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## Research article

# Environmental sustainability assessment of biodegradable bio-based poly(3-hydroxybutyrate-co-3-hydroxyvalerate) from agro-residues: Production and end-of-life scenarios

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## ABSTRACT

In the context of a circular bio-based economy, more public attention has been paid to the environmental sustainability of biodegradable bio-based plastics, particularly plastics produced using emerging biotechnologies, e.g. poly(3-hydroxybutyrate-co-3-hydroxyvalerate) or PHBV. However, this has not been thoroughly investigated in the literature. Therefore, this study aimed to address three aspects regarding the environmental impact of PHBV-based plastic: (i) the potential environmental benefits of scaling up pellet production from pilot to industrial scale and the environmental hotspots at each scale, (ii) the most favourable end-of-life (EOL) scenario for PHBV, and (iii) the environmental performance of PHBV compared to benchmark materials considering both the pellet production and EOL stages. Life cycle assessment (LCA) was implemented using Cumulative Exergy Extraction from the Natural Environment (CEENE) and Environmental Footprint (EF) methods. The results show that, firstly, when upscaling the PHBV pellet production from pilot to industrial scale, a significant environmental benefit can be achieved by reducing electricity and nutrient usage, together with the implementation of better practices such as recycling effluent for diluting feedstock. Moreover, from the circularity perspective, mechanical recycling might be the most favourable EOL scenario for short-life PHBV-based products, using the carbon neutrality approach, as the material remains recycled and hence environmental credits are achieved by substituting recyclates for virgin raw materials. Lastly, PHBV can be environmentally beneficial equal to or even to some extent greater than common bio- and fossil-based plastics produced with well-established technologies. Besides methodological choices, feedstock source and technology specifications (e.g. pure or mixed microbial cultures) were also identified as significant factors contributing to the variations in LCA of (bio)plastics; therefore, transparency in reporting these factors, along with consistency in implementing the methodologies, is crucial for conducting a meaningful comparative LCA.

## 1. Introduction

Within the circular economy action plan for the 2030 Agenda, the European Commission has given priorities to resource-intensive sectors, including plastics, for instance reducing single-use plastics, tackling sources of marine litter, and establishing a clear regulatory framework for plastics with biodegradable properties (European Commission, 2019a). This strategy is aligned with the 2030 Agenda for Sustainable

Development with the 17 Sustainable Development Goals (SDGs), adopted by all United Nations Member States in 2015 (United Nations, 2015), and the European Green Deal (European Commission, 2019b).

The term *bioplastics* refers to either the bio-based origin or the biodegradable nature of plastics, including (i) (partly) bio-based non-biodegradable, e.g. bio-based polyethylene (Bio-PE), (ii) bio-based biodegradable, e.g. polylactic acid (PLA), polyhydroxyalkanoates (PHAs), thermoplastic starch (TPS), and (iii) fossil-based biodegradable,

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e.g. polybutylene adipate terephthalate (PBAT). The global production capacity of bioplastics amounted to 2.2 million tonnes in 2022 and is forecasted to increase about 6.3 times by 2027. In 2022, PLA (21%), starch blends (18%), and Bio-PE (15%) shared the majority; packaging (48%) remains the largest field of application for bioplastics (EUBP, 2023). Amongst *bio-based biodegradable plastics* (BBPs), PHAs contributed 4% in 2022; however, it is forecasted that its production capacity will continue to grow by a factor of 6.5 (about 0.6 million tonnes) over the next 5 years, hence replacing starch blends as one of the two main BBP forms, together with PLA (EUBP, 2022). Among all bioplastics available on the market, PHA occupies a unique and special place by being 100% bio-based, bio-processed and biodegradable in various conditions (soil, aqueous and marine media, home and industrial compost). PHAs are a family of natural polyesters, produced from different bio-based feedstocks by a variety of microorganisms that store this polymer in the cell. After extraction and formulation, PHAs can be shaped into rigid or semi-rigid items using thermomechanical processes, and in their final shape, they can produce containers with interesting functional properties for food packaging applications (Cunha et al., 2016).

Commercial PHAs are currently quasi-exclusively the copolymer polyhydroxy(butyrate-co-valerate), P(HB-co-HV) or PHBV hereafter, the target material of this study. Commercial PHBV is currently produced using pure microbial cultures fed with high-purity substrates that require (1) need for sterility (2) high energy and (3) pure glucose or corn steep liquor as feedstock (48% of total production cost). However, various inexpensive substrates can be potentially used for PHBV bio-production, such as agro-food residues (maize cobs, cheese whey, fruit residues, rice straw, etc.), industrial co- or by-products (crude glycerol, condensed maize soluble, vinasse), industrial wastes, and wastewaters (e.g. from the food industry: rice industry, oil mill), or even methane waste streams, etc. (Fu et al., 2023; Lorini et al., 2022; Policastro et al., 2021). In addition to substrate type, other main aspects of PHBV production, i.e. microorganism (or combination of substrate and microorganism), and processing conditions have been investigated to obtain cost-effective PHBV material (Dev Bairwan et al., 2024). A typical example is the bioconversion of agro-food residues into PHBV using optimized eco-efficient mixed microbial cultures (MMC) (Matos et al., 2021), which enables to decrease in the investments and operating costs of the PHBV conversion in pure culture and facilitates the use of cheaper by-products as feedstock (Silva et al., 2022). Furthermore, with its intrinsically biocompatible and biodegradable properties, PHBV can be biodegraded in various conditions (aerobic, anaerobic, home compost) and nature (soil, marine) (Meereboer et al., 2020). Although mechanical recycling of PHBV is still being assessed, Dedieu et al. (2023, 2022) demonstrated that the mechanical properties of PHBV can be maintained for four cycles using multiple extrusion cycles, which is very promising for further post-usage treatments of PHBV packaging in well-established recycling and recovery streams. PHBV displays thus a broad spectrum of end-of-life (EOL) treatment options.

Various life cycle assessment (LCA) studies have been performed to address the environmental impact of production and EOL treatment of different BBPs, but not PHBV. Several recently published reviews, e.g. Bishop et al. (2021); Mastrolia et al. (2022); Walker and Rothman (2020), discussed this topic covering different perspectives, including methodological choices in LCA, the potential of bioplastics as a long-term solution to plastic pollution, and the environmental sustainability comparison between bio-based and fossil-based plastics. Secondly, variation in the applied methodologies for LCA mainly explains the differences in result interpretation for comparing different types of polymers, e.g. between bio- and fossil-based polymers. This is particularly true for the selection of EOL scenarios and the representation of biogenic carbon. These reviews also proposed suggestions for a comprehensive LCA of bioplastics, e.g. Product Environmental Footprint (PEF) standards were recommended for future comparative LCAs to draw meaningful and reliable conclusions about the environmental

sustainability of bioplastics.

As mentioned earlier, PHBV has not yet been investigated for environmental sustainability in the literature, particularly the PHBV material produced by using MMC from agro-food residues, which is well known to decrease operating costs (Matos et al., 2021). Several published LCAs have quantified the environmental impact of BBPs at the EOL stage, mostly by using the dataset for biowastes available in the LCA database, e.g. ecoinvent. Only Rossi et al. (2015) have quantified and evaluated that for PLA and TPS based on their specific chemical formulas and biodegradability. Moreover, variations in data inventory models and/or used LCA methodologies amongst studies, particularly in the EOL stage, make it infeasible to state which plastic material is the most eco-friendly when comparing the results of different LCA studies (Walker and Rothman, 2020).

To fill the above gaps, this work was performed evaluating the environmental impact of PHBV pellets, which had a high HV molar content (18–19%) and were produced from fruit residues using innovative biotechnology, i.e. the three-stage process using agro-residues as feedstock and MMC as fermentation starter, as described in Matos et al. (2021). This study includes three main objectives. (1) The life-cycle environmental impact of PHBV pellets to be further used for food packaging production was quantified. The production system was investigated through a cradle-to-gate LCA at both pilot and industrial scale: (i) to identify the process stage or the in- and outflows contributing the most to the environmental impact of PHBV pellets, and (ii) to estimate the potential environmental benefits of scaling up the PHBV production process. (2) Several EOL scenarios were compared in terms of their environmental impact. To simulate home and industrial composting conditions as a function of biodegradability, the biodegradability of PHBV films evaluated by respirometry at  $25 \pm 5$  °C to simulate the home composting conditions was presented, in addition to a literature review on this material's biodegradability in other conditions and environment (e.g. industrial composting). These scenarios were compared with incineration that includes energy recovery, and mechanical recycling. (3) Finally, the life-cycle environmental performance of PHBV in the pellet production and EOL treatment was compared with that of some benchmark materials, including both BBPs (PHB, PLA, TPS) and conventional fossil-based plastics: low-density polyethylene (LDPE) and polypropylene (PP).

## 2. Material and methods

The LCA methodology was applied to meet the three aforementioned objectives, focusing on first the PHBV pellet production at both pilot and industrial scale, and then the EOL treatment of PHBV films, following the ISO standards ISO, 2006b and ISO, 2006a. The environmental performance of benchmark materials was also investigated using LCA. More details about goal and scope definition, data inventory and the applied LCA methodologies are presented in the following sections and supplemental information (SI), section A.

### 2.1. PHBV pellet production

#### 2.1.1. Goal and scope definition

The environmental impact of PHBV pellets was quantified at the pilot and industrial scale through a cradle-to-gate LCA, looking at the PHBV pellet production and its link to the production chain (i.e. the technosphere) and the natural environment (see Fig. 1). This aimed at addressing the potential environmental benefit of scaling up the PHBV pellet production from a life cycle perspective, in addition to the identification of the key influencers at both scales. The scope was limited to pellet production as pellets are considered raw material in the plastic industry and can be converted into not only food packaging (films, trays, cups) but also into other applications.

The studied PHBV pellet production was carried out at a pilot scale (FCT-UNL, Portugal and IVV Fraunhofer, Germany) and subsequently

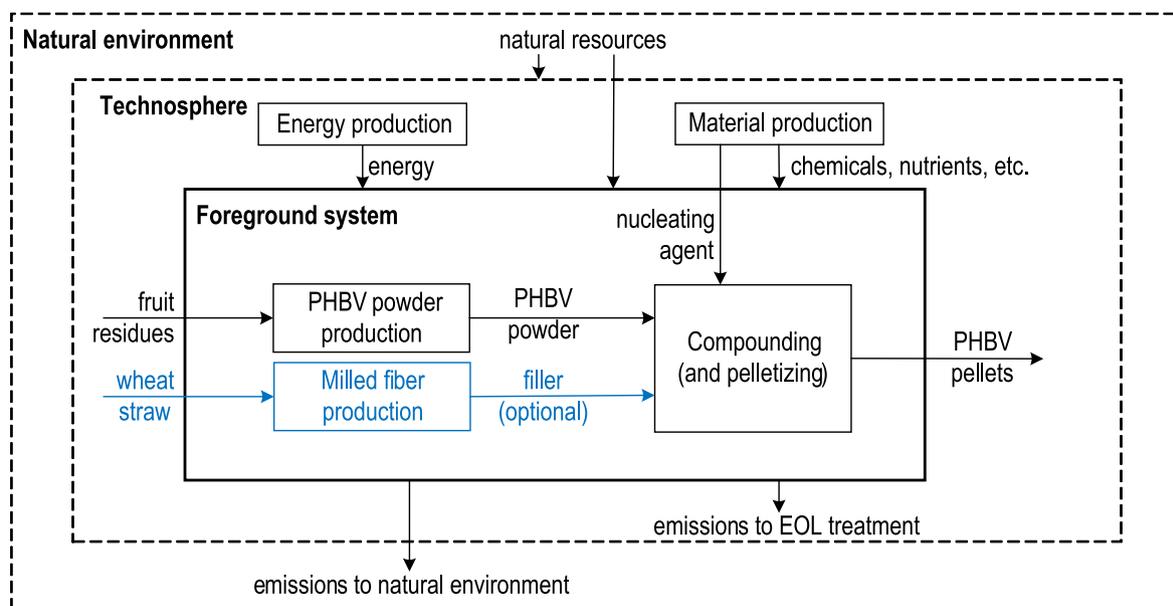


Fig. 1. Overview of the system boundary. The foreground system refers to the PHBV pellet production while the background system includes flows entering or leaving the foreground system (i.e. flows from and to the technosphere or natural environment). The blue refers to the optional input (milled fibre used as filler). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

simulated to an industrial scale (i.e. upscaling of 50 times) within the context of the Horizon project “Granting society with low environmental impact innovative packaging” (GLOPACK). The production at these two scales was assumed to obtain a similar function (or the same functional unit (FU), i.e. 1 kg of PHBV pellets formulated for food packaging to be delivered at the gate, including two main steps. PHBV powder was first produced from fruit residues via microbial synthesis, followed by extraction and purification, namely “PHBV powder production” hereafter. Secondly, the additive (i.e. boron nitride as a nucleating agent) was added to the PHBV powder (0.5 wt%) to increase the PHBV crystallinity, followed by compounding into pellets, referred to as “material processing”. Lignocellulosic fibres, e.g. milled wheat straws) were also considered as an optional filler (up to 20 wt%) (Fig. 1). More details regarding microbial synthesis can be found in Matos et al. (2021).

Four scenarios (see Table 1) were analysed associated with the two formulations, i.e. PHBV pellets without and with filler (20 wt%), produced at pilot and large scale, based on the same mass-based functional unit (FU), i.e. 1 kg of PHBV pellets.

### 2.1.2. Life cycle inventory (LCI)

For each of the four scenarios analysed, both foreground and background systems were considered. The former refers to the PHBV pellet production while the latter covers industrial processes necessary to produce and deliver the inputs to or to treat the emissions from the foreground system.

For the foreground system, the primary data of all inputs (e.g. energy, materials) and outputs (e.g. waste, emissions to air, solid losses, etc.) attributed to the pilot PHBV pellet production were collected from the GLOPACK Consortium partners and then validated by mass flow analysis (MFA). Subsequently, this pilot production was scaled up 50

times to an industrial scale, where a similar functional unit was defined (see section 2.1.1), and the data were simulated via the use of the pilot-scale data together with the following complementary sources: expert knowledge, MFA, databases, and scientific literature on rules of thumb for upscaling processes or similar technologies. Better feasible practices were also considered for industrial PHBV pellet production, e.g. recycling effluents for diluting feedstock, using a closed-water loop for cooling instead of dry ice, etc. The mass and energy data of the PHBV pellet production at both scales can be found in Nhu et al. (2021) and supplemental information SI, section A. However, the electricity use of PHBV extraction was updated using the primary data reported in Rueda et al. (2023) for PHB while energy use in PHBV purification was excluded due to a lack of data. Fruit residues, the feedstock used for PHBV production, do not have any commercial value and were assumed burden-free, following the so-called ‘cut-off approach’ (Baumann and Tillman, 2004). How this assumption affects the results will be discussed further in section 4.2.

The background system included the production of energy and materials (e.g. chemicals, nutrients, etc.), transportation, and treatment systems (Fig. 1). This data originated from the two LCA databases: Agrifootprint (v5) and ecoinvent (v3.8 with cut-off modelling), both embedded in the SimaPro software (v9.4). The analysis excluded machinery, infrastructure, and long-term emissions.

## 2.2. PHBV end-of-life treatment

### 2.2.1. Goal and scope definition

Four EOL scenarios for PHBV material were compared: home composting, incineration with energy recovery, industrial composting and mechanical recycling, using the (household) gate-to-grave LCA. This followed a hypothesis that PHBV films after use can be either home-composted or sorted at household as one of the three waste streams currently existing: (i) residual waste to be incinerated, (ii) vegetable, fruit, garden and food (VFG/F) waste to be industrial-scale composted, and (iii) household plastic packaging waste to be recycled. For recycling, PHBV was assumed to be mechanically recycled (Dedieu et al., 2022). Regarding composting, the environmental impact of both home and industrial composting for PHBV was analysed according to the minimal and maximal biodegradation kinetics of this material in these two

Table 1  
Overview of the four studied PHBV production scenarios.

	Pellets without filler		Pellets with filler	
	Pilot (P1)	Industrial (I1)	Pilot (P2)	Industrial (I2)
PHBV powder (wt%)	99	99	79	79
Additive (wt%)	1	1	1	1
Filler (wt%)	0	0	20	20

conditions. The FU is one kg of materials (polymer) to be treated in the four studied EOL scenarios.

In this work, the term *EOL treatment* refers to the final life cycle stage of a specific product that might be recycled or recovered (ILCD, 2010), but not the final stage of the material after several cycles. It means that the EOL treatment includes not only the mineralization of the polymer to CO<sub>2</sub> but also the conversion to other products (e.g. heat, electricity, compost, or recycled pellets).

### 2.2.2. Materials tested in home composting

For home composting, the biodegradation test was performed on two materials, (i) PHBV18 flat film and (ii) PHBV3 injected cups. Cellulose powder was used as a positive control. PHBV18 refers to the PHBV material produced within the scope of GLOPACK (described in section 2.1) while PHBV3 means the commercial P(3HB-3HV) containing 1–3 mol% of HV monomers, purchased from Natureplast (France). The production of PHBV18 flat film and PHBV3 injected cups was described in detail in SI, section A.

Respirometric tests were performed at  $25 \pm 5$  °C in triplicate under aerobic conditions after mixing the milled test sample with mature compost to evaluate material biodegradability under home-composting conditions. The test was performed by an independent laboratory (CSI S. p.A, Senego, Italy) according to the evaluation method described in the standard (ISO 14855-1, 2005) and using the specification standard from home composting described in AFNOR NF T51 800 (2015). More details regarding the test set-up can be found in SI, section A. The biodegradation rate was determined by the percentage of the initial theoretical carbon in the tested materials converted into CO<sub>2</sub>.

### 2.2.3. Life cycle inventory

Both foreground and background systems were considered for each of the studied EOL scenarios. Similar to the background system of PHBV production, also here ecoinvent database v3.8 was used. For home and industrial composting, the foreground data was simulated using stoichiometric analysis, mass and carbon balances and considering the biodegradation kinetics of PHBV, following the approach applied in Rossi et al. (2015) for PLA and TPS but with modifications (see SI, section A). The biodegradation kinetics of PHBV in home composting were tested in this work (section 2.2.2) but came from literature for industrial composting.

As PHBV owns physical and mechanical properties similar to PP (Policastro et al., 2021), the mechanical recycling of PHBV was modelled using the LCI reported by Civancik-Uslu et al. (2021) for that process of PP. However, the results were updated from ecoinvent v3.6 to v3.8 to keep the LCA model in this work consistent, in addition to the following adaptations. Firstly, the PHBV recyclates can substitute for only virgin PHBV pellets. Secondly, the substitution ratios were considered in the two cases: full substitutability (1:1) and technical quality loss after recycling (1:0.83), assumed similar to PLA (Rossi et al. (2015).

Regarding the incineration with energy recovery, the quantity of energy recovered was modelled specifically for PHBV, using the higher and lower heating values estimated from the elemental composition of PHBV together with the inferred equations proposed in Maksimuk et al. (2020) and Ozyuguran et al. (2018) for biomass. The efficiency of energy recovery was also considered: 10% for electricity and 24% for heat, extrapolated from the municipal incineration of biowaste available in ecoinvent v3.8. The recovered energy was assumed to substitute for energy from the grid at a 1:1 ratio.

## 2.3. Comparison with benchmark materials

### 2.3.1. Goal and scope definition

Finally, the environmental performance of the target material, PHBV pellets (scenario I1), was compared with the following polymers produced at the industrial scale: PHB, PLA, TPS, LDPE and PP pellets. A

similar analysis was performed on the EOL stage to compare these polymers' performance at this stage. PHB, PLA and TPS were selected as they are also BBPs; however, it should be kept in mind that they are different in their production characteristic: microbial synthesis for PHBV and PHB, chemical synthesis for PLA, and mechanical/thermal modification for TPS. LDPE and PP were selected as they are two of the most widespread fossil-based resins.

### 2.3.2. Life cycle inventory

Data on pellet production of PLA, TPS, LDPE and PP was available in ecoinvent v3.8 but was retrieved for PHB from Rueda et al. (2023). However, these authors did not analyse the material processing of PHB pellets and hence this process was assumed to have a similar impact to that of PHBV pellets to keep a fair comparison at the same FU for all selected polymers.

For the EOL stage, a similar approach was adopted as for PHBV (section 2.2.3) and the benchmark materials. However, the difference in biodegradation rates and substitution ratios amongst the studied polymers was considered. The kinetic biodegradation rates of PHB, PLA and TPS in industrial composting came from the review of Hussain et al. (2023) (80, 84, and 85%, respectively). For home composting, the same value, obtained from the experiment on PHBV biodegradability (section 2.2.2) was assumed for all studied BBP materials.

In the case of mechanical recycling, PHB, PLA and TPS, similar to PHBV, were assumed to be mechanically recycled, using the process for LDPE films. The results of Civancik-Uslu et al. (2021) for PP rigid were updated from ecoinvent v3.6 to v3.8 accordingly. Also, for a specific polymer, e.g. PE, the recyclates can substitute for only the PE virgin counterpart. The technical quality loss of recyclates was defined for each studied polymer (1:0.83 for PLA, 1:0.74 for PHB and TPS, 0.98 for LDPE and 0.97 for PP). The values came from Rossi et al. (2015) for PLA and TPS and from Huysveld et al. (2022) for LDPE and PP. In the case of PHBV and PHB, the quality loss was assumed based on the fact that mechanical recyclability is higher for PHBV than for PHB (Fredri and Dorigato, 2021).

To model the incineration impact, the quantity of energy recovered in the case of fossil-based plastics came from ecoinvent v3.8 while the approach described in section 2.2.3 for PHBV was implemented for PHB, PLA, and TPS. The burden of waste collection was considered for the studied EOL scenarios, except for home composting, while post-sorting/screening was included for industrial composting and mechanical recycling as the collected waste needs to be screened to sort out BBPs for better composting or recycling efficiency. The LCI of mechanical recycling of LDPE and PP came from Civancik-Uslu et al. (2021) with an update to ecoinvent v3.8 while the inventory for incineration with energy recovery is available in ecoinvent v3.8.

## 2.4. Life cycle impact assessment

Two midpoint impact categories were discussed: resource use and carbon footprint. The former is particularly relevant to bio-based materials to understand the trade-offs in resource use while the latter was the emission-related category considered in all 44 LCA studies on bio- and fossil-based plastic packaging (Bishop et al., 2021). Resource use was addressed here by the Cumulative Exergy Extraction from the Natural Environment (CEENE v.2013) method (Alvarenga et al., 2013; Dewulf et al., 2007), which uses an inherited property of the material as the basis for resource characterization (European Commission, 2011). This method measures both the quality and quantity of natural resources, including fossil, nuclear, abiotic renewable (wind, geothermal and hydropower), water, mineral, metal and land (including biotic) resources, via a single unit (i.e. Joules of exergy, Jex). CEENE is recommended as the most appropriate method for accounting and characterizing resource use (Berger et al., 2020; European Commission, 2011). In terms of carbon footprint, the Environmental Footprint (EF v3.0), proposed by the European Commission for measuring and

communicating the life cycle environmental performance of products (European Commission, 2013, European Commission, 2021a), is implemented. Moreover, the single score, obtained by aggregation of the results for the 16 environmental impact categories evaluated by the EF method was quantified and discussed in SI, section B. This single score will provide a comprehensive picture of the life-cycle environmental performance of PHBV pellets and hence is easy to interpret and communicate. These two LCIA methods were implemented for all three analyses, described in sections 2.1-2.3.

### 2.5. Sensitivity analysis

A sensitivity analysis was performed to predict the probability of different LCA outcomes intervened by the uncertainty of considered variables, using Oracle Crystal Ball, one of the leading tools for Monte Carlo simulation. All inflows and outflows attributed to the PHBV pellet production were varied (10,000 iterations) within a triangular distribution defined by a range of  $\pm 10\%$  of the original value, which helps to identify the parameter affecting the most the outcomes (Thomassen et al., 2019).

## 3. Results and discussions

The environmental impacts: resource use (i.e., the sum of fossil, nuclear, abiotic renewable (wind, geothermal and hydropower), water,

mineral, metal and land (including biotic) resources) and carbon footprint are presented for the PHBV pellet production to identify the largest contributors to these impact categories and to estimate the potential environmental benefits of scaling up this process (first objective), and then for the EOL stage to determine which EOL option is more favourable to PHBV (second objective). Finally, it is assessed how environmentally sustainable PHBV is compared to its benchmark materials (third objective).

### 3.1. PHBV pellet production

#### 3.1.1. Environmental impact of PHBV pellets

As described in section 2.1, the production of PHBV pellets included PHBV powder production, filler production (optional) and material processing. The environmental impacts of pellets (i.e. resource use and carbon footprint) are presented in Fig. 2 for the four scenarios described in Table 1 (P1, P2 at pilot scale and I1, I2 at industrial scale) and for each of the three processes mentioned above.

Firstly, it can be seen in Fig. 2a that, amongst the three involved processes, PHBV powder production contributes the most at both pilot (96 and 93% of the total impact, respectively) and industrial scale (82 and 76%, respectively), considering each time the case with and without fillers. Electricity consumption, mainly in this process, is the largest contributor to the total resource use of PHBV pellets at the pilot scale (90%, regardless of whether or not the pellets contain fillers). At the

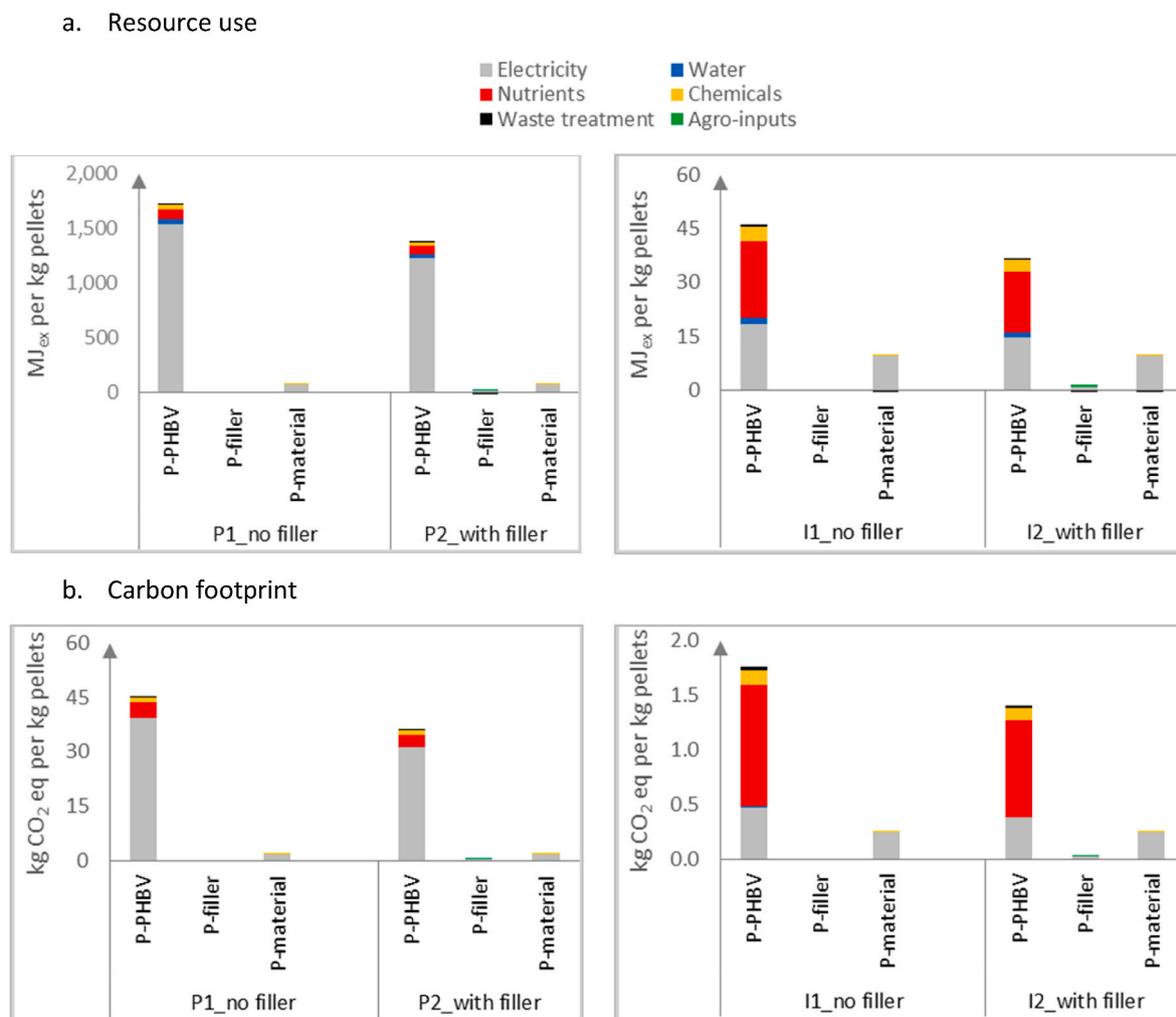


Fig. 2. Cradle-to-gate environmental impacts: (a) resource use and (b) carbon footprint of pilot (left) and industrial (right) scale production of pellets without filler (P1 and I1) and with filler (P2 and I2), produced through the three processes: PHBV powder production (P-PHBV), filler production (P-filler) and material processing (P-material).

industrial scale, in addition to electricity (50 and 52% for pellets without and with fillers, respectively), the use of nutrients (including micro-nutrients) has a high share in the resource footprint of PHBV pellets (38 and 35%, respectively). The resource footprint of PHBV pellets without fillers can decrease significantly (32 times) when the production process is scaled up 50 times (i.e. from 1796 to 56 MJ<sub>ex</sub>/kg pellets at pilot and industrial scale, respectively). The resource use in the case of PHBV pellets with fillers (i.e. dried milled wheat straw) is 1469 and 48 MJ<sub>ex</sub>/kg pellets (decrease by a factor of 30), respectively. This means that the addition of filler at 20% wt. can decrease the resource use of PHBV pellets by 18 or 14% at the pilot or industrial scale, respectively.

The substantial reduction in the resource footprint of PHBV pellets can be mainly explained by the elevated efficiency of the PHBV powder production through a consideration of scaling factors for material and energy use, following judgements of GLOPACK's Consortium experts and scientific literature on rules of thumb for upscaling processes or similar technologies. The data of gas emissions and effluents at the industrial production were estimated using mass flow analysis (for the key elements: C, H, O, N and P). The mass and energy data of the PHBV production at both scales are presented in SI, section A. Moreover, better feasible practices were considered for industrial production. More specifically, thermostatic baths to control the temperature of the reactors are required only for the pilot powder production, resulting in a remarkable energy saving at the industrial scale. Effluents from the culture selection and accumulation reactors can be recycled to dilute the feedstock input. One should keep in mind that PHBV powder production is the process contributing the most to the resource footprint of PHBV pellets at both scales. Next to that, the replacement of dry ice used at the pilot scale with a closed-water loop at the industrial scale for cooling in the material processing is also a reason for more efficient industrial production.

Regarding the carbon footprint, similar results are shown (Fig. 2b); however, the potential benefit from the production upscaling (23 times for pellets with or without fillers) is lower than that of resource use, mainly reasoned by a considerable decrease in electricity use in the PHBV powder production and fossil CO<sub>2</sub> emission due to the use of the closed-water loop instead of dry ice for cooling in the material processing. PHBV pellets also have a higher carbon footprint at both scales if no filler is added (values per kg of pellets and respectively for the case of without and with fillers: 47 and 38 kg CO<sub>2</sub> eq at pilot scale, and 2.0 and 1.7 kg CO<sub>2</sub> eq at industrial scale). In other words, the more fillers are present in the PHBV pellets, the more environmentally beneficial the PHBV pellets are in terms of resource use and carbon footprint. This is explained by the fact that fillers dried and milled from an agro-byproduct (i.e. wheat straw) have a much lower environmental impact than PHBV powder. Indeed, although the powder is produced from agro-residues, the microbial synthesis is energy- and material-intensive. This finding is in line with the work of David et al. (2021) on vine shoots used as fillers for PHB pellets regarding carbon footprint, but is also valid to the resource use category discussed above and the aggregated single score presented in SI, section B. Adding lignocellulosic fibres obtained from agricultural residues as fillers can decrease the environmental impact of the PHBV material while maintaining its functional properties (Berthet et al., 2015) and ultimate biodegradation (David et al., 2019). However, the more filler is added to the PHBV pellets, the more technical constraints might occur during the conversion of PHBV pellets into packaging materials. For instance, the fillers can influence the brittleness and colour (i.e. darker tone) of packaging as well as cause problems with the melt flow during extrusion (Berthet et al., 2015). At both scales, PHBV powder production, especially electricity use (mainly for stirring, pumping, and compressing air), has a significant impact on the carbon footprint of PHBV pellets. However, this impact, at the industrial scale, can be driven by the consumption of not only electricity but also nutrients (including micro-nutrients). The share of electricity and nutrient use is quite similar in both cases: PHBV pellets without fillers (35 and 55%, respectively) and with fillers (38 and 53%, respectively).

### 3.1.2. Sensitivity analysis of the environmental impact of PHBV pellets

To determine to which extent the variations of the input parameters (i.e. all inflows and outflows attributed to the PHBV pellet production) can affect the resource and carbon footprint per kg of PHBV pellets, a sensitivity analysis was conducted. It presents that electricity used in the PHBV powder production is the parameter of which the sensitivity can drive these two impact categories the most at both pilot and industrial scales (Fig. 3). However, at the industrial scale, the uncertainty in nutrient use in the same process can influence the outcomes but to a larger extent (non-filler pellets: 49 and 82%; pellets with filler: 47 and 80% for resource and carbon footprint, respectively). These findings align with the ones shown in section 3.1.1 about the flows (electricity and nutrient use) and the process (PHBV powder production) contributing the most to the environmental impact of PHBV pellets. Attention should be paid to these parameters to improve the environmental sustainability of PHBV pellets.

## 3.2. PHBV end-of-life treatment

### 3.2.1. PHBV biodegradation in home-compost conditions

The biodegradation in home composting conditions of PHBV18 and PHBV3 is presented in Table 2. After 275 days of biodegradation, 91 ± 9 % of the cellulose introduced was converted into CO<sub>2</sub>, whereas only 42 ± 2 % of the PHBV18 and 25 ± 3 % of the PHBV3 were mineralised. The experiment was stopped after 275 days as recommended by the standard since the stationary stage was reached for at least 15 days for all materials. The high conversion rate of cellulose confirms the good microbiological activity of the compost used (76 % of cellulose was converted in 90 days), reactors containing only the inoculum produced between 11 mg of CO<sub>2</sub> during the first 10 days, and the difference between replicates was lower than 20% (i.e. 11%), which validates the experiment.

As the final conversion rate of PHBV is lower than 90% at the end of the experiment, our results show that, at home composting, PHBV material cannot be considered biodegradable, according to ISO 14855. These results are surprising, especially for the commercial polymer PHBV3, which corresponds to the ENMAT Y1000P from Tianan, certified 100% biodegradable in home composting condition by TÜV Austria (OK HOME COMPOST Label). However, in the scientific literature, the home compostability of PHBV has been poorly investigated, with studies mainly focusing on industrial composting conditions using a higher incubation temperature (58 °C). This high temperature stimulates the inoculum enzymatic activity which can degrade a higher quantity of polymer in a shorter time than at a lower temperature ((Al Hosni et al., 2019). However, various degradation rates (24–100%) have also been observed in industrial composting conditions as presented in Table 2. This gap between studies can be explained by environmental factors linked to the inoculum used for the test, such as the use of green or biowaste compost, the microbial community composition, the pH and humidity, or the substrate on the inoculum (S/X) ratio. The material properties also have an impact, such as geometric characteristics (shape, thickness, size), or the methods used to produce the film (casting methods, extrusion, injection, and the parameters used). A higher variation could be observed in soil biodegradation experiments, where Boyandin et al. (2012) observed 7% of PHBV biodegradation in a tropical environment after 197 days, and David et al. (2019) obtained 98% of biodegradation in 80 days during soil biodegradation lab experiment (see SI, section B).

For this reason, it is often hard to compare the biodegradation rates of polymers between different studies, making it difficult to conclude the real biodegradation rate and kinetic of PHBV material in general. It suggests the presence of a worst- and a best-case scenario for the biodegradation of PHBV polymer in all environmental conditions. According to the commercial PHBV3 data, our experiment seems to represent a worst-case scenario where PHBV is not fully biodegradable, maybe because of the absence of efficient microbial PHBV degraders in the compost used.

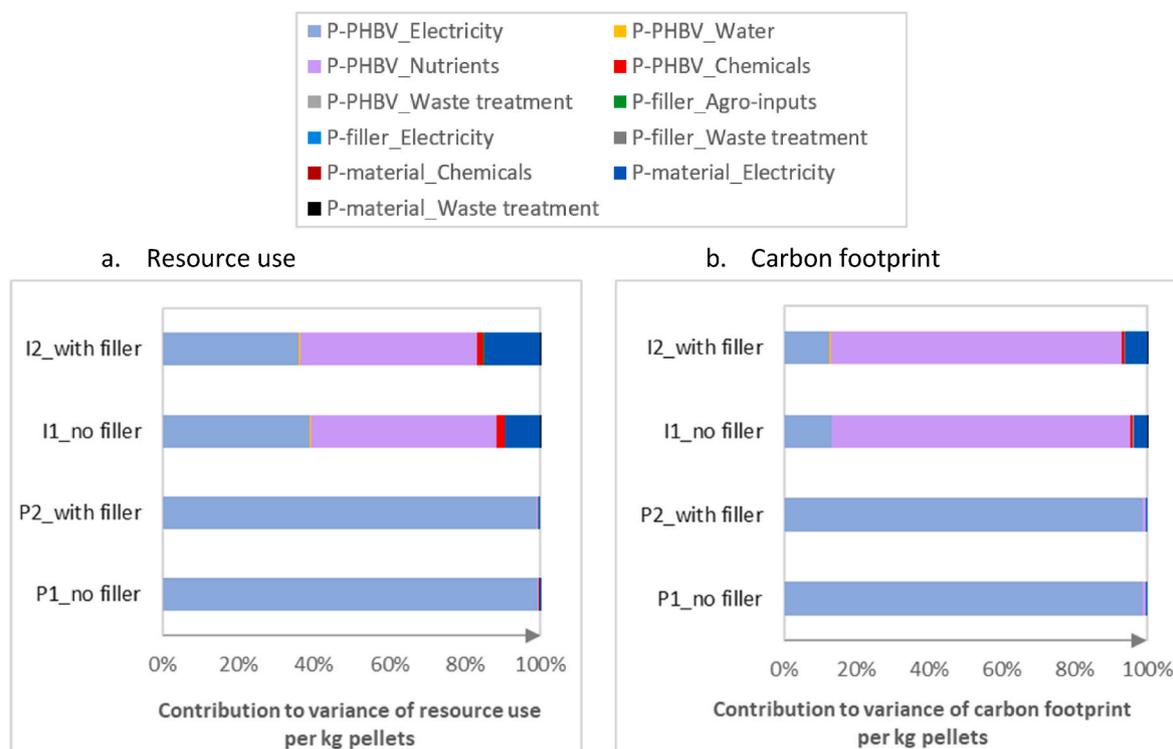


Fig. 3. The relative contribution of the input parameters to the variance of cradle-to-gate environmental impacts: (a) resource use and (b) carbon footprint per kg of PHBV pellets at pilot (P1 and P2) and industrial scale (I1 and I2) produced through the three processes: PHBV powder production (P-PHBV), filler production (P-filler) and material processing (P-material).

Table 2  
Overview of biodegradation results obtained in home and industrial compost for PHBV-based materials.

Condition	Standard	Material	HV content (%)	Shape of material, extruded or cast films	Test conditions	% max of biodegradation	Time to obtain max % (d)	Reference
Home compost	NF T51-800 (specifications); ISO 14855 (method)	PHBV18, fruit waste, MMCs	18	Extruded flat sheet	25 ± 5 °C; 50% humidity; mature compost; 1:6 w:w DM sub/inoculum	42 ± 2	275	This study
	NF T51-800 (specifications); ISO 14855 (method)	PHBV3, glucose syrup, pure culture	3	Injected	25 ± 5 °C; 50% humidity; mature compost; 1:6 w:w dry matter sub/inoculum	25 ± 3	275	This study
Industrial compost	ISO 14855-1:2005	PHBV with different HV contents supplied by Ningbo Tianan Biomaterials Co. Ltd. China and PHBV with 1–3% HV content,	3 20 40	Casting (in chloroform)	58 °C, 58 ± 2 °C; 50% humidity; 2-month mature compost; 1:6 w:w dry matter sub/inoculum	3% HV -> 90.5 20% HV -> 89.3 40% HV -> 80.2	110	Weng et al. (2011)
	ISO 14855-1	Natureplast, a square of 8 × 8 mm	3	Extruded, thermopressed	58 ± 2 °C; 50% humidity; mature green compost; 1:60 w:w dry matter sub/inoculum	100	70	Salomez et al. (2019)
	Inspired by ISO 14855	PHBV, Goodfellow, square of 10 × 10 mm	12	Casting (in chloroform)	58 ± 2 °C; 50% humidity; 3 months old, mushroom composting farm; 1:6 w:w dry matter sub/inoculum	63	100	Muniyasamy et al. (2019)
	ISO 14855-2	PHBV, Tianan, a square of 5 × 5 mm	2	Not mentioned	58 ± 2 °C; 50% humidity; Wastewater treatment plant; 1:6 w:w dry matter sub/inoculum	25	53	Zhao et al. (2019)

However, since the same environmental conditions are applied during a single study, results can be compared to one study. In the present work, we observed that the biodegradation of PHBV18 produced by MMC is twice higher than the one of the commercial PHBV3, even in the worst environmental conditions for biodegradation. This difference is probably due to the higher HV content in the agro-residues-based

polymer (PHBV18). Weng et al. (2011) also observed that a higher ratio of HV (between 3, 20 and 40%) in the PHBV increases the biodegradability of the material, reasoned by the fact that the higher the 3HV content, the lower the crystallinity of the polymer. A high crystallinity is sometimes claimed as an inhibitor of PHBV’s biodegradation (Abbasi et al., 2022; Brdlík et al., 2022). However, there is no clear consensus in

the literature on such impact. For instance, Arcos-Hernandez et al. (2012) observed that increasing the HV amount (from 12 to 72% mol) tended to decrease the biodegradation after 15 weeks but the trend is not linear.

It is important also to highlight the influence of the medium and the ratio of substrate/inoculum. Indeed, in the experiment made in the framework of this study, a high concentration of the material in the substrate is used 1:6 (S/X). This ratio seems to be the worst case and a lot of other studies (Table 2) used different ratios where the material is much more dilute into the substrate. This has probably a predominant impact that deserves to be elucidated in further research.

### 3.2.2. Definition of possible biodegradation patterns for PHBV in composting

The previous analysis highlighted well the variability observed in PHBV biodegradation results in home and industrial composting. Even if standards are used to conduct the tests, slight variations in test conditions and in the material itself from one trial to another make it difficult to conclude the real biodegradation rate of PHBV and the spread of this factor in those different conditions. Because it seems difficult to reach a consensus on the average biodegradation rate, it was decided in the following to consider the best and worst cases of biodegradation for each condition, derived from the present study and literature analysis (Table 2). The resulting compilation of biodegradation patterns is provided in Table 3.

### 3.2.3. Environmental impact of PHBV material at end-of-life treatment

As presented in Table 3, the biodegradation rate during home and industrial composting can vary, depending on the environmental conditions. A scenario analysis was therefore performed (Fig. 4) to identify the effect of (i) the biodegradation rates (minimum, average and maximum) of PHBV material, and (ii) the two different concepts: excluding or including the credit of PHBV-based compost from its substitution for peat (described in SI, section A), on the environmental impact of the two composting options.

Firstly, Fig. 4a indicates that excluding the credit of compost leads to similar net resource and carbon footprints for both composting options regardless of the variety of PHBV biodegradation rates. Secondly, including the credit from substituting PHBV-based compost for peat as a growing medium for plants can significantly affect the net environmental impact of either home or industrial composting, depending on the PHBV biodegradation rates (Fig. 4b). When PHBV degrades less efficiently to biogenic CO<sub>2</sub> (assumed burden-free), more compost is produced and can be substituted for more peat, resulting in a higher environmental credit (or benefit) from this substitution. This finding is in line with the circularity concept that aims to minimise waste and keep products, materials and resources within the economy wherever possible through reuse, repair, remanufacture or recycling, thereby creating further value (European Commission, 2021b). However, the above-mentioned benefit needs to be considered carefully under specific conditions, i.e. in a well-managed dedicated EOL treatment system (e.g. industrial composting), and for short-lived (including single-use) products (e.g. food packaging). Contrarily, from the environmental protection perspective, increasing the biodegradation rate of PHBV could be favourable in the case of mismanaged waste (e.g. marine littering), associated with the long-term benefit for a complete biodegradation of

**Table 3**

Biodegradation patterns of PHBV material used in LCA for home and industrial composting: compilation of min-max kinetic rates from own study and the literature.

EOL condition	Min biodegradation rate (% - time)	Max biodegradation rate (% - time)
Home composting	25% in 275 days	42% in 275 days
Industrial composting	25% in 53 days	89% in 110 days

PHBV in nature.

Of the four analysed EOL options, mechanical recycling is the most favourable, followed by incineration, while industrial composting is the least (see Fig. 5), mainly explained by a remarkably higher credit from substituting the recyclates for the virgin pellets. In other words, recycling maintains a higher value of resources (in the form of recyclates) than incineration or composting (in the form of recovered energy or compost, respectively). This is aligned with the circular economy context (European Commission, 2021b). However, one should keep in mind that only a life cycle or one loop is considered here. Though this finding came from a hypothetical inferred model, it is still interesting when highlighting the possibility that, incineration is supposed to be more environmentally beneficial than composting for BBPs. It is explained by the fact that the PHBV-based compost consists of no main nutrients (NPK) or only in traces and hence cannot substitute for mineral fertilizers like compost originated from conventional biowaste while incinerating PHBV can recover energy which can substitute for energy from the grid at the 1:1 ratio and hence gains a significant credit. This is valid not only for the midpoint impacts: resource use and carbon footprint but also for the endpoint aggregated single score (see SI, section B). The work of Rossi et al. (2015) showed a similar conclusion for PLA and TPS but was limited to only midpoint-level assessment.

### 3.3. Comparison with benchmark materials

The environmental performance of PHBV is compared with the following polymers: PHB, PLA, TPS, LDPE and PP in the two stages: pellet production and EOL scenarios compatible with their characteristics. Fig. 6 shows the resource use and carbon footprint, while the single score is presented in SI (Fig. A2). The PHBV pellets without filler with extrapolation at the industrial scale (scenario I1) were selected for this comparison because, first, the production of other polymers was modelled at the industrial scale, and second, that scenario could be more common and hence representative due to the avoidance of technical constraints from the addition of fillers.

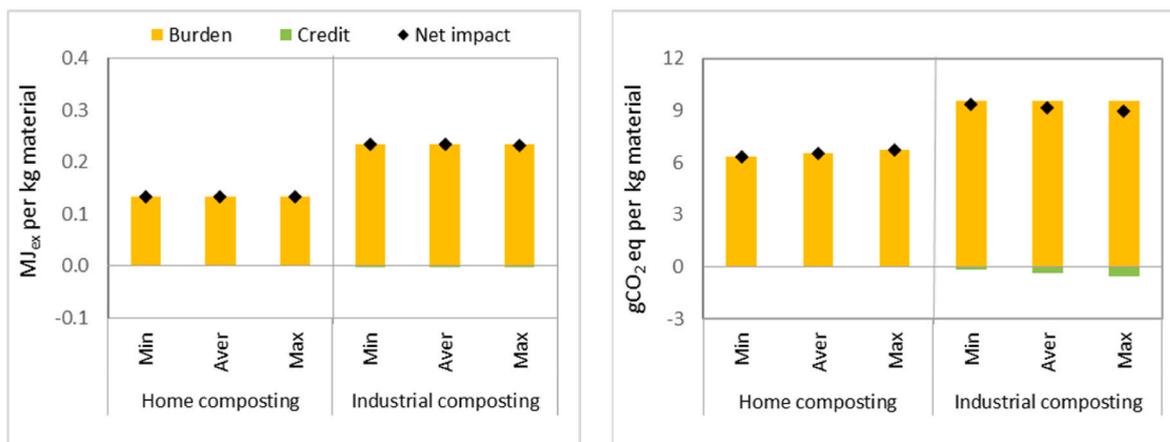
#### 3.3.1. Comparison: pellet production

Fig. 6a (left) first shows that PHBV pellets extract considerably fewer natural resources compared to conventional fossil-based ones (LDPE: 55% and PP: 46%). Such a result can be seen for other studied BBPs, explained mainly by a high fossil resource use of LDPE and PP pellets. Next, amongst the studied BBPs, PHBV pellets obtain a resource use similar to PHB and TPS pellets, and lower than PLA pellets (26%). The better performance of PHBV compared to PLA is mainly explained by the fact that the use of agro-residues (fruit residues for PHBV) instead of agro-products (maize for PLA) requires fewer land resources. The second reason is that the production of PLA pellets is more energy-intensive. This result indicates that both the feedstock source and the technology specifications drive the environmental performance of BBP pellets, especially PHBV and PHB produced through emerging technologies. Rueda et al. (2023) reached a similar conclusion about the technology maturity by comparing their results with others for the PHB material delivered after extraction but not for the PHB pellets.

Additionally, it is noted that the production of these BBP pellets extracted mainly fossil fuel resources for all four studied BBPs, water resources (except for TPS), and land resources for PLA and TPS. However, the reasons for this are different: electricity and nutrients for PHBV, cleaning water and chemicals (chloroform and methanol) for PHB, energy (electricity, heat) and feedstock (maize) for PLA, and energy (natural gas, heat) and feedstock (maize, naphtha) for TPS.

Regarding the carbon footprint (Fig. 6a, right), PHBV pellets have an impact similar to fossil-based LDPE and PP pellets, but lower than PHB (32%) and PLA (41%), explained by a high amount of chemicals or electricity used in PHB and PLA production, respectively. One should keep in mind that the energy production was modelled here using the Europe mix (RER) production processes available in ecoinvent v3.8. The

(a) Excluding the credit of composted material



(b) Including the credit of composted material

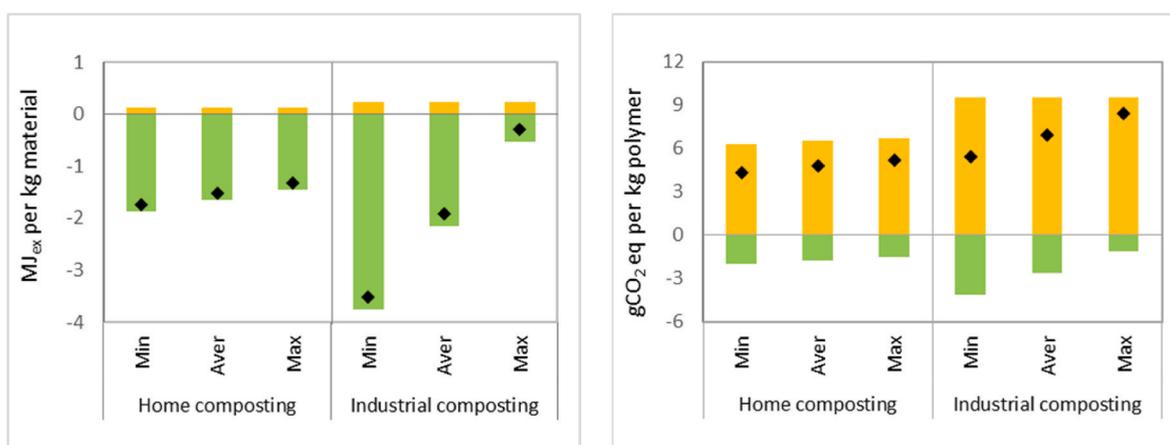


Fig. 4. Cradle-to-gate environmental impacts: resource use (left) and carbon footprint (right) of home and industrial composting per kg of PHBV films in the two scenarios: (a) excluding and (b) including the credit of composted material from its substitution for peat as a growing medium for plants.

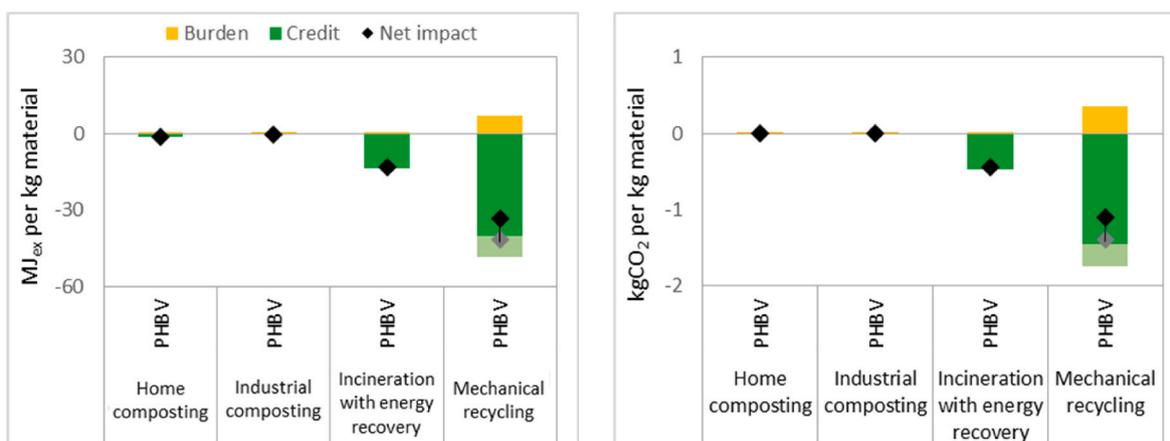


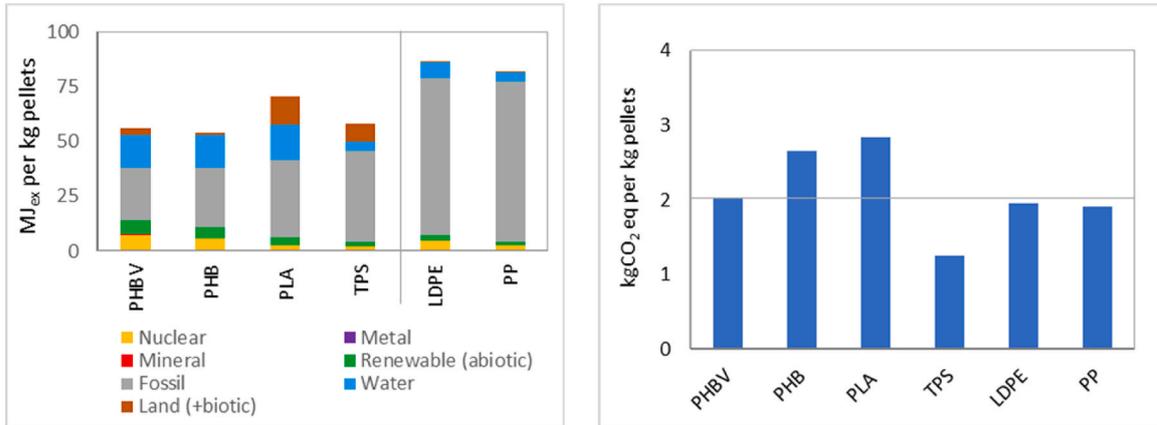
Fig. 5. Cradle-to-gate environmental impacts: resource use (left) and carbon footprint (right) of PHBV at the EOL treatment. For recycling, the black upwardly directed arrows show the corresponding decrease in the credit of recyclates (and the net impact) due to the decrease in substitutability of recyclates (from 1:1 to 0.83:1) for virgin materials for the same type of polymers.

flows aforementioned contribute the most to not only the resource but also the carbon footprint of the studied pellets.

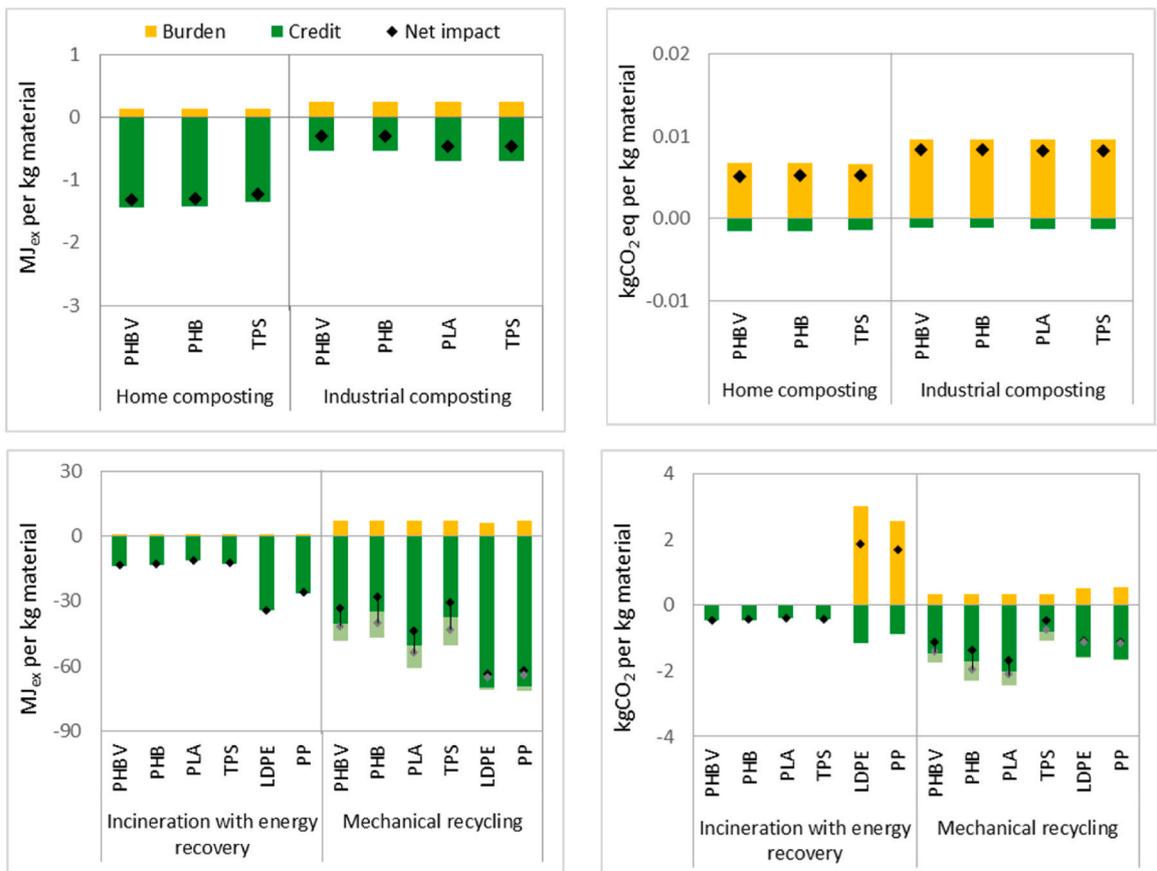
Overall, this comparison indicates that the PHBV pellets, though still under development, might be considered environmentally friendly to some extent compared to the potential alternatives, including both BBPs

and fossil-based plastics. Walker and Rothman (2020) pointed out that, compared to the emerging technologies (e.g. the production of PHBV or PHB pellets), the mature ones (e.g. the production of fossil-based pellets) have been highly optimized and hence have an improved efficiency regarding environmental sustainability. Consequently, there is still

a) Pellet production



(b) EOL treatment



**Fig. 6.** Environmental impacts: resource use (left) and carbon footprint (right) of PHBV compared to benchmark materials in the two stages: pellet production (a) and end-of-life treatment (b). For recycling, the black upwardly directed arrows show the corresponding decrease in the credit of recyclates (and the net impact) due to the decrease in substitutability of recyclates for virgin materials for the same type of polymers. Note that the decrease in the case of LDPE and PP is too small to visualize with an arrow, and the resource use is the sum of all resource categories listed in 5a (left).

room for improvement in PHBV pellet production as discussed in section 3.1.

3.3.2. Comparison: end-of-life treatment

Regarding the EOL treatment (Fig. 6b), as mentioned in section 3.2, there are two perspectives: including or excluding the credit from replacing peat with compost. For this comparison, the former was considered for not only PHBV but also the selected BBPs, as the composted material can still add organic carbon to the soil and hence improve the physical structures and functionality of the soil.

Focusing on resource use (Fig. 6b, left), either home or industrial composting of all analysed BBPs is environmentally beneficial with a similar net impact, mainly explained by their rather similar biodegradation rate in these conditions (presented in SI, section A). As PLA can degrade completely only through industrial composting within 4–6 weeks but is incompatible with home composting (Hussain et al., 2023), this material is not considered for the home composting scenario. For incineration and mechanical recycling, PHBV, like other studied BBPs, is less resource-use beneficial compared to fossil-based materials (LDPE and PP), mainly explained by a lower credit obtained from energy

recovery in incineration and recyclates in mechanical recycling. The latter is reasoned by a higher environmental burden of LDPE and PP and their higher substitutability for virgin materials (0.98 and 0.97, respectively) while the former came from the association of the polymer formulas (the key elemental composition: C, H, O) with their (lower and upper) heating values and hence their capacity in energy recovery (described in sections 2.2-2.3).

Fig. 6b (left) also shows that the incineration of the studied BBPs obtains a similar resource-use benefit while the recycling of PLA is more environmentally beneficial than that of PHBV, PHB, and TPS, which is mainly reasoned by the assumption that a particular recycled polymer can replace only its virgin material but no other virgin or recycled polymers and PLA virgin pellets extract more resource use than the other studied BBPs (Fig. 6a, left). It is additionally noted that the environmental impact, regarding resource use, carbon footprint and single score (see SI, section B), of the mechanical recycling of the studied BBPs, LDPE and PP is predominant by the credit from substituting the recyclates for alternatives. Consequently, a specific polymer of which the pellet production has a high environmental net impact will obtain a high credit (or benefit) correspondingly if it can be recycled and its recyclates can sufficiently or fully replace its virgin pellets.

This reason also explains the carbon footprint results indicating that the mechanical recycling of all six analysed polymers is environmentally beneficial (Fig. 6b, right): the impact of PHBV is similar to LDPE and PP, but is less than the EOL treatment of PHB and PLA while TPS is identified as the worst. Next, a similar net impact on carbon footprint is noted for both composting options. Incineration emits biogenic CO<sub>2</sub> in the case of BBPs but fossil CO<sub>2</sub> in the case of LDPE and PP; therefore, PHBV is environmentally beneficial similar to other studied BBPs while LDPE and PP induce a significant net impact on carbon footprint.

#### 4. Further insights and implications

For a broader view, the methodological choices, including the definition of functional unit, LCI and LCIA aspects relevant to this work, are discussed more in-depth, together with insights and implications of the work.

##### 4.1. Key perspectives on scope definition

For comparative LCA, defining the scope, especially the functional unit could be of importance to deliver scientific-based and reliable outcomes. When focusing on plastic materials, the selected FU (i.e. 1 kg of pellets or 1 kg of films) represents a certain mass of material, which is the most commonly used approach. However, to deliver a comparative LCA of PHBV with other plastic materials (e.g. PLA, PE, etc.), the FU should also reflect the performance of the material in a specific end product or the main relevant properties of the material for a certain application area (Furberg et al., 2021). Consequently, the cradle-to-gate environmental impact of PHBV pellets was also presented per volume (L) of materials in comparison with other plastic pellets in SI, section C. This FU considers not only the mass-based but also the density-based performance of materials.

Compared to mass-based FU, volume-based FU shows the following differences, mainly related to LDPE and PP. Compared to PHBV, the resource use per volume of these two fossil-based polymers is still higher but with a considerable decrease in the difference (17 and 9% instead of 55 and 46%, respectively). Similarly, their carbon footprint per volume decreases and hence is lower than that of PHBV (26 and 29%, respectively). The reason for this is that PHBV has a higher density than LDPE and PP (1221, 923, and 908 kg/m<sup>3</sup>, respectively). Other studied BBPs are also denser than the two polyolefins (LDPE and PP). This indicates that, for packaging having the same volume (same thickness and dimensions), more material is needed (in mass) for denser film (BBPs) than polyolefins. Hence, an LCA including both volume- and mass-based FU can give added value, especially when the entire life cycle of packaging

for a specific application (e.g. food) is analysed as the volume of packaging may drive the energy use at storage while its mass affects the environmental burden during transport (Boone et al., 2023).

Next to that, the scope of a sustainability assessment focusing on emerging technologies should include not only environmental but also economic aspects. Currently, several approaches have been developed for this purpose, e.g. Cost-Benefit Analysis (CBA), Life Cycle Costing (LCC), and Whole Life Cycle Costing (WLCC) (Padilla-Rivera et al., 2023); however, the need for primary data could be one of the challenges. Considering the essence of economic assessment for emerging technologies, the European Commission included it in the scope of many opening calls within the Horizon Work Programme 2023–2024 (European Commission, 2023).

##### 4.2. Key perspectives on life cycle inventory aspects

Focusing on the LCI model for the PHBV pellet production herein reported, it can be seen that the environmental impact of PHBV pellets was estimated using the primary data mainly measured at the pilot-scale production, except the electricity use in extraction came from Rueda et al. (2023), which was then scaled up for industrial application. Energy use in purification was excluded. The upscaling model was established based not only on the expert judgement and data of similar technologies reported in the literature but also on the mass-energy balances and the potential differences between the two production scales. The latter refers to the consideration of e.g. effluent recycling, and cooling technology at the industrial scale (see section 2.1). On the contrary, the estimates reported in the literature, by Ghysels et al. (2018); Tabone et al. (2010); Mozumder et al. (2015), came from the stoichiometric equation or laboratory scale-up or unspecified data source. While those authors focused on PHB produced using pure culture and crop-based feedstock, PHBV produced from agro-residues and using MMCs is the target material of this study. Next to that, there is an important gap in the LCA that the contribution of additives (e.g. boron nitrite) to the overall environmental impact was commonly excluded in the plastic LCA studies due to the quantities and specific substances used as additives being commercially sensitive and widely neglected information (Bishop et al., 2021) while being considered here.

The results presented in section 3.1 were calculated in the case that fruit residues were assumed free-burden at both pilot and industrial production. It means that the impact of storage and transport related to the feedstock was excluded. However, one should keep in mind that, in practice, i.e. at the industrial scale, the need for a high quantity of fruit residues, an easily-fermented feedstock, would increase the environmental impact of PHBV pellets due to extra energy consumption for its transport or storage. Moreover, here agro-residues included fruit residues as the feedstock of PHBV production, and wheat straw as filler. The latter considered a by-product with specific industrial applications and having an economic value, shared the environmental burden with the main product (wheat), using mass allocation, while the former was assumed burden-free as it is currently still a waste to be treated. This waste was used as a feedstock to produce PHBV powder through an innovative technology only recently tested at the pilot scale. In the literature, defining a material as a main, co- or by-product, or residue (waste) is not straightforward (Olofsson and Börjesson, 2018), and hence whether or not residues can be assumed burden-free in LCA becomes a matter for debate. In the context of the circular economy, if waste can be valorised or recycled, Olofsson and Börjesson (2018) suggested, via a review of 31 LCA studies, that the potential value of residues needs to be addressed by analysing the upstream-related impacts to avoid misguided decisions on methodology choices, which can drive the LCA results. Future LCA research on PHBV produced from agro-residues should consider this aspect.

Regarding the EOL stage, though there are other EOL options (e.g. landfill, incineration without energy recovery) existing in Europe, the four selected EOL options have been recommended according to the EU

Waste Framework Directive. These scenarios were also suggested for BBPs in the review of Gioia et al. (2021). Moreover, it should be kept in mind that the PHBV material tested in this study for biodegradability did not contain fillers. It can be anticipated that the addition of natural fillers can pose both positive and negative effects. More specifically, the fillers can potentially improve the biodegradability of PHBV (Meereboer et al., 2021) but can also make the composites more brittle through recycling (Lagazzo et al., 2019). Consequently, it requires further work on testing the biodegradability of PHBV composites in different conditions and investigating the effect of natural fillers on their performance in terms of technical properties and environmental sustainability.

Finally, Morris and Hicks (2022) presented that challenges exist in predicting consumer behaviour at disposal and incorporating it into LCA. This aspect was considered to some extent here through modelling the potential EOL options for PHBV associated with the existing disposal routes of household waste (see section 2.2). However, the subsequent effects of improper sorting or disposing of PHBV packaging in nature (e.g. marine) were not covered here but were investigated in Boone et al. (2023). These authors presented that PHBV has lifetime costs on marine ecosystem services about seven times lower than PP despite a higher mass of PHBV for a similar packaging volume.

#### 4.3. Key perspectives on life cycle impact assessment methodological aspects

As PHBV material was the focus of this work, the implemented approach to account for biogenic carbon becomes important. This aspect is currently considered in various plastic LCA studies through one of the two following approaches: (temporary) carbon storage and carbon neutrality, without a clear motivation. In the carbon storage approach, the sequestered biogenic carbon is assumed to be temporarily captured in the bio-based material and hence has a credit at the production stage but a burden at the EOL stage. When considering the second approach, the biogenic carbon gas is assumed to be released back into the natural environment without any climate-forcing effect (i.e. carbon neutral) and has neither credit nor burden (Pawelzik et al., 2013). Through a review of 44 LCA plastic studies, Bishop et al. (2021) concluded that one can apply the carbon storage approach but needs to carefully do that under explicitly accounting for the EOL stage of carbon release. Otherwise, the carbon neutrality approach would be recommended. The European Commission (2022) emphasized similarly: only long-lifetime bio-based plastic products, which are not incinerated at the EOL, can have beneficial effects from carbon storage, while the atmospheric carbon, initially taken up by short-lived products (food, beverage or single-use packaging predominantly made of bio-based plastics), is quickly released back. Following the guidelines of the European Commission, the carbon neutrality approach was implemented in this work to analyse the short-term environmental impact of PHBV production and EOL in only a life cycle.

However, as this study compares PHBV with not only other BBPs but also LDPE and PP, which are very persistent and pervasive particulate matters, regarding the environmental impact, it could be crucial to further investigate the long-term fates (e.g. mismanagement, leakage, marine littering) of these plastics. Currently, there is still a lack of advanced LCA approaches to address this topic (Gontard et al., 2022). Specifically for PHBV, marine littering was investigated by Boone et al. (2023) but only at the LCI level (through estimating the costs on marine ecosystem services).

Lastly, to adequately represent the environmental sustainability of plastics, the review of Bishop et al. (2021) recommended the implementation of a comprehensive LCIA methodology such as EF or at least selecting impact categories that capture priority environmental challenges. In this work, the CEENE and EF methodologies were adopted to address two of the main impacts of plastics on the environment: resource use and carbon footprint, in addition to the aggregated single score.

## 5. Conclusions

PHBV is considered a promising future BBP material; however, its production has only been tested at lab and pilot scale and the assessment of its environmental sustainability is still a gap in the literature. Therefore, this study was conducted using the LCA methodology to address the three following concerns: (i) what would be the potential benefit of scaling up the PHBV pellet production, which process or flows are the main contributors to the impact of PHBV pellets, (ii) which EOL scenario is favourable for PHBV in future, and (iii) how environmentally sustainable is PHBV versus its alternative polymers at the two stages: pellet production and EOL treatment.

Firstly, the analysis showed a substantial environmental benefit, i.e. resource use, and carbon footprint, achieved from a 50-time production upscaling through the implementation of scaling factors for materials and energy use and mass flow analysis for emissions and effluents. Some possibilities for optimising the processes at the industrial scale were considered in this work (e.g. reusing effluent for diluting feedstock and replacing dry ice with a closed-loop water system for cooling). However, it should be kept in mind that the bioconversion of agro-food residues into PHBV using mixed microbial cultures (MMC) is still at the start of the maturity/optimisation curve, the estimated benefit might be uncertain to some extent, including the potential of room for improvement, such as a reduction in the use of electricity and nutrients (including micro-nutrients) mainly in microbial synthesis. Agro-residues can be optionally added as natural filler, which weighs up the pros and cons in different life cycle stages of PHBV material (e.g. pellet production, packaging processing and EOL).

Secondly, it was concluded that mechanical recycling is the most favourable EOL option for PHBV. However, one should keep in mind that this finding comes from the analysis considering the short-term impact of the EOL stage after one cycle of use. The technical challenges related to the mechanical recycling of PHBV were also not considered. Furthermore, it became clear that the environmental impact of the two composting options depends largely on considering whether or not the credit of compost and the biodegradation rates of PHBV films in these conditions.

Thirdly, PHBV material shows a good environmental performance both at pellet production and EOL stages, comparable to or even to some extent better than other studied BBPs (PHB, PLA, TPS) and fossil-based plastics (LDPE, PP). Consistent methodological choices were made for this comparative LCA. More importantly, a key message was also highlighted: feedstock sources and technology specifications, in addition to methodological choices, can affect the answer to the question of which polymer type is more environmentally sustainable. Transparency in describing the studied technologies, the LCI model and the LCIA methodology should be most important when comparing the PHBV investigated in this work with the PHBV produced from other technologies, and also PHBV versus other polymer types in terms of environmental sustainability. Also, further research is recommended on not only the technical and environmental aspects discussed above but also on economic aspects throughout the entire life cycle of PHBV products, in addition to attention on the long-term fates of plastics.

#### CRedit authorship contribution statement

**Trang T. Nhu:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Validation, Visualization, Writing – original draft, Writing – review & editing. **Lieselot Boone:** Conceptualization, Data curation, Methodology, Validation, Writing – review & editing. **Valérie Guillard:** Investigation, Methodology, Validation, Writing – review & editing. **Lucile Chatellard:** Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft. **Maria Reis:** Writing – review & editing, Validation. **Mariana Matos:** Data curation, Validation, Writing – review & editing. **Jo Dewulf:** Conceptualization, Methodology, Supervision,

Validation, 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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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvman.2024.120522>.

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