Prospective LCA to provide environmental guidance for developing waste-to-PHA biorefineries

Saavedra del Oso, M.; Mauricio-Iglesias, M.; Hospido, A.; Steubing, B.R.P.

Citation

Saavedra del Oso, M., Mauricio-Iglesias, M., Hospido, A., & Steubing, B. R. P. (2023). Prospective LCA to provide environmental guidance for developing waste-to-PHA biorefineries. *Journal Of Cleaner Production*, *383*. doi:10.1016/j.jclepro.2022.135331

Journal of Cleaner Production 383 (2023) 135331

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Prospective LCA to provide environmental guidance for developing waste-to-PHA biorefineries

Mateo Saavedra del Oso [a,b,c,*], Miguel Mauricio-Iglesias [a], Almudena Hospido [a], Bernhard Steubing [b]

[a] Cross-disciplinary Research Center in Environmental Technologies (CRETUS), Department of Chemical Engineering, Universidade de Santiago de Compostela, 15782, Santiago de Compostela, Spain
[b] Institute of Environmental Sciences (CML), Leiden University, 2300, Leiden, the Netherlands
[c] Centre of Excellence for Packaging Sustainability (CEPS), Division of Packaging Materials, Stora Enso, 00180, Helsinki, Finland

ARTICLE INFO

Handling Editor: Kathleen Aviso

Keywords:
Polyhydroxyalkanoates
Mixed culture fermentation
Life cycle assessment
Environmental assessment
Bioprocess scale-up

ABSTRACT

Polyhydroxyalkanoates (PHA) production from waste streams using mixed microbial cultures (MMC) can unlock the potential of PHA to substitute oil-based plastics. However, these processes are still at low technology readiness level (4–6). Demonstrating a better environmental performance would boost their deployment at industrial scale. Hence, including environmental guidance during their development, when there are still opportunities for major alterations, is essential. To the best of our knowledge, this work elucidates for the first time how waste-to-PHA biorefineries could develop in the future by combining prospective LCA with scenario methodology and where the attention of stakeholders should be focused. Four future scenarios were derived considering both surrounding (e.g., scale, environmental or bioeconomy policies) and technological parameters (e.g., acidification yield, PHA content in biomass or recovery yield). Those scenarios derived under ambitious environmental and bioeconomy policies shop up to 50% lower environmental impacts than those under business-as-usual policies. These differences are caused by the different background processes' environmental burdens (e.g., electricity mix with low renewable energies share) and the higher consumption of chemicals and utilities. However, the environmental impacts caused by lower yields can be partially mitigated by valorizing the intermediate waste streams into biogas. Sensitivity analysis results pointed out recovery yield and PHA content as the parameters that influence most the environmental performance, being responsible for up to 60% of variance in environmental performance. These parameters determine the chemicals and utilities consumption in PHA downstream processing, which is confirmed as the main environmental hotspot. This work goes beyond previous LCA studies on PHA production and quantifies the influence of different parameters on the environmental performance.

1. Introduction

Bioeconomy is expected to play a significant role in the mitigation and adaptation to climate change across the European Union, targeting at reducing the pressure on biological resources as well as reducing CO_2 emissions and fossil fuel use in the chemical sectors according to its action plan launched by the European Commission in 2018 (Bell et al., 2018). In this endeavor, using biomass and especially organic wastes as feedstock to produce these chemicals and materials is a priority. For instance, polyhydroxyalkanoates (PHA), which are biodegradable polymers produced through microbial fermentation from diverse feedstocks (Khatami et al., 2021), could substitute petrochemical plastics in multiple applications (Dietrich et al., 2017). However, their high production cost and uncertain environmental performance hamper their industrialization (Tan et al., 2021). Demonstrating a better environmental performance than conventional plastics would foster PHA deployment (Yadav et al., 2020). High energy requirement during the feedstock cultivation, sterilization in pure culture fermentation and PHA downstream processing were reported as the main hotspots from both environmental and economic perspectives (Saavedra del Oso et al., 2021). Coupling PHA production with the carboxylate platform, where

---

[*] Corresponding author. Cross-disciplinary Research Center in Environmental Technologies (CRETUS), Department of Chemical Engineering, Universidade de Santiago de Compostela, 15782, Santiago de Compostela, Spain.

E-mail address: msaavedra.deloso@usc.es (M. Saavedra del Oso).

https://doi.org/10.1016/j.jclepro.2022.135331
Received 5 August 2022; Received in revised form 8 November 2022; Accepted 24 November 2022

Abbreviations and nomenclature

BAU	business-as-usual
CHP	cogeneration unit plant
COD	chemical oxygen demand
FDP	fossil depletion potential
FEP	freshwater eutrophication
FETPinf	freshwater ecotoxicity
FU	functional unit
GDP	gross domestic product
GMA	general morphological analysis
GWP100	climate change
HTPinf	human toxicity
LCA	life cycle assessment
LCI	life cycle inventory
MMC	mixed microbial culture
OFMSW	organic fraction of the municipal solid waste
PHA	polyhydroxyalkanoates
R&D	research and development
RES	Renewable energies share
SSP	shared socio-economic pathway
TAP	terrestrial acidification
TRL:	technology readiness level
VFA	volatile fatty acids
$Y_{PHA/VFA}$	accumulation yield
$Y_{VFA/S}$	acidification yield
$Y_{X/VFA}$	selection yield

organic waste streams are converted into volatile fatty acids (VFA) as PHA precursors, would significantly reduce the cost and could improve the environmental performance of PHA production (Atasoy et al., 2018).

H2020 project USABLE Packaging (H2020 USABLE Packaging, 2019) seeks the development of new value chains from food industry wastes by solving the bottlenecks in the PHA production. This project proposes cost-effective and sustainable production routes, such as the one based on the mixed microbial cultures (MMC) systems which comprises a 3-step system: (1) an anaerobic fermentation, where feedstock organic carbon is converted into VFA, (2) an enrichment of mixed culture, where PHA-storing bacteria are selected by imposing a feast/famine regime, and (3) an accumulation step, where enriched biomass is fed VFA until sufficient PHA has been accumulated (Nguyenhuynh et al., 2021). The performance of these systems at pilot scale has been evaluated for a diverse range of feedstocks, such as wastewater (Morgan-Sagastume et al., 2020), household waste (Moretto et al., 2020) or food industry side-streams (Silva et al., 2022). However, scaling up these processes to a commercial full-scale PHA production still faces multiple challenges, from low substrate conversion and poor mechanical properties to deploying a cost-effective and sustainable extraction strategy at pilot scale (Estévez-Alonso et al., 2021).

Demonstrating a better environmental performance than petrochemical plastics would promote PHA development and market expansion. Previous LCA studies on the production of PHA by MMC assessed the environmental benefits of integrating the PHA production within urban (Morgan-Sagastume et al., 2016) and industrial wastewater treatment plants (Fernández-Dacosta et al., 2015; Roibás-Rozas et al., 2020). Fernández-Dacosta et al. (2015) evaluated the environmental performance of different PHA recovery technologies from wastewater. However, none of these articles addressed the inherent uncertainties of upscaling processes at lab- and pilot-scale or quantified the influence of process parameters on the environmental performance (Igos et al., 2019). Prospective life cycle assessment (LCA) can play a key role in the development and optimization of MMC systems by providing environmental guidance (Arvidsson et al., 2018). Prospective LCA facilitates the upscaling of emerging processes employing scenarios of future performance at industrial scale, and the comparison of the future process with the incumbent processes (Cucurachi et al., 2018). However, its prospective character entails, to some degree, lack of data and considerable uncertainty (Igos et al., 2019). For instance, the market share of these biopolymers and the feedstock availability will determine the production scale, or the environmental policies will influence how electricity is produced (Saavedra del Oso et al., 2021). Yet, both future market share and environmental policies entail a high uncertainty. A feasible approach to evaluate the environmental performance in a future framework is to propose scenarios projecting how emerging processes will develop in the future (Arvidsson et al., 2018; Bergerson et al., 2020). Contributions on how to combine scenario methodology and prospective LCA have been published (Delpierre et al., 2021; Thomassen et al., 2019) but did not cover biobased emerging technologies, which face different challenges regarding the choice of functional unit, allocation approaches, or process upscaling. Only recently, Langkau et al. (Unpublished work) have proposed a scenario methodology framework on biobased products, which enables the systematic and documented development of scenarios, rather than the implicit approach that is often behind prospective LCA studies. Analyzing the LCA results of the proposed scenarios allows inferring the relationship between the characteristics of the scenarios (e.g., type of substrate, yield or renewable energies share) and the environmental performance. Thus, it is possible to identify the most relevant parameters to optimize during the technology development, enabling effective environmental guidance.

The objective of this work is to elucidate how waste-to-PHA biorefineries could develop in the future and ensure that environmental guidance is included in their development. To do so, influencing parameters were firstly identified based on literature review as well as both individual meetings and a workshop with stakeholders. Based on the information collected, scenarios were then developed and validated with stakeholders. Finally, their environmental impacts were quantified and analyzed, allowing the identification and quantification of the parameters influence on the environmental performance.

2. Methodology

In this section, prospective LCA (section 2.1) and scenario methodology (section 2.2) are described.

2.1. Prospective LCA

Prospective LCA is a systematic methodology which determines the environmental impacts of an emerging/incumbent product/process in a future framework where the production system is modelled (Cucurachi et al., 2018). Like conventional LCA, prospective LCA is comprised by four steps (see Fig. A.1): (i) the goal and scope state of the system function, functional unit, system boundaries, technology readiness level (TRL), temporal boundaries, foreground and background data source, identification of alternative systems, impact assessment method and impact categories; (ii) the inventory analysis involves the data collection and the development of scenarios and its implementation as inputs and outputs of a product system; (iii) the impact assessment transforms the inventory results into potential environmental impacts; and (iv) the interpretation phase, which involves a critical review, results presentation and determination of data uncertainty and sensitivity (Cucurachi et al., 2022).

2.2. Scenario methodology

Scenario development is a systematic and documented methodology (Thomassen et al., 2019) that involves both the goal & scope definition and inventory analysis phases. The key choices for scenario development are made during the goal & definition, i.e., the time horizon, the current and expected TRL and the scenario approach (i.e. whether a predictive, explorative, or normative approach is followed; it aims to answer "how will/could/should the future develop" respectively (Langkau et al., Unpublished work)). During the inventory analysis, a 4-step procedure is followed to develop the scenarios: (i) identification of influencing parameters, (ii) construction of sub-scenarios for each parameter, (iii) creation of scenarios from sub-scenarios and (iv) implementation of scenarios in the life cycle inventory (LCI) model.

2.2.1. Identification of influencing parameters

Scopus web search engine was employed to screen scientific papers on waste-to-PHA biorefineries. To do so, the following query string was used:

TITLE-ABS-KEY ((pha OR polyhydroxyalkanoates OR polyhydroxybutyrate OR phb OR phbv OR p3hb OR poly3-hydroxybutyrate) AND ((mixed AND microbial AND cultures) OR (mixed AND culture) OR (open AND mixed AND culture) OR (mmc)) AND (pilot-scale OR scale-up OR (large AND scale))).

Among the 77 resulting elements, only studies on PHA production by MMC at pilot scale were selected (i.e., the 11 peer-review studies listed in appendix A). The insights and data gathered in this review were used to develop the scenarios, both identifying the influencing parameters and upscaling the processes.

2.2.2. Individual meetings with stakeholders

Individual meetings with stakeholders (feedstock providers, technology developers, packaging producers and end users) were carried out after the literature review to support the scenario development. The procedure was the following: (i) presentation of the PHA production flowchart, (ii) identification and validation of the process influencing parameters, (iii) identification and validation of the parameters influencing the surroundings, (iv) creation and validation of sub-scenarios for each parameter. The stakeholders feedback enabled the influencing parameters and sub-scenarios definition.

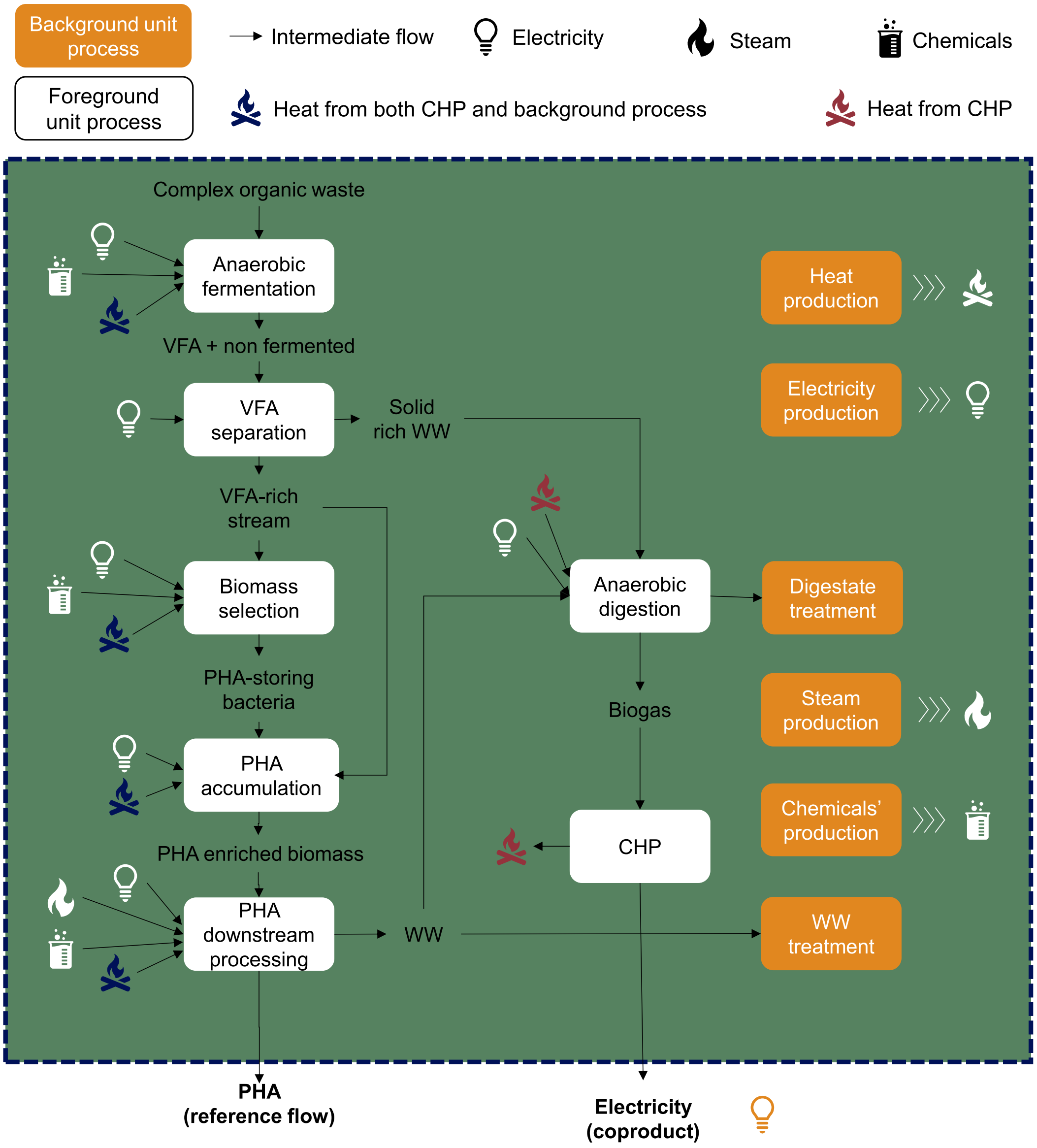

Fig. 1. Waste-to-PHA biorefinery system boundaries.

2.2.3. *Workshop with stakeholders*

An on-line workshop with the previously identified stakeholders was organized to gather their individual feedback and to jointly create, develop, and validate the scenarios derived. The workshop was structured in the following steps: (i) presentation of the prospective LCA and scenario methodology, (ii) overview of the waste-to-PHA biorefinery system, (iii) validation of the influencing parameters, (iv) validation of parameters' sub-scenarios, (iv) creation and validation of scenarios from sub-scenarios, (v) discussion and final remarks.

3. Scenario development

This section depicts the goal and scope definition (section 3.1) and the inventory analysis (section 3.2).

3.1. *Goal and scope definition*

The main goal of this prospective LCA is to quantify the environmental impacts of the waste-to-PHA biorefinery in a future context when its large-scale production is already implemented. As the system function is to produce PHA from complex organic wastes, the functional unit (FU) was defined as 1 kg of PHA powder (a common approach in previous LCA of PHA (Roibás-Rozas et al., 2022)). A cradle-to-gate approach covering all the unit processes within the waste-to-PHA biorefinery is defined in Fig. 1; including within the system boundaries the PHA production (i.e. anaerobic fermentation, the VFA separation, the PHA enrichment and accumulation, as well as the downstream processing) and the further energy valorization of the residual intermediate streams by anaerobic digestion (AD) and cogeneration unit plant (CHP); and excluding then the gate-to-grave phases (i.e., compounding and shaping, use and end-of-life).

The multifunctionality of the system was addressed by allocating the environmental burdens from VFA separation and from CHP to PHA and electricity production, respectively. This choice was made following the guidelines on the LCA of alternative feedstock for plastics production (Nessi et al., 2021). Given the actual TRL, i.e., from 5 to 6, the temporal boundaries were established for 2030, when market level maturity is expected to be reached (Lorini et al., 2022).

An explorative scenario approach (Börjeson et al., 2006) was followed with the aim of envisioning how the PHA production by MMC could develop. The foreground system was modelled based on the data collected from the literature review and using an upscaling framework for emerging technologies (Tsoy et al., 2020). For the background system, a futurized version of the ecoinvent 3.7.1 database was used that includes scenario data derived from the IMAGE integrated assessment model (Stehfest et al., 2014). The latter model's global future scenarios are based on the Shared Socio-Economic Pathway (SSP) scenarios (O'Neill et al., 2014) and representative concentration pathways (van Vuuren et al., 2011). Two scenarios, the SSP2-base and the SSP2-RCP2.6 were derived here via the PREMISE framework (Sacchi et al., 2022). Both scenarios represent the SSP "middle of the road", although they differ substantially in terms of climate change mitigation. In the SSP-base scenario a warming of 3.5 °C is modelled by 2100, while in the SSP2-RCP2.6 the temperature increase is limited to just below 2 °C (Sacchi et al., 2022). The foreground scenarios were modelled together with the background scenarios using the superstructure approach (Steubing and de Koning, 2021), as implemented in the open source LCA software Activity Browser (Steubing et al., 2020).

The analysis of the environmental impacts followed a midpoint approach, being climate change (GWP100), terrestrial acidification (TAP), freshwater eutrophication (FEP), human toxicity (HTPinf), freshwater ecotoxicity (FETPinf) and fossil depletion (FDP), for the selected impact categories, according to previous LCA on PHA production (Roibás-Rozas et al., 2022). All impact categories were assessed using the Hierarchist ReCiPe (H) v1.13.

3.2. *Inventory analysis*

The core part of the applied methodology concerns the scenario construction during the inventory analysis phase (Langkau et al., Unpublished work). Scenarios represent both descriptions of possible future states and descriptions of developments (Börjeson et al., 2006); affecting both foreground and background data of the LCI. The main results of the 4-step procedure are presented in the next subsections (3.2.1, 3.2.2, 3.2.3 and 3.2.4).

3.2.1. *Identification of influencing parameters*

The identified influencing parameters are listed in Table B.1. A 60% of them are surrounding parameters, which are related to political actions, societal concerns, technological and environmental aspects, but are not intrinsically present in the LCI model. The rest of identified parameters are technological parameters (foreground and background), i.e., those that affect directly the LCI model flows. Foreground parameters affect the mass and energy balances, while background parameters modify the background processes.

The hybrid causal loop flowchart diagram shown in Fig. 2 depicts the influences and correlations between the identified influencing parameters and the LCI model. This diagram helps the analyst to consider surrounding parameters (left side), but also to understand how the technological parameters can be modelled within the LCI model (right side), and contains both quantitative/qualitative parameters, unit processes and intermediate/elementary flows for the LCI model.

The process performance parameters (i.e., the acidification yield, productivity, PHA-storing biomass selection and PHA accumulation yield, PHA content in enriched biomass and extraction yield) depend on the R&D on these processes, but also on the feedstock employed (Saavedra del Oso et al., 2022). The choice of wastes as feedstock, e.g., organic fraction of the municipal solid waste (OFMSW) or sewage sludge, may hinder the applicability of the polymer for high purity applications such as food contact materials.

The implementation of ambitious bioeconomy policies (Fritsche et al., 2020), fostered by higher environmental awareness and the impact of climate change, may increase the demand of PHA as well as the use of waste or industrial side-streams as feedstocks. Ambitious bioeconomy policies would increase not only the production scale but the R&D funding, whose outcomes can contribute to the reduction of the production costs. A positive feedback loop between the production scale and costs may be possible, i.e., by decreasing the production costs, the production scale increases, which in turn decreases them. The production of PHA by MMC from waste streams can benefit from the adoption of new environmental policies. The implementation of these legislations may increase the RES and introduce new emissions regulations, modifying then the background processes such as electricity production or chemicals production.

This causality diagram allows not only to develop the model by identifying correlations between parameters and deal with their complexity, but also to communicate to stakeholders the influence of parameters. Thus, this tool adds transparency to the study. As the number of identified influencing parameters is high and the effects of some surrounding parameters are either intrinsically present in others or cannot be easily implemented in the LCI model, only environmental policies, bioeconomy policies and feedstocks were chosen, among those for the construction of sub-scenarios.

3.2.2. *Construction of sub-scenarios for each parameter*

For all the selected surrounding parameters (bioeconomy policies, environmental policies, feedstock, production scale) a set of three levels or sub-scenarios are defined, except for the feedstock type where only two options were fixed. Business-as-usual (BAU), moderately ambitious and ambitious were defined as the three sub-scenarios for both environmental and bioeconomy policies. Related the former, since environmental policies determine both background parameters, i.e., RES and

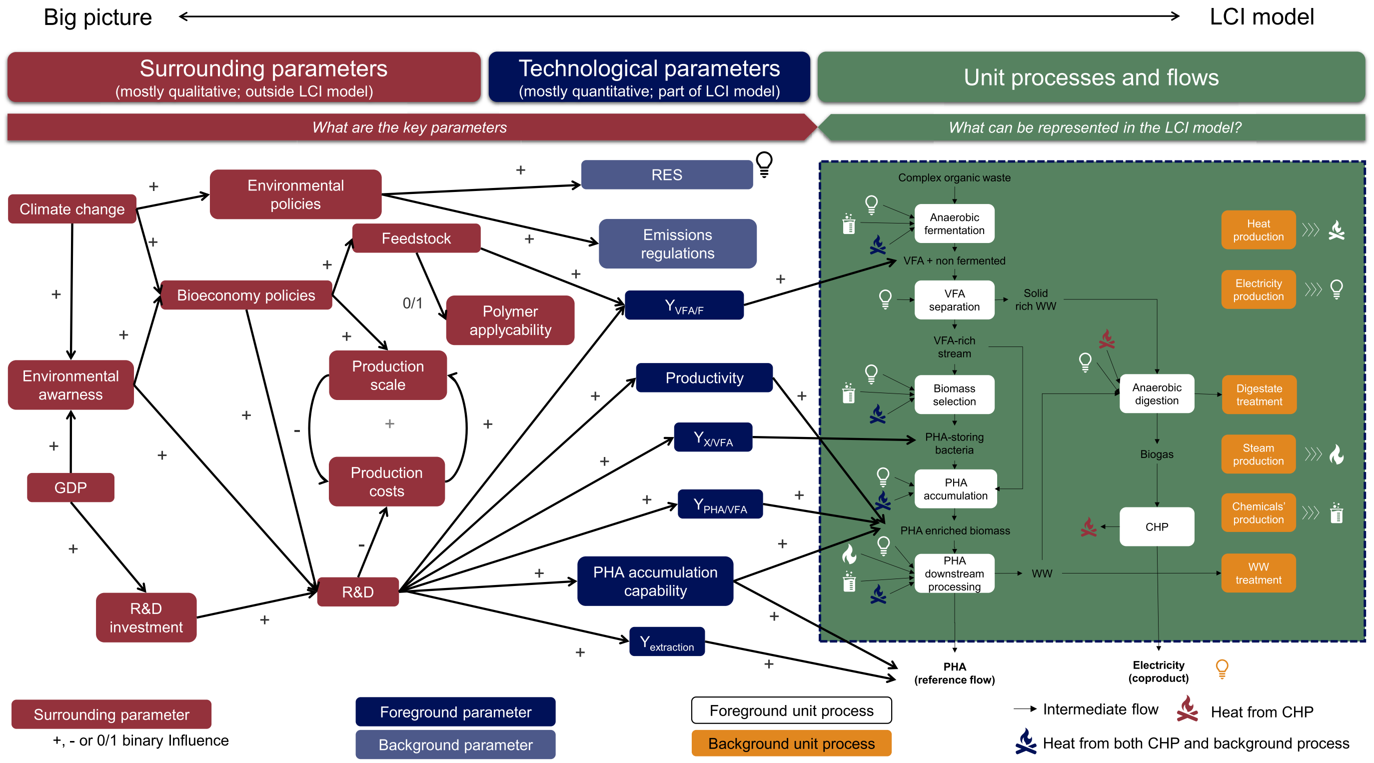

Fig. 2. Hybrid causal loop flowchart diagram depicting the interrelations between surrounding and technological parameters and the LCI model.

emissions regulations, these three sub-scenarios were implemented in the LCI model according to the RES and the SSP (O'Neill et al., 2014): BAU (current RES and SSP2-base), moderately ambitious (50% RES and SSP2-RCP2.6) and ambitious (60% RES and SSP2-RCP2.6). Concerning the later, BAU at bioeconomy policies means focus mainly on waste treatment and resource recovery, but the market share of these biobased products is still small. Moderately ambitious and ambitious bioeconomy policies consider waste as resources that can be transformed widely into added-value products with a big market share. These sub-scenarios are qualitative and are not directly implemented in the LCI model.

Regarding the production scale, the sub-scenarios were proposed according to waste availability and technological developers' knowledge: 500, 2000 and 7500 t PHA/y. The sub-scenarios for the feedstock and process performance parameters were chosen according to the literature review on PHA production by MMC at pilot scale. Only two sub-scenarios were created for the feedstock, i.e., fruit waste and the mixture of OFMSW and sewage sludge, since they are widely available (Yadav et al., 2020) and were employed in most of the waste-to-PHA pilot scale studies (Moretto et al., 2020; Silva et al., 2022).

The sub-scenarios for the selected technological parameters are listed in Table 1. The sub-scenarios for the acidification yield, selection yield, accumulation yield, productivity and PHA content in biomass were created based on the literature review and feedback provided by the stakeholders in the individual meetings and the workshop. The recovery yield values were selected according to previous authors' work on the optimization of the environmental performance of the PHA downstream processing (Saavedra del Oso et al., 2021).

3.2.3. Creation of scenarios from sub-scenarios

Since the total number of combinations of sub-scenarios is very high, creating scenarios from those sub-scenarios is not a straightforward task. A cross-consistency assessment was performed to discard combinations of sub-scenarios that were not consistent (Appendix B.2). Regarding the environmental policies, it is inconsistent to have an ambitious bioeconomy policy or medium and large-scale production under the SSP2-base sub-scenario, as oil-based chemicals and materials would still have a higher market share compared to biobased alternatives. Likewise, the PHA production from OFMSW & sewage sludge seems unfeasible at industrial scale under BAU bioeconomy policies, as conventional waste treatments such as energy valorization would be preferred. However, the use of concentrated, carbon-rich, easily fermentable, and localized feedstocks such as side-streams of food processing facilities could be feasible, e.g., fruit waste from juice beverage industries.

Table 1
Sub-scenarios created for selected technological parameters.

Parameter	Sub-scenario 1	Sub-scenario 2	Sub-scenario 3	Units
$Y_{VFA/F}$	0.35	0.55	0.75	g COD/g COD
$Y_{X/VFA}$	0.40	0.45	0.55	g COD/g COD
$Y_{PHA/VFA}$	0.50	0.65	0.80	g COD/g COD
Productivity	3.00	6.00	8.00	g PHA/(L·d)
PHA content in biomass	0.46	0.59	0.80	g/g
Recovery yield	0.65	0.75	0.85	g/g

As showed in Fig. 2, the implementation of ambitious bioeconomy policies would lead to a higher investment in the R&D of these technologies and thus, to higher values of process performance parameters. Nevertheless, these parameters are influenced by the type of feedstock too. For instance, a value of 0.75 g COD-VFA/g COD-F for the anaerobic fermentation yield of OFMSW & sewage sludge seems inconsistent, as these substrates are not easily hydrolyzed and fermented into VFA. The cross-consistency check also allows disregarding combinations that would lead to technological inconsistencies, such as low PHA-storing biomass selection yield together with high PHA accumulation yield.

As the remaining number of consistent scenarios is still high, general morphological analysis (GMA) was applied (see Table B.2). GMA is a method for structuring and investigating the total set of relationships contained in multi-dimensional, nonquantifiable problem complexes [51]. Different narratives for feasible scenarios were derived from the GMA. The eight narratives derived from the GMA, which are summarized in Appendix B.2, were presented, and discussed at the stakeholders' workshop.

As a result of the discussion, four of them (i.e., scenarios A, D, E and H) were chosen to be implemented in the LCI, as they can be considered as the cornerstone scenarios and thus, it may allow assessing scenarios at the edges of the solution space. From now, scenarios A, D, E and H will be referred as scenarios 1, 2, 3 and 4; and Table 2 lists their specific parameters sub-scenarios.

3.2.4. *Implementation of scenarios in the LCI model*

The foreground system, i.e. the waste-to-PHA biorefinery, was upscaled using detailed process calculations (Tsoy et al., 2020). The process calculations, which follow a scale-up framework for chemical processes in LCA studies (Piccinno et al., 2016), are detailed in the Appendix C.

Background process "market for electricity, high voltage (RER)" was chosen for scenario 1 from the ecoinvent 3.7.1 IMAGE database, which is the database employed for the rest of background processes. Electricity production was modelled for scenarios 2, 3 and 4 according to the project report Roadmap 2050 (included in Appendix D). Regarding the lack of specific background data, the following considerations were made: (i) the production process for sodium dodecyl sulphate (SDS), used in downstream processing, was assimilated to another chemical with similar function: non-ionic surfactant; (ii) the wastewater stream produced in the downstream processing was assumed as urban wastewater, (iii) potassium phosphate was assumed as sodium phosphate and (iii) the digestate treatment was assumed as treatment of raw sewage sludge by municipal incineration with fly ash extraction. Finally, the following assumptions related to process system (Fig. 1) were formulated:

- The electricity produced by the AD is not locally consumed in the process and, instead, it is exported to the net. Thus, all the electricity consumed in the system comes from the European Union electricity mix.
- The heat produced by the AD is internally consumed for heating the digestor, and should heat surplus be produced, it is assumed to be used by the PHA production section. All the environmental burdens of AD have been allocated to electricity production, which is the function of the waste valorization section.
- The residual gas stream produced in the anaerobic fermentation, with negligible flowrate and composed by (biogenic) carbon dioxide with a small fraction of hydrogen, is assumed to be released to the atmosphere.

Inputs of raw material and energy as well as emissions to air, water, and wastewater per kg of obtained PHA powder (i.e., defined functional unit) are summarized in Table 3.

4. Results

This section presents the results of scenarios life cycle impact assessment and analyzes the parameters influence on the environmental performance (sections 4.1 and 4.2). Furthermore, the allocation methods are compared in Appendix E.2.

4.1. *Environmental evaluation*

Results from the characterization stage for PHA production, where are displayed on Fig. 3.

Heat production is the main contributor to GWP100 (52 and 51%) and FDP (62 and 58%) in scenarios 1 and 2. However, its contribution is negligible to all impacts categories in scenarios 3 and 4. This discrepancy is caused by the heat employed in these scenarios, which comes from the CHP (scenario 3, 100%; scenario 4, 90%), and has no environmental burdens as explained earlier. Thus, the heat source determines the related environmental impact, as shown in Fig. 3.

Electricity contributes significantly to GWP100, HTPinf and FDP categories, especially in scenario 3. Regarding the electricity consumption, the downstream processing, the PHA accumulation and the biomass selection are pointed out as the unit processes that consume the most electricity (see Fig. E.1 and E.2). PHA downstream processing is caused by the high-pressure homogenization employed for disrupting the cells and the aeration systems used in both biomass selection and PHA accumulation. Significant differences were found between scenarios, as the values for extraction, selection and accumulation yield were different. For instance, extreme electricity consumption values in PHA downstream processing are 0.620 and 1.608 kWh/FU for scenarios 2 and 3 respectively (see Table 3), being the correspondent recovery yield 0.85 and 0.65 g/g respectively, and the biomass PHA content 0.80 and 0.46 g/g.

Electricity consumption values in biomass selection and PHA accumulation for scenarios 2 and 3 are 0.140 and 1.209 and 0.414 and 1.260 kWh/FU respectively (see Table 3). These differences are explained by the differences in biomass selection (0.55 vs 0.4 g COD/g COD) and PHA accumulation yield (0.8 vs 0.5 g COD/g COD) as well as productivity (8 vs 3 g PHA/(L·d)), which affect the reactors dimensions and the required aeration. However, background parameters such as the RES must be considered, as they have a significant influence on the environmental

Table 2
Description of selected parameter sub-scenarios for the created scenarios.

Parameter	Scenario 1	Scenario 2	Scenario 3	Scenario 4	Units
Environmental policy	SSP2-Base Current RES	SSP2-RCP2.6 60% RES	SSP2-RCP2.6 50% RES	SSP2-RCP2.6 60% RES	
Bioeconomy policy	BAU	Ambitious	Ambitious	Ambitious	
Production scale	500	7500	7500	7500	t PHA/y
Feedstock	Fruit waste	Fruit waste	OFMSW & SS	OFMSW & SS	
$Y_{VFA/F}$	0.55	0.75	0.35	0.55	g COD/g COD
$Y_{X/VFA}$	0.45	0.55	0.40	0.45	g COD/g COD
$Y_{PHA/VFA}$	0.65	0.80	0.50	0.65	g COD/g COD
Productivity	6.00	8.00	3.00	6.00	g PHA/(L·d)
PHA content in biomass	0.59	0.80	0.46	0.59	g/g
Recovery yield	0.75	0.85	0.65	0.75	g/g

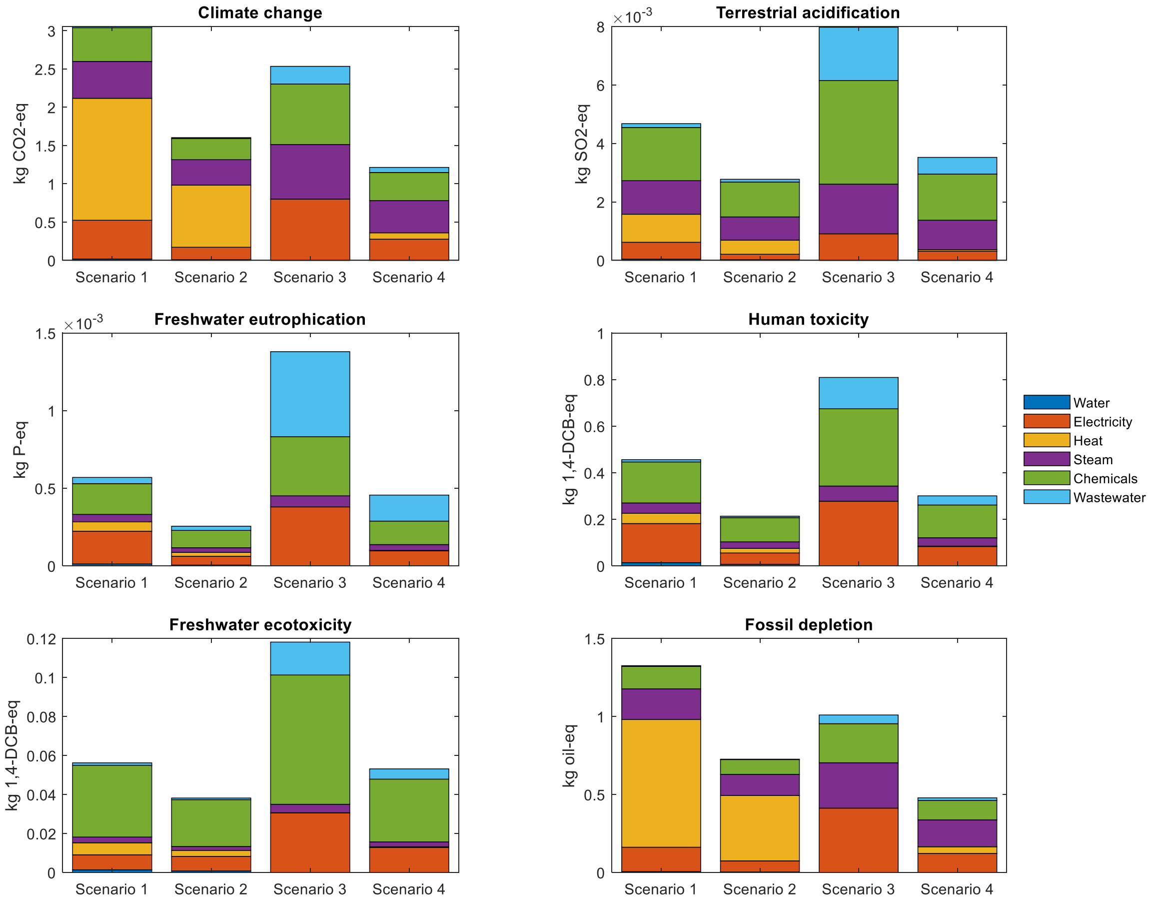

Fig. 3. Characterization of PHA production within a waste-to-PHA biorefinery and contributions of background processes (FU: 1 kg of PHA).

impacts too. The GWP100 environmental burdens for the current, 50% and 60% RES are 0.268, 0.154 and 0.126 kg CO_2-eq/kWh.

The steam production is responsible for up to 25% environmental impact in GWP100, FEP and FDP. The steam is consumed in the last step of the PHA downstream processing, where the PHA cake is spray dried into powder. Regarding the consumption, scenario 1 employs a 44% more steam than scenario 2, while scenario 3 doubles scenario 1 and is 70% higher than scenario 4. These notable differences can be explained by the different PHA content and the recovery yield (see Tables 2 and 3).

Chemicals contribute considerably to TAP, FEP, HTPinf and FETPinf. SDS, which is employed to disrupt the biomass in the downstream processing, has the highest contribution within chemicals (up to a 60% chemicals' environmental burdens). Its consumption, which is related with the PHA content in the biomass, is substantially higher in scenario 1 than scenario 2, although scenario 3 has the highest. The other chemicals are used as additives in the biomass selection and their rates per FU depend essentially on the selection yield, being scenario 3 the one with the highest consumption (see Tables 2 and 3).

Table E.1 summarizes the technological parameters influence on the background processes' environmental burdens. With regards to the background parameters, the RES is determinant for the electricity production environmental burdens as previously stated. However, the environmental burdens reduction caused by using the SSP2-RCP2.6 background scenario instead SSP2-Base for 2030 is almost negligible (less than a 5% in all impact categories), as these scenarios consider a slow progress in achieving the sustainable development goals until 2050. Regarding the foreground parameters, recovery yield and PHA content in biomass are the parameters that influence most the environmental burdens of electricity, steam, and chemicals.

4.2. Uncertainty and sensitivity analysis

Applying the scenario methodology allows the characterization of model structure and context uncertainty (Igos et al., 2019), i.e., accounting the uncertainty in the representativeness of the reality of the LCA model and the normative choices of LCA analysts respectively. Uncertainty analysis, shown in.

Fig. 4, was performed to evaluate both epistemic (i.e. lack of knowledge) and ontic (i.e. deterministic and randomness) uncertainty related to the process performance parameters, considering a 10% variation for the lower and upper limits. The pressure drop in the high-pressure homogenizer was included in the analysis, as there is a significant uncertainty related to this parameter (variance interval is 500–1500 kPa) and this step is one of the hotspots regarding the electricity. Uncertainty analysis was carried out through a Monte Carlo analysis (1000 iterations). Then, a global sensitivity analysis (GSA) (Cucurachi et al., 2022) was performed to evaluate the influence of each parameter on the results uncertainty. The GSA, shown in Fig. 5, was carried out following the standardized regression coefficients method.

Significant differences were found among scenarios and categories when analyzing the uncertainty and GSA results together (Figs. 4 and 5). However, recovery yield is the parameter responsible for at least 50%

Table 3
LCI per kg of PHA powder for scenarios 1, 2, 3 and 4.

Items	Scenario 1	Scenario 2	Scenario 3	Scenario 4	Ecoinvent process
Anaerobic fermentation					
Intermediate products					
VFA + non fermented (kg COD)	14.05	5.81	39.95	12.71	
Technosphere inputs					
Feedstock (kg COD)	14.41	5.97	39.97	12.72	
Water (m^3)	0.07	0.01	0.02	0.01	market for tap water (Europe without Switzerland)
Electricity (kWh)	0.11	0.04	0.40	0.13	market for electricity, high voltage[a] (RER[b])
Heat (MJ)	30.35	12.70	0.00	1.27	heat production, natural gas, at industrial furnace >100 kW (Europe without Switzerland)
Heat (MJ)	0.45	0.00	42.79	12.42	
Sodium hydroxide (kg)	0.10	0.06	0.17	0.09	chlor-alkali electrolysis, membrane cell (RER)
VFA separation					
Intermediate products					
VFA rich stream (kg COD)	6.13	3.46	10.8	5.41	
Solid-rich wastewater stream (kg COD)	7.92	2.35	29.13	7.30	
Technosphere inputs					
VFA + non fermented (kg COD)	14.05	5.81	39.95	12.71	
Electricity (kWh)	0.17	0.07	0.72	0.23	
Biomass selection					
Intermediate products					
Biomass (kg)[c]	1.13	0.57	2.04	0.99	
Technosphere inputs					
VFA rich stream (kg COD)	2.90	1.00	6.37	2.56	
Electricity (kWh)	0.36	0.14	1.21	0.39	market for electricity, high voltage (RER)
Heat (MJ)	1.74	3.81	0.00	4.00	heat production, natural gas, at industrial furnace >100 kW (Europe without Switzerland)
Heat (MJ)	0.03	0.00	7.81	0.41	
Ammonium chloride (kg)	0.04	0.02	0.08	0.03	market for ammonium chloride (GLO[d])
Potassium phosphate (kg)	0.03	0.02	0.07	0.03	market for sodium phosphate (RER)
Calcium chloride (kg)	0.02	0.01	0.04	0.01	market for calcium chloride (RER)
Emissions to air					
Biogenic CO_2 (kg)	1.07	0.37	2.35	0.95	
PHA accumulation					
Products					
PHA enriched biomass (kg)	2.26	1.57	3.34	1.99	
Technosphere inputs					
VFA rich stream (kg COD)	3.23	2.46	4.46	2.85	
Electricity (kWh)	0.49	0.41	1.26	0.61	market for electricity, high voltage (RER)
Heat (MJ)	0.06	0.00	0.00	0.00	heat production, natural gas, at industrial furnace >100 kW (Europe without Switzerland)
Heat (MJ)	0.00	0.00	0.04	0.02	
Emissions to air					
Biogenic CO_2 (kg)	1.19	0.91	2.55	1.05	
PHA downstream processing					
Products					
PHA (kg)	1.00	1.00	1.00	1.00	
Technosphere inputs					
PHA enriched biomass (kg)	2.26	1.57	3.34	1.99	
Electricity (kWh)	0.93	0.62	1.61	0.83	market for electricity, high voltage (RER)
Steam (MJ)	4.77	3.31	7.06	4.21	steam production, as energy carrier, in chemical industry (RER)
Sodium hydroxide (kg)	$1.20\cdot10^{-4}$	$8.37\cdot10^{-5}$	$1.78\cdot10^{-4}$	$1.06\cdot10^{-4}$	chlor-alkali electrolysis, membrane cell (RER)
Sulfuric acid (kg)	$7.77\cdot10^{-5}$	$5.39\cdot10^{-5}$	$1.15\cdot10^{-4}$	$6.86\cdot10^{-5}$	market for sulfuric acid (RER)
SDS (kg)	0.06	0.04	0.09	0.05	market for non-ionic surfactant (GLO)
Waste to treatment					
Wastewater (m^3)	0.04	0.52	0.98	0.47	market for wastewater, average (Europe without Switzerland)
Anaerobic digestion					
Intermediate products					
Biogas **(m^3)**	2.12	0.40	13.98	2.76	
Technosphere inputs					
Solid-rich wastewater stream (kg COD)	7.92	2.35	29.13	7.30	
Wastewater (kg COD)	1.89	0.86	3.49	1.46	
Electricity (kWh)	0.43	0.18	1.81	0.58	
Heat (MJ)	0.00	9.10	0.00	0.00	heat production, natural gas, at industrial furnace >100 kW (Europe without Switzerland)
Heat (MJ)	23.49	4.57	40.04	14.83	
Waste to treatment					
Digestate (kg)	35.35	15.16	32.38	20.91	
Emissions to air					
CH_4 (g)	0.87	0.16	5.72	1.13	
H_2S (g)	$1.41\cdot10^{-2}$	$2.7\cdot10^{-3}$	$9.35\cdot10^{-2}$	$1.85\cdot10^{-2}$	
NH_3 (g)	$1.41\cdot10^{-3}$	$2.7\cdot10^{-4}$	$9.35\cdot10^{-3}$	$1.85\cdot10^{-3}$	
CHP[e]					

(*continued on next page*)

Table 3 (*continued*)

Items	Scenario 1	Scenario 2	Scenario 3	Scenario 4	Ecoinvent process
Products					
Electricity (kWh)	5.33	1.02	35.20	4.66	
Heat (MJ)	23.97	4.57	158.38	31.27	
Technosphere inputs					
Biogas (m^3)	2.12	0.40	13.98	2.76	
Emissions to air					
CH_4 (g)	1.12	0.21	3.91	0.98	
CO_2 (kg)	4.04	0.77	26.69	5.27	
CO (g)	2.37	0.45	15.65	3.09	
N_2O (g)	0.12	0.02	0.80	0.16	
NOx (g)	0.73	0.14	4.81	0.95	
NMVOC (g)	0.10	0.02	0.65	0.13	
SO_2 (g)	1.24	0.24	8.22	1.62	

[a] Refers to current, 50% and 60% RES electricity mix production processes.

[b] RER = Europe.

[c] Biomass contains a 15% weight of PHA.

[d] GLO = Global.

[e] Note that these values are reported per FU, 1 kg of PHA powder.

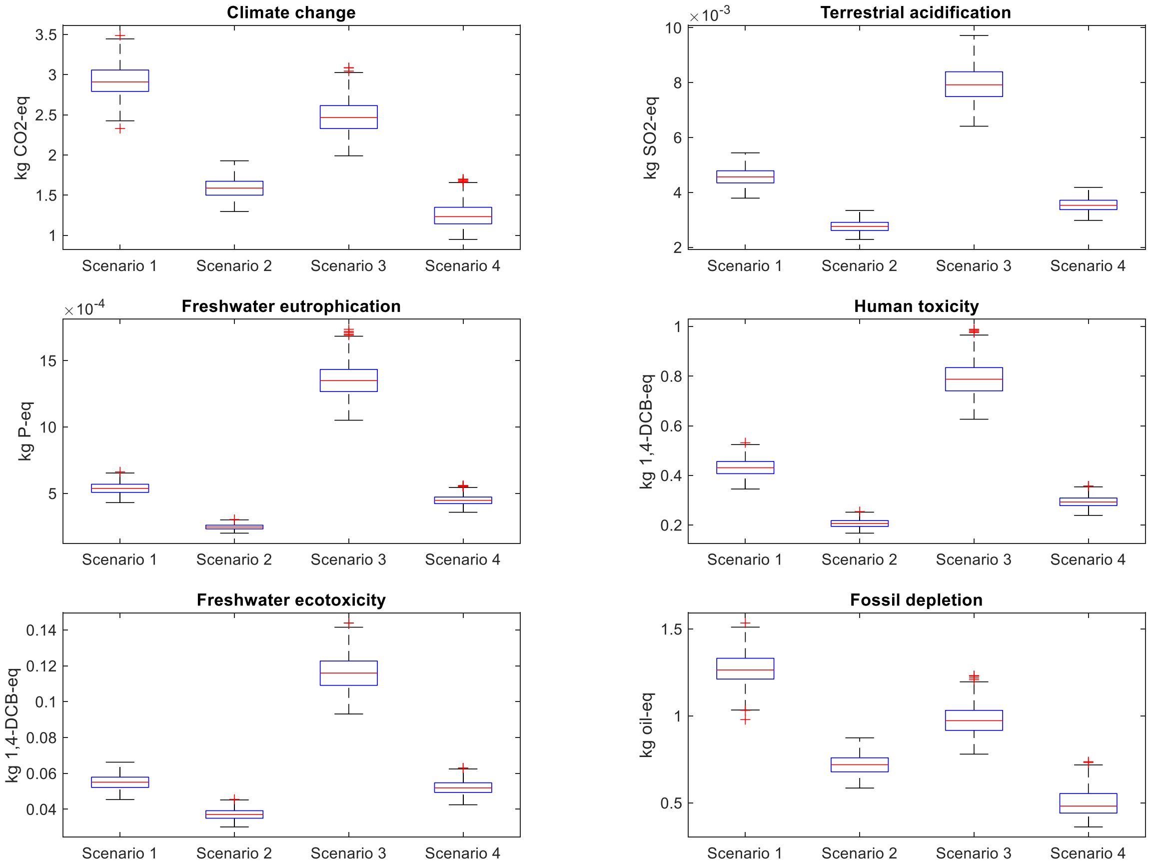

Fig. 4. Uncertainty results for PHA production in the four scenarios defined and across the evaluated impact categories.

and up to a 70% of results variance. As previously stated in the section 4.1, this parameter is strongly related to the steam, electricity, and chemicals consumption and thus, it influences considerably the environmental performance of the process. Similarly, the PHA content is responsible for an average 25% of results variance, as it is related to both chemicals and electricity consumption. The pressure drop in the high-pressure homogenizer has a negligible effect on the variance of the environmental performance results, except for scenario 1 in freshwater eutrophication and human toxicity. Other parameters such as acidification yield are responsible for up to 30% variance in GWP100 and FDP, as this parameter determines the amount of intermediate waste that is sent to anaerobic digestion and further transformed into heat (free of environmental burdens). Therefore, it affects the environmental burdens of the employed in the process.

5. Discussion

In this section, the current LCA results are compared with the available literature on LCA of PHA production from organic wastes. Those LCA studies on PHA production from organic wastes published recently (2014 onwards) were selected (Table F.1). Five out of eight were based on MMC, while the rest employed pure culture. Half of

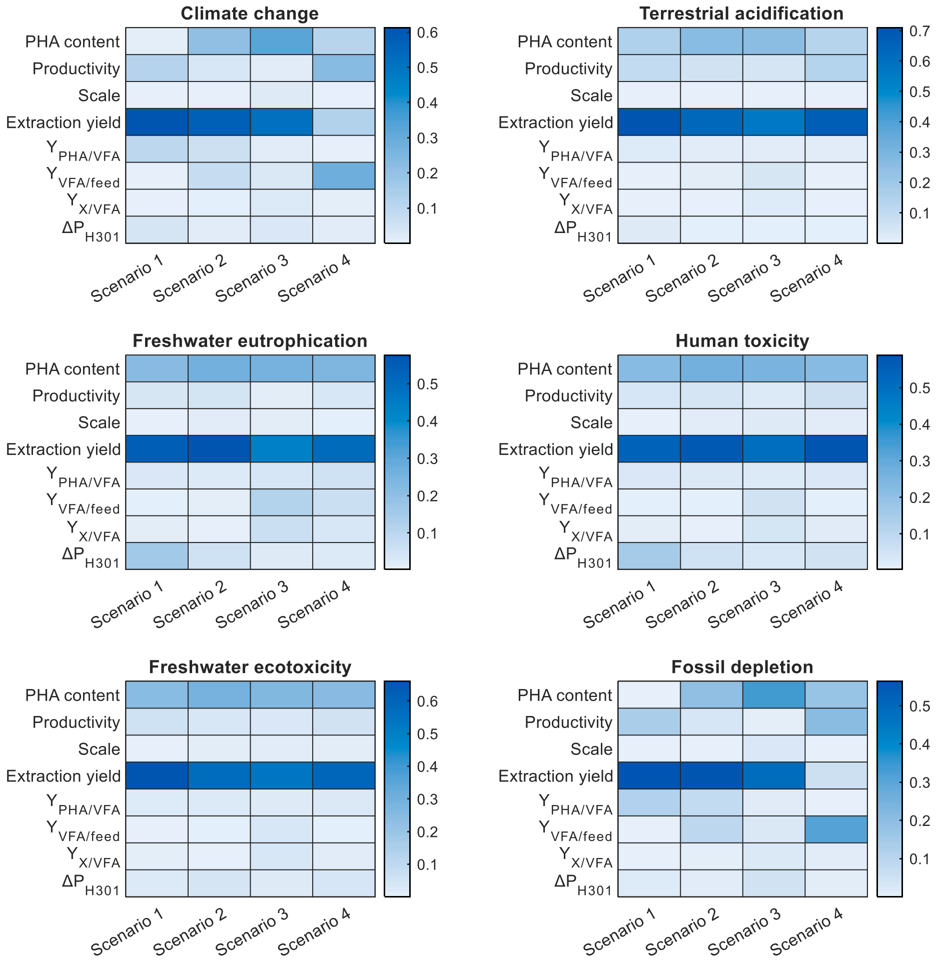

Fig. 5. GSA contribution analysis to the selected impacts categories and developed scenarios.

studies employed urban or industrial wastewater as feedstock, while the rest employed food processing byproducts and wastes such as cheese whey, molasses or glycerol. Regarding the goal and scope, half of the studies followed a consequential approach and half an attributional one. Among these ones, three out of four assessed PHA production as an alternative to conventional waste (Asunis et al., 2021) or wastewater treatment approaches (Morgan-Sagastume et al., 2016; Roibás-Rozas et al., 2020). Only Vea et al. (2021), who followed a cradle-to-grave approach, compared a PHA-based packaging material with other peers. Within the attributional approach, all the studies choose 1 kg of PHA as the functional unit, covering cradle-to-gate system boundaries. Nitkiewicz et al. (2020) evaluated the environmental performance, following an end-point approach, of medium-length and short-length PHA from crude oil, glycerol, and cooking waste oil (Fernández-Dacosta et al., 2015). compared different PHA recovery methods from MMC and provided some insights on the optimization of these processes. Heimersson et al. (2014) discussed the methodological issues of LCA on PHA production from organic wastes. However, none of these studies assess simultaneously the influence of parameters such as substrate, acidification yield, recovery yield or electricity mix. Only local sensitivity analysis, where different ratios of chemicals, electricity mixes or process configurations, were included.

The current study presents a different goal from any of previous studies. The systematic development of scenarios adds transparency and enables dealing with the uncertainty by covering the cornerstone scenarios. Hence, it is possible to determine semi-quantitatively (characterization analysis) and quantitatively (UA and GSA) the influence of both foreground and background parameters on the environmental performance. Besides, this work addresses the uncertainty related to handling a multifunctionality system. The results obtained are not straightforward comparable to those from the available literature on LCA of PHA from wastes, as the temporal boundaries are different. However, scenarios 2 and 4 outperform other LCA studies results (Asunis et al., 2021; Fernández-Dacosta et al., 2015).

6. Limitations of the study

In this work, it is proven the usefulness of prospective LCA by projecting how PHA production by MMC could develop in the future and identifying the key parameters for the environmental performance. However, limitations regarding the process upscaling and the system modelling should be considered.

Upscaling was performed using Piccinno et al. (2016) framework and pilot scale data. Elginoz et al. (2022) tested the application of this framework in anaerobic digestion and found that the main differences between industrial scale data and upscaled experimental data is caused by the difference in the methane production yield (Elginoz et al., 2022). Using mathematical models to estimate the yields combined with lab-scale data can decrease the level of uncertainty (Elginoz et al., 2022) as models impose restrictions (e.g., mass, elemental, electron balances) on the range of yield values. In this sense, the computer-aided design tool previously developed by Saavedra del Oso et al. (2022) can be employed to estimate the anaerobic fermentation yield and assess the environmental performance of other waste streams.

The potential competition amongst technologies for valorizing these organic wastes has not been addressed here. For instance, when overall PHA production yield is low (scenario 3: 0.025 kg PHA/kg COD feedstock), other options such as anaerobic digestion might be preferred from both technoeconomic and environmental perspective. Besides, feedstock availability can be also affected by future contexts, whose location and seasonality in some cases might be crucial. Another challenge that prospective LCA should address is the long-term environmental impacts caused by plastic litter. In this sense, Plastic Leak Project (Quantis, 2020) has developed a methodology to estimate an inventory for plastic leakage in the environment. MariLCA project has developed midpoint and endpoint characterization factors for the impact assessment of microplastics generation in marine environments (Corella-Puertas et al., 2022; Woods et al., 2021). Thus, future work should assess the whole value chain including the environmental impacts caused by mismanaged plastic.

7. Conclusions

In this research, we have applied the scenario methodology within a prospective LCA. To the best of our knowledge, this work elucidates for the first time how waste-to-PHA biorefineries could develop in the future and where the attention of stakeholders should be focused. Developing scenarios with a detailed description of how data were derived added transparency to the study and allowed dealing with uncertainties. The impact assessment, uncertainty assessment and the GSA quantified the influences of these parameters and identified the hotspots within the system for a more sustainable environmental performance.

As conclusions, valuable insights are extracted from this work:

- The type of substrate influences the process environmental performance, as it determines the acidification, biomass selection, and PHA accumulation yields. These parameters determine the amount of heat, electricity and chemicals employed in the system, which are the environmental hotspots. However, valorizing the intermediate waste streams into electricity and residual heat mitigates a lower yield in acidification and improves the overall system's environmental performance.
- The background scenarios influence the environmental performance. A higher RES would decrease the environmental impacts, as this process is energy intensive.
- Recovery yield and PHA content are key parameters for the environmental performance, accounting for up to 50% variance in environmental performance. These parameters are strongly related to the electricity, steam, and chemicals consumption. PHA downstream processing remains the main hotspot within the waste-to-PHA biorefinery.

This work contributes to the development of cleaner PHA production within a circular economy approach. Future work should cover the whole value chain from cradle to grave and address the long-term environmental impacts caused by plastic leakage.

CRediT authorship contribution statement

Mateo Saavedra del Oso: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Supervision, Visualization, Writing – original draft, Writing – review & editing. **Miguel Mauricio-Iglesias:** Conceptualization, Funding acquisition, Project administration, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. **Almudena Hospido:** Conceptualization, Funding acquisition, Methodology, Project administration, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. **Bernhard Steubing:** Conceptualization, Methodology, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing.

Declaration of competing interest

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

Data availability

Data will be made available on request.

Acknowledgments

This work has been financially supported by the USABLE Packaging project (Call: H2020-BBI-JTI-2018, EU ID: 836884). Mateo Saavedra del Oso, Miguel Mauricio-Iglesias and Almudena Hospido belong to a Galician Competitive Research Group (GRC ED431C 2021/37), programme co-funded by FEDER (UE). Mateo Saavedra del Oso acknowledges CRETUS for supporting his research stay at the Institute of Environmental Sciences (CML). Finally, USABLE Packaging project partners are deeply recognized for their time and valuable knowledge both at individual meetings and at the workshop.

Appendix G. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2022.135331.

Appendix A. Methodology

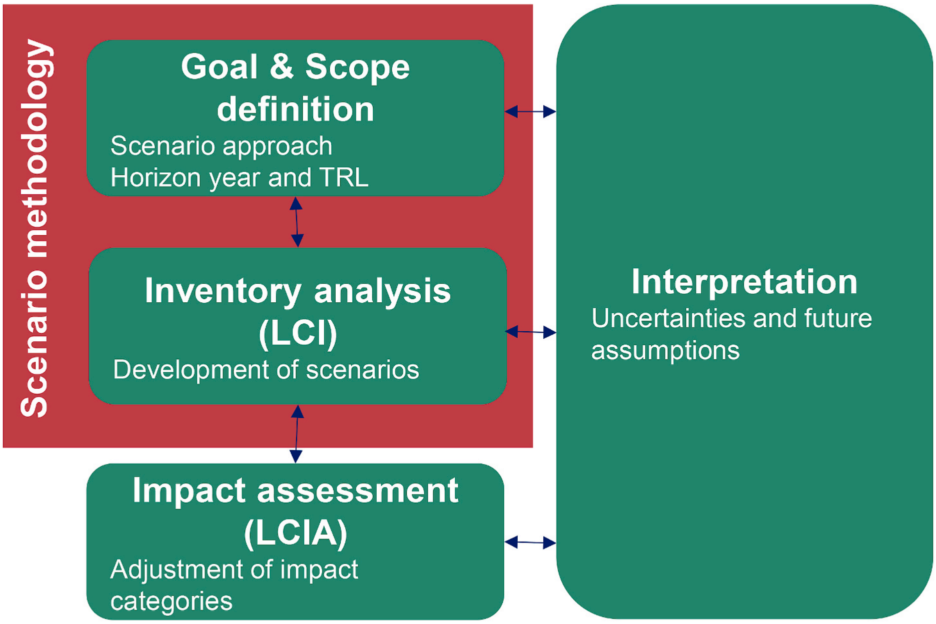

Fig. A.1. Prospective LCA stages (green boxes) and its intersection with the scenario methodology (red box).

Table A.1
Summary of peer-review studies on PHA production by MMC at pilot scale, presented in chronological order.

Number	Year	Title	Reference
1	2015	Integrated production of polyhydroxyalkanoates (PHAs) with municipal wastewater and sludge treatment at pilot scale	Morgan-Sagastume et al. (2015)
2	2017	A process for polyhydroxyalkanoate (PHA) production from municipal wastewater treatment with biological carbon and nitrogen removal demonstrated at pilot-scale	Bengtsson et al. (2017)
3	2018	Organic Fraction of Municipal Solid Waste Recovery by Conversion into Added-Value Polyhydroxyalkanoates and Biogas	Valentino et al. (2018)
4	2018	Consistent production of high quality PHA using activated sludge harvested from full scale municipal wastewater treatment – PHARIO	Werker et al. (2018)
5	2019	Pilot-Scale Polyhydroxyalkanoate Production from Combined Treatment of Organic Fraction of Municipal Solid Waste and Sewage Sludge	Valentino et al. (2019)
6	2020	Pilot-scale production of poly-3-hydroxybutyrate-co-3-hydroxyvalerate from fermented dairy manure: Process performance, polymer characterization, and scale-up implications	Guho et al. (2020)
7	2020	Biopolymers from urban organic waste: Influence of the solid retention time to cycle length ratio in the enrichment of a Mixed Microbial Culture (MMC)	Moretto et al. (2020)
8	2020	Mixed-culture polyhydroxyalkanoate (PHA) production integrated into a food-industry effluent biological treatment: A pilot-scale evaluation	Morgan-Sagastume et al. (2020)
9	2021	Combined Strategies to Boost Polyhydroxyalkanoate Production from Fruit Waste in a Three-Stage Pilot Plant	Matos et al. (2021a)
10	2021	Sludge retention time impacts on polyhydroxyalkanoate productivity in uncoupled storage/growth processes	Matos et al. (2021b)
11	2022	An integrated process for mixed culture production of 3-hydroxyhexanoate-rich polyhydroxyalkanoates from fruit waste	Silva et al. (2022)

Table A.2
Technological analysis of peer-reviewed studies on PHA by MMC at pilot scale.

Number		1	2	3	4	5	6	7	8	9
Year		2018	2018	2019	2020	2020	2020	2021	2021	2022
Feedstock		OFMSW	Industrial wastewater + SS	OFMSW + SS	Manure	OFMSW + WAS	Potato starch wastewater^3	Fruit waste	Fruit waste	Fruit waste
Acidification	Reactor configuration	CSTR	Batch	Batch	SBR	Batch		UASB	UASB	UASB
	pH	5	5.5	5		5		5	5	5
	HRT (d)	3		6	4	6		1	1	1
	OLR (gCOD/(L d))	20.5^1		6.6^1	6.8^1	15.0^1		29.2	30.0	27.9
	$Y_{VFA/S}$ (g COD/ g COD))	0.31	n.a.	0.25	n.a.	0.65^2	n.a.	0.74	0.80	0.57

(continued on next page)

Table A.2 (*continued*)

Number		1	2	3	4	5	6	7	8	9
Enrichment	Reactor configuration	SBR	SBR	SBR	SBR	SBR	SBR	SBR	SBR	SBR
	SRT (d)	1	7	1	4	1	7	4	4	4
	Cycle Length (h)	6	n.a.	6	24	12	8	12	12	12
	OLR (gCOD/(L d))	2.50	n.a.	4.00	0.52	4.00	2.20	5.40	12.30	3.00
	$Y_{X/VFA}$ (g COD/g COD))	0.20	n.a.	0.26	n.a.	0,55	n.a.	n.a.	n.a.	
Accumulation	$Y_{PHA/VFA}$ (on COD basis)	0.57	0.40	0.50	0.58–0.87	0.62	0.69	0.70	0.74	1.00
	PHA content (g/g)	0.52	0.37	0.46	0.40	0.59	0.50	0.80	0.69	0.71
Reference		Valentino et al. (2018)	Werker et al. (2018)	Valentino et al. (2019)	Guho et al. (2020)	Moretto et al. (2020)	Morgan-Sagastume et al. (2020)	Matos et al. (2021a)	Matos et al. (2021b)	Silva et al. (2022)

n.a. = not available. [1] kg TVS·m^{-3}/d. [2] kg COD/kg VS. [3] No acidification step.

Appendix B. Scenario development

B.1 Identification of influencing parameters

Table B.1
Identified influencing parameters, including description, and classification as surrounding or technological parameters.

Name	Description	Classification
Bioeconomy policy	Adoption (or not) of policies towards the deployment of biobased technologies and resource recovery from wastes	Surrounding
Alternative feedstocks	Use of waste or side-streams as feedstock to produce PHA	Surrounding
Climate change	Intensity of climate change	Surrounding
Environmental awareness	Environmental concern of both society and companies	Surrounding
Environmental policy	Adoption (or not) of policies towards the deployment of cleaner technologies	Surrounding
Gross domestic product	Total value of produced goods and provided services in a region for one year	Surrounding
Polymer applicability	Polymer applicability according to its origin and functional characteristics	Surrounding
Production costs	Operational and capital costs	Surrounding
Production scale	Amount of product (PHA) produced in a facility for one year	Surrounding
Research & Development	R&D activities regarding the PHA production by MMC that could lead to technological improvements	Surrounding
Research & Development Investment	Investments in R&D activities	Surrounding
Renewable energies share (RES)	Renewable energies (wind, solar, hydropower, biomass) share in electricity mix	Technological (Background)
Emissions regulations	Emissions regulations in the production of energy, materials and chemicals	Technological (Background)
Acidification yield	Yield of VFA from the anaerobic fermentation of feedstock	Technological (Foreground)
Biomass selection yield	Yield of PHA-storing biomass from VFA	Technological (Foreground)
Recovery yield	Yield of PHA powder from PHA enriched biomass	Technological (Foreground)
PHA accumulation yield	Yield of PHA in PHA accumulation from VFA	Technological (Foreground)
PHA content in biomass	PHA content in PHA enriched biomass after accumulation	Technological (Foreground)
Productivity	Productivity in the selection and accumulation steps	Technological (Foreground)

B.2 Creation of scenarios from sub-scenarios

For the cross-consistency check we refer to the provided excel file (APPENDIX B-1).

Table B.2
GMA for the creation of scenarios from sub-scenarios. The color coding specifies the drivers (in gold) and remaining consistent sub-scenarios (in blue).

Bioeconomy policy	Environmental policy	Feedstock	Production scale	$Y_{VFA/F}$	$Y_{X/VFA}$	$Y_{PHA/VFA}$	Productivity	PHA content in biomass	Recovery yield
(a)									
BAU	SSP2-Base Current RES	Fruit waste	500	0.35	0.40	0.50	3.00	0.46	0.65
Moderately ambitious	SSP2-RCP2.6 50% RES	OFMSW & SS	2000	0.55	0.45	0.65	6.00	0.59	0.75

(continued on next page)

Table B.2 (*continued*)

Bioeconomy policy	Environmental policy	Feedstock	Production scale	$Y_{VFA/F}$	$Y_{X/VFA}$	$Y_{PHA/VFA}$	Productivity	PHA content in biomass	Recovery yield
Ambitious	SSP2-RCP2.6 60% RES		7500	0.75	0.55	0.80	8.00	0.80	0.85
(b)									
BAU	SSP2-Base Current RES	Fruit waste	500	0.35	0.40	0.50	3.00	0.46	0.65
Moderately ambitious	SSP2-RCP2.6 50% RES	OFMSW & SS	2000	0.55	0.45	0.65	6.00	0.59	0.75
Ambitious	SSP2-RCP2.6 60% RES		7500	0.75	0.55	0.80	8.00	0.80	0.85
(c)									
BAU	SSP2-Base Current RES	Fruit waste	500	0.35	0.40	0.50	3.00	0.46	0.65
Moderately ambitious	SSP2-RCP2.6 50% RES	OFMSW & SS	2000	0.55	0.45	0.65	6.00	0.59	0.75
Ambitious	SSP2-RCP2.6 60% RES		7500	0.75	0.55	0.80	8.00	0.80	0.85
(d)									
BAU	SSP2-Base Current RES	Fruit waste	500	0.35	0.40	0.50	3.00	0.46	0.65
Moderately ambitious	SSP2-RCP2.6 50% RES	OFMSW & SS	2000	0.55	0.45	0.65	6.00	0.59	0.75
Ambitious	SSP2-RCP2.6 60% RES		7500	0.75	0.55	0.80	8.00	0.80	0.85

The eight narratives derived from the GMA, which are summarized below, were presented and discussed at the stakeholders' workshop:

- Scenario A: a juice beverage company decides to replace its present waste treatment by a waste-to-PHA system, where this polymer and energy are produced, under the premise that this new process entails lower costs and environmental impacts. This occurs under BAU environmental policies (Current RES and SSP2-Base), and bioeconomy policies and the values of process performance parameters correspond to the Sub-scenario 2. The production scale is small (500 t/y).
- Scenario B: same narrative as A, but under moderately ambitious environmental policies (50% RES and SSP2-RCP2.6).
- Scenario C: under ambitious bioeconomy policies and thus, a higher market share and wider applicability of PHA, a PHA manufacturing company employs fruit waste as feedstock. The environmental policies are moderately ambitious (50% RES and SSP2-RCP2.6), and the values of process performance parameters are the Sub-scenario 2. The production scale is large, i.e., 7500 t of PHA/y.
- Scenario D: same narrative as C but under ambitious environmental policies (60% RES and SSP2-RCP2.6) and high process performance parameters (Sub-scenario 3).
- Scenario E: under ambitious bioeconomy policies and thus, a higher market share of biobased plastics, a waste management consortium decides to produce PHA from organic fraction of solid urban waste and urban wastewater. This occurs under moderately ambitious environmental policies (50% RES and SSP2-RCP2.6) and low process performance parameters (Sub-scenario 1). The production scale is large, i.e., 7500 t PHA/y.
- Scenario F: same narrative as E, but under ambitious environmental policies (60% RES and SSP2-RCP2.6).
- Scenario G: same narrative as E, but the values of process performance parameters are from Sub-scenario 2.
- Scenario H: same narrative as F, but the values of process performance parameters are from Sub-scenario 2.

Appendix C. Process upscaling

C.1 Calculation basis

The process to estimate the calculation basis was the following:

(1) PHA and biomass obtained in PHA on a t/y and t of chemical oxygen demand (COD) basis was calculated considering influencing parameters: scale, recovery yield and PHA content in biomass (Eq. C.1 and C.2).

$$m_{PHA,basis}\left[\frac{t\ COD}{year}\right]=\frac{Scale}{Y_{extraction}}\cdot COD_{PHA} \tag{Eq. C.1}$$

$$m_{X,basis}\left[\frac{t\ COD}{year}\right]=\frac{Scale}{Y_{extraction}}\bullet\left(\frac{1}{PHA_{content}}-1\right)\bullet COD_{X} \tag{Eq. C.2}$$

(2) Based on the amount of PHA produced in the accumulation and enrichment step and the amount of PHA-storing biomass required, the amount of volatile fatty acids to fed in both enrichment and accumulation reactors was calculated based on influencing parameters: $Y_{X/VFA}$ and $Y_{PHA/VFA}$ (Eq. C.3 and C.4). The biomass productivity (XPR) and the organic loading rate (OLR) in the enrichment reactor are calculated in Eq. C.5 and C.6 considering the productivity.

$$m_{VFA,basis}\left[\frac{t\ COD}{year}\right]=\frac{m_{PHA,basis}\cdot\left(1-PHA_{selector}\right)}{Y_{PHA/VFA}}+\frac{m_{X,basis}}{Y_{X/VFA}} \tag{Eq. C.3}$$

$$f_{VFA,selector} = \frac{m_{X,basis} \Big/ Y_{X/_{VFA}}}{m_{VFA,basis}} \tag{Eq. C.4}$$

$$XPR\left[g\ X \cdot L^{-1} \cdot d^{-1}\right] = Productivity \cdot \frac{m_{X,basis} \big/ COD_X}{m_{PHA,basis} \big/ COD_{PHA}} \tag{Eq. C.5}$$

$$OLR\left[g\ VFA \cdot L^{-1} \cdot d^{-1}\right] = \frac{XPR \cdot COD_X}{Y_{X/_{VFA}}} \tag{Eq. C.6}$$

(3) Finally, considering the efficiency on the VFA separation step and the acidification yield, the amount of feedstock required was estimated (Eq. C.7).

$$m_{feed,basis}\left[\frac{t\ COD}{year}\right] = \frac{m_{VFA,basis}}{Y_{VFA/_{feed}} \cdot Y_{VFA,separation}} \tag{Eq. C.7}$$

C.2 Process design

Operational units' conditions and design parameters are summarized in Table C.1. When estimating reactors' volume, a design factor of 1.2 was applied.

Table C.1
Process specifications

	Operating Conditions/Design Basis	Reference
R101	Continuous stirred tank reactor (CSTR) HRT = 1 d (FW) or 6 d (OFMSW) OLR = 30 g/(L·d) (FW) or 10 g/(L·d) (OFMSW) T = 25 °C V_{gas} = 7.4 $m^3/(m^3 \cdot d)$ H_2 = 15.9 %vv CH_4 = 0 %vv k = 0.034 $W/(m^2 \cdot K)$ (applies to all reators) s = 0.05 m	(Matos et al., 2021a,b; Moretto et al., 2020; Silva et al., 2022)
FT101	$\eta_{VFA,separation}$ = 0.7735	(Moretto et al., 2020, 2020)
R201	Sequence batch reactor (SBR) HRT = 1 CL = 0.5 d T = 25 °C	(Moretto et al., 2020; 2022)
R202	Fed batch reactor (FBR) CL = 0.5 d T = 25 °C	(Matos et al., 2021a,b; Moretto et al., 2020; Silva et al., 2022)
FT301	Microfiltration TSS = 150 g/L	(Saavedra del Oso et al., 2021)
R301	CSTR HRT = 0.25 h pH = 4	(Saavedra del Oso et al., 2021)
H301	High pressure homogenizer ΔP_H = 1500 bar η_{Hm} = 0.9 [c] η_{he} = 0.9 [c]	(Saavedra del Oso et al., 2021)
R302	CSTR HRT = 1 h pH = 10	(Saavedra del Oso et al., 2021)
FT302	Microfiltration TSS = 150 g/L	(Saavedra del Oso et al., 2021)
R-301	CSTR Vigorous agitation [b] Residence time = 1 h [e] Stream/water dilution ratio = 1:4	(Saavedra del Oso et al., 2021)
FT303	Microfiltration TSS = 600 g/L	(Saavedra del Oso et al., 2021)
S301	Spray dryer [a] TSS = 990 g/L	(Saavedra del Oso et al., 2021)
R401	Upflow anaerobic sludge blanket HRT = 19 d OLR = 2.4 g/(L·d) Y_{biogas} = 0.243 m^3/kg COD CH_4 = 65 %vv	(Rodriguez-Verde et al., 2014)

C.3 Electricity consumption

Upscaling methods used in ex-ante LCA of emerging technologies were followed to estimate utilities consumption (Piccinno et al., 2016; Tsoy et al., 2020).

C.3.1 Stirring energy

Stirring electric energy is required by the reactors (Eq. ((8)), where N_p is a dimensionless number specific to a certain type of impeller (axial or radial) and constant at turbulent flow. N_p is 0.79 in the case of axial flow (our case) and the density of the reaction mixture (ρ_{mix}) is assumed equal to 1000 kg/m^3.

$$E_{stir}[J] = \frac{N_p \cdot \rho_{mix} \cdot N^3 \cdot d^5 \cdot t}{\eta_{stir}} \tag{8}$$

N is the rotational velocity of stirring, calculated as in Eq. (9), where v_t is the tip speed assumed equal to 1.66 m/s.

$$N\left[s^{-1}\right] = \frac{v_t}{\pi \cdot d} \tag{9}$$

The diameter of the impeller (d) is calculated based on the assumption that it measures one third of the reactor diameter (D). Since some energy loss occurs, for example through friction, from the electricity input to the actual stirring, an efficiency value (η_{stir}) is included and assumed equal to 0.9.

C.3.2 Aeration energy

Aeration is required in both enrichment and accumulation reactors. Energy required by the aeration is calculated in Eq. (10), where $W_{\Delta S}$ is the minimum isentropic gas work and n_{air} is the molar flow and the compressor efficiency (η_{aer}), assumed equal to 0.75. The required air molar flow was estimated assuming an extra 25% of the required air for the VFA consumed in the enrichment and accumulation reactors. The minimum isentropic gas work is calculated in Eq. C.11, where γ is the adiabatic compression factor (dimensionless, 1.4 for ideal gases), Z the compressibility factor (1 for ideal gas), T_1 the temperature before compression (K) and, P_1 and P_2 the in an out pressure of the gas respectively (Pa)

$$E_{aer}[J] = \frac{W_{\Delta S} \cdot \dot{n}_{air} \cdot t}{\eta_{aer}} \tag{Eq. C.10}$$

$$W_{\Delta S}\left[J \cdot mol^{-1}\right] = \frac{\gamma \cdot Z \cdot R \cdot T_1}{(\gamma - 1)} \cdot \left[\left(\frac{P_2}{P_1}\right)^{(\gamma-1)/\gamma} - 1\right] \tag{Eq. C.11}$$

C.3.3 Filtration

An electricity consumption of 10 kWh per ton of dry material was considered in all filtration units (Piccinno et al., 2016).

C.3.4 Pumping

An electricity consumption of 50 kWh per ton of dry material was considered in PHA production section (Vea et al., 2021), while a 0.155 kWh per cubic meter of stream was considered in the anaerobic digestion (Rodriguez-Verde et al., 2014).

C.4 Heat consumption

The heating energy required in the reactors is the energy necessary to raise the reaction mixture to a certain temperature and to keep it for the duration of the reaction (Q_{react}). This is calculated as the sum of the energy for raising the temperature (Q_{heat}) and the heat loss on the reactor surface (Q_{loss}) divided by the efficiency of the heating device (η_{heat}) (Eq. C.12). To calculate the Q_{heat} (Eq. C.13), the specific heat capacity (C_p), the mass of the reaction mixture (m_{mix}) and the reactor (T_r) and inlet (T_0) temperature of the stream are required. To calculate the heat loss (Eq. C.14), he surface area of the reactor (A), the thermal conductivity of the insulation material (k_a), the thickness of the insulation (s), the temperature difference between the inside and outside of the reactor ($\Delta T = T_r - T_{out}$) and the time of the reaction (t) are needed.

$$Q_{react} = \frac{Q_{heat} + Q_{loss}}{\eta_{heat}} \tag{Eq. C.12}$$

$$Q_{heat} = C_P \cdot m_{mix} \cdot (T_r - T_0) \tag{Eq. C.13}$$

$$Q_{loss} = A \cdot \frac{k_a}{s} \cdot (T_r - T_{out}) \cdot t \tag{Eq. C.14}$$

C.5 Steam consumption

The required steam (Q_{dry}) in the spray dryer is calculated based on Eq. C.15. For that, the drying efficiency (η_{dry}) the liquid's specific heat capacity ($C_{p,liq}$), the mass of the liquid (m_{liq}), the temperature difference between the boiling and the initial temperature ($\Delta T = T_{boil} - T_{out}$), the enthalpy of vaporization (ΔH_{vap}) and the mass of vaporised liquid are required (m_{vap}).

$$Q_{dry} = \frac{C_{P,liq} \cdot m_{liq} \cdot (T_{boil} - T_0) + \Delta H_{vap} \cdot m_{vap}}{\eta_{dry}} \tag{Eq. C.15}$$

C.6 Chemical consumption

Chemical consumption is summarized in Table C.2. Sodium hydroxide is consumed in acidogenic fermentation to maintain the pH and it is estimated according to Gálvez-Martos et al. (2021). Chemical consumption in enrichment reactor is calculated according to Serafim et al. (2004). Chemical consumption in PHA downstream processing (DSP) is estimated according to Saavedra del Oso et al. (2021).

Table C.2
Summary of chemical consumption.

Operational unit	Chemical	Conditions/Amount	Units	Reference
Acidification	NaOH	0.880	mol NaOH per mol C consumed	Gálvez-Martos et al. (2021)
Enrichment	NH4Cl	0.160	g/L	Serafim et al. (2004)
	K2HPO4	0.137	g/L	Serafim et al. (2004)
	CaCl2	0.070	g/L	Serafim et al. (2004)
DSP	H2SO4	pH 4		Saavedra del Oso et al. (2021)
	NaOH	pH 10		Saavedra del Oso et al. (2021)
	SDS	4.00	g/L	Saavedra del Oso et al. (2021)

C.7 Emissions

Emissions are listed in Table C.3. Biogenic carbon dioxide emissions in PHA production were estimated based on COD consumed, while emissions due to leakage in the anaerobic digestion were calculated based on the volume of biogas produced. Emissions in the cogeneration unit.

Table C.3
Summary of emissions.

Operational unit	Flow	Conditions/Amount	Units	Reference
Enrichment & Accumulation	CO_2	0.37	g/g COD	Vea et al. (2021)
Anaerobic digestion	CH_4	$6.50\cdot10^{-2}$	%vv biogas	Rodriguez-Verde et al. (2014)
	H_2S	$5.00\cdot10^{-4}$	%vv biogas	Rodriguez-Verde et al. (2014)
	NH_3	$1.00\cdot10^{-4}$	%vv biogas	Rodriguez-Verde et al. (2014)
CHP	CO_2	1.91	kg/m^3 biogas	Rodriguez-Verde et al. (2014)
	CO	$1.10\cdot10^{-3}$	kg/m^3 biogas	Rodriguez-Verde et al. (2014)
	N_2O	$5.74\cdot10^{-5}$	kg/m^3 biogas	Rodriguez-Verde et al. (2014)
	CH_4	$5.29\cdot10^{-4}$	kg/m^3 biogas	Rodriguez-Verde et al. (2014)
	NO_x	$3.44\cdot10^{-4}$	kg/m^3 biogas	Rodriguez-Verde et al. (2014)
	NMVOC	$4.62\cdot10^{-5}$	kg/m^3 biogas	Rodriguez-Verde et al. (2014)
	SO_2	$5.88\cdot10^{-4}$	kg/m^3 biogas	Rodriguez-Verde et al. (2014)

Appendix D. Electricity mix life cycle inventories

For the electricity mix LCI we refer to the provided excel file (APPENDIX D).

Appendix E. Environmental evaluation

E.1 Characterization

Fig. E.1. Electricity consumption within the PHA production.

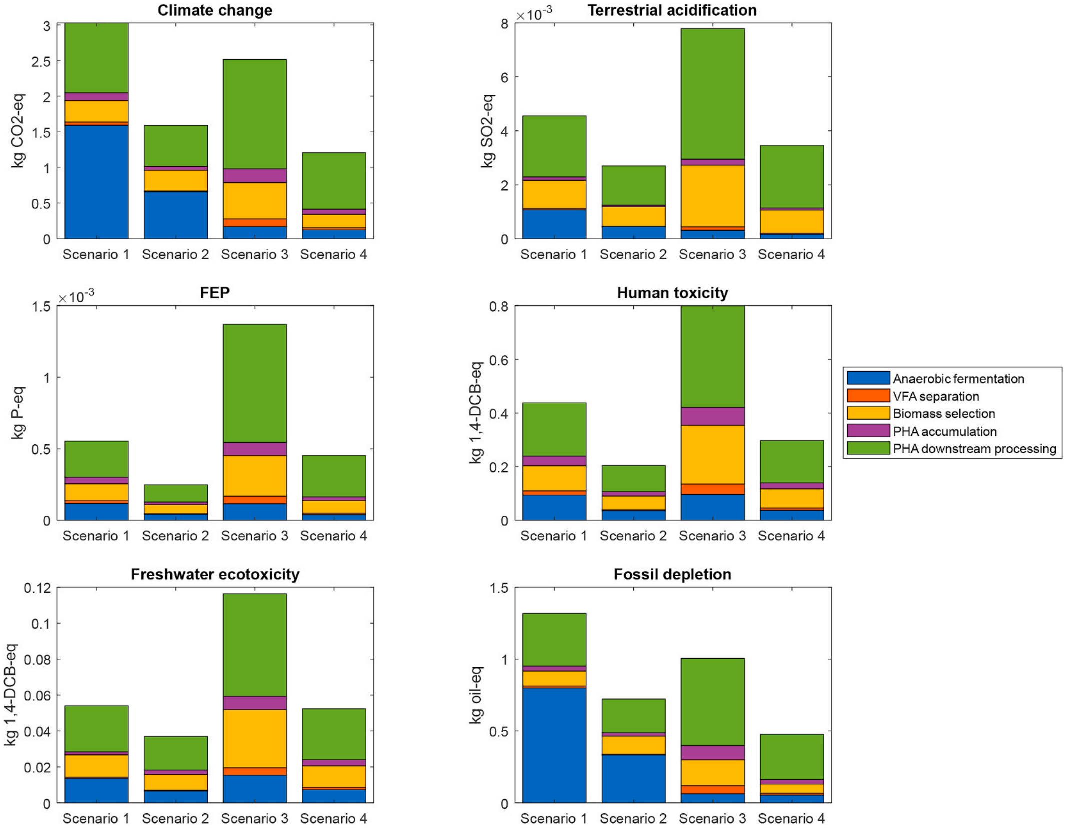

Fig. E.2. Characterization of PHA production within a waste-to-PHA biorefinery and contributions of each process unit (FU: 1 kg of PHA).

Table E.1
Qualitative assessment of the technological parameters influences on the background processes' environmental burdens. Empty cells mean negligible influence.

Parameter	Electricity	Heat[1]	Steam	Chemicals
RES	++			
Emissions regulations				
$Y_{VFA/F}$	+	+		
$Y_{X/VFA}$	+	+		++
$Y_{PHA/VFA}$	+			
Productivity	+			
PHA content in biomass	++		++	++
Recovery yield	+++		+++	+++

[1] Heat environmental burdens depend mainly on the heat source.

E.2 Comparison of allocation methods

Allocation method selection can also affect the results. To evaluate the uncertainty related to this selection, the baseline scenarios (100% allocation to PHA and electricity) were compared with substitution and other allocation approaches: COD and primary energy savings (PES) (Frangopoulos, 2012) allocation in VFA separation and CHP respectively, and only minor differences were found among allocation approaches (see Fig. E.3). As biogas production and combustion CO_2 emissions are biogenic, the related environmental impacts are caused mainly by the digestate incineration and the N_2O, NOx and NMVOC emissions. The related environmental impacts of heat production in the CHP are seven times lower than heat district production from natural gas. Thus, PES allocation results present a significant variance compared to the baseline. Similarly, as VFA separation related environmental impacts are low compared to other units, differences with the baseline allocation method are lower than a 10%. However, substitution method results differ significantly compared to allocation methods. The high amount of avoided electricity produced in scenario 3 favors its environmental performance in both GWP100 and FDP, although the emissions in the AD and CHP hamper its environmental performance in the rest of impact categories. Indeed, the environmental impacts are strongly related to these emissions. Substitution may lead to misunderstandings in decision-making. For instance, if policy makers prioritize the GWP100, scenario 3 could be chosen over other scenarios even though the PHA production environmental performance is worse. Thus, this approach should be avoided.

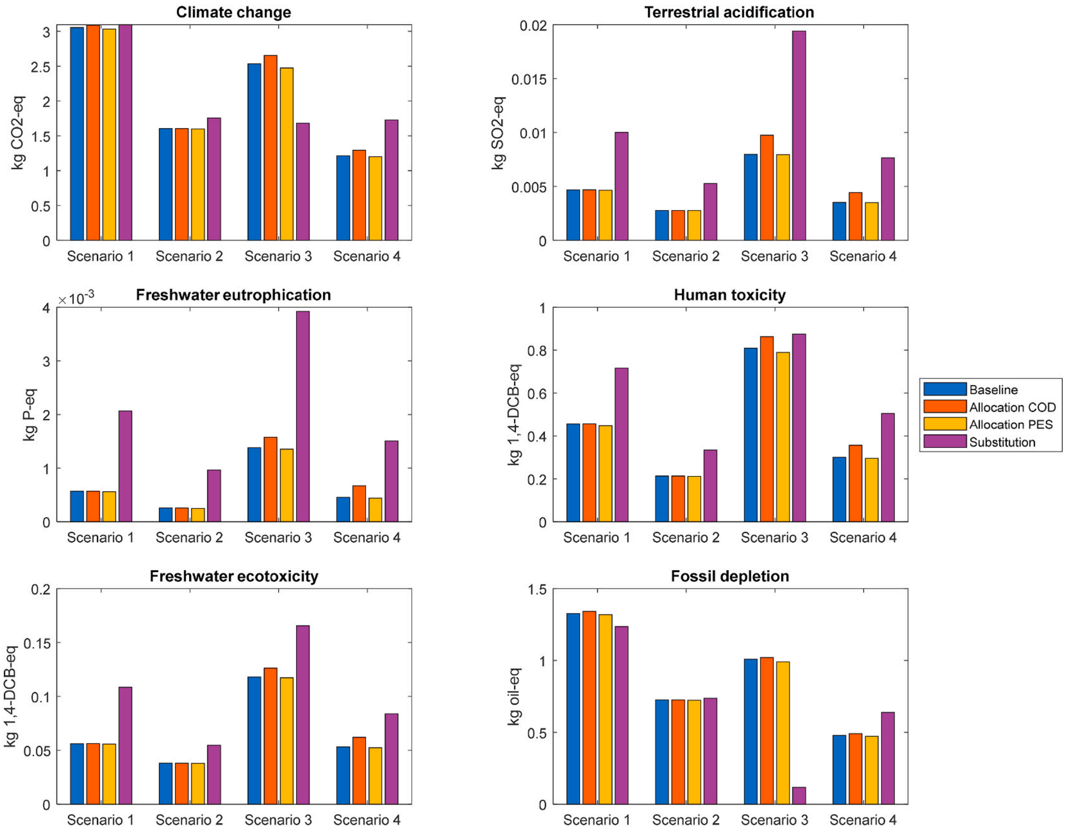

Fig. E.3. Comparison of different approaches to handle multifunctionality: baseline (solid waste stream and heat have no environmental burdens), allocation COD (on VFA separation), allocation PES (on CHP) and substitution (avoided electricity production).

Appendix F. Discussion

Table F.1
Summary of LCA studies on PHA production from organic wastes published 2014 onwards.

Number	Year	Title	Reference
1	2014	Methodological issues in life cycle assessment of mixed-culture polyhydroxyalkanoate production utilising waste as feedstock	Heimersson et al. (2014)
2	2015	Microbial community-based polyhydroxyalkanoates (PHAs) production from wastewater: Techno-economic analysis and ex-ante environmental assessment	Fernández-Dacosta et al. (2015)
3	2016	Techno-environmental assessment of integrating polyhydroxyalkanoate (PHA) production with services of municipal wastewater treatment	Morgan-Sagastume et al. (2016)
4	2020	How sustainable are biopolymers? Findings from a life cycle assessment of polyhydroxyalkanoate production from rapeseed-oil derivatives	Nitkiewicz et al. (2020)
5	2020	Environmental assessment of complex wastewater valorization by polyhydroxyalkanoates production	Roibás-Rozas et al. (2020)
6	2021	Environmental life cycle assessment of polyhydroxyalkanoates production from cheese whey	Asunis et al. (2021)
7	2021	Techno-economic evaluation and life-cycle assessment of poly(3-hydroxybutyrate) production within a biorefinery concept using sunflower-based biodiesel industry by-products	Kachrimanidou et al. (2021)
8	2021	Inclusion of multiple climate tipping as a new impact category in life cycle assessment of polyhydroxyalkanoate (PHA)-based plastics	Vea et al. (2021)

References

Arvidsson, R., Tillman, A.M., Sandén, B.A., Janssen, M., Nordelöf, A., Kushnir, D., Molander, S., 2018. Environmental assessment of emerging technologies: recommendations for prospective LCA. J. Ind. Ecol. 22, 1286–1294. https://doi.org/10.1111/JIEC.12690.

Asunis, F., de Gioannis, G., Francini, G., Lombardi, L., Muntoni, A., Polettini, A., Pomi, R., Rossi, A., Spiga, D., 2021. Environmental life cycle assessment of polyhydroxyalkanoates production from cheese whey. Waste Manag. 132, 31–43. https://doi.org/10.1016/J.WASMAN.2021.07.010.

Atasoy, M., Owusu-Agyeman, I., Plaza, E., Cetecioglu, Z., 2018. Bio-based volatile fatty acid production and recovery from waste streams: current status and future challenges. Bioresour. Technol. 268, 773–786. https://doi.org/10.1016/J.BIORTECH.2018.07.042.

Bell, J., Paula, L., Dodd, T., Németh, S., Nanou, C., Mega, V., Campos, P., 2018. EU ambition to build the world's leading bioeconomy—uncertain times demand innovative and sustainable solutions. Nat. Biotechnol. 40, 25–30. https://doi.org/10.1016/J.NBT.2017.06.010.

Bengtsson, S., Karlsson, A., Alexandersson, T., Quadri, L., Hjort, M., Johansson, P., Morgan-Sagastume, F., Anterrieu, S., Arcos-Hernandez, M., Karabegovic, L., Magnusson, P., Werker, A., 2017. A process for polyhydroxyalkanoate (PHA) production from municipal wastewater treatment with biological carbon and nitrogen removal demonstrated at pilot-scale. N Biotechnol 35, 42–53. https://doi.org/10.1016/J.NBT.2016.11.005.

Bergerson, J.A., Brandt, A., Cresko, J., Carbajales-Dale, M., MacLean, H.L., Matthews, H. S., McCoy, S., McManus, M., Miller, S.A., Morrow, W.R., Posen, I.D., Seager, T., Skone, T., Sleep, S., 2020. Life cycle assessment of emerging technologies: evaluation techniques at different stages of market and technical maturity. J. Ind. Ecol. 24, 11–25. https://doi.org/10.1111/JIEC.12954.

Börjeson, L., Höjer, M., Dreborg, K.H., Ekvall, T., Finnveden, G., 2006. Scenario types and techniques: towards a user's guide. Futures 38, 723–739. https://doi.org/10.1016/J.FUTURES.2005.12.002.

Corella-Puertas, E., Guieu, P., Aufoujal, A., Bulle, C., Boulay, A.M., 2022. Development of simplified characterization factors for the assessment of expanded polystyrene and tire wear microplastic emissions applied in a food container life cycle assessment. J. Ind. Ecol. https://doi.org/10.1111/JIEC.13269.

Cucurachi, S., Blanco, C.F., Steubing, B., Heijungs, R., 2022. Implementation of uncertainty analysis and moment-independent global sensitivity analysis for full-scale life cycle assessment models. J. Ind. Ecol. 26, 374–391. https://doi.org/10.1111/jiec.13194.

Cucurachi, S., van der Giesen, C., Guinée, J., 2018. Ex-ante LCA of emerging technologies. Procedia CIRP 69, 463–468. https://doi.org/10.1016/j.procir.2017.11.005.

Delpierre, M., Quist, J., Mertens, J., Prieur-Vernat, A., Cucurachi, S., 2021. Assessing the environmental impacts of wind-based hydrogen production in The Netherlands using ex-ante LCA and scenarios analysis. J. Clean. Prod. 299, 126866 https://doi.org/10.1016/J.JCLEPRO.2021.126866.

Dietrich, K., Dumont, M.J., del Rio, L.F., Orsat, V., 2017. Producing PHAs in the bioeconomy — towards a sustainable bioplastic. Sustain. Prod. Consum. 9, 58–70. https://doi.org/10.1016/j.spc.2016.09.001.

Elginoz, N., Owusu-Agyeman, I., Finnveden, G., Hischier, R., Rydberg, T., Cetecioglu, Z., 2022. Application and adaptation of a scale-up framework for life cycle assessment to resource recovery from waste systems. J. Clean. Prod. 355, 131720 https://doi.org/10.1016/J.JCLEPRO.2022.131720.

Estévez-Alonso, Á., Pei, R., van Loosdrecht, M.C.M., Kleerebezem, R., Werker, A., 2021. Scaling-up microbial community-based polyhydroxyalkanoate production: status and challenges. Bioresour. Technol. 327, 124790 https://doi.org/10.1016/J.BIORTECH.2021.124790.

Fernández-Dacosta, C., Posada, J.A., Kleerebezem, R., Cuellar, M.C., Ramirez, A., 2015. Microbial community-based polyhydroxyalkanoates (PHAs) production from wastewater: techno-economic analysis and ex-ante environmental assessment. Bioresour. Technol. 185, 368–377. https://doi.org/10.1016/j.biortech.2015.03.025.

Frangopoulos, C.A., 2012. A method to determine the power to heat ratio, the cogenerated electricity and the primary energy savings of cogeneration systems after the European Directive. Energy 45, 52–61. https://doi.org/10.1016/J.ENERGY.2011.12.044.

Fritsche, Uwe, Brunori, Gianluca, Chiaramonti, David, Galanakis, C.M., Hellweg, S., Matthews, Robert, Panoutsou, Calliope, 2020. Future Transitions for the Bioeconomy towards Sustainable Development and a Climate-Neutral Economy - Knowledge Synthesis Final Report. Publications Office of the European Union, Luxembourg.

Gálvez-Martos, J.L., Greses, S., Magdalena, J.A., Iribarren, D., Tomás-Pejó, E., González-Fernández, C., 2021. Life cycle assessment of volatile fatty acids production from protein- and carbohydrate-rich organic wastes. Bioresour. Technol. 321, 124528. https://doi.org/10.1016/J.BIORTECH.2020.124528.

Guho, N.M., Pokhrel, D., Abbasi, M., McDonald, A.G., Alfaro, M., Brinkman, C.K., Coats, E.R., 2020. Pilot-scale production of poly-3-hydroxybutyrate-co-3-hydroxyvalerate from fermented dairy manure: Process performance, polymer characterization, and scale-up implications. Bioresour. Technol. Rep. 12, 100588. https://doi.org/10.1016/j.biteb.2020.100588.

H2020 USABLE Packaging, 2019. USEABLE packaging – bio-based raw materials [WWW Document]. USABLE Packaging – Bio-Based Raw Materials. URL. https://www.usable-packaging.eu/. accessed 10.1.21.

Heimersson, S., Morgan-Sagastume, F., Peters, G.M., Werker, A., Svanström, M., 2014. Methodological issues in life cycle assessment of mixed-culture polyhydroxyalkanoate production utilising waste as feedstock. Nat. Biotechnol. 31, 383–393. https://doi.org/10.1016/j.nbt.2013.09.003.

Igos, E., Benetto, E., Meyer, R., Baustert, P., Othoniel, B., 2019. How to treat uncertainties in life cycle assessment studies? Int. J. Life Cycle Assess. 24, 794–807. https://doi.org/10.1007/S11367-018-1477-1/TABLES/3.

Kachrimanidou, V., Ioannidou, S.M., Ladakis, D., Papapostolou, H., Kopsahelis, N., Koutinas, A.A., Kookos, I.K., 2021. Techno-economic evaluation and life-cycle assessment of poly(3-hydroxybutyrate) production within a biorefinery concept using sunflower-based biodiesel industry by-products. Bioresour. Technol. 326, 124711 https://doi.org/10.1016/J.BIORTECH.2021.124711.

Khatami, K., Perez-Zabaleta, M., Owusu-Agyeman, I., Cetecioglu, Z., 2021. Waste to bioplastics: how close are we to sustainable polyhydroxyalkanoates production? Waste Manag. 119, 374–388. https://doi.org/10.1016/J.WASMAN.2020.10.008.

Langkau, S., Steubing, B., Mutel, C., Permana Aije, M., Erdmann, L., Janssen, M., n.d. Guidelines for the Construction of Scenarios for Prospective LCA.

Lorini, L., Munarin, G., Salvatori, G., Alfano, S., Pavan, P., Majone, M., Valentino, F., 2022. Sewage sludge as carbon source for polyhydroxyalkanoates: a holistic approach at pilot scale level. J. Clean. Prod. 354, 131728 https://doi.org/10.1016/J.JCLEPRO.2022.131728.

Matos, M., Cruz, R.A.P., Cardoso, P., Silva, F., Freitas, E.B., Carvalho, G., Reis, M.A.M., 2021a. Combined strategies to boost polyhydroxyalkanoate production from fruit waste in a three-stage pilot plant. ACS Sustain Chem Eng 9, 8270–8279. https://doi.org/10.1021/acssuschemeng.1c02432.

Matos, M., Cruz, R.A.P., Cardoso, P., Silva, F., Freitas, E.B., Carvalho, G., Reis, M.A.M., 2021b. Sludge retention time impacts on polyhydroxyalkanoate productivity in uncoupled storage/growth processes. Sci. Total Environ. 799, 149363. https://doi.org/10.1016/j.scitotenv.2021.149363.

Moretto, G., Lorini, L., Pavan, P., Crognale, S., Tonanzi, B., Rossetti, S., Majone, M., Valentino, F., 2020. Biopolymers from urban organic waste: influence of the solid retention time to cycle length ratio in the enrichment of a Mixed Microbial Culture (MMC). ACS Sustain. Chem. Eng. 8 https://doi.org/10.1021/acssuschemeng.0c04980.

Morgan-Sagastume, F., Bengtsson, S., de Grazia, G., Alexandersson, T., Quadri, L., Johansson, P., Magnusson, P., Werker, A., 2020. Mixed-culture polyhydroxyalkanoate (PHA) production integrated into a food-industry effluent biological treatment: a pilot-scale evaluation. J. Environ. Chem. Eng. 8, 104469 https://doi.org/10.1016/j.jece.2020.104469.

Morgan-Sagastume, F., Heimersson, S., Laera, G., Werker, A., Svanström, M., 2016. Techno-environmental assessment of integrating polyhydroxyalkanoate (PHA) production with services of municipal wastewater treatment. J. Clean. Prod. 137, 1368–1381. https://doi.org/10.1016/j.jclepro.2016.08.008.

Morgan-Sagastume, F., Hjort, M., Cirne, D., Gérardin, F., Lacroix, S., Gaval, G., Karabegovic, L., Alexandersson, T., Johansson, P., Karlsson, A., Bengtsson, S., Arcos-Hernández, M. v., Magnusson, P., Werker, A., 2015. Integrated production of polyhydroxyalkanoates (PHAs) with municipal wastewater and sludge treatment at pilot scale. Bioresour. Technol. 181, 78–89. https://doi.org/10.1016/J.BIORTECH.2015.01.046.

Nessi, S., Sinkko, T., Bulgheroni, C., Garcia-Gutierrez, P., Giuntoli, J., Konti, A., Sanye-Mengual, E., Tonini, D., Pant, R., Marelli, L., Ardente, F., 2021. Life Cycle Assessment (LCA) of Alternative Feedstocks for Plastics Production Part 1: the Plastics LCA Method. Publications Office of the European Union. https://doi.org/10.2760/693062.

Nguyenhuynh, T., Yoon, L.W., Chow, Y.H., Chua, A.S.M., 2021. An insight into enrichment strategies for mixed culture in polyhydroxyalkanoate production: feedstocks, operating conditions and inherent challenges. Chem. Eng. J. 420, 130488 https://doi.org/10.1016/J.CEJ.2021.130488.

Nitkiewicz, T., Wojnarowska, M., Sołtysik, M., Kaczmarski, A., Witko, T., Ingrao, C., Guzik, M., 2020. How sustainable are biopolymers? Findings from a life cycle assessment of polyhydroxyalkanoate production from rapeseed-oil derivatives. Sci. Total Environ. 749 https://doi.org/10.1016/j.scitotenv.2020.141279.

O'Neill, B.C., Kriegler, E., Riahi, K., Ebi, K.L., Hallegatte, S., Carter, T.R., Mathur, R., van Vuuren, D.P., 2014. A new scenario framework for climate change research: the concept of shared socioeconomic pathways. Clim. Change 122, 387–400. https://doi.org/10.1007/S10584-013-0905-2/TABLES/2.

Piccinno, F., Hischier, R., Seeger, S., Som, C., 2016. From laboratory to industrial scale: a scale-up framework for chemical processes in life cycle assessment studies. J. Clean. Prod. 135, 1085–1097. https://doi.org/10.1016/J.JCLEPRO.2016.06.164.

Quantis, 2020. The plastic Leak project guidelines [WWW Document]. URL. https://quantis.com/report/the-plastic-leak-project-guidelines/. accessed 10.13.22.

Rodriguez-Verde, I., Regueiro, L., Carballa, M., Hospido, A., Lema, J.M., 2014. Assessing anaerobic co-digestion of pig manure with agroindustrial wastes: The link between environmental impacts and operational parameters. Sci. Total Environ. 497–498, 475–483. https://doi.org/10.1016/J.SCITOTENV.2014.07.127.

Roibás-Rozas, A., Mosquera-Corral, A., Hospido, A., 2020. Environmental assessment of complex wastewater valorisation by polyhydroxyalkanoates production. Sci. Total Environ., 140893 https://doi.org/10.1016/j.scitotenv.2020.140893.

Roibás-Rozas, A., Saavedra del Oso, M., Zarroli, G., Mauricio-Iglesias, M., Mosquera-Corral, A., Fiore, S., Hospido, A., 2022. How can we validate the environmental profile of bioplastics? Towards the introduction of polyhydroxyalkanoates (PHA) in the value chains. Assessing Progress Towards Sustainability 405–429. https://doi.org/10.1016/B978-0-323-85851-9.00010-9.

Saavedra del Oso, M., Mauricio-Iglesias, M., Hospido, A., 2021. Evaluation and optimization of the environmental performance of PHA downstream processing. Chem. Eng. J. 412, 127687 https://doi.org/10.1016/J.CEJ.2020.127687.

Saavedra del Oso, M., Regueira, A., Hospido, A., Mauricio-Iglesias, M., 2022. Fostering the valorization of organic wastes into carboxylates by a computer-aided design tool. Waste Manag. 142, 101–110. https://doi.org/10.1016/J.WASMAN.2022.02.008.

Sacchi, R., Terlouw, T., Siala, K., Dirnaichner, A., Bauer, C., Cox, B., Mutel, C., Daioglou, V., Luderer, G., 2022. PRospective EnvironMental Impact asSEment (premise): a streamlined approach to producing databases for prospective life cycle assessment using integrated assessment models. Renew. Sustain. Energy Rev. 160, 112311 https://doi.org/10.1016/J.RSER.2022.112311.

Serafim, L.S., Lemos, P.C., Oliveira, R., Reis, M.A.M., 2004. Optimization of polyhydroxybutyrate production by mixed cultures submitted to aerobic dynamic feeding conditions. Biotechnol. Bioeng. 87, 145–160. https://doi.org/10.1002/BIT.20085.

Silva, F., Matos, M., Pereira, B., Ralo, C., Pequito, D., Marques, N., Carvalho, G., Reis, M.A.M., 2022. An integrated process for mixed culture production of 3-hydroxyhexanoate-rich polyhydroxyalkanoates from fruit waste. Chem. Eng. J. 427 https://doi.org/10.1016/j.cej.2021.131908.

Stehfest, E., van Vuuren, D., Kram, T., Bouwman, L., Alkemade, R., Bakkenes, M., Biemans, H., Bouwman, A., den Elzen, M., Janse, J., Alkemade, R., Bakkenes, M., Biemans, H., Bouwman, A., den Elzen, M., Janse, J., Lucas, P., van Minnen, J., Muller, C., Prins, A.G., 2014. Integrated Assessment of Global Environmental Change with IMAGE 3.0, Model Description and Policy Applications. PBL Netherlands Environmental Assessment Agency, The Hague (The Hague).

Steubing, B., de Koning, D., 2021. Making the use of scenarios in LCA easier: the superstructure approach. Int. J. Life Cycle Assess. 26, 2248–2262. https://doi.org/10.1007/S11367-021-01974-2/FIGURES/7.

Steubing, B., de Koning, D., Haas, A., Mutel, C.L., 2020. The Activity Browser — an open source LCA software building on top of the brightway framework. Software Impacts 3, 100012. https://doi.org/10.1016/J.SIMPA.2019.100012.

Tan, D., Wang, Y., Tong, Y., Chen, G.Q., 2021. Grand challenges for industrializing polyhydroxyalkanoates (PHAs). Trends Biotechnol. 39, 953–963. https://doi.org/10.1016/J.TIBTECH.2020.11.010.

Thomassen, G., van Dael, M., van Passel, S., You, F., 2019. How to assess the potential of emerging green technologies? Towards a prospective environmental and techno-economic assessment framework. Green Chem. 21, 4868–4886. https://doi.org/10.1039/C9GC02223F.

Tsoy, N., Steubing, B., van der Giesen, C., Guinée, J., 2020. Upscaling methods used in ex ante life cycle assessment of emerging technologies: a review. Int. J. Life Cycle Assess. 25, 1680–1692. https://doi.org/10.1007/S11367-020-01796-8/FIGURES/3.

Valentino, F., Gottardo, M., Micolucci, F., Pavan, P., Bolzonella, D., Rossetti, S., Majone, M., 2018. Organic fraction of municipal solid waste recovery by conversion into added-value polyhydroxyalkanoates and biogas. ACS Sustain. Chem. Eng. 6, 16375–16385. https://doi.org/10.1021/acssuschemeng.8b03454.

Valentino, F., Moretto, G., Lorini, L., Bolzonella, D., Pavan, P., Majone, M., 2019. Pilot-Scale polyhydroxyalkanoate production from combined treatment of organic fraction of municipal solid waste and sewage sludge. Ind. Eng. Chem. Res. 58, 12149–12158. https://doi.org/10.1021/acs.iecr.9b01831.

van Vuuren, D.P., Edmonds, J., Kainuma, M., Riahi, K., Thomson, A., Hibbard, K., Hurtt, G.C., Kram, T., Krey, V., Lamarque, J.F., Masui, T., Meinshausen, M., Nakicenovic, N., Smith, S.J., Rose, S.K., 2011. The representative concentration pathways: an overview. Clim. Change 109, 5–31. https://doi.org/10.1007/S10584-011-0148-Z/TABLES/4.

Vea, E.B., Fabbri, S., Spierling, S., Owsianiak, M., 2021. Inclusion of multiple climate tipping as a new impact category in life cycle assessment of polyhydroxyalkanoate (PHA)-based plastics. Sci. Total Environ. 788, 147544 https://doi.org/10.1016/J.SCITOTENV.2021.147544.

Werker, A., Bengtsson, S., Korving, L., Hjort, M., Anterrieu, S., Alexandersson, T., Johansson, P., Karlsson, A., Karabegovic, L., Magnusson, P., Morgan-Sagastume, F., Sijstermans, L., Tietema, M., Visser, C., Wypkema, E., van der Kooij, Y., Deeke, A., Uijterlinde, C., 2018. Consistent production of high quality PHA using activated sludge harvested from full scale municipal wastewater treatment -. PHARIO. Water Sci. Technol. 78, 2256–2269. https://doi.org/10.2166/wst.2018.502.

Woods, J.S., Verones, F., Jolliet, O., Vázquez-Rowe, I., Boulay, A.M., 2021. A framework for the assessment of marine litter impacts in life cycle impact assessment. Ecol. Indicat. 129, 107918 https://doi.org/10.1016/J.ECOLIND.2021.107918.

Yadav, B., Pandey, A., Kumar, L.R., Tyagi, R.D., 2020. Bioconversion of waste (water)/residues to bioplastics- A circular bioeconomy approach. Bioresour. Technol. 298, 122584 https://doi.org/10.1016/J.BIORTECH.2019.122584.