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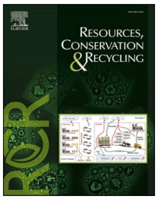

Full length article

Comparative life cycle analysis of PHA-based consumer items for daily use

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ABSTRACT

USABLE Packaging project has developed an innovative value chain based on mixed microbial cultures using organic wastes as feedstock to produce prototypes of PHA-based items that were compared to their commercial counterparts, so the expected better environmental performance can be checked. To do so, a cradle-to-grave life cycle assessment was carried out. The system was modeled integrating all available knowledge, from pilot-scale data to process simulators, and following upscaling frameworks. PHA-based items outperform their commercial counterparts since they show environmental benefits thanks to the avoided electricity obtained in the cogeneration heat and power unit within the PHA production; however, these environmental benefits are very sensitive to the substituted electricity environmental burdens. The different levels of development and market implementation are seen as critical and therefore attention should be paid on how upscaling affects the results and interpretation of the LCA while serving as a guide for process and product development.

1. Introduction

Polyhydroxyalkanoates (PHA) are biodegradable polymers produced by microorganisms from renewable sources (Yadav et al., 2020). Their biobased character and wide range of properties conforms them as an outstanding material to substitute oil-based plastics in food packaging applications (Khatami et al., 2021). Recent studies have shown environmental awareness in consumers and willingness to pay for more sustainable food packaging (Herrmann et al., 2022). But the high uncertainty related to bioplastics' environmental performance has also raised greenwashing concerns among consumers (Herrmann et al., 2022).

USABLE Packaging project (USABLE Packaging, 2019) aims to overcome former PHA-based products environmental bottlenecks (Roibás-Rozas et al., 2022) by developing new value chains from food industry wastes and byproducts. The proposed production route relies on a mixed microbial culture (MMC) system where (1) organic wastes are anaerobically fermented into volatile fatty acids (VFA), (2) culture is enriched in PHA-storing biomass by imposing a feast/famine regime and (3) PHA-storing biomass is fed VFA to accumulate PHA (Silva et al., 2022). As resulted polymer has a high hydroxyvalerate content (up to 70 % content molar basis), which hinders its downstream processing by mechanical/chemical means (Vermeer et al., 2022), a solvent-based approach was developed within USABLE Packaging Project by Bio Base Europe Pilot Plant[1] to extract the PHA (Nair, 2022). The obtained PHA, compounded with other biopolymers, resulted successfully into functional food packaging prototypes (Pardo Figuerez et al., 2022): reusable plates, frozen bags, bag in box and bread and biscuit packaging.

USABLE Packaging project was able to achieve pilot-scale performance, and the implementation of its circular approach at industrial scale depends on proving lower (or comparable) costs and environmental impacts than the commercial counterparts. In this sense, the number of studies that explore the environmental feasibility of producing PHA by MMC has grown along in recent years (Estévez-Alonso et al., 2021), with different substrates evaluated: urban wastewater (Fernández-Dacosta et al., 2015; Morgan-Sagastume et al., 2016)

Abbreviations: CHP, combined heat and power; EoL, end-of-life; FU, functional unit; LCA, life cycle assessment; LDPE, low-density polyethylene; LR, loss rate; MMC, mixed microbial culture; OTR, oxygen transfer rate; PBS, polybutylene succinate; PDF, potentially disappeared fraction of species; PEF, product environmental footprint; PHA, polyhydroxyalkanoates; PP, polypropylene; PS, polystyrene; RedL, initial release rate to the ocean; RedR, redistribution rates to the ocean; TRL, technology readiness level; VFA, volatile fatty acids; WVTR, water vapor transmission rate; ε_b, elongation at break; σ_y, tensile strength.

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[1] https://www.bbeu.org/

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industrial wastewater and side streams (Asunis et al., 2021; Roibás-Rozas et al., 2020), food waste and organic fraction of municipal solid waste (Saavedra del Oso et al., 2023), among others. Most life cycle assessment (LCA) studies pointed out PHA downstream processing as the main hotspot and, in that sense, Saavedra del Oso et al. (2021) evaluated the environmental performance of eight different processes, including mechanical, chemical, and solvent-based technologies, and proposed optimization insights. However, there is a lack of LCA studies on alternative production of PHA by MMC that cover the whole life cycle and compare its performance at product level with their commercial counterparts. Previous studies followed a cradle-to-gate or a gate-to-gate approach, excluding the final stages of the bioplastic life cycle, namely, the shaping and compounding, the use, and the end-of-life (EoL). Even when food packaging use normally adds negligible burdens to the life cycle impacts, as it does not involve significant energy consumption nor emissions to the environment (Nessi et al., 2021); excluding the EoL disregards the benefits derived from PHA biodegradability and ignores the long-term environmental impacts caused in marine ecosystems by conventional plastic pollution (Roibás-Rozas et al., 2022). Recent developments on plastic and microplastics leakage modeling (Quantis, 2020), estimation of impact methods (Corella-Puertas et al., 2022) and characterization factors (Maga et al., 2022) make possible the inclusion of the EoL phase in the LCA of plastics and bioplastics (even though these methodologies are at early development level).

The objective of this work is to analyze the environmental performance of a selection of PHA-based food packaging and items food contact (reusable plates, frozen bag, and biscuits bag) and compare it with their commercial counterparts, under a cradle-to-grave approach (i.e., from the feedstock provision to the final disposal of both managed and mismanaged plastic waste), with the aim of validating the expected better environmental performance of the selected bio-based products. By using upscaling frameworks, pilot-scale data, and process simulation, the whole PHA value chains were modelled, and life cycle impacts analysis enabled pointing out the environmental hotspots and barriers for the further development of these value chains.

2. Methodology

LCA is a systematic and standardized methodology which quantifies the potential environmental impacts of a product system throughout its life cycle (International Standard Organisation, 2006a). LCA is comprised by four steps (International Standard Organisation, 2006b): i) the goal and scope states the intended application, the system, its function and the related functional unit, the system boundaries, the impact categories selected and the impact assessment methodology; ii) the inventory analysis involves data collection and calculation procedures to quantify relevant 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 results presentation, data sensitivity analysis and critical review (if applicable).

2.1. Goal and scope

The goal and scope of the current LCA is the analysis of PHA-based items selected within the USABLE packaging consortium intended for food contact and the comparison to their commercial counterparts. The system function to be covered by the products are listed below, being in all cases the chosen functional unit (FU) one unit for each item.

- Reusable plates: food container during up to 2 h at least in hot and cold food and liquid up to 100 °C, contact with fat and dry food and microwave use, able to keep its strength (shelf life for 2 years). Diameter: 25 cm. Mechanical properties: elongation at break (ε_b) = 3 %, tensile strength (σ_y) = 3300 MPa.
- Frozen bags: protection of 900 g of frozen spinach (shelf life for 730 days), against ice and water during transport and storage (water vapor transmission rate (WVTR) < 11 $g/m^2 \cdot d$).
- Biscuits bag: protection of 250 g of biscuits (shelf life for 6 months), against migration of oxygen, water, and aromas during transport and storage (WVTR < 0.5 $g/m^2 \cdot d$ and oxygen transfer rate (OTR) = 950 $cc/m^2 \cdot d$).

The system boundaries (Fig. 1) follow a cradle-to-grave approach, covering from the production of the polymers' feedstock to the items' EoL. Some considerations regarding the system boundaries are listed below:

- During the PHA production, intermediate waste streams generated in from separating the rests of biomass from PHA are valorized into biogas via anaerobic digestion. This biogas is further transformed into electricity and heat at a cogeneration heat and power (CHP) unit, where following Nessi et al. (2021), system expansion was applied so electricity generation provides environmental credits, while no environmental credits are given to heat generation which is fully used within the system.
- The use phase is excluded from the system boundaries as it is equivalent for both USABLE prototype and commercial item and it does not involve significant energy consumption nor emissions to the environment (Nessi et al., 2021).
- The generation and transfer of microplastic to marine environments during the manufacturing and the EoL is also included and labelled as mismanaged EoL.

The ecoinvent v3.9 database (Moreno Ruiz et al., 2022) was used for background processes such as chemicals and energy production. Regarding the geographical boundaries, all processes are located in the EU, including background processes –whenever possible–. The model was implemented in the open source LCA software Activity Browser (Steubing et al., 2020), using the Environmental Footprint (Zampori and Pant, 2019) impact assessment method (EF 3.1). Further details are provided in Section 2.3.

2.2. System modeling and upscaling

Upscaling lab and pilot data to industrial scale entails a certain level of uncertainty. Following Tsoy et al. (2020) recommendations for upscaling emerging technologies and considering the available data and the technology readiness level (TRL), 4 to 6, advanced process calculations and chemical process simulation were combined here for upscaling the foreground system.

USABLE Packaging tested the feasibility of different organic wastes (regrind pasta, wheat bran, bread crust, vinasses, tuna canning wastewater, household organic waste) as substrates (Villano et al., 2021), being regrind pasta the chosen feedstock for pilot-scale testing (Rodríguez Gamero, 2022) and all the final products evaluated here. Regrind pasta is a byproduct of pasta production currently used for animal feed; pasta production was modelled according to Bevilacqua et al. (2007), and Barilla Environmental Product Declaration was used for the allocation factors (EPD International AB, 2021) for the main product (pasta) and the main byproduct (regrind pasta) following a mass-based allocation procedure stated at (Zampori and Pant, 2019).

PHA production was evaluated at pilot scale by Innoven[2] while the conversion of intermediate waste streams to biogas by anaerobic digestion was tested only at lab-scale (Rodríguez Gamero, 2022). Experimental data provided by Innoven on mass and energy flows, including chemicals consumptions, were used as the basis for upscaling the process following Piccinno et al. (2016) framework. Propionic acid

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[2] https://www.innoven.it/

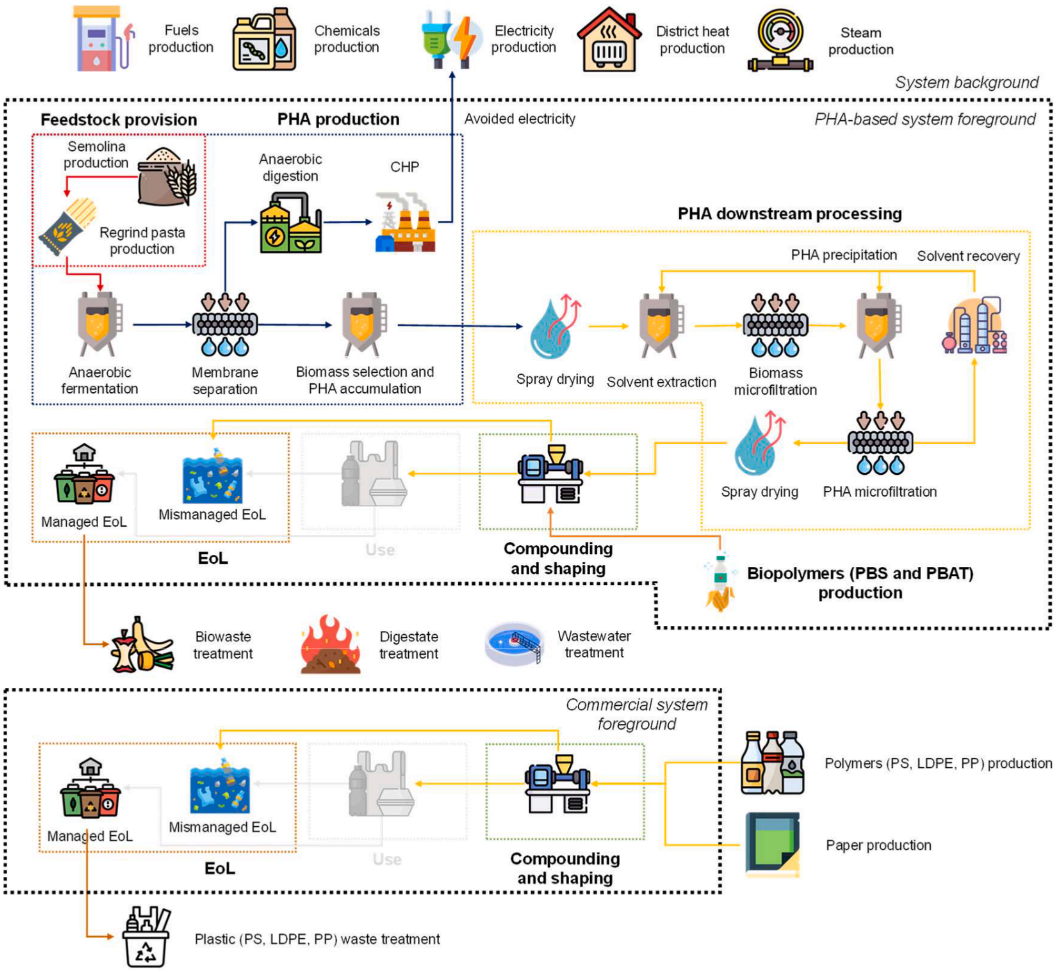

Fig. 1. System boundaries of the PHA-based items and their commercial peers. Note that most of background processes are not connected to the foreground system to enhance the visibility.

addition was considered in the upscaling for reaching the required 40 % hydroxyvalerate content (molar basis) in the polymer . Anaerobic digestion was upscaled using lab-scale and literature (Rodriguez-Verde et al., 2014) data, while the combined heat and power (CHP) unit modeling was also taken from Rodriguez-Verde et al. (2014), considering electricity conversion efficiency of 40 %, heat use efficiency of 50 %, respectively, and 10 % losses.

PHA downstream processing was assessed at pilot scale by Bio Base Europe Pilot Plant, where, following their technology developers' advice, a higher extraction yield (90 %) was considered in the upscaling. As solvent recovery was not implemented there while it has been reported as one of the environmental and economic hotspots (Saavedra del Oso et al., 2021), process simulation by Aspen Plus was used. Mass balances and the utilities requirements results were then validated with Bio Base Europe Pilot Plant, and a lower solvent-biomass ratio (9:1 instead 40:1 used at pilot scale) was finally considered as the values reported in the literature (Saavedra del Oso et al., 2021).

PHA-based items compounding and shaping also took place at pilot scale by CSIC[3] and Bio-mi[4]; however, due to confidential disclosure agreements the exact composition was not provided but a threshold composition (Pardo Figuerez et al., 2022), and all PHA-based items were assumed to have the same amount of material as their commercial peers. Polybutylene succinate (PBS), used as a copolymer for the frozen bag and the biscuit bag, was modeled according to Broeren et al. (2017). Composition for commercial items was provided by the end-users. Their shaping was modeled using that composition, and electricity consumption and polymer losses were estimated according to Broeren et al. (2017).

The managed EoL of PHA-based items and their counterparts was characterized by existing ecoinvent processes for waste treatment mixes. PHA-based items were represented as biowaste and the existing biowaste treatment mix process for Switzerland was chosen, as there was not any available process for the EU. This ecoinvent process considers the following biowaste treatment mix: 35.9 % industrial composting, 19.4 % anaerobic digestion and 44.7 % incineration. Regarding the commercial items, the existing background processes for polystyrene (PS), polypropylene (PP), low-density polyethylene (LDPE) and paper waste treatment mixes in Europe were chosen.

The generation of microplastics during the compounding and shaping and the mismanaged EoL of both PHA-based and commercial items system was modeled using the Plastic Leak Project guidelines (Quantis, 2020). A short summary of the procedure applied is described here and further calculation details can be found in the Appendix A:

- The loss rate of macroplastics (*LR*) represents the percentage of macroplastic mass that is released to the environment, was calculated according to Eq. (1), where *Littering* refers to the percentage of plastic that is littered in-home (non-flushable and flushable) and on-the-go, and the second addend refers to the mismanaged waste, which depends on the country status on waste management. The leak

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[3] https://www.csic.es/en

[4] https://www.bio-mi.eu/index.php/en/

- rate of macroplastics to the ocean is calculated according to Eq. (2), and depends on the *LR*, the initial release rate to the ocean (*RelR*), and the redistribution rates to the ocean (*RedR*).

$$LR = Littering + (1 - Littering)\cdot\left(LR_{dirpat} + Fly\ tipping + Dumping + Landfill\right) = Littering + (1 - Littering)\cdot(Unspecified\ landfills + Open\ dump + Unaccounted\ for)\ [=]\ \% \tag{1}$$

$$Leak\ macro\ ocean = LR\cdot RelR\cdot RedR\ [=]\ \% \tag{2}$$

- The resulted microplastics formation is the product of the leak of macroplastics to the ocean and the fragmentation rate.
- With regards to the microplastics generated at the manufacturing stage, the product of the estimated loss rate for manufacturing stage and final release to the ocean determines the result.

2.3. *Impact assessment*

The analysis of the environmental impacts uses the Environmental Footprint (EF) 3.1 (Zampori and Pant, 2019), as recommended by the Joint Research Center guidelines for comparative LCA of alternative feedstock for plastics production (Nessi et al., 2021). To obtain a single overall score, the characterization results are normalized and weighted according to Sala et al. (2018). The normalization factors were developed based on a technical report by the Joint Research Center, which describes the territorial production-based inventory for the European Union (Benini et al., 2014). The weighting factors were developed as hybrid evidence and judgment based weighting set that includes also aspects of the robustness of the results (Sala et al., 2018).

The long-term environmental impacts caused by microplastics generated during the mismanaged EoL and the shaping and compounding cover the physical effects on biota [CTUe/kg emitted]. It pretends to depict the physical impacts of plastic litter on organisms, including ingestion and entanglement/smothering pathways (Woods et al., 2021). The characterization factors (midpoint) (Eq. (3)) are calculated combining the developed fate factors (FF) (Corella-Puertas et al., 2022),the recommended exposure (XF) (Verones et al., 2017) and effect (EF) factors (Lavoie et al., 2021). The characterization factors for endpoint approach were estimated according to Eq. (4) (Corella-Puertas et al., 2022), with a severity factor of 0.5 PFD/PAF (PDF: Potentially disappeared fraction of species) (Bulle et al., 2019) and a continental seawater depth of 100 m (Fantke et al., 2017). The endpoint scores are further aggregated to the ecosystem quality endpoint category of IMPACT World method (Bulle et al., 2019) to measure the impact of microplastics. Further explanations on the estimation of characterization factor are provided in Appendix A.

$$CF(midpoint) = FF\cdot EF\cdot XF\ [=]\ \frac{PAF\cdot m^3}{kg\ in\ marine\ compartment} \tag{3}$$

$$CF(endpoint) = \frac{FF\cdot EF\cdot XF\cdot Severity\ factor}{Ocean\ depth}\ [=]\ \frac{PDF\cdot m^2\cdot year}{kg\ emitted} \tag{4}$$

3. Inventory analysis

Inputs of raw material and energy as well as emissions to air, water, and waste treatment for PHA production (cradle-to-gate biopolymer value chain) per declared unit (1000 kg of PHA-powder) are provided in Table 1, while the life cycle inventory (LCI) of both PHA-based and commercial items per FU (i.e. one unit for each item) are provided in Table 2. The declared unit is equivalent to 79,365, 80,000 and 45,249 PHA-based reusable plates, frozen bags and biscuit bags, respectively. LCI for PBS production is included in Appendix B.

4. Results

In this section, results of the environmental assessment are presented: characterization, normalization, and weighting of environmental impacts (Section 4.1) and contribution analysis (Section 4.2).

4.1. *Characterization, normalization, and weighting*

Table 3 summarizes: (i) the characterization results for EF impact categories and physical effects on biota, (ii) the relevance of the impact category on the normalized and weighted results and (iii) the EF single score for each item. Normalized and weighted results per impact category are included in Appendix C.

PHA-based items outperform their commercial peers from an environmental perspective, even showing environmental benefits. Ionizing radiation, freshwater eutrophication, water use, and use of mineral and metal resources are the most relevant categories for PHA-based items. Climate change and particulate matter only show relevance for PHA-based biscuit bags. Nevertheless, climate change and use of fossil resources are the most relevant categories for their commercial counterparts. Besides, PHA-based items show substantially lower physical effects on biota caused by microplastics than their commercial counterparts.

4.2. *Contribution analysis*

Contribution analysis of processes for PHA-based items is showed in Fig. 2. PHA-based items show similar contribution analysis results as they have equivalent EoL and are composed of PHA. Differences are caused by the different composition of PHA and PBS. The (avoided) electricity produced at the CHP provides environmental benefits in all impact categories, but specially in ionizing climate change, human toxicity cancer effects, ionizing radiation, acidification, freshwater eutrophication, freshwater ecotoxicity, water use and use of fossil resources. Utilities (district heat, steam and cooling energy) production contribute substantially to climate change, ozone depletion, human toxicity cancer effects, particulate matter, acidification, freshwater ecotoxicity, water use, use of fossil resources, and use of mineral and metals resources. Solvents (DMC and ethanol) production cause a relevant environmental burden in climate change, human toxicity non-cancer effects, particulate matter, acidification, terrestrial eutrophication, marine eutrophication, land use, freshwater ecotoxicity and use of mineral and metal resources. Chemicals (propionic acid, sodium bicarbonate, sodium hydroxide) production shows a relevant environmental burden in ozone depletion, particulate matter, acidification, terrestrial eutrophication and use of mineral and metal resources. Own emissions are the main contributor to photochemical ozone formation. Copolymer (PBS) production has a significant environmental burden in climate change, particulate matter and use of fossil resources for biscuit bags. Other processes such as electricity production, feedstock provision, waste treatment or end-of-life treatment have negligible contribution to overall impact categories.

Regarding the commercial items, polystyrene production is the main contributor within reusable plates production to climate change (67 %),

Table 1
Life cycle inventory of PHA production (cradle-to-gate: from feedstock provision until PHA powder obtention) per declared unit: 1000 kg of PHA.

Stage	Unit process	Product	Amount	Unit
Feedstock production	Semolina production	*Intermediate products*[*]		
		Semolina	79.8	t
		Technosphere inputs		
		Wheat	105.7	t
		Tap water	17.6	m^3
		Natural gas	17,613.6	m^3
		Electricity	68,105.9	kWh
		Truck	5284.1	tkm
		Emissions to air		
		Particulate Matter, < 2.5 mm	$1.19\cdot10^{-3}$	kg
		Sulfur dioxide	$3.64\cdot10^{-2}$	kg
		Nitrogen oxides	$1.58\cdot10^{-2}$	kg
		Dinitrogen monoxide	$6.17\cdot10^{-3}$	kg
		Carbon monoxide, fossil	$1.09\cdot10^{-3}$	kg
		Carbon dioxide, fossil	8.4	t
		Hydrocarbons	$1.26\cdot10^{-3}$	kg
	Durum wheat pasta	*Intermediate products*		
		Regrind pasta	79.1	t
		Technosphere inputs		
		Semolina production	79.8	t
		Tap water	24.5	m^3
		Natural gas	2000.2	m^3
		Electricity	12,649.2	kWh
		Heavy fuel oil	1.3	t
		Emissions to air		
		Particulate Matter, < 2.5 um	$1.19\cdot10^{-3}$	kg
		Sulfur dioxide	$3.64\cdot10^{-2}$	kg
		Nitrogen oxides	$1.58\cdot10^{-2}$	kg
		Dinitrogen monoxide	$6.17\cdot10^{-3}$	kg
		Carbon monoxide, fossil	$1.09\cdot10^{-3}$	kg
		Carbon dioxide, fossil	8.4	t
		Hydrocarbons	$1.26\cdot10^{-3}$	kg
PHA production	Anaerobic fermentation	*Technosphere inputs*		
		Regrind pasta	79.1	t
		Tap water	268.8	m^3
		Heat	379.8	MJ
		Electricity	53.5	kWh
		Sodium bicarbonate	2489.8	kg
	VFA separation	*Technosphere inputs*		
		Electricity	1212.0	kWh
	Biomass selection	*Technosphere inputs*		
		Tap water	3334.7	m^3
		Heat	831.7	MJ
		Electricity	2049.0	kWh
		Sodium hydroxide	2742.8	kg
	PHA accumulation	*Intermediate products*		
		PHA-enriched biomass	2000.0	kg
		Technosphere inputs		
		Heat	84.7	MJ
		Electricity	1380.2	kWh
		Propionic acid	841.9	kg
	Anaerobic digestion	*Technosphere inputs*		
		Heat	1022.8	MJ
		Electricity	72.0	kWh
		Emissions to air		
		Methane, non-fossil	25.0	kg
		Hydrogen sulfide	0.4	kg
		Ammonia	$4.08\cdot10^{-2}$	kg
		Waste to treatment		
		Digestate	40.7	t
	CHP	*Byproducts and coproducts*		
		Heat (byproduct)	678,962.7	MJ
		Electricity (coproduct)	150,880.6	kWh
		Emissions to air		
		Methane, non-fossil	33.2	kg
		Carbon monoxide, non-fossil	70.4	kg
		Carbon dioxide, non-fossil	120.0	t
		Nitrogen oxides	3.6	kg
		NMVOC	2.9	kg
		Nitrogen oxides	21.6	kg
		Sulfur oxides	36.9	kg
PHA downstream processing	Pretreatment	*Technosphere inputs*		
		Heat	1310.0	MJ
		Electricity	50.3	kWh

(*continued on next page*)

Table 1 (*continued*)

Stage	Unit process	Product	Amount	Unit
	Solvent extraction	*Technosphere inputs*		
		Heat	15.8	MJ
		Electricity	10.3	kWh
		Dimethyl carbonate	903.1	kg
	PHA recovery	*Products*		
		PHA	1000.0	kg
		Technosphere inputs		
		Steam	770.2	MJ
		Electricity	11.6	kWh
		Cooling energy	13,890.4	MJ
		Ethanol	2709.2	kg
	Solvent recovery	*Technosphere inputs*		
		Steam	131,039.8	MJ
		Cooling energy	130,387.5	MJ
		Emissions to air		
		Dimethyl carbonate	903.1	kg
		Ethanol	2709.2	kg

[*] Intermediate products refer to those products that are consumed internally in further life cycle stages and do not interact with external system.

Table 2

Life cycle inventory of both PHA-based (P) and commercial (C) items (gate-to-grave) for food contact: reusable plates, frozen bags and biscuits bag.

Stage	Product	Reusable plates (P)	Reusable plates (C)	Frozen bags (P)	Frozen bags (C)	Biscuits bag (P)	Biscuits bag (C)	Unit
Compounding and shaping	*Products*							
	Item	1	1	1	1	1	1	unit
	Technosphere inputs							
	PHA	$1.26\cdot10^{-2}$		$1.25\cdot10^{-2}$		$2.21\cdot10^{-2}$		kg
	PS		$1.26\cdot10^{-2}$					kg
	PBS			$5.34\cdot10^{-3}$		$5.15\cdot10^{-2}$		kg
	LDPE				$1.78\cdot10^{-2}$			kg
	PP						$5.88\cdot10^{-2}$	kg
	Paper						$1.48\cdot10^{-2}$	kg
	Electricity	0.53	0.53	0.53	0.53	0.53	0.53	kWh
	Emissions to water							
	Microplastics	$1.52\cdot10^{-7}$	$1.52\cdot10^{-7}$	$2.13\cdot10^{-7}$	$2.13\cdot10^{-7}$	$8.83\cdot10^{-7}$	$7.06\cdot10^{-7}$	kg
EoL	*Waste to treatment*							
	Biowaste	$1.26\cdot10^{-2}$						kg
	PS waste		$1.26\cdot10^{-2}$					kg
	PP waste						$5.88\cdot10^{-2}$	kg
	LDPE waste				$1.78\cdot10^{-2}$			kg
	Paper waste					$1.48\cdot10^{-2}$	kg	
	Emissions to water							
	Microplastics	$9.68\cdot10^{-7}$	$9.68\cdot10^{-7}$	$3.41\cdot10^{-5}$	$3.41\cdot10^{-5}$	$2.32\cdot10^{-4}$	$1.86\cdot10^{-4}$	kg

carcinogenic human toxicity (37 %), particulate matter formation (96 %), photochemical oxidant formation (96 %), acidification (97 %), terrestrial eutrophication (93 %) and marine eutrophication (81 %). Waste polystyrene treatment contributes to climate change (32 %), carcinogenic human toxicity (36 %), non-carcinogenic human toxicity (53 %) and freshwater ecotoxicity (63 %). Similarly, polyethylene production is the main contributor within frozen bags manufacturing to most impact categories, being waste management especially relevant to climate change (37 %). Environmental burdens within biscuit bags production are shared by paper production, propylene production and propylene waste management. Paper and its waste management dominate some categories such as terrestrial eutrophication (41 %), land use (41 %) or freshwater ecotoxicity (36 %).

An endpoint approach (ecosystem quality) was chosen to measure the contribution of physical effects on biota. As shown in Fig. A.1, microplastic impacts in marine ecosystem contribute to up to 81 % of overall environmental impacts in ecosystem quality for commercial frozen bags and biscuit bags. Microplastic impacts in marine ecosystems contribution to ecosystem quality for prototypes are almost negligible (reusable plates and frozen bags) and low (biscuits bags) due to lower CF ($2.49\cdot10^{-4}$ - $2.93\cdot10^{-2}$ vs $4.12\cdot10^{-2}$–$9.98\cdot10^{-2}$), as PHA-based items are composed by biodegradable polymers. For further details see Appendix A.

5. Discussion

In this section, the environmental hotspots of PHA-based items' value chain (Section 5.1) and the limitations of the environmental assessment (Section 5.2) are discussed.

5.1. Hotspot analysis

Ionizing radiation, freshwater eutrophication, water use, and use of mineral and metal resources were pointed out as the most relevant impact categories in Section 4.1. Fig. 3 depicts the hotspot analysis of the life cycle stages of PHA-based items for these impact categories. Climate change was included to enable the comparability with other research studies. CHP is excluded from PHA production to enable the visibility of PHA production environmental burdens.

Independent of the item assessed, PHA downstream processing is the main environmental hotspot in climate change, water use and use of minerals and metal resources. These environmental burdens are due to solvent production and solvent recovery, as high amounts of steam (131 MJ per kg PHA) and cooling energy are required there (Table 1). These results are aligned with previous LCA on PHA downstream processing (Saavedra del Oso et al., 2023, 2021).

When disregarding avoided electricity credits, PHA production

Table 3
Characterization, impact category relevance after normalization and weighting, and overall single scores of PHA-based and commercial items for food contact (bold values indicates the best performance between pairs).

Midpoint indicator	Reusable plates (P)	Reusable plates (C)	Frozen food packaging (P)	Frozen food packaging (C)	Biscuits bag (P)	Biscuits bag (C)	Unit
Characterization							
Climate change	**$-2.86\cdot10^{-4}$**	$6.83\cdot10^{-2}$	**$5.85\cdot10^{-2}$**	$8.30\cdot10^{-2}$	0.57	**$9.25\cdot10^{-2}$**	kg CO_2-eq
Ozone depletion	$3.24\cdot10^{-8}$	**$3.03\cdot10^{-11}$**	$3.32\cdot10^{-8}$	**$3.59\cdot10^{-10}$**	$6.88\cdot10^{-8}$	**$1.50\cdot10^{-9}$**	kg CFC-11-eq
Human toxicity, cancer effects	$1.54\cdot10^{-10}$	**$1.08\cdot10^{-11}$**	$2.00\cdot10^{-10}$	**$2.43\cdot10^{-11}$**	$7.27\cdot10^{-10}$	**$6.67\cdot10^{-11}$**	CTUh
Human toxicity, non-cancer effects	$3.51\cdot10^{-9}$	**$1.11\cdot10^{-10}$**	$3.77\cdot10^{-9}$	**$4.19\cdot10^{-10}$**	$9.11\cdot10^{-9}$	**$9.16\cdot10^{-10}$**	CTUh
particulate matter	$1.85\cdot10^{-8}$	**$1.88\cdot10^{-9}$**	$1.98\cdot10^{-8}$	**$2.21\cdot10^{-9}$**	$4.74\cdot10^{-8}$	**$4.38\cdot10^{-9}$**	disease incidence
Ionizing radiation, human health	**-0.32**	$1.20\cdot10^{-5}$	**-0.32**	$4.87\cdot10^{-3}$	**-0.52**	$1.06\cdot10^{-2}$	kbq u235-eq
Photochemical ozone formation - human health	$6.93\cdot10^{-4}$	**$1.28\cdot10^{-4}$**	$8.12\cdot10^{-4}$	**$2.29\cdot10^{-4}$**	$2.45\cdot10^{-3}$	**$4.20\cdot10^{-4}$**	kg NMVOC-eq
Acidification	$4.64\cdot10^{-4}$	**$1.75\cdot10^{-4}$**	$6.39\cdot10^{-4}$	**$2.28\cdot10^{-4}$**	$2.56\cdot10^{-3}$	**$4.09\cdot10^{-4}$**	mol H+-eq
Eutrophication, terrestrial	$2.30\cdot10^{-3}$	**$3.07\cdot10^{-4}$**	$2.62\cdot10^{-3}$	**$4.86\cdot10^{-4}$**	$7.45\cdot10^{-3}$	**$1.18\cdot10^{-3}$**	mol N-eq
Eutrophication, freshwater	**$-3.74\cdot10^{-4}$**	$5.56\cdot10^{-7}$	**$-3.56\cdot10^{-4}$**	$1.40\cdot10^{-5}$	**$-5.38\cdot10^{-4}$**	$1.09\cdot10^{-4}$	kg N-eq
Eutrophication marine	$3.95\cdot10^{-4}$	**$3.22\cdot10^{-5}$**	$4.18\cdot10^{-4}$	**$5.15\cdot10^{-5}$**	$9.63\cdot10^{-4}$	**$1.99\cdot10^{-4}$**	kg N-eq
Land use	3.77	**0.03**	3.82	**0.15**	7.58	**0.48**	CTUe
Ecotoxicity freshwater	4.78	**$5.05\cdot10^{-3}$**	4.82	**0.19**	**9.41**	11.11	pt
Water use	0.79	$2.92\cdot10^{-2}$	0.82	**$3.79\cdot10^{-2}$**	1.78	**$4.84\cdot10^{-2}$**	m^3
Resource use, fossils	**-3.71**	1.05	**-2.90**	1.53	**0.89**	1.74	MJ
Resource use, minerals and metals	$4.55\cdot10^{-6}$	**$6.88\cdot10^{-9}$**	$4.65\cdot10^{-6}$	**$1.89\cdot10^{-7}$**	$9.60\cdot10^{-6}$	**$2.97\cdot10^{-7}$**	kg Sb-eq
physical effects on biota*	**$2.03\cdot10^{-5}$**	$8.14\cdot10^{-3}$	**0.04**	0.25	**0.50**	1.36	CTUe
Normalization and weighting							
Climate change	0.0 %	45.3 %	3.2 %	31.3 %	34.0 %	17.7 %	
Ozone depletion	0.2 %	0.0 %	0.2 %	0.0 %	0.4 %	0.0 %	
Human toxicity, cancer effects	0.2 %	0.1 %	0.2 %	0.2 %	0.9 %	0.3 %	
Human toxicity, non-cancer effects	0.3 %	0.1 %	0.3 %	0.2 %	0.8 %	0.3 %	
particulate matter	4.9 %	6.6 %	5.7 %	4.4 %	15.0 %	4.5 %	
Ionizing radiation, human health	100.1 %	0.0 %	109.0 %	11.4 %	193.2 %	12.7 %	
Photochemical ozone formation - human health	1.5 %	3.7 %	1.9 %	3.7 %	6.3 %	3.5 %	
Acidification	1.0 %	4.8 %	1.4 %	3.5 %	6.3 %	3.2 %	
Eutrophication, terrestrial	0.9 %	1.6 %	1.1 %	1.4 %	3.4 %	1.7 %	
Eutrophication, freshwater	26.4 %	0.5 %	27.3 %	7.4 %	45.0 %	29.3 %	
Eutrophication marine	0.8 %	0.8 %	0.9 %	0.7 %	2.2 %	1.5 %	
Land use	0.2 %	0.0 %	0.2 %	0.1 %	0.5 %	0.1 %	
Ecotoxicity freshwater	1.7 %	1.3 %	2.2 %	3.3 %	7.0 %	5.5 %	
Water use	10.8 %	5.3 %	12.2 %	3.9 %	29.0 %	2.5 %	
Resource use, fossils	7.9 %	29.7 %	6.7 %	24.6 %	2.3 %	14.2 %	
Resource use, minerals and metals	12.1 %	0.2 %	13.4 %	3.8 %	30.3 %	3.0 %	
EF single score	$-7.85\cdot10^{-15}$	$5.94\cdot10^{-16}$	$-7.21\cdot10^{-15}$	$1.05\cdot10^{-15}$	$-6.61\cdot10^{-15}$	$2.06\cdot10^{-15}$	

* Physical effects on biota are not included in the normalized and weighted EF single score results.

contributes to climate change, ionizing radiation, freshwater eutrophication and use of mineral and metal resources. Propionic acid, sodium hydroxide and sodium bicarbonate, which are consumed in biomass selection and anaerobic fermentation respectively, are the main contributors within PHA production.

The shaping and compounding stage contribution to these impact categories depends on the item and its copolymer content. Namely, frozen bags and (especially) biscuit bags' shaping, and compounding contribution is relevant to climate change, freshwater eutrophication and use of mineral and metal resources due to PBS production.

Feedstock provision and EoL contribution to these impact categories is negligible. Physical effects on biota are caused by microplastics generated from the plastic leakage in the EoL (see Appendix A). However, it is important to quantify their environmental burdens of these two life cycle stages. Another consideration is that the environmental impacts caused by chemicals and additives during the mismanaged EoL of these items is not addressed.

5.2. Limitations and recommendations

In this work, the environmental performance of PHA-based items for food contact is analyzed and compared to their commercial peers. However, limitations regarding the process upscaling and the system modeling should be considered. PHA-based items outperform their commercial peers in some impact categories such as climate change, where the lower environmental impacts (or even environmental benefits) are due to the avoided electricity produced at the CHP and, therefore, are very sensitive to the substituted background activity. Here the European average electricity ("electricity production (high voltage) | RER") was selected and other electricity mixes with a higher share on renewable energies (e.g., Sweden) may affect the overall result.

Although PHA downstream processing was carried out at pilot scale, the solvent-biomass ratio was not optimized (ratio of 40:1), and solvent recovery was not tested. Considering previous findings on the environmental performance sensitivity of downstream processes to extraction yield (Saavedra del Oso et al., 2023), the results uncertainty is high as the low solvent-biomass ratio (ratio of 9:1) and extraction yield (95 %) assumed in upscaling has not been proved at pilot scale. However, measures can be taken to minimize the environmental impacts. For instance, using part of the biogas obtained in the anaerobic digestion in a boiler to produce steam would decrease the environmental impacts associated to solvent recovery. Another reported solution would be using acetone and water as solvent and antisolvent (Vermeer et al., 2022), as solvent recovery would entail lower utilities consumption and it would avoid using ethanol, whose production has high environmental burdens (Saavedra del Oso et al., 2021).

Finally, shaping and compounding were modelled assuming that both PHA-based and commercial items weight the same. However, the

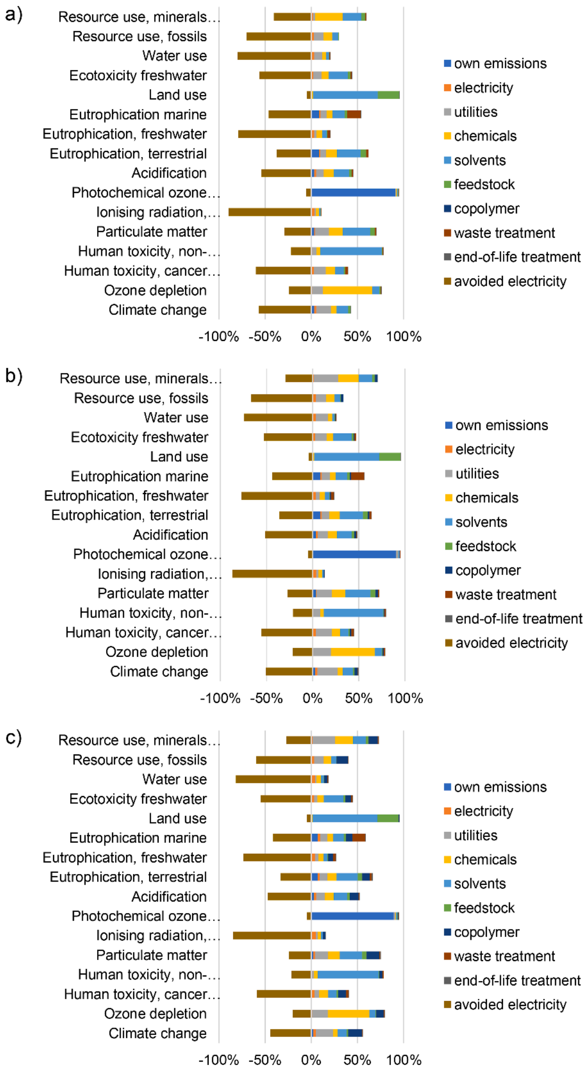

Fig. 2. Contribution analysis of PHA-based items for food contact: reusable plates (a), frozen bags (b) and biscuit bags (c).

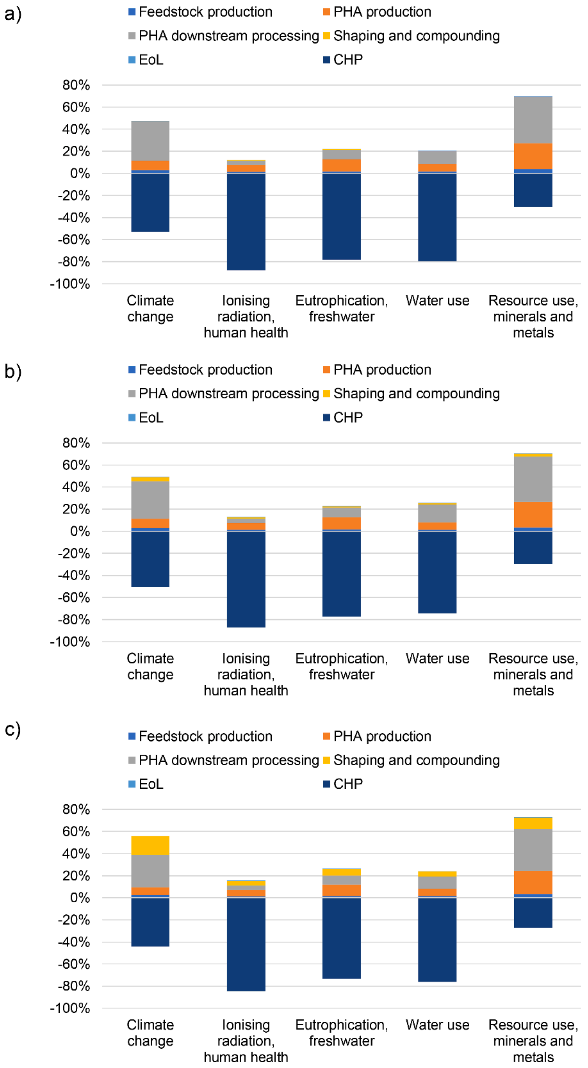

Fig. 3. Hotspot analysis of PHA-based items for food contact: reusable plates (a), frozen bags (b) and biscuit bags (c).

weight of PHA-based prototypes depends on the items' required thickness and the grammage of the compounds. Thus, there is certain uncertainty regarding this assumption that unfortunately could not be solved here due to confidentiality issues.

6. Conclusions

Under this research, the environmental performance of PHA-based items for food contact is evaluated and compared to their conventional pairs, and recommendations to improve it are provided. To the best of our knowledge, this study evaluates for first time the environmental performance of PHA-based items produced at pilot-scale using mixed microbial cultures. Besides, physical effects on biota caused by microplastics generation from plastic leakage into marine environments are included.

As conclusions, valuable insights are extracted from this work:

- PHA-based items outperform their commercial peers from an environmental perspective. Indeed, they show environmental benefits.
- Those environmental benefits are caused by avoided electricity produced at CHP. Thus, these environmental benefits are very sensitive to the substituted electricity's environmental burdens.
- PHA downstream processing is the main environmental hotspot due to solvents and utilities usage. Results are also very uncertain due to assumptions and sensitiveness to solvent biomass ratio and extraction yield.
- Recommendations are provided: using the obtained biomass to produce utilities consumed in the solvent recovery and changing DMC-ethanol by acetone-water as solvent and antisolvent.
- Due to their biodegradable character, PHA-based items show lower physical effects on biota caused by microplastics generated in the manufacturing and EoL. These impacts could contribute up to 75 % of environmental impacts in ecosystem quality endpoint impact category for commercial items.

By evaluating a selection of PHA-based food packaging items and integrating the information provided by key stakeholders, this work contributes to the development of a sustainable PHA value chain within a circular economy approach.

CRediT authorship contribution statement

Mateo Saavedra del Oso: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Supervision, Visualization, Writing – original draft, Writing – review & editing. **Rakesh Nair:** Data curation, 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.

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.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2023.107242.

Appendix A. Long-term environmental impacts caused by microplastics

Modeling of microplastics generation during the manufacturing process and the mismanaged EoL is detailed in Microplastics_LCI_CF.xls (Fig. A1)

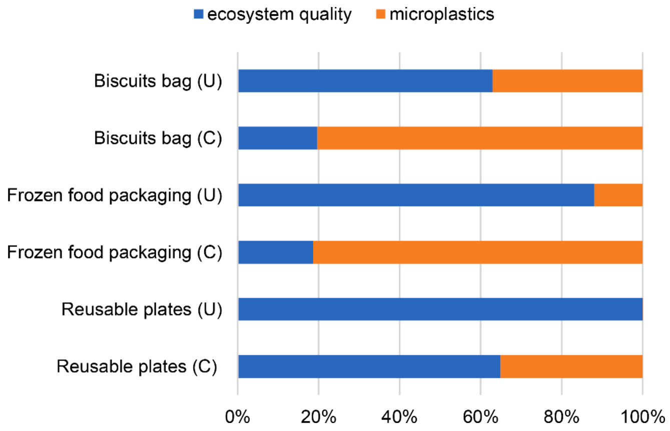

Fig. A1. Contribution of physical effects on biota to ecosystem quality endpoint impact category.

Appendix B. Inventory analysis

The inventory analysis for PBS production is summarized in Table B1.

Table B1
Life cycle inventory for PBS production (Broeren et al., 2017).

Product	Amount	Unit
PBS	1	kg
Technosphere inputs		
adipic acid	0.69	kg
butane-1,4-diol	0.52	kg
steam	3.30	MJ
electricity	0.92	kWh

Appendix C. Characterization, normalization and weighting results

Characterization, normalization and weighting results can be found in EF_Impact_assessment.xls. Contribution analysis and hotspot analysis are also included in this file.

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