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Inclusion of multiple climate tipping as a new impact category in life cycle assessment of polyhydroxyalkanoate (PHA)-based plastics

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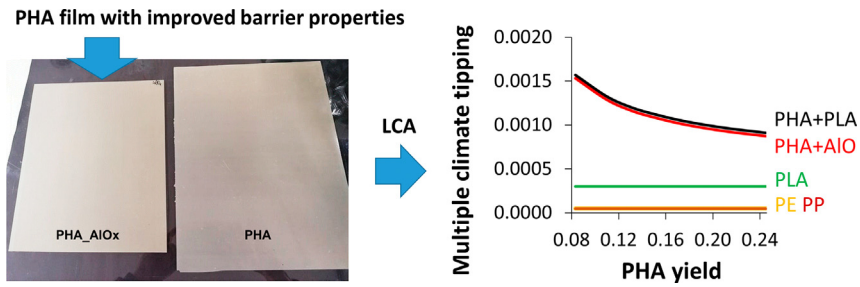
HIGHLIGHTS

- LCA applied to PHA with improved barrier properties.
- Inclusion of new impact category, the multiple climate tipping.
- PHA films with high biodegradability perform best.
- Multiple climate tipping is a relevant impact category for LCA of PHA.

GRAPHICAL ABSTRACT



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ABSTRACT

The merits of temporary carbon storage are often debated for bio-based and biodegradable plastics. We employed life cycle assessment (LCA) to assess environmental performance of polyhydroxyalkanoate (PHA)-based plastics, considering multiple climate tipping as a new life cycle impact category. It accounts for the contribution of GHG emissions to trigger climate tipping points in the Earth system, considering in total 13 tipping elements that could pass a tipping point with increasing warming. The PHA was either laminated with poly(lactic acid), or metallized with aluminum or aluminum oxides to lower permeability of the resulting plastics toward oxygen, water vapor and aromas. The assessments were made accounting for potential differences in kinetics of evolution of greenhouse gases (CO_2, CH_4) from bioplastic degradation in the end-of-life. Results show that: (1) PHA films with high biodegradability perform best in relation to the climate tipping, but are not necessarily the best in relation to radiative forcing increase or global temperature change; (2) sugar beet molasses used as feedstock is an environmental hot spot, contributing significantly to a wide range of environmental problems; (3) increasing PHA production scale from pilot to full commercial scale increases environmental impacts, mainly due to decreasing PHA yield; and (4) further process optimization is necessary for the PHA-based plastics to become attractive alternatives to fossil-based plastics. Our study suggests that multiple climate tipping is a relevant impact category for LCA of biodegradable bioplastics.

1. Introduction

Bioplastics are a diverse group of materials which have been in the focus of research as alternative to conventional plastics (Spierling et al., 2018; García et al., 2019; Kookos et al., 2019; Pavan et al., 2019; Rameshkumar et al., 2020). Bioplastics consist of three sub-categories and can either be (1) "fossil-based and biodegradable", (2) "bio-based

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and biodegradable" or (3) "bio-based and non-biodegradable". The last two categories can also be combined with the term bio-based plastics (Endres and Siebert-Raths, 2011). The market share of bioplastics is predicted to continue to grow within the next years to 2.8 Mio. tonnes in 2025, an increase of 35% compared to the production capacities in 2020 (European Bioplastics, 2020b).

This study focuses on production of polyhydroxyalkanoate (PHA) from sugar beet molasses. PHA is a bio-based and biodegradable polyester which can be produced by bacterial fermentation (Chen et al., 2020; Moretto et al., 2020). The PHAs are produced as intracellular high-molecular inclusion bodies, which have a role as carbon- and energy storage compounds within the bacteria. The selection of different microbial production strains as well as adaptions of the bioprocess allows the composition of different PHA types (homo-, co-, ter-, and quad-polyesters), resulting in up to 150 different PHA structures which have been identified so far. Based on their carbon atoms used for the monomeric unit PHAs can be differentiated in two main groups: (1) short-chain-length PHAs with 3–5 carbon atoms and (2) medium-chain-length PHAs with 6–14 carbon atoms. The most well-known and common PHA type is poly-β-hydroxybutyrate (PHB) (Koller, 2017; Kourmentza et al., 2017; Troschl et al., 2018). Additionally, for the production of PHA, a wide range of feedstocks can be utilized. E.g. renewable materials like sugar, industrial waste and by-product streams, as well as CO_2, which can be utilized by cyanobacteria types (Koller, 2017; Kookos et al., 2019; Moretto et al., 2020; Wongsirichot et al., 2020). This makes PHA a versatile plastic type within the bioplastics and bio-based plastics.

A number of international patents about plastic materials based on PHA have been obtained (Elvers et al., 2016 and references therein). Yet, global production of PHA is relatively low (25,320 tons in year 2019), accounting for only 1.2% of the bioplastics market (Rameshkumar et al., 2020). The reason for the low market share of PHAs is mainly due to high production costs (García et al., 2019; Kookos et al., 2019; Pavan et al., 2019), that are estimated to be 5–10 times higher than the cost of traditional polymers (Kookos et al., 2019). The latest market data for PHA predicts an increase to 11.5% of the bioplastics market till 2025 (European Bioplastics, 2020a). Currently PHA is manufactured worldwide at both pilot and industrial scale. Manufacturers are based in Canada, Germany, Italy, China, USA, Japan as well as Malaysia. On these scales mainly first-generation feedstock like sugar (e.g. from sugar beet) as well as vegetable oil (e.g. canola oil or palm oil) is dominant (Kourmentza et al., 2017). Molasses, a co-product of sugar beet, is often considered as feedstock for PHA production (Baei et al., 2009; Keunun et al., 2018; Kiran Purama et al., 2018; Remor Dalsasso et al., 2019). Molasses contains about 50% of the disaccharide sucrose and is commonly used as an energy supplement to livestock feed. PHA exhibits physical and mechanical properties similar to those of conventional plastics, such as polyethylene (PE) and polypropylene (PP) (Kookos et al., 2019).

One potential application of PHA is as food packaging material (Khosravi-Darani and Bucci, 2015). For example, bioplastic is considered for packaging of high quality bakery products, replacing fossil based polypropylene. Yet, high permeability toward water, oxygen and aromas makes PHA a rather poor packaging material (Kassavetis et al., 2012). It is thus necessary to improve the barrier properties to lower permeability if PHA is to be used in contact with food (Struller et al., 2014). This can be done by lamination with (poly)lactic acid (PLA) or metallization with aluminum (Al) or aluminum oxides (AlOx) (Kassavetis et al., 2012). Until now, nothing was known about sustainability implications of lamination or metallization of PHA, which improve barrier properties but may impair biodegradability in their end-of-life. Environmental assessments of PHA production from molasses generally focused on the fermentation and PHA recovery steps, without considering other important process in the PHA-based plastic film life cycle like post-treatment and end-of-life (Leong et al., 2017; Kookos et al., 2019).

The purpose of this paper was to assess the environmental performance of the whole value chain of PHA-based plastics with improved barrier properties. We considered lamination using PLA or metallization using Al or AlOx as two viable surface treatment options. The environmental performance was assessed using life cycle assessment (LCA). To provide additional insights to the metrics of climate change recommended in the EU Commission's ILCD Handbook (ISO, 2006; EC-JRC, 2010) (the global warming potential, GWP_{100}, and the global temperature change potential, GTP_{100}), we present the inclusion of a new life cycle impact category, the multiple climate tipping (Fabbri et al., 2021). It accounts for the contribution of GHG emissions to trigger multiple climate tipping points in the earth system (up to 13 tipping points).

2. Methods

2.1. Scenarios

Molasses, a by-product from sugar beet refinery rich in the carbohydrate sucrose, was chosen as a feedstock and the Gram-negative bacterium *Ralstonia eutropha* as fermenting microbe because they were found promising for full scale applications (Baei et al., 2009; BioBarr, 2019; Remor Dalsasso et al., 2019). Both pilot and large scale were modelled and compared. They differ in capacity (about 100 and 9000 tones PHA recovered per year in the pilot and large scale plants, respectively), and in means of how feedstock is collected, pre-treated, fermented, recovered and purified. In addition to testing the influence of plant scale, we considered differences in (i) geographic location of the PHA plant, (ii) conventional use of molasses, (iii) composition of PHA-based films and other packaging materials, (iv) yield of PHA in the fermentation process, (v) thickness of the PHA layer, and (vi) fate of the improved bioplastic in its end-of-life. Production of poly-β-hydroxybutyrate (PHB) was modelled.

Italy and Germany were chosen as two representatives of countries where production of PHA is currently conducted. Differences in electricity grid mixes and waste management systems between the countries were considered. The waste management systems were modelled according to country-specific rates for recycling, incineration and landfilling of plastic packaging (Eurostat, 2017). Bioplastic packaging is currently not recyclable. Thus, it was assumed that the remaining fraction was treated proportionally to the treatment of non-recovered plastic waste (that is, 50% landfilled and 50% incinerated in Italy and 100% incinerated in Germany). The conventional use of molasses is as animal feed, however, it can be also used for ethanol production (Takriti et al., 2017) and these two scenarios were also explored. We investigated optimization potentials for PHA-based plastics made from molasses, which lie in selection of the material for lowering permeability of the PHA (either PLA or Al or AlOx), optimizing PHA production yield and reducing thickness of the plastic layers. The PHA-based plastics were compared with fossil-based alternatives, namely PP and PE. Comparisons were also made with PLA. Merits of temporary carbon storage are often debated for bioplastics, and the end-of-life stage of the plastics life cycle is the only stage where temporal carbon storage can occur. PHA is generally considered as readily biodegradable (the actual duration of degradation depends on product dimensions as well as environmental parameters like temperature), but it is currently unknown how improving barrier properties influences biodegradability during landfilling (Emadian et al., 2017; Meereboer et al., 2020). Thus, fast degradation was assumed in the baseline scenario. Delayed biodegradation may be caused by differences in availability of water and oxygen during landfilling (Meereboer et al., 2020). Moreover, combining PHA with PLA has shown to reduce biodegradability of PHA (Meereboer et al., 2020). To explore sensitivities toward mineralization rates, different biodegradation rates and extents of lag phases were explored for the landfilling scenarios. We conservatively assumed that no PHA plastic is lost to the environment owing to generally sound

management of plastic waste in Europe (Ryberg et al., 2019). In total, 53 scenarios were considered (refer to Table S3S3, Section S2 of SI for an overview of all scenarios).

2.2. Literature review

Parameters and installations for fermentation, recovery and purification of PHA at pilot and large scales were modelled based on parameters retrieved from scientific literature, identified through a systematic literature review (see Section S1 of the SI for further details). The review encompassed studies focusing on both technical and environmental aspects of PHA production. It was carried out using Scopus in March 2020, applying a set of keyword strings. We retrieved those studies, which: (i) report parameters relevant for the PHA production (feedstock type and its water content, producing microorganism, plant scale and capacity), (ii) are either at pilot (as defined by the study itself or between 10 and 1000 L fermenter volume) or large (above 1000 L) scales, and (iii) either report PHA yield ($kg_{PHA}/kg_{feedstock}$), or sufficient data to estimate the yield. In total, 25 studies were retrieved. We found four studies which use disaccharides, and report data on resource consumptions (e.g. electricity, water and chemicals) (see Table S2 in Section S1). These four studies were used for extraction of parameters and bills of materials needed to model PHA production installations in our LCA.

2.3. Overview of PHA installations

The pilot and large scale systems differ in how feedstock collection, pre-treatment and fermentation, recovery and purification are carried out (see Section S2 of the SI, Fig. S1 for an overview of their installations). The remaining steps are the same and represent large-scale systems. The feedstock is transported by truck at pilot scale, whereas at large scale the feedstock is transported using pipes. At both scales, the feedstock is sterilized by steam and the sterilized feedstock is cooled down using a heat exchanger. At pilot scale, the sterilized feedstock is fermented in one 10-m^3 reactor for 80 h. At large scale, three 102-m^3 reactors for 54 h are used. Electricity input for aeration and agitation are different for the two scales. Fermentation yield is higher at pilot than at large scale (0.360 and 0.268 $kg_{PHA}/kg_{substrate}$, respectively). At both scales, PHA is extracted from fermenting cells and purified in a sequence of steps, involving centrifugation and spray drying, but electricity inputs are higher and consumption of materials generally lower at the pilot scale. Hydrochloride is used for extraction at pilot scale, while hydrogen peroxide and enzymes are used at large scale. The obtained PHA powder is compounded and blended with additives (plasticize, nucleating agent, stabilizer and reinforcing filler) before extruding it into PHA pellets. These pellets are subsequently extruded into a PHA film. This film is either laminated with a layer of PLA, or metallized using aluminum. The aluminum layer can be optionally oxidized to aluminum oxide (AlOx) to make the resulting film transparent. Details of the parameters underlying LCA model are presented in the SI, Section S2.

2.4. Life cycle assessment

The environmental performance was assessed using life cycle assessment (LCA) conducted in accordance with the requirements of the ISO 14044 standard and the guidelines of the EU Commission's ILCD Handbook (ISO, 2006; EC-JRC, 2010).

2.4.1. Functional unit and reference flow

The primary function of the PHA-based bioplastic in the context of this study is to protect dry food against environment during transport and storage. We choose a croissant as an exemplar of dry food product. The functional unit was therefore defined as "Protection of one average croissant (ca. 40 g) against migration of oxygen, water and aromas (according to global and specific migration standards BS EN 1186 and UNE-EN 13130 for migration of aromatic primary amines, pthalic acid, crotonic acid, acrylic acid and the elements Al, B, Ba, Cu, Co, Fe, Li, Mn, Ni and Zn) during transport and storage for 30 days". This functional unit was chosen as it allows a consistent comparison with alternative plastics used as packaging materials. The reference flow is equal to 0.06384 m^2 of PHA-based plastic film with improved barrier properties, and the same reference flows apply to other plastics fulfilling this functional unit. Yet, differences in thicknesses of the PHA based plastic films and other plastics result in different reference flows when expressed on a mass basis.

2.4.2. Modelling framework and system boundaries

Production of PHA-based bioplastic with improved barrier properties and its use in food supply is a relatively new technology and its implementation is not expected to cause large scale market consequences (for example the need to install new power plants). Therefore, consistent with ILCD's recommendations, the current LCA is considered a microlevel decision support situation (type A) (EC-JRC, 2010). This implies that: (i) system expansion is the preferred way to solve multifunctionality, and (ii) average processes are to be used to model the background system of the study. The consequential version of the ecoinvent v3.5 database was employed to model the background system because it prioritizes system expansion rather than allocation (Bjørn et al., 2017). However, this consequential database systematically uses marginal processes rather than average ones. Therefore, to make the database more consistent with the attributional approach, some processes were adapted to be based on average rather than marginal mixes. Details on these adaptations are presented in SI, Section S2 (Table S9). For example, as in (Bohnes, 2020), the marginal electricity grid mix originally included in the consequential database was adapted to represent the average mix of 2018. The use of marginal data was considered negligible for other processes in the bioplastic life cycle and their adaptation was not deemed necessary. The product systems were modelled in SimaPro, version 8.3.0.0 (PRé Consultants B.V., the Netherlands).

An overview of system boundaries, specifying processes included in the LCA, is presented in Fig. 1. Background processes include (avoided) conventional use of the feedstock, production of energy and chemicals, construction and disposal of equipment, and treatment of biological waste. The use stage includes transport from production site to the customer. The end-of-life stage comprises waste management processes according to the waste management of system in the country of interest. The foreground system comprises all processes, as presented in Fig. 1. Molasses is a residual product from production of sugar and therefore no burdens are attributed to its production. However, environmental burdens occur when the molasses waste stream is diverted for production of PHA, rather than its conventional use as animal feed. Consistently with system expansion being prioritized over allocation when handling multifunctional processes, this animal feed has to be produced from other sources, like barley grains.

2.4.3. Life cycle impact assessment

Environmental impact scores were mainly calculated using ReCiPe 2016 as LCIA methodology, applying midpoint indicators and hierarchist perspective. Impact scores were calculated for all ReCiPe impact categories, except climate change which was replaced by the approach of ILCD (2011) combined with updated GWP100 values from IPCC AR5 (IPCC, 2014). The ILCD (2011) approach was preferred as it gives credits to delayed emissions of greenhouse gases (GHGs), which are particularly relevant for the end-of-life stage of the PHA-based plastics. In addition to the GWP100, which is the default metric in LCA and addresses short/medium term climate impacts, we employed the global temperature change potential (GTP100) and the multiple climate tipping points potentials (MCTPs) as characterization factors (CFs). The GTP100 is recommended for use in LCA, next to the GWP100, as it focuses on long-term impacts, representing global average temperature increase of the atmosphere at 100 years that results from the emission

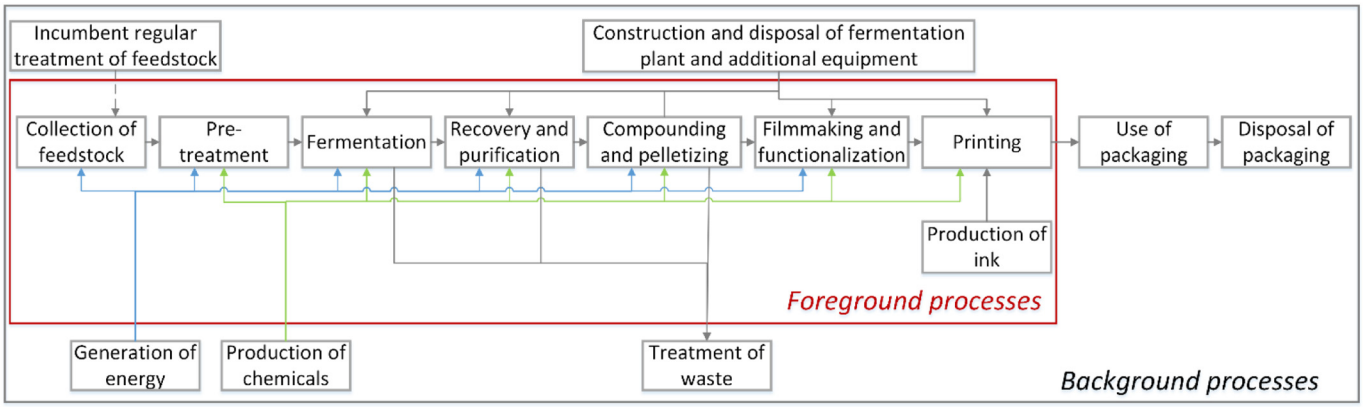

Fig. 1. System boundaries of the PHA-based plastics with improved barrier properties.

(Shine et al., 2005; Levasseur et al., 2016). The MCTP is a recently developed metric for climate tipping impacts (Fabbri et al., 2021), building on earlier work of Jørgensen et al. (2013). It specifically addresses the potential contribution of GHG emissions to trigger multiple climate tipping points in the earth system (like loss of Arctic summer sea ice or the El Niño-southern oscillation intensification), considering in total 13 tipping elements that could pass a tipping point with increasing warming. The contribution to tipping is measured as the share of remaining carrying capacity up to each tipping point that is consumed by the emissions, using Eq. (1) (Fabbri et al., 2021), and is expressed as fraction of depleted remaining capacity in parts per trillion, ppt_{rc}, per kg GHG emission:

$$MCTP_i(T_{\text{emission}}) = \sum_{j=1}^{m} \frac{I_{\text{emission},i,j}\,(T_{\text{emission}})}{CAP_j\,(T_{\text{emission}})} \quad (1)$$

where $MCTP_i(T_{\text{emission}})$ is the characterization factor for GHG i emitted at time T_{emission}, j is the jth out of m potentially exceeded tipping points, $I_{\text{emission},\,i,\,j}$ is the increase in CO_2-equivalent concentration caused by the emission with respect to tipping point j, and CAP_j is the remaining capacity of the atmosphere to absorb this concentration increase without triggering tipping point j (Fabbri et al., 2021). Given that the MCTP is sensitive to the timing of emissions, the metric is particularly relevant for the end-of-life of PHA-based plastics, as emissions are distributed over time and could contribute to crossing tipping points (Fabbri et al., 2021). The three climate-related sets of indicators are complementary to each other and represent three different impact categories. Details of calculation of impact scores using these three approaches are presented in Section S3 of the SI.

2.5. Sensitivity and uncertainty analyses

Sensitivities of the LCA results to discrete parameters were evaluated in a scenario analysis (see Section 2.1). Sensitivities to PHA yield, which is a continuous parameter, was also considered for selected scenarios from Table S3S3, Section S2 of SI. Quantification of inventory uncertainties is currently not possible to carry out with the consequential version of the ecoinvent database as attached to SimaPro. To compensate for this limitation, we conducted a qualitative uncertainty analysis discussing limitations of the study considering the specificity of the inventory data.

3. Results and discussion

In the following sections, we present an overview of life cycle impact assessment results for selected scenarios, identify factors which determine overall environmental performance of the PHA plastics, and identify optimization potentials.

3.1. Environmental hot-spots in the PHA value chain

To identify processes with the largest contribution to this burden, process contribution analysis was carried out on PHA laminated with PLA at pilot and large scale systems in Italy (Fig. 2). Refer to Table S13, Section S4 of the SI for tabulated impact scores for the two scenarios. Irrespective of the plant scale, incumbent management of feedstock had the highest contribution to environmental burden for most, but not all, impact categories (up to 94% of total impact, depending on the impact category). As explained in Section 2.4.2, we consider that when molasses is used for PHA production rather than its incumbent use as animal feed, this animal feed has to be produced from other sources, like barley grains. Thus, relatively high contribution of incumbent management of feedstock is explained by burdens associated with production of animal feed from barley grains. Negative impact scores (indicating environmental benefits) are observed for the climate change impact category. They are a result of fixation of CO_2 during cultivation of barley grains. These environmental benefits are, however, outweighed by the burden stemming from the fermentation itself which uses energy and emits CO_2, treatment of wastewater, and incineration of plastic waste in the end-of-life treatment.

The fermentation had relatively small contribution (up to 8% of total impact), except the three climate-related impact categories where its contribution ranged from 21 to 64% of the total impact. The post-treatment processes (recovery, purification, compounding and pelletizing), however, altogether contributed up to 60% of the total impact, depending on the impact category. Previous studies on PHA production from sucrose (including collection, pre-treatment, fermentation and PHA recovery), reported global warming impacts which were higher (1.96 kg CO_2 eq/kg $PHA_{recovered}$ in Harding et al. (2007)) and lower (−2.58 kg CO_2 eq/kg $PHA_{recovered}$ in Kookos et al. (2019) owing to energy recovery from bagasse), compared to 0.76 CO_2 eq/kg $PHA_{recovered}$ in the large scale system of this study. As in Kookos et al. (2019), direct emissions of CO_2 during fermentation were important contribution to climate change burdens in our study. Harding et al. (2007), on the other hand highlighted steam and electricity use as the most contributing process. Further, while our study showed that surfactant had a high contribution to global warming impacts (in our LCA modelled as non-ionic surfactant), neither Harding et al. (2007) nor Kookos et al. (2019) found that the surfactant was the hot spot. Compared to the former study, the consumption of surfactant in our study was 16 times higher, while Kookos et al. (2019) applied a negative GHG emission factor for surfactant based on data from Akiyama et al. (2003).

Relatively high contribution of recovery and purification was mainly caused by the use of steam for spray drying in the pilot system and

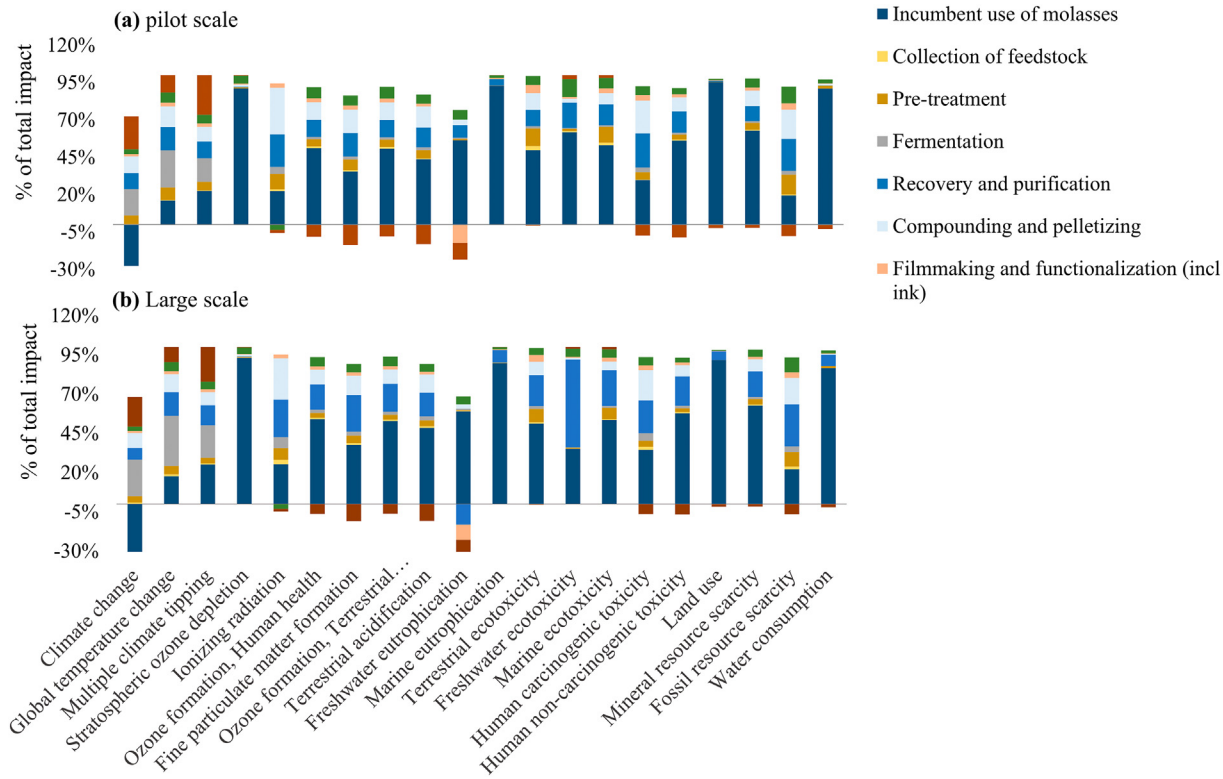

Fig. 2. Contribution of life cycle processes to total impacts from PHA-based packaging at pilot and large scale. The scores for each impact category are scaled to 100%.

surfactant in the large scale system. Surfactant contributed to 55, 24 and 15% of total freshwater ecotoxicity, fossil resource scarcity and climate change impacts. Negative contributions to total impact scores on freshwater eutrophication observed in our study for recovery and purification and filmmaking and functionalization are unexpected, but can be explained by system expansion mechanisms occurring in non-ionic surfactant applied during recovery and purification and ink applied in filmmaking and functionalization processes.

Electricity consumption for processing of the recovered PHA into PHA pellets explains 23 and 19% of total impact for climate change and human carcinogenic toxicity. Negative impact scores for waste management systems for several other impact categories, indicating environmental benefits, were due to incineration with energy recovery (61% of the packaging is incinerated in Italy), substituting production of energy (in this case, electricity and heat for reuse in municipal waste incineration).

3.2. Effects of upscaling

The large scale system has slightly higher impact scores than the pilot scale one consistently for all impact categories, except climate change and freshwater eutrophication (Table S13, Section S4). The largest differences were observed for freshwater ecotoxicity followed by, water consumption, land use and marine eutrophication, where large scale production shows impacts from ~1.5 to ~2.5 times higher than at pilot scale, respectively. This finding was unexpected, because upscaling of technologies is often associated with decreasing environmental impacts perf unit of output (although generalization across different technologies cannot be made) (Gavankar et al., 2015; Owsianiak et al., 2016). The different result in our case can be explained by differences in environmental performance of; (i) recovery and purification steps (all impact categories, except climate change and stratospheric ozone depletion), (ii) fermentation (all impact categories), (iii) collection of feedstock fermentation (all impact categories) and (iv) incumbent use of molasses (all impact categories, except climate change). Increased impacts for recovery and purification were due to higher consumption of surfactant in the large scale system, particularly so for freshwater ecotoxicity where the large scale systems shows 7.7 times higher impact. This increase outweighed benefits from a lower electricity and steam consumption in the large scale. Increasing impacts from fermentation were mainly due to a higher electricity consumption for aeration. Furthermore, slightly lower yield in the large scale system resulted in higher consumption and collection of molasses per unit of PHA output, increasing impacts. Similar, the lower yield at large scale increased incumbent use of molasses and impacts for all categories except climate change where increased amount of CO_2 fixated reduced impact scores. By contrast, reduced impacts from pre-treatment were due to lower consumption of steam, but these reductions were generally insufficient make the large-scale system perform better.

3.3. Influence of geographic location and incumbent use of feedstock

Impact scores decreased for 12 out of 20 impact categories when (large-scale) PHA production and functionalization took place in Germany instead of in Italy (see Section S4 of the SI, Fig. S2). The largest differences were for the climate change and multiple climate tipping impact categories (decrease by 32 and 20%, respectively) followed by fine particulate matter formation and terrestrial acidification (decrease by 17 and 16%, respectively). For climate change, the reductions were due differences in waste management systems between the two countries (the majority of plastics is incinerated in Germany, while landfilling is the dominant treatment option in Italy). Incineration is seen beneficial over landfilling because it does not result in emission of potent GHG, methane (71% of carbon is assumed to be released as methane during landfilling (Rossi et al., 2015)). For fine particulate matter and terrestrial acidification, lower impacts in Germany can be explained by a lower portion of oil in the electricity grid mix in Germany (3.7% and 0.9% in Italy and Germany, respectively), which has a high contribution to these impact categories.

Impact scores increased for 13 out of 20 impact categories when molasses was used as feedstock for ethanol production (scenario 5) rather than for animal feed (scenario 2) in Italy. The largest increase was observed for impacts related to mineral resources, human non-carcinogenic toxicity, terrestrial ecotoxicity and global temperature change (increase by 64, 36, 31 and 28%, respectively) (see Section S4 of the SI, Fig. S3). This was due to generally higher environmental impacts from production of ethanol than production of animal feed (per unit of molasses). Substantial reductions in impact scores were seen for freshwater eutrophication, land use and marine eutrophication (decrease by 365, 138 and 94%, respectively), with negative impact scores for the first two categories (-1.6×10^{-5} kg P eq and -4.0×10^{-2} m^2a crop eq, respectively) and low impacts for marine eutrophication (1.1 × 10–5 kg N eq) in scenario 5. These negative scores were due to handling a waste product from ethanol production from maize by system expansion, replacing soybean meal. Similar observations were made for Germany, where both increases and decreases in impact scores were observed when the incumbent treatment of molasses was as feedstock for ethanol production.

3.4. Influence of PHA stability

Impact scores of the PHA value chain for the three climate-related impact categories are influenced by mineralization kinetics and extent of the mineralization lag phase in landfilling (Table 1). For all three indicators, lowest impact scores were consistently identified for the very slow degradation scenario (scenario 51). This was mainly due to incomplete degradation over 100 year time (GWP and GTP) and over 94 years (MCTP), where only 1% of initial plastic degraded in this scenario, resulting in lower impact scores. Plastics with fast and medium mineralization kinetics generally performed worse according to GWP as credits given for temporary carbon storage are lower compared to more stable plastics. By contrast, climate tipping impact scores increased with decreasing mineralization rates, because the probability that a significant portion of emissions is released in proximity to tipping points, where MCTP values are the largest, was higher for the more stable plastics. This was even more pronounced for cases where a mineralization lag phase of 20 and 40 years was assumed (scenarios 52 and 53 in Table 1). In those cases, a larger share of the emissions was released close to the year 2050, where MCTPs are the highest. Mineralization kinetics was not found to matter for the GTP metric, because this approach disregards any benefits from temporary carbon storage and does not account for when GHG emissions occur in the life cycle.

3.5. Making PHA-based plastics more sustainable

PHA-based plastics can be made more sustainable by optimizing PHA yield, thickness of the PHA layer, and choice of material for ensuring barrier properties. Fig. 3 shows the effects of these parameters on environmental performance for selected impact categories. Comparisons were also made with pure PLA or pure fossil-based PE, and pure fossil-based PP. Increasing PHA yield generally improves environmental performance of the PHA-based packaging. For MCTP, fossil resource scarcity and land use, impacts decreased from 87 to 28% if yield increases from the minimum to the maximum values reported in the literature for PHA made from molasses (i.e. from 0.083 to 0.245 kg $PHA_{raw}/kg_{molasses}$; scenarios 3–18 in Table S3S3, Section S2). However, only a small increase was observed for climate change (by 2%).This relatively small increase was due to the fact that the decreasing fixation of CO_2 (hence increasing impacts with increasing yield), was outweighed by decreased emissions of CO_2 from fermentation and reduced amount of carbon-containing wastewater to be treated (per unit of PHA output).

The results also showed that PHA combined with either Al or AlOx (scenarios 7 and 8) were more sustainable than the PHA combined with PLA (scenario 2). Impact scores were consistently reduced for all impact categories, except for ionizing radiation (Fig. 3 and Table S14 in the SI, Section S4). The reduction was, however, modest (up to 11% for fossil resource scarcity). Despite relatively large differences in environmental impacts per kg of each alternative material (e.g. higher impacts for Al when compared to PLA), significantly less Al or AlOx (10-nm layer) than PLA (20-μm) is needed to fulfill the functional unit, explaining small differences between PLA and Al (or AlOx).

PHA-based plastics can also be made more sustainable if thickness of underlying materials is reduced (while still allowing the packaging to fulfill the function). However, the extent of required improvements is relatively large. For example, thickness of the PHA layer in the PHA/PLA alternative needs to be reduced to ca. 20 μm for this alternative to be able to compete with pure PLA of 91 μm (in terms of climate change and multiple climate tipping). If PHA yield increases, these PHA-based films would be able to compete with PLA of 50 μm thickness (again, assuming that their functional performance parameters are the same). Irrespective of yield and assumed PHA thickness, however, packaging made of PHA generally does not perform as good as PP- and PE- based packaging does (scenarios 11 and 12 in Table S3) (Fig. 3). The differences were by factor of 2 to 5, depending on the impact category, even if high yield and low thickness of PHA were assumed.

4. Limitations and data gaps

This study presents full life cycle inventory and impact assessment results for PHA-based plastics with improved barrier properties. The main limitations of the study relate to: (1) variability and uncertainty in parameters used for modelling life cycle inventories, (2) the choice of LCI database for modelling background system, and (3) deficiencies in impact assessment method.

Table 1
Impact scores per functional unit (f.u.) of the PHA value chain as depending on stability of the PHA plastics in landfilling conditions (mineralization rate constant and extent of mineralization lag phase) and the climate-related impact category. Color shades from green, yellow to red, indicate increasing impact (per impact category). The scenarios tested for stability and degradation are; fast kinetics: 90% degradation in 2 years (100% degraded in 100 years), medium kinetics: 90% degradation in 31 years (99.9% degraded in 100 years), slow kinetics: 90% degradation in 105 years (89% degraded in 100 years), very slow kinetics: 90% degraded in 22,798 years (1% degraded in 100 years), delayed (20): degradation delayed by 20 years, fast kinetics, delayed (40): degradation delayed by 40 years, fast kinetics (see a full overview of scenarios in Table S3, Section S2).

	GWP_{100} (kg CO_2eq/f.u.)	$MCTP_{RCP6}$ (ppt_{rc}/f.u.)	GTP_{100} (kg CO_2eq/f.u.)
Fast (scenario 2)	5.25E-02	1.22E-03	1.04E-01
Medium (scenario 49)	4.53E-02	1.40E-03	1.04E-01
Slow (scenario 50)	3.57E-02	1.45E-03	1.03E-01
Very slow (scenario 51)	2.81E-02	9.40E-04	9.31E-02
Fast with 20-yr lag phase (scenario 52)	4.17E-02	4.95E-03	1.04E-01
Fast with 40-yr lag phase (scenario 53)	3.08E-02	4.93E-03	1.04E-01

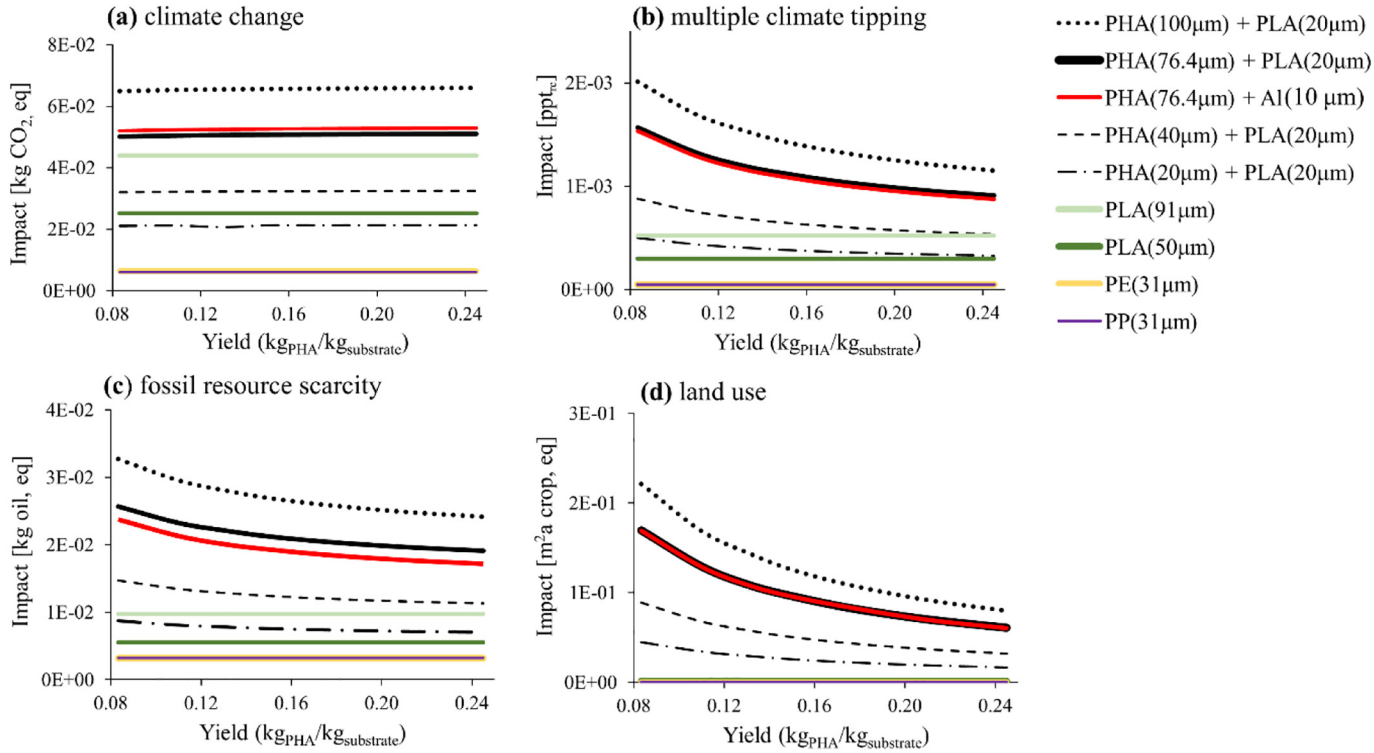

Fig. 3. Impact scores for climate change, multiple climate tipping, fossil resource scarcity and land use as influenced by PHA yield, and type and thickness underlying materials (scenarios 7–48 in Table S3S3, Section S2 of SI). Yields are based on literature data, where the minimum yield is from Kookos et al. (2019) and the maximum yield is estimated from a theoretical yield from Yamane (1993) and assuming that 95% of the accumulated biomass is PHA. The yield in the x-axis refers to PHA-based plastics only. Impacts of PLA, PE and PP are shown in the figure for comparison, but are not influenced by PHA yield.

First, we modelled pilot and large scale PHA production systems basing on data retrieved from the literature, but several parameters are known to be variable or uncertain. This may influence comparisons between scales. For example, PHA yield varies, but is an important parameter which determines performance of the PHA vale chain, and the large scale system would generally perform better than the pilot scale if the PHA yield was in higher range of possible values (0.245 $kgPHA_{raw}/kg_{molasses}$) (data not shown).

Second, biodegradation kinetics of the PHA-based plastics in the environment is highly uncertain (Emadian et al., 2017; Meereboer et al., 2020), and furthermore it is unknown how surface treatment may influence biodegradation kinetics in landfilling conditions. Our sensitivity analyzes show that this parameter is important not only for the end-of-life, but for the performance of the whole PHA value chain (in terms of climate change and multiple climate tipping impacts).

Third, the surfactant in the current study was modelled as a generic non-ionic surfactants, which consists of ethylene oxide (66%) and fatty acid (33%) derivatives. Impacts of surfactants vary considerably (Schowanek et al., 2018). For example, if the fatty acid derivate was used, freshwater eutrophication and ecotoxicity impacts would decrease by 114% and 45%, respectively (data not shown). It is therefore important to address this data gap in future studies on PHA.

Fourth, the consequential background database was consistently applied for background processes, with the exception of electricity processes, which were adapted to average grid mixes rather than marginal mixes. The sensitivity of this was tested for incumbent use of molasses and found to have a high influence on the overall results. Although contribution from other processes of the background system is expected to be smaller when compared to energy and avoided incumbent use of molasses, there is some uncertainty as average mixes (rather than marginal mixes) should ideally be used consistently for all processes in the background system.

Finally, owing to the limitations of the ecoinvent database, indirect land use changes (ILUC) were not considered in the PLA value chain (PLA is made from maize). If they were considered, impacts of those PHA-based plastics which include PLA would increase. According to Ögmundarson et al. (2020), e.g., climate change impact for lactic acid from corn could increase by 14% if ILUC are included. Hence, including ILUC would further favor those alternatives which use either Al or AlOx as barrier materials.

5. Implications for PHA value chain

We showed that PHA-based plastics with improved barrier properties have higher environmental impacts than alternative packaging made from PE, PP and potentially even PLA. These results are not surprising given that PHA production is still relatively immature when compared to the aforementioned alternatives. The largest optimization potentials (which are also challenges to PHA technology developers), are: 1) reduction of PHA thickness while maintaining functional properties of the PHA plastic, 2) increase PHA production yield, 3) increase the energy efficiency during compounding and pelletizing, 4) decrease amount and change type of surfactant used in recovery and purification processes, 5) consider feedstock other than molasses, that do not have a highly beneficial alternative treatment and use. Industrial wastewater could be considered as feedstock, as it avoids incumbent management of the wastewater (Heimersson et al., 2014). Furthermore CO_2 could be a promising alternative feedstock for PHA production (Troschl et al., 2018), but separate LCA would be needed to evaluate performance of PHA made from other feedstock. 6) consider alternative end-of-life options. Although the biodegradability of PHA offers aerobic and anaerobic end-of-life pathways in comparison to conventional plastics, recent research results for PLA show that also recycling (e.g. mechanical recycling) is a potential option which can offer additional benefit from an LCA as well as circular economy perspective (Maga et al., 2019; Spierling et al., 2020). Our study may suggest that that Al (or AlOx) is the preferred material to ensure barrier properties. However, unknown influence of the Al layers on biodegradability of PHA in the environment

warrants further studies. Finally, we stress that we have addressed environmental aspects of sustainability, but economic and socially-oriented analyses are required to make more informed decisions about implementing PHA with improved barrier properties as alternative packaging materials.

CRediT authorship contribution statement

Eldbjørg Blikra Vea: Methodology, Investigation, Data curation, Visualization, Writing – original draft. **Serena Fabbri:** Methodology, Data curation, Writing – review & editing. **Sebastian Spierling:** Writing – review & editing, Validation. **Mikołaj Owsianiak:** Conceptualization, Writing – review & editing, Validation, Supervision.

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.

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Appendix A. Supplementary data

Details of literature review, data underlying LCA model, details of life cycle impact assessment, additional results (Sections S1–S5), Simpro model processes (Section S6) Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.147544.

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