402 contractors and customers, as well as impacts on society at a large scale [10]. When evaluating
403 theoretical scenarios, such as the ones in the current paper, it is difficult and of limited relevance
404 to hypothesize on internal employment conditions and the level of compliance of workers and
405 consumers. Therefore, social impact assessment was focused on the different social
406 acceptability of bioproducts (PHA) and bioenergy (CHP), and local community acceptance of
407 facilities dedicated to biopolymer or energy production.
408 Although biogas is positively regarded as a renewable energy source, its social acceptance
409 remains controversial. Local opposition towards industrial facilities that are considered
410 beneficial for society has been widely studied and is typically referred to as *Nimbyism* (*Not In*
411 *My Back-Yard*). Public opposition towards biogas production, and towards waste processing
412 facilities in general, is based on odor, noise and other nuisance [37, 38]. A recent study
413 demonstrated that the construction of several biogas facilities did not significantly affect
414 property value in neighboring communities. However, a slight negative effect was observed on
415 low quality houses constructed near plants dedicated to CHP [39]. A reduction in odor emission

416 can be expected in PHA producing facilities compared to CHP plants, given the minimized
417 emission of VOCs and VSCs.
418 Additionally, some studies have pointed out that social acceptance can be represented as a result
419 of a personal or social cost/benefit analysis [40]. Increasing social demand for bioproducts (80%
420 of European consumers is willing to buy products with minimal environmental impact) and
421 sustainable technologies, could potentially help minimizing *Nimbyism* by evoking more
422 positive emotions on consumers than biofuel production [41, 42]. In this sense, local
423 communities are more likely to be positively affected by benefits of the local production of
424 biopolymers (such as the development of a secondary biopolymer-based industry, improved job
425 opportunities, increased local tax revenues or indirect boost to local economic activities), than
426 from energy production [37, 38]. A recent report from the EU have estimated in 23,000 the
427 number of jobs related to bioplastic production in 2013, and forecasted an increase to 300,000
428 direct jobs by 2030 [42]. Additionally, there is a huge potential for creation of indirect jobs
429 associated to the new markets for these innovative biobased products, their future
430 commercialization and distribution within the circular bio-economy.
431

432 **4. Conclusions**

433 This study demonstrated that biogas valorization into PHA in waste treatment plants is a
434 competitive alternative to its current utilization for heat and power production (CHP). PHA
435 production entails a significant reduction of atmospheric acidification and odour emissions
436 compared to traditional CHP. Both processes showed similar impacts on global warming and
437 water eutrophication and acidification. In contrast, biopolymer production exhibited higher
438 ecotoxicity to aquatic life and ozone depletion potentials, and demanded more land, water,
439 energy and chemical reagents than CHP. On the other hand, higher investment and operational
440 costs are necessary for PHA production compared to CHP, although the higher market value of

PHA overcame this limitation and both processes supported similar NPV and IRR. PHA can be already produced from biogas at a competitive market price (8.6-8.8 €·kg$^{-1}$ PHA) in medium-size waste treatment plants, regardless of the economy of scale and the level of technology readiness. In this context, this study revealed that the optimal scenario for biogas valorization within a waste treatment plant involves the utilization of biogas-fueled CHP units for providing the power and heat necessary for PHA production, extraction and purification, which would result in PHA market prices of 4.2-4.6 €·kg$^{-1}$ PHA. Finally, the increasing public demand for bioproducts and the job creation associated to this new biopolymer industry could potentially enhance social and local acceptance of waste treatment facilities, traditionally facing *Nimbyism* issues.

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