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Life cycle greenhouse gas emissions and energy use of polylactic acid, bio-derived polyethylene, and fossil-derived polyethylene

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ABSTRACT

Bioplastics recently have become an attractive, viable, and popular alternative to conventional petroleum-based plastics, with the hope that replacing fossil-derived plastics with renewable alternatives will reduce greenhouse gas (GHG) emissions and fossil energy consumption (FEC). The bioplastic industry is encouraging creative designs and enhanced properties such as biodegradability, which is considered a sustainable solution for waste plastic management. However, biodegradability also means that carbon in the product is emitted to the atmosphere as GHG emissions. In this paper, a life cycle analysis (LCA) of biodegradable polylactic acid (PLA) and bio-polyethylene (bio-PE) plastics was conducted to understand the environmental effects of these bioplastics from feedstock production to product end-of-life (EOL). In particular, emissions from biodegradability (EOL emissions) are accounted for. The results were compared to those of conventional fossil-based plastics such as high-density polyethylene (HDPE) and low-density polyethylene (LDPE). Results showed that the lowest GHG emissions (−1.0 and 1.7 kg CO_2e per kg for bio-PE and PLA with no biodegradation, respectively) and FEC (29 and 46 MJ per kg of bio-PE and PLA, respectively) were achieved with bio-derived plastics, particularly bio-PE plastic. However, despite the benefits of biogenic carbon uptake, when landfill and composting emissions were considered for the PLA pathway, the life cycle emissions of PLA increase significantly, from 16% to 163% depending on the biodegradation condition, compared to the case where there is no degradation in the landfill. This study also contributed to understand the effects on the GHG emissions of biodegradability in landfill and composting scenarios, regional electricity mix, and plastics manufacturing technologies.

1. Introduction

Plastics play an important role in modern society. Their versatility, durability, and light weight allow them to have innumerable applications — from building and construction to food packaging and clothing. Global plastics manufacturing has grown from about 1.5 million metric tons (Mt) per year in 1950 to more than 322 million Mt per year in 2015 (Plastics Europe, 2017). In 2017, the United States (U.S.) was responsible for 35% of global plastic production, or 112 million Mt (American Chemistry Council, 2017). Growth of the plastics industry is expected to continue into the near future, with an estimated global production of about 1.63 billion Mt by 2050 (Ryan, 2015). This has driven a significant increase in plastic consumption worldwide, to the point where disposal of plastic waste is creating considerable problems for humans, wildlife, and the environment (Jambeck et al., 2015; Lebreton et al., 2018). The U.S. Environmental Protection Agency (EPA) estimated that the amount of plastic found in municipal solid waste (MSW) in the U.S. in 1960 was around 0.35 million Mt, which increased to about 32 million Mt by 2015 (U.S. Environmental Protection Agency, 2018).

Plastic production in the U.S. accounts for 1% of total U.S. greenhouse gas (GHG) emissions and 3% of total energy use annually (Posen et al., 2017). A major contributor to GHG emissions during plastics production is the consumption of fossil resources, which are used as both feedstock and energy inputs in the process. This has spurred the development of new products that can

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contribute to a more sustainable plastics industry. New plastics are being designed with special features and improved characteristics, such as biodegradability, compostability, recyclability, or an oxygen barrier. Some of these characteristics are important for determining the proper end-of-life (EOL) and disposal of the plastics, which will be beneficial for achieving a more sustainable plastic industry. Today, plastics can be produced from fossil-derived sources, such crude oil and natural gas, as well as bio-derived resources including traditional agricultural products (e.g., corn, soybeans, sugar cane), agricultural waste (e.g., corn stover or wood waste), or other alternatives (e.g., algae). Now, depending on the source of feedstock, plastics can be classified as either conventional fossil-based plastics or bio-derived plastics (bioplastics). Polyethylene (PE) including high-density polyethylene (HDPE) and low-density polyethylene (LDPE), polypropylene (PP), polybutylene (PB), polyethylene terephthalate (PET), and polystyrene (PS) are polymers commonly produced using fossil resources. However, it is believed that as much as 90% of global plastics could be physically replaced by bioplastics (Shen et al., 2010). Bioplastics currently account for approximately 1% (2.1 million Mt) of global plastics production (European Bioplastics, 2017). Global production capacity for bioplastics is expected to increase by 19% to 2.5 million Mt by 2022 (European Bioplastics, 2017), continuing a trend of 20%–30% growth per year (Nampoothiri et al., 2010).

Bioplastics can potentially benefit the environment by replacing conventional fossil-derived feedstocks with renewable alternatives, reducing GHG emissions and utilizing renewable sources (Adom et al., 2014; Benavides et al., 2018). Life cycle analysis (LCA) can be used to evaluate the environmental benefits associated with the production of bioplastics along the supply chain. The bioplastics LCA results depend on multiple factors, such as feedstock type and sourcing, process design and conversion technology, and the choice of final disposal. In this paper, an LCA comparison between fossil-derived HDPE and LDPE and the bioplastics PLA and bio-derived polyethylene (bio-PE) is presented.

PLA is a linear aliphatic thermoplastic polyester with three stereochemical forms: poly-L-lactide (PLLA), poly-D-lactide (PDLA), and poly-D, L-lactide (PDLLA) (Nampoothiri et al., 2010). It currently accounts for more than 10% of global bioplastic production (European Bioplastics, 2017), and production capability is expected to increase significantly in the near future (Detzel et al., 2013). PLA can be substituted for PP, PE, acrylonitrile butadiene styrene (ABS), and others (Guo and Crittenden, 2011; Groot and Boren, 2010) because its optical, mechanical, thermal, and barrier properties are comparable to those commercially available plastics (Castro-Aguirre et al., 2016). PLA is a potential replacement in conventional plastics applications such as cups, bottles, to-go containers, packaging, films, and textiles (Vink et al., 2003; Henton et al., 2005; NERC, 2020; Sarlee et al., 2006). Bio-PE is obtained by polymerization of bio-based ethylene gas. Ethylene can be produced from catalytically dehydrated ethanol, which can be obtained from various biomass sources such as corn stover. Bio-ethylene can then be used in traditional polyethylene polymerization processes to make various grades of PE, such as HDPE and LDPE.

Several authors have studied the environmental and energy impacts of bioplastics, especially EOL strategies such as incineration, recycling, composting, and landfill, compared with those of conventional fossil fuel counterparts. For instance, Gironi and Piemonte (2011) compared the environmental impact of the PLA vs. PET bottles for drinking water. The authors presented the benefits of PLA for bottle production in terms of saving fossil resources and the drawbacks in term of causing major damage of human health and ecosystem quality. Piemonte (2011) compared the potential energy and GHG savings of PLA and Novamont's MATER-BI bioplastic to conventional plastic production (PET and PE). In his research, he concluded that replacing fossil PET and PE with PLA and MATER-BI bioplastics could significantly reduce energy demand and GHG emissions. Piemonte (2011) also showed that to maximize energy savings and reduce demand for renewable resources, the best solution for bio-based products disposal is mechanical recycling. While Hottle et al. (2017) compared the life cycle environmental metrics of eight fossil-based plastics with their bio-derived counterparts, including PLA and thermoplastic starch (TPS). They concluded that composting has some environmental benefits over landfill, but those benefits are only applicable to biodegradable plastics. Recycling provides the greatest environmental benefits, as it would be applicable to all. However, currently recycling is limited to fossil-derived plastics because of the lack of recycling infrastructure for biodegradable plastics. Although these references provide an overview of environmental impact assessments, especially for different EOL strategies for several plastics, very few provide details on the key drivers that affect GHG emissions and energy from the supply chain point of view.

In this present work, LCAs of two common bioplastics, bio-PE and PLA, are performed to show the contribution to GHG emissions and fossil-energy consumption (FEC) at each stage of the plastic supply chain, from feedstock production to EOL. The results are compared to those of conventional fossil-based plastics, such as HDPE and LDPE. With the scope of this analysis being the U.S., this paper focuses on the impact of disposing of plastics in landfills to understand EOL tradeoffs between biodegradability and carbon sequestration in these plastic pathways. The fate of waste plastic varies in different regions around the world: While in the U.S. the most common waste management method for waste plastic is landfill, in Europe it is mostly combustion with energy recovery. However, in eleven European Union member states there is still a significant share (about 50%) of waste plastic ending up in landfills (Plastics Europe, 2017). It is understood that there are environmental benefits to recycling plastic, as the previous literature has concluded, but not all plastics can be efficiently recycled.

In 2015, only 9% of total plastic in MSW was recycled in the U.S., while the rest was either combusted (16%) or landfilled (75%), even though efforts to increase recycling rates have been improved in the last few years in the U.S. (U.S. Environmental Protection Agency, 2018). If bioplastics replace conventional fossil-derived plastics in single-use or short shelf life applications, it is likely they will be landfilled after use (Siracusa et al., 2008). Thus, it is clearly important to understand the environmental impact of bioplastics' EOL, especially emissions from landfilled biodegradable plastics, compared to that of fossil-derived plastics. Therefore, the objective of this study is not only to quantify the life cycle GHG emissions and FEC from each in the supply chain of plastic production, but also to consider EOL tradeoffs between biodegradability and carbon sequestration in these plastic pathways. Composting scenarios were also included for PLA, because it can be designed to be biodegradable in industrial composting conditions.

In addition, to have a better understanding of the key factors that affect the life cycle GHG emissions and fossil energy consumption of plastic production pathways studied here, sensitivity analyses were conducted. These sensitivity analyses help us evaluate the effect on GHG emissions of biodegradability in landfill and composting scenarios, parameters in the landfill management such as landfill gas (LFG) collection efficiency, regional electricity mix, and the plastics manufacturing technologies — aspects that have not been explored in great detail in the past literature. An expanded version of the Greenhouse gases, Regulated Emissions, and Energy use in Technologies (GREET®) model (Argonne National Laboratory, 2019) was used. This tool was developed to evaluate the energy and environmental impacts of advanced fuels and products from fossil and bio-derived sources.

This paper is organized in the following order: First, the LCA methodology is described, providing information on the selected plastic pathways studied here. Then the biodegradation of PLA is discussed, and a list of key parameters for PLA biodegradation in landfill and composting conditions is also provided. LCA results and discussion are presented in Section 3, where a comparison with previous LCAs of PLA and the sensitivity analysis results are presented.

2. Methodology

2.1. Life cycle analysis

An LCA was performed for four plastic pathways using two waste management scenarios to identify the key drivers that affect their environmental footprint. Two forms of fossil-derived PE (HDPE and LDPE) were selected for this analysis due to their broad range of applications, many of which overlap with the potential or actual applications of the bioplastics under consideration. The bioplastics selected for this study are bio-PE from corn stover and PLA from corn via lactic acid fermentation. Fig. 1 presents the system boundary of both fossil- and bio-derived plastic production pathways for this analysis. The system boundary covers all material and energy flows associated with all stages of plastics production: feedstock production, conversion, plastic manufacturing, and EOL. Fig. 1 also presents the net emissions of the plastic pathways. Details of the calculation of these emissions are presented in Section 2.4 and Section 3. Two EOL strategies are studied here: landfill, due to its popularity in the U.S., and composting. The composting scenario is applicable only to PLA, due to its biodegradability properties. It was assumed that plastics in this study were limited to single-use applications such as cutlery, food containers, cups, and the like. As mentioned earlier, PE is considered to be displaced by bioplastics like bio-PE and PLA because of similar mechanical characteristics and performance profiles. The LCA results were compared in terms of the amount of material without specifying the application (as it can be anything from cutlery to food container to cup) and reported the results per one kg of waste plastics that ends up in the landfill or composting. This functional unit was consistent with what has been presented in the literature for the LCA of plastic (Hottle et al., 2017; Carus, 2017; Castro-Aguirre et al., 2016). However, other functional units could be chosen if the weight of plastic for a specific application is different depending on the type of plastic.

The GREET model was used to calculate the life cycle GHG emissions (gCO_2e/kg of plastic) and FEC (MJ/kg of plastic) (Argonne National Laboratory, 2019). The GHG emissions include carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) emissions, calculated on the basis of the 100-year global warming potentials for CO_2, CH_4, and N_2O emissions, which are 1, 30, and 265, respectively (IPCC, 2014). For this analysis, the U.S. electricity generation mix, and natural gas produced in the U.S. were assumed (Argonne National Laboratory, 2019).

2.2. Plastic production pathways

PE is obtained by the polymerization of ethylene from either fossil or biomass feedstock. Fossil-derived ethylene is produced from hydrocarbons by steam cracking, a technique that uses high temperature and pressure to produce lighter hydrocarbons, such as ethylene. Crude oil and natural gas are the primary feedstocks used to produce ethylene. The cradle-to-gate impacts for conventional HDPE and LDPE are included in the GREET model, and data sources and methodology are documented in Keoleian et al. (2012) and Argonne National Laboratory (2019). Bio-ethylene is produced from catalytically dehydrated ethanol produced from corn stover. Because corn stover is the post-harvest residue in cornfields, the only energy consumption and emission burdens tied to corn stover are those associated with stover collection and supplemental fertilizer application (Canter et al., 2016 and Argonne National Laboratory, 2019). The cradle-to-gate impacts for bio-PE can be found in Adom et al. (2014) and Argonne National Laboratory, 2019. Bio-ethylene can then be used in traditional polyethylene polymerization processes. Regardless of the types of feedstock used (bio- or fossil-derived), bio-PE and fossil PE are compatible because they are identical polymers (Tsiropoulos et al., 2015). On the other hand, PLA is produced from lactic acid (LA) by polymerization

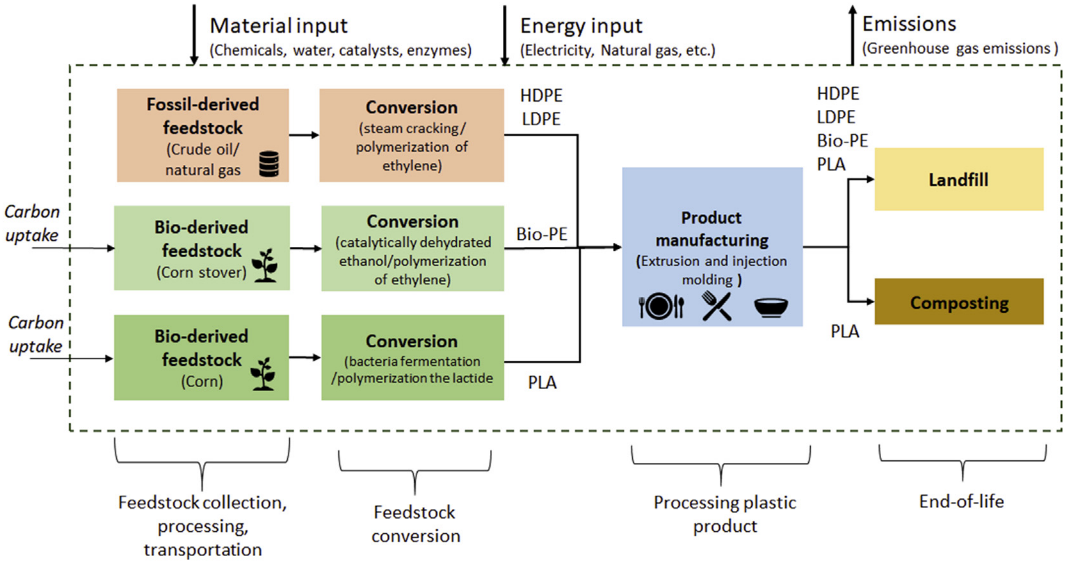


Fig. 1. System boundary of plastic pathways (fossil-derived and bioplastic) for this analysis. Note: Detailed plastic production stages can be found in previous work (Benavides et al., 2019 for PLA, Adom et al., 2014 for bio-PE, and Keoleian et al., 2012 and Franklin Associates, 2011 for HDPE and LDPE fossil-based plastics). All these pathways are also available in the GREET model (Argonne National Laboratory, 2019).

through the lactide intermediate. There are two methods of producing LA: bacterial fermentation of carbohydrates and chemical synthesis. The former is used by NatureWorks, the major worldwide producer of PLA (Castro-Aguirre et al., 2016). In this process, corn is used as the initial feedstock. Details of the energy and emission burdens of corn agriculture and the method of allocation between grain and stover are discussed in previous work by Canter et al. (2016) and Argonne National Laboratory (2019). Data concerning PLA production is based on published work by NatureWorks (Vink et al., 2007; Vink and Davies, 2015). This information was adapted and modeled in GREET 2019 (Argonne National Laboratory, 2019). The details of this process can be found in Benavides et al. (2019).

The material and energy inputs used for the four plastic pathways are shown in Table 1. Natural gas plays an important role for each plastic pathway and represents the majority of the energy input, ranging from 65% of the total energy input (for PLA) to 90% (for LDPE). Electricity plays a smaller role in general, while PLA has the highest electricity demand among all four pathways. For the fossil-based plastic pathways, Table 1 shows both the amount of natural gas and crude oil directly used as feedstock as well as the natural gas used for heat in the LDPE and HPDE production process. For bioplastic, in addition to the feedstock, there are other materials used in the pretreatment, conversion, and recovery of the final product, including sodium chloride, sulfuric acid, and others. The upstream processing inputs of these are also available in GREET (Argonne National Laboratory, 2019). While there are other chemicals for PLA production, their contribution is not significant compared to feedstock or energy input (Benavides et al., 2019).

Table 1
Material and energy used to produce one kg of HDPE, LDPE, bio-PE and PLA.

	HDPE[a,d]	LDPE[a,d]	Bio-PE[b,d]	PLA[c,d]
Energy Inputs: MJ/kg				
Total Energy Input	12.97	16.92	9.85	30.01
Natural gas	11.12	15.18	7.68	19.70
Electricity	1.27	1.32	2.11	6.68
Residual oil	0.49	0.32	0.06	3.39
Diesel fuel	0.05	0.07		
Coal				0.24
LPG	0.01		9.54E-04	
Gasoline	0.03	0.03		
Naphtha				
Hydrogen				0.09
Mass Inputs: kg/kg				
Natural gas as feed (MJ/kg)	39.84	40.59		
Crude oil as feed (MJ/kg)	8.15	7.12		
Corn				1.28
Oxygen				1.74E-04
Bio ethylene			1.01	
Sulfuric acid (H_2SO_4)				0.25
Nitrogen gas				0.01
Sodium chloride (NaCl)				0.11
Bauxites (aluminum salts)				6.0E-06
Barite (barium sulfate, $BaSO_4$)				0.001
Iron (Fe)				3.3E-04
Lead (Pb)				2.0E-06
Limestone ($CaCO_3$)				0.79
Sand (SiO_2)				0.01
Phosphate (as P_2O_5)				0.01
Sulfur (elemental)				0.01
Dolomite				4E-06
Olivine				3E-06
Potassium chloride (KCl)				0.02
Kaolin				0.02
Recycled steel				6.7E-05

Note that natural gas as feed and crude oil as feed represent the amount of these that are used as feedstocks, while the energy inputs represent the amount needed for heat requirements during the conversion process.

[a] Keoleian et al. (2012).

[b] Adom et al. (2014).

[c] Benavides et al. (2019).

[d] Argonne National Laboratory (2019) .

2.3. Processing and manufacturing of plastics

The third section of Fig. 1 is product manufacturing. Manufacturing of plastic products requires a variety of transformation methods, including extrusion, injection molding, blow molding, and compression molding, among others (American Chemistry Council, 2014). Extrusion and injection molding are considered the most important and well-established conversion techniques used by the thermoplastics processing industry in the transformation of PE or PLA (Castro-Aguirre et al., 2016). In injection molding, plastic pellets are fed into the injection unit, melted, and injected directly into the mold cavity. The plastic is cooled and solidified in the shape of the cavity before it is finally ejected (American Chemistry Council, 2014). The extrusion process is similar to injection molding except that extrusion of the melted polymer is continuous. The plastic pellets are fed (via gravity) from a top mounted hopper into the barrel of the extruder where the pellets come in contact with the screw. The screw rotates, forcing the pellets to move forward into the heated barrel where the plastic melts. The plastic is pushed through to the other side where it exits the barrel and enters the die. The die is what gives the final product its shape (Plastic technology, 2019). Other polymers or additives may be added to the plastic to improve its functional properties, and they are not included in this study.

The material and energy inputs for injection molding and extrusion molding of plastics used in this analysis, as determined by Keoleian et al. (2012), are summarized in Table 2. Injection molding requires 7.2 MJ of electricity per kg of plastic transformed into final product, with a mass input factor of 0.95 kg of plastic per final transformed plastic product. The energy requirement for extrusion (2 MJ/kg) is lower than for injection molding, and the mass input factor is 1.01. Methods for processing and manufacturing PLA are the same well-established methods used for other commercial polymers (Castro-Aguirre et al., 2016). Therefore, the input information in Table 2 was assumed to be the same for all pathways considered in this study. The environmental effects of both plastic manufacturing technologies are presented in Section 3.

2.4. Biodegradability of PLA in landfill and composting

The last section of Fig. 1 is the EOL strategies. Despite increasing efforts to promote recycling and the use of recycled materials in plastics production, lack of infrastructure for recycling PLA limits its waste management treatment methods. PLA cannot be recycled with conventional plastics. Adding PLA to a "mixed" recycling stream is not a viable option for now, due to concerns related to the cost of separation and processing, increased contamination, and reduced quality of the recycled materials (Castro-Aguirre et al., 2016). Therefore, PLA is likely to end up in landfills. Conventional plastics, including some types of bioplastics, are chemically stable in landfills and may take over a century to degrade. For this analysis, it was assumed that no degradation of landfilled HDPE, LDPE, or even bio-PE would occur in less than 100 years. Thus, the carbon within these plastic products is effectively captured, meaning no CO_2 should be released to the atmosphere. To be consistent with our previous studies (Adom et al., 2014; Benavides et al., 2018) and what has been commonly used in the literature to assess global warming mitigation calculations (Fearnside, 2001), 100 years was chosen as a time horizon.

In contrast, PLA is biodegradable in the proper conditions and breaks down into smaller compounds such as CO_2, CH_4, and water.

Table 2
Mass of plastic input and energy input used to transform 1 kg of plastic by injection molding and extrusion (Keoleian et al., 2012).

	Injection molding	Extrusion
Material input		
kg of plastic needed per kg of final transformed plastic (kg/kg)	0.95	1.01
Energy input		
Electricity (MJ/kg)	7.2	2.0

Therefore, biodegradation of PLA could result in the release of GHGs such as CO_2 and CH_4 into the ambient environment, contributing to PLA's total GHG emissions. Biodegradation of PLA occurs in two stages: hydrolysis or oxidative reaction of PLA into oligomers and monomers, followed by ultimate degradation via metabolization by microorganisms (aerobic or anaerobic organisms) that break it down into CO_2, H_2O, and CH_4 (Castro-Aguirre et al., 2016; Lyu et al., 2007). In this study, it was assumed that the degradation of PLA can occur in anaerobic (similar to landfill) or aerobic (similar to composting) conditions. Although there has been intensive research on PLA degradation (Table 3), there is still uncertainty regarding the specific landfill/composting degradation condition of PLA, because biodegradation happens in specific environmental conditions, and multiple EOL scenarios are possible.

Therefore, based on current knowledge, biodegradability ranges are presented for different landfill and composting conditions, as found in the literature, and summarized in Table 3. Temperature, type of microorganism, and humidity are important factors influencing biodegradability. For example, at mesophilic temperatures (21°C–35 °C), like those assumed to exist in landfills, PLA can be considered non-biodegradable (Kolstad et al., 2012; Krause and Townsend, 2016). However, at higher temperatures or thermophilic conditions (i.e., 50 °C–65 °C), like those that exist in an industrial composting facility, PLA can biodegrade. Biodegradability is also dependent on the polymer's molecular structure, which ranges from semi-crystalline structures, which exhibit organized and tightly packed molecular chains, to amorphous structure, in which molecular chains are more disorganized. Amorphous structures have been found to emit CH_4 in landfill conditions, while semi-crystalline PLA does not have significant degradation, as it is more difficult to hydrolyze (Kolstad et al., 2012; Itavaara et al., 2002). Landfills usually do not promote biodegradation, as is shown in the first two cases in Table 3. Nevertheless, changing conditions, especially increasing temperatures, can start to degrade PLA by accelerating the rate of hydrolysis of the PLA "backbone," breaking high molecular weight molecules down into low molecular weight monomers that can be assimilated by microorganisms (Itavaara et al., 2002). The information in Table 3 was used to calculate the lower and upper bounds of the emissions from biogas formation in landfill or composting conditions for PLA biodegradation.

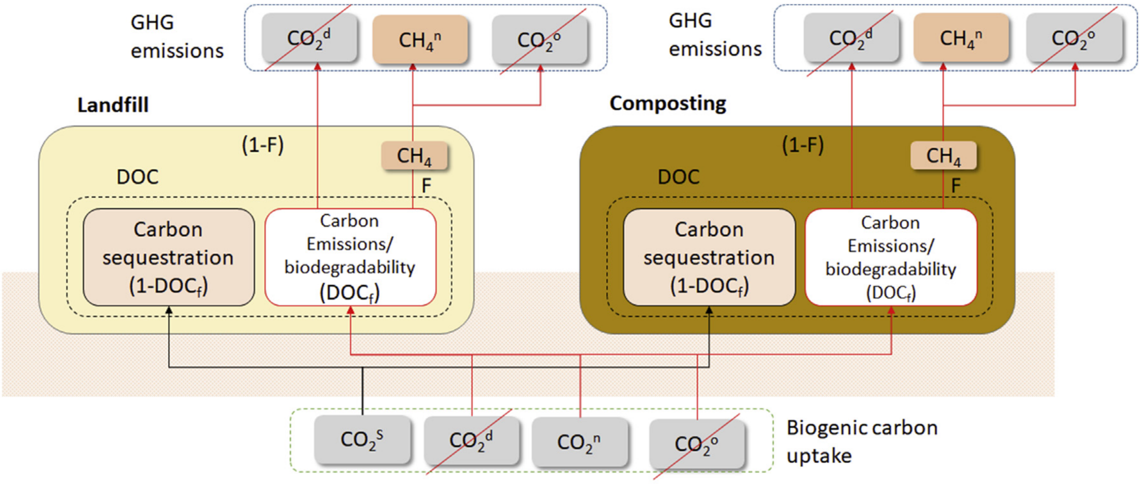
Fig. 2 describes the fate of carbon from landfilled and composting PLA. In this figure, a fraction of the degradable carbon (DOC_F) is used to represent how much carbon is eventually converted into gaseous form (either CO_2 or CH_4), which is the same as the percentage of biodegradation listed in Table 3 for both landfill and composting scenarios. The remainder ($1\text{-}DOC_F$), represents carbon sequestration. The biogas generated through decomposition in landfill, known as landfill gas (LFG), is composed of roughly 50% CH_4, which is represented by F in Fig. 2, and 50% CO_2 (Krause and Townsend, 2016). On the other hand, aerobic degradation is expected to fully oxidize carbon to CO_2 and H_2O, with little (about 5%) or no CH_4 (Itavaara et al., 2002; Gironi and Piemonte, 2011; Hottle et al., 2017; Piemonte, 2011).

Table 3
Parameters for PLA biodegradation in landfill and composting conditions.

% Biodegradability (BD)	Conditions	Source of Information
Landfill (Anaerobic Conditions)		
No biodegradation	Mesophilic temperatures: 35 °C Over 60 days	Krause and Townsend (2016)
No biodegradation	Mesophilic (21 °C, varying moisture contents) and found no significant degradation in the equivalent of one year	Kolstad et al. (2012)
48%	Simulated conditions: 130 years at 35 °C Methane/carbon content in biogas: 58.3% of methane in biogas	Kolstad et al. (2012)
60%–72%	Mesophilic temperature: 37 °C in 100 days Thermophilic temperatures: 52 °C in 40 days In aquatic environment which benefits hydrolysis and increases the rate of biodegradability	Itavaara et al. (2002)
85%		Piemonte (2011) Gironi and Piemonte (2011)
30%–80%[a]	Thermophilic temperatures: 55 °C Samples from cups, lids, cutlery and straws, moisture content from 0.4 to 1.8 Over 60 days	Krause and Townsend (2016)
Composting (Aerobic Conditions)		
60%	Methane/carbon ratio: 95% of the degraded carbon converse into CO_2, the remaining 5% is assumed to evolve into CH_4	Piemonte (2011) Detzel and Kruger (2006) Gironi and Piemonte (2011) Hottle et al. (2017)
10%[b]	Mesophilic conditions 200 days at 25 °C and 37 °C Performed in an aquatic aerobic/microbial condition Microbial	Itavaara et al. (2002)
90%[b]	Thermophilic temperatures at 120 days at 60 °C	Itavaara et al. (2002)

[a] The biodegradability varies depending the biochemical methane potential (BMP) used to predict anaerobic degradation, as well as PLA application and moisture content. (Krause and Townsend, 2016).

[b] Biodegradability was calculated as the mineralization degree on the basis of CO_2 = (gCO_2 resealed/g theoretical CO_2) x 100 (Itavaara et al., 2002).



DOC_F: The fraction of the degradable carbon (or %BD see Table 3)
F: CH_4 concentration. (landfill 50%, composting 5%)
CO_2^s: CO_2 absorbed by biomass used for plastic production that remains as carbon in soil (carbon sequestration)
CO_2^d: CO_2 absorbed by biomass used for plastic production that ends up being decomposed as CO_2
CO_2^n: CO_2 absorbed by biomass used for plastic production that ends up in non-captured CH_4 emission
CH_4^n: non-captured CH_4 emission
CO_2^o: CO_2 absorbed by biomass used for plastic production that ends up being oxidized (including combustion from LFG capture)

Fig. 2. Carbon fate of PLA from landfill and composting.

Part of the generated CH_4 can be collected with landfill gas collectors and flared (Lee et al., 2017). A typical collection efficiency of 75% (U.S. Environmental Protection Agency, 2019), which represents the amount of LFG collected and flared, was assumed. As the rest of the CH_4 passes through the landfill covers, it is partially oxidized into CO_2. It was assumed that 36% of non-captured CH_4 was oxidized (Lee et al., 2017). Thus, LFG emissions consist of CO_2 from decomposition (CO_2^d), CO_2 from the oxidation of non-captured CH_4 (CO_2^o), and non-captured CH_4 emissions (CH_4^n). Since all this carbon comes from biogenic sources (because PLA is produced from corn), CO_2^d and CO_2^o are offset by the carbon uptake occurring earlier during biomass growth, while having an unbalanced uptake of biogenic CO_2 in non-captured CH_4 emissions (CO_2^n) and carbon that remains in the soil (CO_2^s). Note that emissions during transportation from the customer to EOL scenarios were not included in this analysis; however, these emissions may not have significant impact for comparing the results among different EOL conditions, assuming that similar transportation modes, travel distances, and fuels are used among all pathways.

3. Results and discussion

Life cycle GHG emissions of all pathways studied in this paper are presented in Fig. 3. This figure also includes three EOL scenarios for PLA: landfill with no biodegradation (0% BD), landfill with 60% BD, and composting with 60% BD. EOL emissions for HDPE, LDPE, and bio-PE are zero, because it is assumed that all the carbon in the plastic product is sequestered, and no degradation occurs. Fossil-based plastics have the highest GHG emissions (2.6 and 2.9 kg CO_2e per kg of HDPE and LDPE, respectively) while bio-PE has the lowest GHG emissions (−1.0 kg CO_2e/kg). These results are consistent with previous studies, which have also found higher GHG emissions for fossil-based HDPE and LDPE than for bio-based sources (Castro-Aguirre et al., 2016; Hottle et al., 2013). In the case in which PLA does not degrade (0% BD), the GHG emissions are lower — about 35% and 42% lower than HDPE and LDPE, respectively. However, when the biodegradability of PLA is considered, life cycle GHG emissions of PLA increase significantly, especially in the landfill scenario. For example, life cycle GHG emissions of PLA are 27% higher than those of HDPE in composting conditions (PLA composting 60% BD) and 40% higher in landfill conditions (PLA landfill 60% BD), and are 15% and 26% higher than LDPE in the composting and landfill scenarios, respectively.

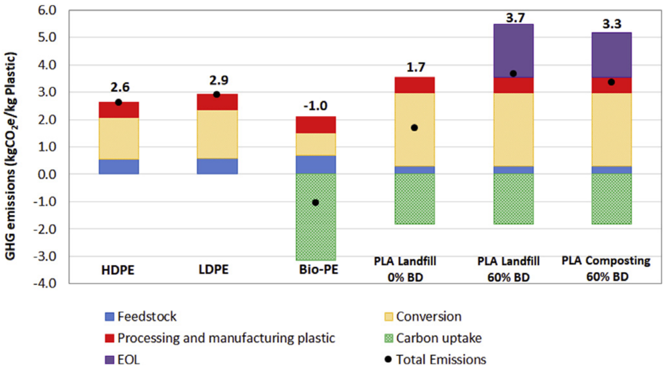


Fig. 3. Life cycle GHG emissions comparison between HDPE, LDPE, bio-PE and PLA, including different EOL scenarios for PLA.

The major reason that bioplastics, in particular bio-PE, have significantly lower GHG emissions than fossil-based plastic is biomass carbon uptake, which represents the key benefit of using renewable resources. The biogenic carbon in biomass is either emitted during PLA or bio-PE production or stored in the final bioproduct. The biogenic carbon emissions during bioplastic production are canceled by the biogenic carbon absorbed during the biomass growth. This is not, of course, the case for fossil-based plastics. Therefore, the net CO_2 uptake from the atmosphere is calculated based on the carbon content of each bioplastic. The CO_2 uptake for PLA is 1.8 kg CO_2/kg PLA — lower than bio-PE (3.1 kg CO2e/kg bio-PE) because of its lower carbon content (50% for PLA compared to 86% for bio-PE).

Emissions during feedstock production and conversion differ in each pathway. For instance, more than half of the HDPE and LDPE

GHG emissions come from conversion of feedstocks (57% and 61% for HDPE and LDPE, respectively) while 21% and 20% for HDPE and LDPE, respectively, are emissions from feedstock processing. These plastics rely on GHG-intensive fossil-based resources such as diesel, residual oil, gasoline, and liquefied petroleum gas (LPG) to convert crude oil and natural gas into plastics (Argonne National Laboratory, 2019). These plastics production processes also have air pollutant emissions including volatile organic carbon (VOC), carbon monoxide (CO), particulate matter (PM10 and PM2.5), and nitric and sulfur oxide (NOx and SOx), as well as GHG emissions such as carbon dioxide, methane, and nitrous oxide (Keoleian et al., 2012 and Argonne National Laboratory, 2019). In the case of bioplastics, about 39% and 76% of the emissions for bio-PE and PLA, respectively, are conversion emissions without accounting for biogenic credits and EOL emissions. The conversion of bio-PE results in GHG emissions lower than those from fossil-derived plastics and PLA because less energy is needed to convert corn stover ethanol-based bio-ethylene (see Table 1). In contrast, emissions during conversion of corn to PLA are significantly higher than those of fossil-derived plastics and bio-PE because of the high energy requirements, especially natural gas and electricity inputs (see Table 1).

PLA is a relatively new type of plastic compared to fossil-derived plastics, which have been industrially produced since the 1930s. As PLA is in its (relatively) early stages of development, it is expected to have higher carbon intensity than those well-known conventional plastics due to less optimized processing compared to more mature fossil-derived plastics (Carus, 2017). NatureWorks, for example, has been making Ingeo PLA (their branded PLA product) for around 15 years, and they have improved the conversion technology to reduce resource demands such as natural gas, electricity, water consumption, etc., which adds significant benefits to PLA's environmental profile (Vink and Davies, 2015). Therefore, it is likely that conversion emissions of PLA can be decreased from those presented in Fig. 3. The PLA results presented here are based on the work by Benavides et al. (2019) that relied on life cycle inventory (LCI) from Vink et al. (2007). New studies by NatureWorks on Ingeo PLA show improved cradle-to-gate inventories and GHG emissions that have decreased to 0.6 gCO_2e/kg (Vink and Davies, 2015). However, it was not possible to use this value because details on LCI for the most recent PLA developments were not publicly available.

Other authors have also studied the environmental impacts of the production of plastics, including HDPE, LDPE, and PLA, that include global warming potential (GWP). It is important to note that LCA results from other publications might be different because of the upstream input information used, such as fuel and energy sources (e.g., different electricity grids, alternative energy scenarios like renewable energy certificates), the use of different LCA tools to evaluate these impacts, or different system boundaries. For example, Hottle et al. (2013) reported cradle-to-gate GHG emissions for PLA of about 3.2 kg CO_2e/kg based on an Ecoinvent v2.2 dataset and LCI from Vink et al. (2007). Castro-Aguirre et al. (2016) reported cradle-to-gate GHG emissions of about 2.8 kg CO_2e/kg of PLA based on Ecoinvent v3.2, while Carus (2017) reported cradle-to-gate GHG emissions of 2.4 kg CO_2e/kg for PLA based on Vink and Davies (2015). These studies involve feedstock and conversion emissions. They did not mention taking into account biogenic credits, and only Carus (2017) included plastic processing and manufacturing. Therefore, if the PLA GHG emissions presented here are compared for only the feedstock and conversion steps (3.0 kg CO_2e/kg from Fig. 3), they are in agreement with the studies mentioned above. The emissions related to processing and manufacturing are similar for all types of plastics, since the same input values were used for all pathways (Table 2). Both injection molding and extrusion are popular as transformation techniques for plastics like PE and PLA (Castro-Aguirre et al., 2016); therefore, a 50% share for each technology was assumed. Note that the impact of using these technologies is presented separately in Fig. 4.

Fig. 4a presents different sensitivity cases for PLA life cycle GHG emissions. As presented in Section 2.4, there are significant variations in the biodegradability of PLA depending on the conditions and data sources; for example, different temperature, moisture, oxygen requirement, and type of microorganism might increase or decrease the PLA biodegradation rate. Therefore, the first two cases refer to the different percentages of biodegradability (%BD) in landfill and composting scenarios based on the information presented in Table 3. In the landfill scenarios for PLA, a lower bound of 0% BD (no biodegradation) and an upper bound of 85% BD are assumed. For composting scenarios, 10% BD and 85% BD were chosen. Fig. 4a shows how biodegradability greatly influences PLA GHG results. For example, it can be seen that as the percentage of biodegradation increases, the total GHG emissions also increase, from a minimum of 1.7 g CO_2e/kg for 0% BD to a maximum of 4.5 g CO_2e/kg with 85% BD in landfill scenarios. However, for composting scenarios, the total GHG emissions are slightly lower than those from the landfill scenarios, with GHG emission values ranging from 2 to 4 g CO_2e/kg of PLA for 10% BD and 85% BD, respectively. One of the reasons for this is that less CH_4 is released from biogas generated during composting (5% of biogas is CH_4) than is released from LFG, which it is assumed to be 50% of the landfill biogas with a 75% collection efficiency. The share of CH_4 in the biogas is important for determining the GHG emissions, because CH_4 has 30 times more GWP than CO_2. Although biodegradation of PLA can be seen as an advantage and an opportunity for plastic waste management options, GHG emissions released to the environment on its degradation into CO_2 and CH_4 could be a disadvantage from the environmental sustainability point of view. Therefore, biodegradability is an important property to consider when evaluating GHG emissions of biodegradable bioplastics on a cradle-to-grave basis.

Fig. 4a also presents a sensitivity case for PLA GHG emissions with regard to LFG collection efficiency. The impact of lower and higher LFG collection efficiency on PLA GHG emissions was evaluated. There are different LFG collection scenarios, depending on the landfill operation conditions, according to the U.S. EPA's WARM model (U.S Environmental Protection Agency, 2019). For this analysis, 79% was used as the highest LFG collection efficiency, corresponding to the California regulatory scenario, and 60% as the lowest, corresponding to *worst-case collection* in the WARM model. As shown in Fig. 4a, if a landfill has a collection efficiency lower than, for example, the typical 75% used in this paper as the base case, the GHG emissions of PLA will be higher than emissions in a landfill with a higher collection efficiency, due to more non-captured CH_4 emissions. The collection efficiency will depend on different factors, including when collectors are installed, location, and climate conditions (Lee et al., 2017).

The impacts of the regional electricity grid and plastics manufacturing techniques for all plastic pathways are also shown in Fig. 4. Electricity generation mix varies region by region within the U.S. For instance, on the West Coast, the electricity mix is mostly represented by low GHG-energy resources, such as hydroelectric, geothermal, solar, and wind. On the East Coast, on the other hand, coal and nuclear power are the main sources for electricity. For this analysis, the Western Electricity Coordinating Council (WECC) mix was chosen, representing 14 western states in the U.S. (California, Oregon, Washington, and others), and the Reliability First Corporation (RFC) mix, including New Jersey, Pennsylvania, and Maryland, among others. These are two sub-regions defined by the North America Electric Reliability Corporation (NERC, 2020). In the WECC mix, about 51% of the energy comes from renewable sources, while in the RFC mix, 39% and 30% come from coal and nuclear

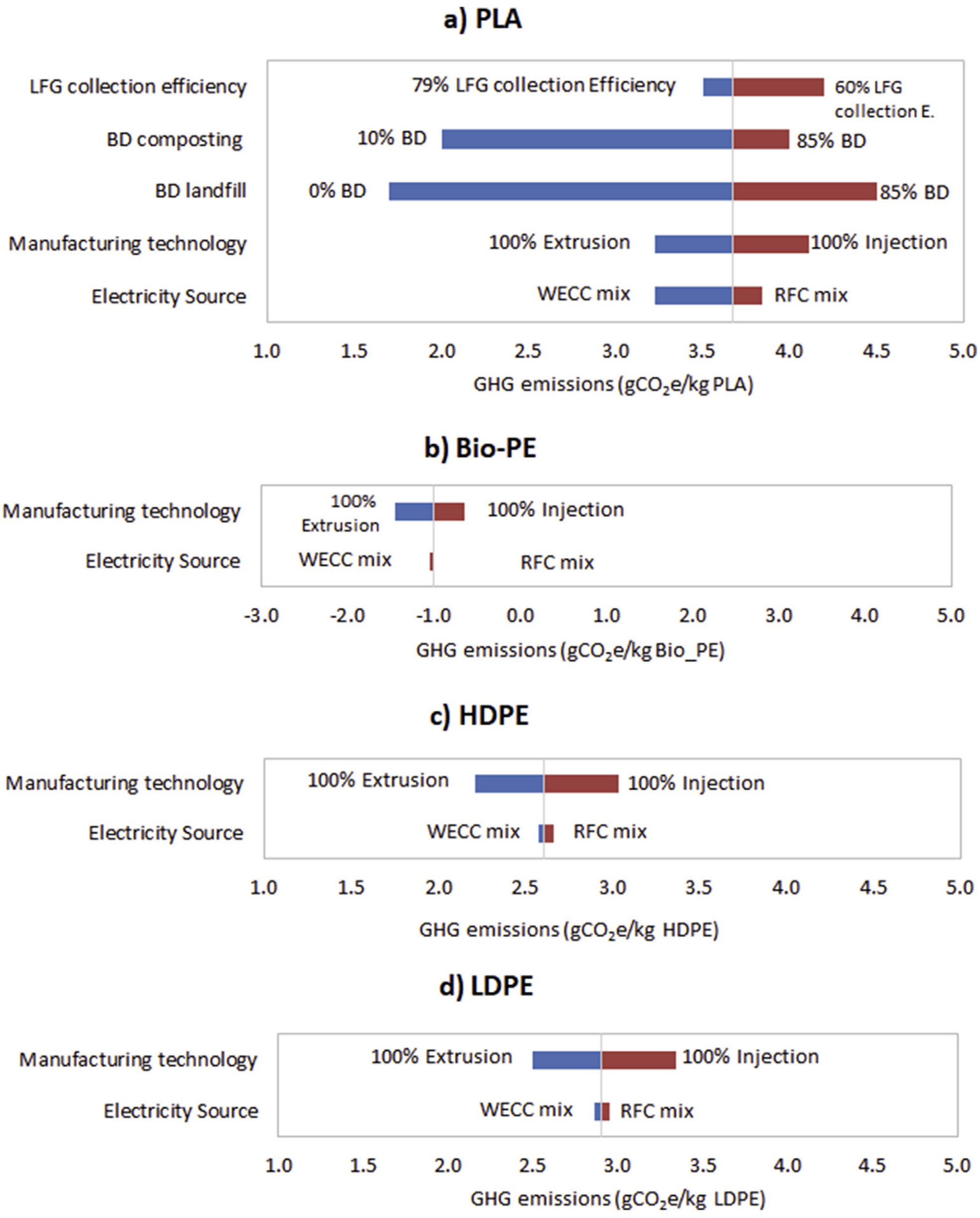


Fig. 4. Sensitivity cases for plastic pathways GHG emissions.

power resources, respectively. For the U.S. mix, the base case, 33% comes from natural gas, 29% from coal, and 20% from nuclear power (Argonne National Laboratory, 2019). The carbon intensities for the U.S. mix, RFC mix, and WECC mix are 483, 574, and 343 g CO2e/kWh of electricity, respectively (Argonne National Laboratory, 2019). For HDPE and LDPE, the GHG emissions decrease only 2% for both pathways when a more renewable-based electricity mix is used, compared to the base case, and increase only 1% in the case using the RFC electricity mix. For bio-PE, moving to either the WECC or RFC electricity mix will not have an effect on GHG emissions. However, for the PLA pathway, increasing the renewable energy resource in the electricity mix will decrease the GHG emissions by 12% compared to the case in which the U.S. electricity mix is used. This is because electricity is one of the major contributors to the GHG emissions in PLA production, while in the other plastic pathways natural gas consumption during conversion plays a more significant role than electricity.

Changing the transformation technique has a greater effect on the GHG emissions than the regional electricity grid scenario. When the transformation technique was switched from 50% injection molding and 50% extrusion to 100% one or the other, the GHG emissions increase or decrease depending on the technique used. For instance, the GHG emissions decrease 38%, 16%, 14%, and 12% for bio-PE, HDPE, LDPE, and PLA, respectively, when only extrusion was chosen as the transformation technique, compared to the base case, as shown in Fig. 3 for HDPE and LDPE, and assuming PLA landfill with 60% BD. The major reason why GHG emissions are reduced in this scenario is that extrusion requires less energy (electricity) to transform one kg of plastic than injection molding, according to the information provided in Table 2.

Finally, Fig. 5 compares FEC — the use of petroleum, natural gas, and coal — for all four plastics pathways. As in the GHG emission results, bio-PE has the lowest FEC value, followed by PLA, while LDPE and HDPE have the highest FEC values (73 and 79 MJ/kg,

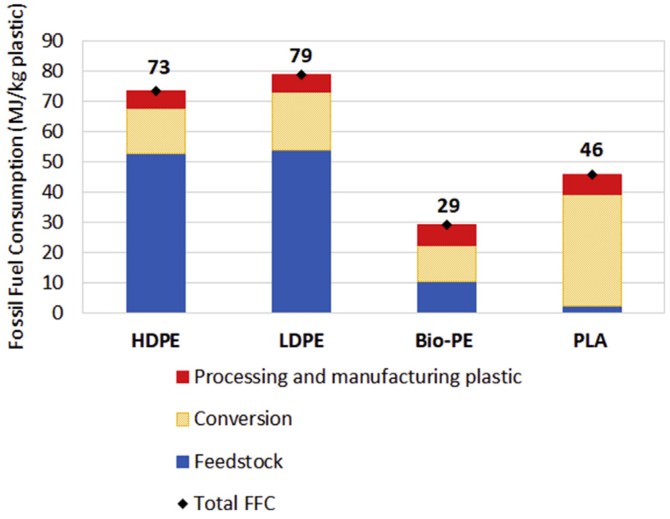


Fig. 5. Life cycle fossil energy consumption comparison of HDPE, LDPE, bio-PE and PLA.

respectively) because of their high fossil fuel dependency. As shown in Fig. 5, feedstock production has the largest FEC for both HDPE and LDPE, representing 72% and 68% of the total FEC for HDPE and LDPE, respectively. Natural gas contribution during feedstock production is about 44 MJ/kg of HDPE and 45 MJ/kg of LDPE, while crude oil only contributes 9 MJ/kg for both fossil-based plastics. Energy consumption for conversion for these plastics represents 20% and 25% of the total FEC for the HDPE and LDPE cases, respectively. Energy-intensive resources such as diesel, residual oil, gasoline, and LPG are used in the conversion of crude oil and natural gas into plastics (Argonne National Laboratory, 2019). In contrast, bioplastic bio-PE and PLA have low FEC values (29 and 46 MJ/kg, respectively) mainly because of their low FEC for feedstock production (10 and 2 MJ/kg, respectively). However, the FEC values are higher in the conversion step, contributing to 41% and 81% of the total FEC for bio-PE and PLA, respectively. In the case of PLA, this result is consistent with its high-energy-demand conversion process. For instance, natural gas consumption accounts for 59% of the fossil energy used during conversion, while 30% is due to electricity and the remaining 11% represents energy from coal, hydrogen and residual oil.

4. Conclusions

In this work, LCAs for bio-PE and PLA bioplastic were conducted and compared to the LCAs of two fossil—derived plastics (HDPE and LDPE). Bio-PE and PLA were chosen because of their great potential to displace fossil-based plastics in single-use and short shelf life applications — products often landfilled after use. The analysis included GHG contributions from feedstock production, conversion, and plastics manufacturing. In addition, the effects of landfilling or composting PLA on GHG emissions were investigated.

The results presented here compare the LCA metrics (GHG emissions and FEC) of four plastic pathways, showing the benefits of the bio-based plastic pathways over fossil-based, as bio-based plastics (bio-PE and PLA) use renewable carbon feedstocks to produce plastic material instead of fossil carbon feedstock, as in the case of HDPE and LDPE. However, when the biodegradability of PLA was considered in EOL scenarios, the life cycle emissions results for PLA changed significantly depending on the degradation conditions, increasing from 16% to 163% when different EOL strategies were compared to a landfill scenario with no biodegradation. EOL emissions were lower in composting scenarios than in landfill scenarios, because less CH_4 is emitted in the composting gas. An important parameter in landfill scenario was the LFG collection efficiency, which varies from landfill to landfill and depends on regulatory programs around the U.S. GHG emissions of PLA can improve if landfills have high collection efficiency. Therefore, biodegradability can greatly influence life cycle GHG emissions of bioplastics, such as PLA, that are designed to break down in anaerobic digestion or composting conditions, even if biogenic credits from carbon uptake during feedstock growth are included. It was also found that regional electricity mix does not have a great effect on the life cycle GHG emissions for HDPE, LDPE, and bio-PE pathways. In contrast, having a more renewable-based electricity mix will benefit the GHG emissions of producing PLA, because electricity is an important contributor to GHG emissions, as can be seen on the U.S. West Coast. One interesting factor analyzed in this work was the effect of transformation techniques to manufacture the plastics. Using a less energy-intensive processing method such as extrusion will benefit the GHG emissions of the plastic pathways studied in this work.

5. Key sources of uncertainty

The best data available was used for this analysis; however, it is important to mention that the results are not necessarily representative of the current state of technology in the plastic industry, especially newly developed plastics with characteristics designed to handle their EOL fate. Moving into a new era of plastics design, there will be more plastics designed with exclusive properties to better manage the EOL, and this analysis could be expanded to address the environmental impacts of the newly developed plastics in the circular economy. One interesting point for further analysis is to investigate the environmental impacts of new plastic pathways that do not depend on food crops as their feedstock source. For example, PLA is typically produced from corn (maize), which competes for land with food production. This is not the case for a second-generation feedstock, such as corn stover, or even waste feedstock, such as sludge from wastewater treatment plants. It is expected that the results of this study can be used to compare life cycle GHG emissions and FEC of various plastics, especially PLA, in different biodegradation conditions. However, plastic or bioplastic production inputs/outputs can be different, as feedstock type, production, and conversion technology can vary from pathway to pathway. It is also understandable that there is no single case that represents biodegradability; therefore, the results presented here do not represent the entire scenario for plastic degradation. Although the analysis presented here focused on the assessment of GHG emissions and FEC to provide some insights into the environmental effects of four plastic pathways, including two common bioplastics, there are other sustainability metrics that can be explored to help quantify the environmental benefits of bioplastic with a broader perspective, including EOL scenarios, which could be the subject of our future research. As we concluded in this paper, as biodegradable properties influence the life cycle emissions of PLA, waste management strategies for bioplastics will be one important issue in developing a plastic circular economy.

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CRediT authorship contribution statement

Pahola Thathiana Benavides: Conceptualization, Investigation, Formal analysis, Writing - original draft, Supervision. **Uisung Lee:** Conceptualization, Formal analysis, Writing - original draft. **Omid Zarè-Mehrjerdi:** Writing - original draft, Investigation, 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.

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