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Full length article

Valorisation of polylactic acid (PLA) waste: A comparative life cycle assessment of various solvent-based chemical recycling technologies

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ABSTRACT

To understand the environmental impacts of various chemical recycling solutions for handling end-of-life (EoL) post-consumer (PC) PLA waste, a comparative environmental assessment was conducted. A life cycle assessment (LCA) was applied the ISO standards 14040/44. The functional unit of the study was the treatment of one tonne PC-PLA waste. The four EoL scenarios investigated were: (1) hydrolysis, (2) alcoholysis using methanol, (3) alcoholysis using ethanol and (4) the direct incineration of PC-PLA waste. The scope of the study considers environmental impacts emanating from: (a) the collection and transport of the PC-PLA waste, (b) its pre-treatment, (c) respective EoL treatment for the production of recyclates and (e) the substitution of conventional products with recyclates. The results show that all three chemical recycling technologies perform better from an environmental perspective as compared to the direct incineration of the PLA waste. In particular, the chemical recycling processes using alcoholysis show considerable environmental benefits across various impact categories such as global warming, acidification, eutrophication, ionising radiation, photochemical ozone formation, resource use (energy carriers; mineral and metals) and respiratory inorganics. On the other hand, the valorisation of PLA to lactic acid via hydrolysis shows the highest benefits concerning land use and water scarcity. In the case of potential impacts on ozone depletion, alcoholysis-using methanol performs worst. Through the results obtained and subsequent discussions, this study establishes the need for instituting a recycling strategy for PLA to create a resource-efficient future involving different sectors along the value chain.

1. Introduction

Mismanaged plastic waste has attracted increasing attention across the world, leading to an influx of policies to combat plastic waste, that range from mandates to increase recycled content (Steensgaard et al., 2017) to an outright ban on single use plastics (Xanthos and Walker, 2017). Geyer (Geyer, 2020) estimates that around 343 million tonnes of plastic waste was generated worldwide in 2018, of which only 20% was recycled globally. With a a steep increase in the production of plastics that is expected to reach 1,100 million tonnes in the next 20 years (Geyer, 2020), development of cost-effective and scalable plastic waste recycling technologies is necessary. At the same time, it is equally important to reduce our reliance on fossil-based resources in order to lower the impact on climate change. One way to achieve this is to substitute conventional, fossil-based plastics with bio-based alternatives. However, studies show that significant greenhouse gas (GHG) savings can only be achieved if second-generation resources such as cellulosic crops and forest residues are used as feedstock for their production (Patel et al., 2018, Posen et al., 2017).

Currently, the global production of bioplastics stands around 2.1 million tonnes (European-Bioplastics, 2020) which accounts for less than 1% of the global plastics market. Currently, bio-plastics are predominantly produced from first generation feedstock such as corn, cane sugar, sugar beet, etc. Poly-lactic acid (PLA) is a promising bio-based polymer that can also be produced from second generation feedstock like corn stover, bagasse and straw (Alves De Oliveira, R. et al., 2020). The physical and chemical properties of PLA enable its application over a wide range of sectors such as disposable cutlery, packaging, 3D printing, textiles, drug delivery, implantable biomedical devices, etc. (DeStefano et al., 2020, Jamshidian et al., 2010).

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However, despite these advantages, the end-of-life (EoL) management of PLA-waste is a key area for research as the share of bioplastics (PLA in particular) is expected to grow in the near future. Furthermore, criticisms also include the lack of comprehensive life cycle assessments (LCA) that take both direct and indirect land use change (LUC) emissions into account (Escobar et al., 2018). In addition to this, converting rainforests, grasslands or other natural landscapes to cultivate feedstock for bio-based products can lead to a net GHG emissions of an order of 90 to 120 times higher than the current annual GHG savings achieved by replacing fossil-based products (Piemonte and Gironi, 2010). Hence, significant GHG reductions can only be achieved if non-food crops or waste residues (also referred to as second-generation bio-based feedstock) are used for the production of bioplastics and provided that, these plastics are duly recycled at their end-of-life (EoL). Use of second generation feedstock and/or appropriate recycling strategies (mechanical/chemical) at the end-of-life of the product can reduce the emissions from LUC and yield net positive environmental benefits (Piemonte and Gironi, 2012).

Different EoL treatment options for PLA waste include composting (Castro-Aguirre et al., 2018), mechanical recycling (Beltrán et al., 2019, Maga et al., 2019), solvent-based purification via the CreaSolv® process (Maga et al., 2019), pyrolysis (Cornelissen et al., 2008), hydrolysis (Siparsky et al., 1998), alcoholysis (McKeown and Jones, 2020, Petrus et al., 2016) and incineration (Maga et al., 2019). Though mechanical recycling promises the highest environmental benefits amongst all recycling strategies (Cosate de Andrade et al., 2016, Maga et al., 2019), it represents a finite recycling strategy, since the obtained recyclate has a lower quality than its virgin counterpart (Carné Sánchez and Collinson, 2011). Composting in general is not recommended option since it requires energy and does not allow for any material recovery (Rossi et al., 2015). In addition to this, PLA in particular, cannot be integrated in the conventional recycling infrastructure (Hong and Chen, 2017, Spierling et al., 2018). Thus, the inability to handle different grades of PLA while retaining their value in the supply chain (Cristina et al., 2016) coupled with the small market share of bio-based plastics (Spierling et al., 2018) poses a major challenge towards instituting pertinent mechanical recycling strategies for PLA.

The production of virgin PLA is highly raw material and energy intensive. To put this in perspective, according to Total Corbion PLA, every tonne of PLA produced at its production facility in Thailand consumes 19.5 kg sugarcane and utilizes 28.8 MJ energy (Morão and Bie, 2019). Fermentation of lactic acid and its subsequent purification primarily accounts for 79% of the total energy required for PLA production (Morão and Bie, 2019) and contributes to 40% of its total production cost (Cristina et al., 2016). This presents chemical recycling as a plausible option for recycling EoL PLA-waste from different value chains into its constituent monomer and/or other valuable chemicals. Though chemical recycling of plastics is a prevalent method of waste management, it is often touted as a “false solution” due to insufficient information about its operational performance and environmental impact (Rollinson and Oladejo, 2020, Spierling et al., 2018).

Therefore, the scope of this research involves a comprehensive life cycle assessment of two different chemical recycling strategies, namely alcoholysis and hydrolysis, instituted for recycling PLA-waste. These results are then compared with the environmental impacts resulting from direct incineration of PLA-waste which is the most common method of municipal waste management in Germany (Maga et al., 2019)

1.1. Technologies under study

Fig. 1 illustrates the different technologies investigated in this study, wherein, scenarios 1, 2 and 3 represent the chemical recycling of PLA-waste, while scenario 4 focusses on energy recovery through incineration.

Scenario 1 refers to the hydrolysis of PLA waste to lactic acid at 180°C and 1.5 MPa for a period of 50 mins. This results in the production of lactic acid (Piemonte et al., 2013). The reaction is autocatalytic and therefore, sustains itself on its own. However, the insolubility of PLA in water necessitates the use of large quantities of water, high temperature and elevated pressure (Grewell et al., 2014). This leads to racemization of the product. In addition to this, it has also been reported that, hydrolysis has not been proven to be an effective depolymerisation strategy for post-consumer PLA substrates (Spierling et al., 2018). These limitations strongly hinders its implementation on an industrial scale

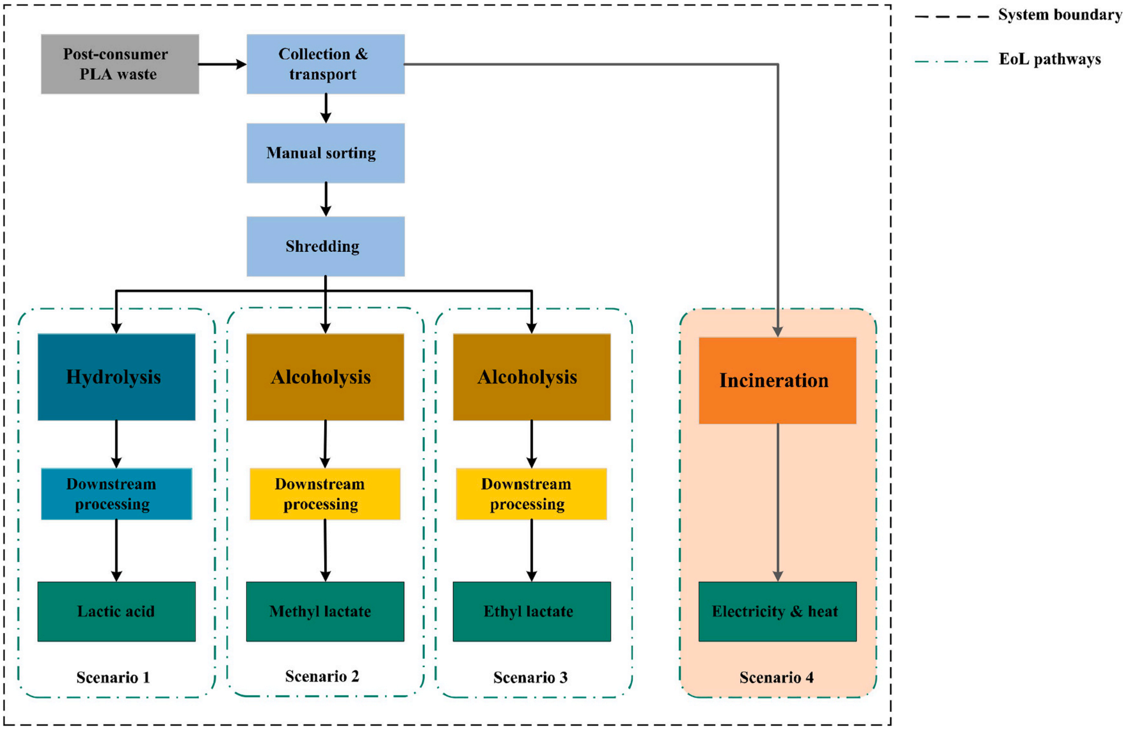


Fig. 1. Technologies under study.

(Spierling et al., 2018)

Scenario 2 represents the alcoholysis of PLA waste to produce methyl lactate. In this approach, chloroform is used as a solvent in order to dissolve the macro-molecular PLA, while methanol is employed as a nucleophile. The reaction is carried out at 57°C in the presence of stannous (II) octoate as a catalyst for seven hours in order to obtain the product, methyl lactate. (Anneaux et al., 2018). However, the use of a chlorinated, carcinogenic solvent and a toxic metal-based catalyst reduces the green credentials of this process.

Scenario 3 emphasises on a holistic strategy for recycling PC-PLA waste by upcycling it for the production of ethyl lactate. This involves the use of acetone as a relatively sustainable solvent as opposed to the previous attempt employing a toxic halogenated solvent (scenario 2). Bio-based ethanol which can also be procured from second-generation feedstock is used as a nucleophile. The reaction is carried out at 50°C and ambient pressure in the presence of an organic catalyst (1,5,7-Triazabicyclo[4.4.0]dec-5-ene) for a period of 30 min in order to obtain ethyl lactate as the product (Majgaonkar et al., 2021). This strategy was not only investigated for virgin PLA material but was also extended to other post-consumer PLA substrates such as, 3D printing filaments, yoghurt cups, etc.

2. Methodology for the life cycle assessment

The life cycle assessment (LCA) methodology applied in this study was structured and conducted according to the ISO 14040:2006 and ISO 14044:2006 standards (ISO, 2006a, ISO, 2006b). The LCA was modelled using the commercial software GaBi ts (version 10) and follows an attributional approach.

2.1. Goal and scope

The prime goal of this study was to analyse the environmental impacts of four end-of-life (EoL) handling and processing pathways instituted for post-consumer (PC) PLA waste. The investigated scenarios are presented in Table 1, wherein three novel solvent-based chemical recycling technologies are benchmarked against a typical thermal treatment. Scenario 1 evaluates the process developed by University of Rome (Piemonte et al., 2013), which uses water as a solvent to valorise PC-PLA waste into lactic acid. Scenario 2 explores the technology developed by Zeus Industrial Products Inc. (Anneaux et al., 2018), which uses chloroform as a solvent and stannous octoate as a catalyst to produce methyl lactate from PC-PLA waste. Scenario 3 analyses the technology developed by Fraunhofer ICT (Majgaonkar et al., 2021) to produce ethyl lactate from PC-PLA waste using acetone as a solvent and 1,5,7-Triazabicyclo[4.4.0]dec-5-ene as a catalyst. Scenario 4 deals with the treatment of the PC-PLA waste in a municipal waste incineration (MSWI) plant.

Although the three solvent-based recycling scenarios are on a similar technology readiness level (TRL 2 – TRL 4), it must be noted that a one-to-one comparison between them is not possible since the final product obtained from each of this scenario is different. However, all of them use PC-PLA waste as a primary input and consequently can be analysed for the potential environmental impacts resulting from its treatment. In addition to this, a further goal of this paper is to identify potential hotspots for future optimisation of these recycling technologies, especially considering the fact that they have a varying degree of tolerance to the impurities present in the feedstock.

2.1.1. Functional unit

The functional unit (FU) for this study is defined as "*the treatment of 1 tonne post-consumer PLA waste*". The selection of this functional unit allows for comparison of different EoL treatment scenarios based on PC-PLA waste as the primary input.

2.1.2. Geographical and technological scope

The geographical scope of the present study was limited to Europe. However, products based on PLA are produced and consumed across the world with the leading producer based in the United States with a nameplate capacity of 150,000 metric tonnes in 2020 (NatureWorks LLC, 2020) and the largest consumer being Taiwan with an import value of 37 million US dollars in 2016 (Plastics Insight, 2016). The solvent-based recycling technologies investigated in this study lie between the TRLs 2 and 4 and employ typical solvents, nucleophiles and catalysts. In order to allow a fair comparison to thermal treatment of PC-PLA waste, which occurs at an industrial scale, theoretically computed data was modelled prospectively in order to address the scale-up effects. With 170 nations pledging to "significantly reduce" the use of fossil-based plastic by 2030 (UNEP, 2019), the investigated technologies aimed at recycling bio-based plastics have a potential to be implemented across the globe.

2.1.3. System boundaries

The LCA follows an attributional approach focusing on the "EoL" stage. It considers the environmental impacts derived from (1) the collection and transport of the PC-PLA waste to a treatment plant, (2) pre-treatment operations, (3) respective EoL treatment, and (4) the production of recyclates (recycling output). The system boundary for all the three solvent-based recycling scenarios starts with the collection and transport of the PC-PLA waste to a pre-treatment facility for sorting and shredding, following which, this pre-treated waste is subjected to solvolysis (see Fig. 2). For scenarios 1, 2 and 3, the extended system boundary includes the application of recyclates, i.e., lactic acid, methyl lactate and ethyl lactate for the substitution of equivalent reference products. It is assumed that lactic acid (scenario 1) can replace virgin lactic acid obtained from biomass, while the solvents, methyl lactate (scenario 2) and ethyl lactate (scenario 3), can replace N,N'dimethylformamide (DMF).

For the reference scenario (incineration), the collected waste was assumed to be directly transported to a MSWI facility (see Fig. 3). The extended system boundary includes utilising the products obtained from incineration products, i.e., electricity and heat, for substituting equivalent fossil-based energy sources representing an average European mix.

2.1.4. Data collection and data quality

The primary data required in order to establish a mass balance for the investigated recycling technologies was evaluated from previous results disseminated in open source literature for scenarios 1 and 2 (Anneaux et al., 2018, Piemonte et al., 2013). The technology developer (Fraunhofer Institute for Chemical Technology) obtained a similar dataset through experimental investigations for scenario 3 (Majgaonkar et al., 2021) (see Tab.A.1, Tab.A.5 and Tab. A.13 in the supplementary material). In order to establish a prospective LCA, the energy demand for the different recycling strategies investigated in this study was evaluated

Table 1
Investigated PLA waste recycling scenarios.

Scenarios	Process	Nucleophile	Solvent	Catalyst	Final product from recycling
1	Hydrolysis	-	Water	Autocatalytic	Lactic acid
2	Alcoholysis	Methanol	Chloroform	Stannous octoate	Methyl lactate
3	Alcoholysis	Ethanol	Acetone	1,5,7-Triazabicyclo[4.4.0]dec-5-ene (TBD)	Ethyl lactate
4	Incineration	-	-	-	Heat, electricity

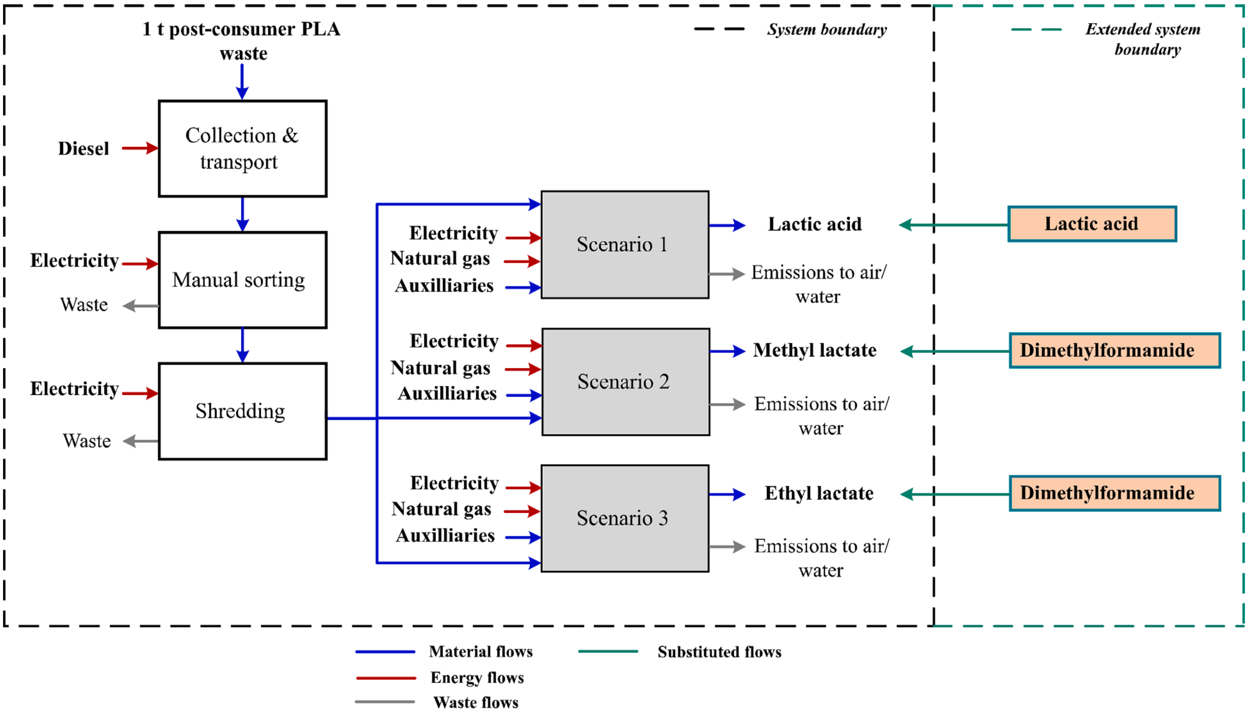


Fig. 2. System boundary of scenario 1, 2 and 3.

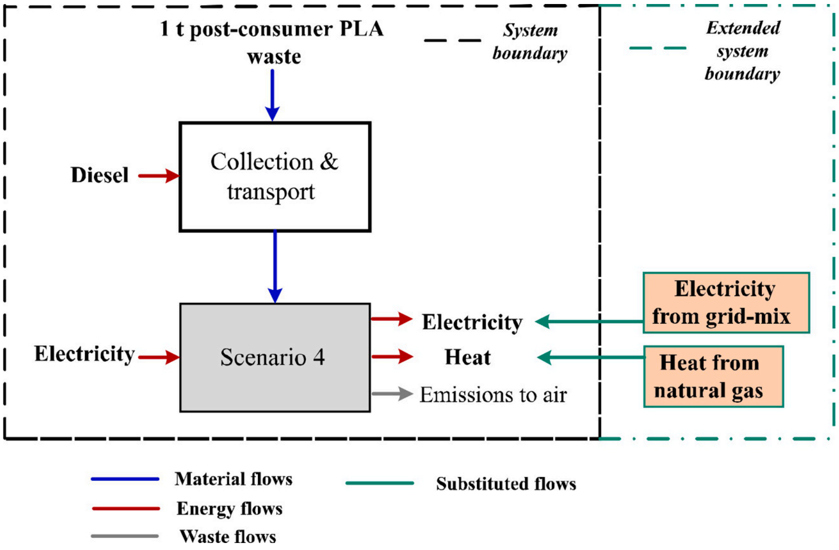


Fig. 3. System boundary of scenario 4.

based on the energy required for production (reaction) followed by subsequent purification (down streaming). The energy required for the former step was evaluated theoretically by calculating the heat input required to increase the temperature of the reaction mixture and that required to maintain the temperature until the completion of the reaction (see Tab.A.2, Tab.A.6 and Tab.A.14 in the supplementary information). In order to determine the energy required for down streaming, process simulations were carried out in Aspen Plus^{TM}. A single distillation column was needed for the binary separation in scenario 1 (see Tab. A.3, tab.A.4). Formation of a low boiling azeotrope amongst the components present in the resulting reaction mixture necessitated the use of a sequence of three distillation columns for scenario 2. (see Tab. A.7-A.12). Scenario 3 required a sequence of two distillation columns in order to affect the ternary separation between the product, solvent and the unreacted nucleophile. (see Tab.A.15-A.18).

The background data used for conducting the LCA analysis was based on the GaBi database GaBi (thinkstep, 2020). For few exceptional cases where no representative data was available in the GaBi database, suitable datasets from the ecoinvent database (ecoinvent v. 3.6, cut-off system model) were used.

2.1.5. *Choice of reference system*

In this study, PC-PLA waste was assumed to be obtained from disposable cutlery (e.g. drinking cups and food plates) used at festivals and other recreational events. This waste is typically directed to MSWI plants, either with or without energy recovery. As a conservative estimation, we assumed that 98% of the PC-PLA waste is PLA, while the remaining fraction resembles/is comprised of conventional municipal waste. For scenario 4 (direct incineration of PC-PLA waste), we chose the Gabi-dataset "EU-28: Polylactic acid (PLA) in a waste incineration plant" which represents the incineration of 100% PLA assuming a net calorific value of 17.9 MJ/kg. The total (biogenic) carbon content of the PLA is defined as 0.5 kg biogenic carbon/kg product. The incineration takes place in a waste-to-energy plant with dry flue gas cleaning and selective catalytic reduction for NO_x-removal. The efficiency of energy recovery is assumed to be 38% of the heating value of PLA. 28% of this energy output is electricity while the remaining 72% is steam. The recovered energy is assumed to replace the European-electricity grid mix and the European-district heating mix. As background data for the treatment of the 2% municipal waste, the thinkstep dataset "EU-28: Municipal waste in waste incineration plant" was chosen which also considers energy recovery.

2.1.6. *Multifunctionality*

The issue of multifunctionality is needed to be addressed when a system produces multiple product outputs or uses inputs that originate from another product life cycle. In this study, PC-PLA waste is used as main input for all EoL treatment scenarios. It was assumed that the waste has no environmental burdens, i.e., zero burden approach (Ekvall et al., 2007), since all burdens are allocated to the PLA product itself. In the case of thermal treatment of process residues or complete incineration of the PC-PLA waste (reference scenario), system expansion was applied and credits were given for replacing electricity and thermal energy. In addition, credits were also assigned to the final products obtained from the three chemical recycling strategies.

2.1.7. *Impact assessment methodology*

The life cycle impact assessment (LCIA) methods chosen and applied in this study are based on the recommendations published by the International Reference Life Cycle Data System (ILCD) and the Product Environmental Footprint (PEF) method developed by the European Commission (EC) (EC, 2013, JRC, 2010). With regards to global warming, the global warming impact (GWI) was calculated considering a 100 year timeframe, by using the characterization factors published by the fifth assessment report of the Intergovernmental Panel on Climate Change IPCC (IPCC, 2014).

The selected impact categories are terrestrial and freshwater acidification (A), global warming (GWI) including and excluding biogenic carbon, freshwater, marine and terrestrial eutrophication (EU_F, EU_M, EU_T), ionising radiation (IR), land use (LU), ozone depletion (OD), photochemical ozone formation (POF), resource use for energy carriers (RU_E), resource use for minerals and metals (RU_M), respiratory inorganics (RI) and water scarcity (WS). Environmental Footprint EF 3.0 was used for calculating the environmental impacts.

3. Systems under study and life cycle inventories

In this section, the various investigated EoL scenarios for the handling of PC-PLA waste, along with their respective life cycle inventory data is provided in detail.

3.1. *Pre-treatment of end-of-life PC-PLA waste*

After collection, the PLA waste was assumed to be transported to a treatment facility for initial sorting. The sorting was done manually to remove impurities. Table 2 provides an overview on the composition of the collected and analysed PC-PLA waste from a music festival in the year 2019. The waste stream was relatively clean with about 98.8% PLA and 1.2% impurities. The PLA fraction primarily consisted of drinking cups, while the impurities represented a mixed fraction of paper, plastic, metals and other inert substances.

Although manual sorting was performed in a laboratory for the purpose of this study, continuous waste treatment on an industrial scale can involve higher amounts of impurities, thereby, necessitating the use of near infrared (NIR)- based sorting techniques (Zheng et al., 2018). In further steps, the sorted PLA waste fraction is washed with a 2% aqueous NaOH solution at 70°C and is dried. This results in the removal of remnant impurities (e.g. imprints, printing inks, etc.) that could not be separated by the previous steps. This is followed by the shredding of the dried PC-PLA waste which is a final step in its pre-treatment. It was assumed that, chemical recycling of PC-PLA waste as investigated in scenarios 1, 2 and 3 is preceded by the aforementioned common pre-treatment step.

Table 2
Composition of one tonne post-consumer PLA waste collected (Ginter, 2019).

Material	Weight in kg
PLA (cups and plates)	988
Paper (mainly cellulosic products like handkerchiefs and napkins but also bottle labels and tickets)	4.68
Plastic (drinking straws and packaging films made out of PE and PP and caps and beverage bottles made out of PET)	3.12
Metal (crown caps)	1.92
Other impurities (cigarette packets, leaves, soil, fruit peels and food leftovers)	2.28

For scenario 4 (direct incineration of PC-PLA waste), there was no requirement for any pre-treatment. It was assumed that the collected PLA waste is transported by a truck driven by diesel engine from the collection point to the thermal treatment plant. Table 3 provides the inventory data for the pre-treatment of PC-PLA waste for all three solvent-based recycling scenarios. Table 4 shows the inventory data for the collection and transportation of PC-PLA waste as addressed in scenario 4.

Table 3
Assumptions and data used for pre-treatment of post-consumer PLA waste for scenarios 1, 2 and 3.

Mass/energy flow or process	Amount	Unit	Foreground data source	LCI data set
INPUT FLOWS				
EoL PLA waste	1000	kg	FU (assumed)	
Transport of PLA waste to pre-treatment facility	25	km	assumed	GLO: Truck PE (payload - 11.4 tonnes)
Diesel demand for PLA waste collection and transportation	1.98	kg	amount of fuel calculated with GaBi ts	EU-28: Diesel mix at filling station ts
Water demand for washing	200	kg/t_{PLA}	measured	EU-28: Water (deionised) ts
Washing agent demand (2% aq. NaOH solution at 70°C)	4	kg/t_{PLA}	measured	2% aq. NaOH solution <u-so>
Thermal energy demand for drying	43	kWh/t_{PLA}	(Maga et al., 2019, NEUE HERBOLD, 2017)	EU-28: Process steam from natural gas 95% ts
Electricity demand for shredding	63.53	kWh/t_{PLA}	measured	EU-28: Electricity grid mix (average power plants) ts
OUTPUT FLOWS				
Residues after sorting stage	12	kg	measured	EU-28: Municipal waste in waste incineration plant ts
Residues after washing stage	27	kg	measured	EU-28: Disposal of plastics (landfill/incineration) ts
Residues after shredding stage	18	kg	measured	
Net amount of PLA waste after pre-treatment	943	kg	calculated	

Table 4
Assumptions and data used for collection and transportation of post-consumer PLA waste for scenario 4.

Mass/energy flow or process	Amount	Unit	Foreground data source	LCI data set
INPUT FLOWS				
EoL PLA containing waste	1000	kg	FU (assumed)	
Transport of PLA waste to the municipal waste incineration (MSWI) plant	25	km	assumed	GLO: Truck PE (payload - 11.4 tonnes)
Diesel demand for PLA waste collection and transportation	1.98	kg	amount of fuel calculated with GaBi ts	EU-28: Diesel mix at filling station ts
OUTPUT FLOWS				
PLA waste for incineration	1000	kg	Calculated	

3.2. *Scenario 1 – hydrolysis of pre-treated PC-PLA waste*

Scenario 1 involves the hydrolysis of PC-PLA waste at high temperature and is based on the technology developed by the University of Rome (Piemonte et al., 2013). The product obtained from the reaction is the monomer, lactic acid. The reaction is autocatalytic and has a yield of approximately 1225 kg lactic acid per tonne of processed PLA waste. The inventory data for recycling PLA to produce lactic acid is shown in Table 5.

3.3. *Scenario 2 – alcoholysis of pre-treated PC-PLA waste*

Scenario 2 involves the alcoholysis of PC-PLA waste at 57°C to produce methyl lactate and is based on the technology developed by Zeus Industrial Products Inc. (Anneaux et al., 2018). The reaction yields about 1444 kg methyl lactate per tonne of processed PLA waste. The solvent used is chloroform while the catalyst employed is stannous octoate. Methanol is used as a nucleophile for this reaction. The inventory data for recycling PLA to produce the methyl lactate is shown in Table 6.

3.4. *Scenario 3 – alcoholysis of pre-treated PC-PLA waste*

Table 5
Assumptions and data used for Scenario 1.

Mass/energy flow or process	Amount	Unit	Foreground data source	LCI data set
INPUT FLOWS				
PLA waste after pre-treatment	943	kg	measured	
Electricity demand for recycling	11	kWh/t_{PLA}	measured and scaled-up	EU-28: Electricity grid mix (average power plants) ts
Thermal energy demand for recycling	5406	MJ/t_{PLA}	measured and scaled-up	EU-28: Process steam from natural gas 95% ts
Solvent demand for recycling | Solvent: Water	5000	kg/t_{PLA}	measured and scaled-up	EU-28: Water deionised ts
Thermal energy demand for the entire downstream processing	15960	MJ/t_{PLA}	measured and scaled-up	EU-28: Process steam from natural gas 95% ts
OUTPUT FLOWS				
Spent solvent for reuse	4900	kg/t_{PLA}	calculated	EU-28: Water deionised ts
Amount of lactic acid produced	1225.18	kg/t_{PLA}	measured	EU-28: Lactic acid ts

Table 6
Assumptions and data used for Scenario 2.

Mass/energy flow or process	Amount	Unit	Foreground data source	LCI data set
INPUT FLOWS				
PLA waste after pre-treatment	943	kg	measured	
Electricity demand for recycling	11	kWh/t_{PLA}	measured and scaled-up	EU-28: Electricity grid mix (average power plants) ts
Thermal energy demand for recycling	1848	MJ/t_{PLA}	measured and scaled-up	EU-28: Process steam from natural gas 95% ts
Solvent demand for recycling | Solvent: Chloroform	5730	kg/t_{PLA}	measured and scaled-up	RER: trichloromethane production ecoinvent 3.6
Catalyst demand for recycling | Catalyst: Tin(II) 2-ethylhexanoate	10	kg/t_{PLA}	measured and scaled-up	
Nucleophile demand for recycling | Nucleophile: Methanol	2370	kg/t_{PLA}	measured and scaled-up	EU-28: Methanol mix ts
Thermal energy demand for the entire downstream processing	8523	MJ/t_{PLA}	measured and scaled-up	EU-28: Process steam from natural gas 95% ts
OUTPUT FLOWS				
Spent solvent for reuse	5615.4	kg/t_{PLA}	calculated	RER: trichloromethane production ecoinvent 3.6
Spent catalyst for reuse	10	kg/t_{PLA}	calculated	
Spent nucleophile for reuse	2322.6	kg/t_{PLA}	calculated	EU-28: Methanol mix ts
Amount of methyl lactate produced	1444.62	kg/t_{PLA}	measured	

Scenario 3 involves alcoholysis of PC-PLA waste to produce ethyl lactate at 50°C and is based on the technology developed by Fraunhofer Institute for Chemical Technology (Majgaonkar et al., 2021) This strategy results in the production of about 1330 kg ethyl lactate per tonne of processed PC-PLA waste. The solvent used is acetone and the catalyst employed is 1,5,7-Triazabicyclo[4.4.0]dec-5-ene (TBD). However, though TBD is less toxic than zinc acetate, the latter was chosen in order to model this scenario owing to the lack of inventory data pertaining to TBD. Ethanol is used a nucleophile for this reaction. The inventory data for the recycling of PLA to produce ethyl lactate is summarized in Table 7.

3.5. *Scenario 4 – incineration of pre-treated PC-PLA waste*

Scenario 4 addresses the thermal treatment (direct incineration) of PC-PLA waste. It involves the transportation of collected PC-PLA waste by a truck driven on diesel engine from the point of collection to the thermal treatment plant for incineration. The energy (heat and electricity) recovered from thermal treatment of the PLA waste was assumed to substitute the energy required by an average European heating grid. Hence, the environmental benefits resulting from the avoided burdens arising from the use of fossil fuels for conventional energy production were included in the LCA as credits. The datasets used to assign the heat and electricity credits in scenario 4 are shown in Table 8.

4. Life cycle impact assessment – results and interpretation

4.1. *Comparison of global warming impact*

The GHG emissions resulting from biogenic sources including the

Table 7
Assumptions and data used for Scenario 3.

Mass/energy flow or process	Amount	Unit	Foreground data source	LCI data set
INPUT FLOWS				
PLA waste after pre-treatment	943	kg	measured	
Electricity demand for recycling	11	kWh/ t_{PLA}	measured and scaled-up	EU-28: Electricity grid mix (average power plants) ts
Thermal energy demand for recycling	627	MJ/ t_{PLA}	measured and scaled-up	EU-28: Process steam from natural gas 95% ts
Solvent demand for recycling | Solvent: Acetone	5000	kg/ t_{PLA}	measured and scaled-up	EU-28: Acetone ts
Catalyst demand for recycling | Catalyst: Zinc acetate	20	kg/ t_{PLA}	measured and scaled-up	
Nucleophile demand for recycling | Nucleophile: Ethanol	2040	kg/ t_{PLA}	measured and scaled-up	EU 28: Bioethanol from sugar beet, at filling station
Thermal energy demand for the entire downstream processing	5550	MJ/ t_{PLA}	measured and scaled-up	EU-28: Process steam from natural gas 95% ts
OUTPUT FLOWS				
Spent solvent for reuse	4900	kg/ t_{PLA}	calculated	EU-28: Acetone ts
Spent catalyst for reuse	10	kg/ t_{PLA}	calculated	
Spent nucleophile for reuse	1999.2	kg/ t_{PLA}	calculated	
Amount of ethyl lactate produced	1329.45	kg/ t_{PLA}	measured	

Table 8
Assumptions and data used for Scenario 4.

Mass/energy flow or process	Amount	Unit	Foreground data source	LCI data set
INPUT FLOWS				
PLA waste to incineration	988	kg	Calculated	EU-28: Polylactic acid (PLA) in waste incineration plant ts
Municipal waste to incineration	12	kg	Calculated	EU-28: Municipal waste in waste incineration plant
OUTPUT FLOWS				
Credits for electricity produced from incineration	718.3	kWh	Calculated according to the used GaBi data sets	EU-28: Electricity grid mix (average power plants) ts
Credits for heat produced from incineration	4655	MJ	Calculated according to the used GaBi data sets	EU-28: District heating mix ts

fixation of carbon in biomass from all four investigated scenarios are represented in Fig. 4. The results can be further categorised in two parts, namely, the actual emissions generated from the core recycling process (including collection, transportation, pre-treatment and production) and the normalised emissions obtained after incorporating the credits obtained from substitution of equivalent reference products. Table 9 provides an overview of the final products produced from the different recycling scenarios, their applications and their targeted substitution.

If GHG emissions from the core recycling process are considered, scenario 4 has the highest GHG emissions, amounting to 1834 kg CO_2-eq./FU across all scenarios. This can be attributed to the fact that the collected PC-PLA waste is directly subjected to incineration, owing to which the biogenic carbon contained in PLA is subsequently released into the environment. If scenarios 1, 2 and 3 aimed at chemical recycling of PC-PLA waste are compared, it can be observed that scenario 3 has the lowest impact at - 49 kg CO_2-eq./FU, This can be credited to the overall savings attained from the use of bio-based ethanol as a nucleophile. In case of scenario 1, the GHG emissions stood at 1553 kg CO_2-eq./FU, primarily stemming from the high energy demand required for downstreaming. Scenario 2 was found to have the highest GHG emissions of about 1721 kg CO_2-eq./FU on account of the high energy demand as well as the use of a fossil-based nucleophile (methanol).

For the expanded system, credits or avoided burdens associated with the substitution of the reference products by the products obtained from the investigated scenarios were considered. In the expanded system, scenario 3 still has the highest GHG savings amounting to - 2910 kg CO_2-eq./FU followed by scenario 2 at - 1390 kg CO_2-eq./FU. Contrary to this, scenario 1 has the highest emissions at 1670 kg CO_2-eq./FU, which is even higher than scenario 4 with net GHG emissions equivalent to 1250 kg CO_2-eq./FU. This can be attributed to the fact that the reference products in scenario 2, 3 (DMF) and scenario 4 (energy) that are substituted contribute to GHG reductions while the substitution of virgin lactic acid in scenario 1 leads to additional GHG emissions due to its negative net GWI. In addition to this, the use of the bio-based nucleophile (ethanol) in scenario 3 accounts for additional GHG savings. In contrast to this, incineration of PLA waste as addressed in scenario 4 leads to higher GHG emissions than GHG savings that is realised from the recovered energy.

4.2. *Comparison of impact assessment results of all scenarios*

The impact assessment results calculated in this study are summarised as a heat map in Table 10. The lowest and the highest net impacts generated from the EoL treatment of each of the four scenarios are highlighted in green and red respectively. The alcoholysis of PC-PLA waste (scenarios 2 and 3) shows clear benefits as compared to its hydrolysis (scenario 1) or incineration (scenario 4). The hydrolysis of PLA performs better than its incineration in some impact categories such as acidification, freshwater and terrestrial eutrophication, land use, photochemical ozone formation, respiratory inorganics and water scarcity. The impacts in these categories can be mainly attributed to the agricultural activities needed for the conventional production of lactic acid. However, the alcoholysis of PLA using methanol as a nucleophile (scenario 2) shows the lowest net environmental impacts in several categories, namely acidification, eutrophication, global warming excluding biogenic carbon, ionising radiation, photochemical ozone formation, resource use (energy carriers; mineral and metals) and respiratory inorganics. On the contrary, the alcoholysis of PLA using ethanol (scenario 3) shows similar benefits, while performing best in the categories focussed on global warming including biogenic carbon, ozone depletion and resource use (energy carriers, minerals and metals). Since the utilisation and the EoL phases of the recyclates (e.g. lactic acid, methyl lactate, ethyl lactate) produced in scenarios 1-3 is out of the scope of this study, the GWI including the influence ofthe fixation of CO_2 by biomass presents a more accurate picture. Accordingly, alcoholysis of PLA using ethanol (scenario 3) shows considerable advantages with regards to global warming.

Fig. 5 illustrates the gross emissions resulting from each EoL option, the savings realized by their substitution potential as well as the net emissions. As represented in Fig. 5, the net emissions, strongly depend on the product that is substituted. In particular, the substitution of the solvent DMF by either ethyl or methyl lactate results in clear benefits in several impact categories, namely acidification, freshwater, marine and terrestrial eutrophication, global warming, ionising radiation, photochemical ozone formation, resource use (energy carriers, mineral and metals), respiratory inorganics and water scarcity.

It is particularly interesting to note that scenario 3 shows the highest

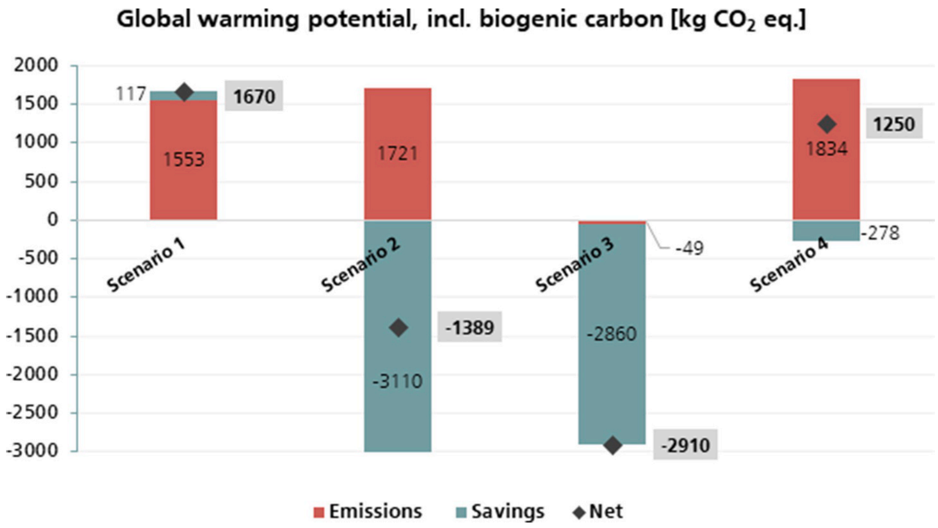


Fig. 4. Greenhouse gas emissions from the investigated scenarios.

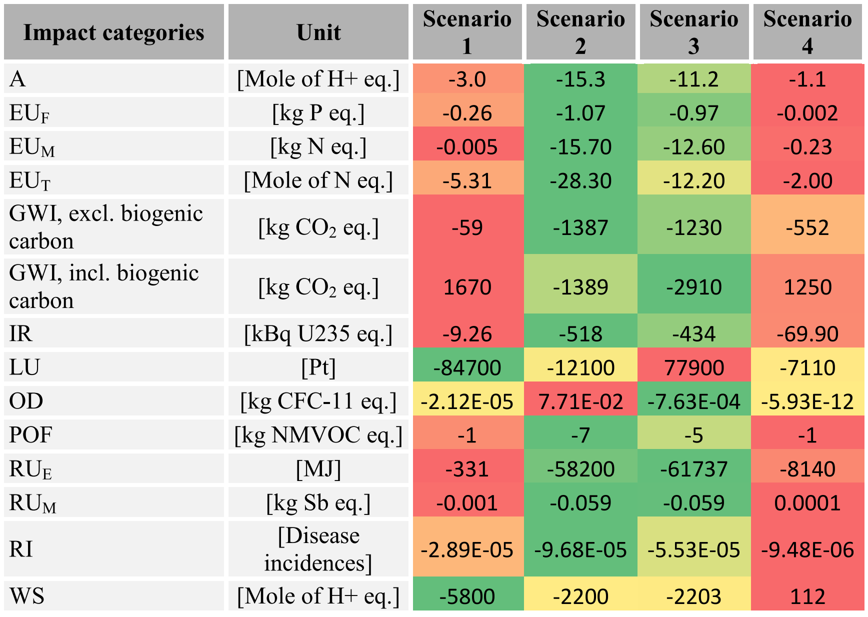
Table 9
Recycling products, their applications and the targeted substituted products.

Scenarios	Final product	Product yield per tonne PLA waste	Application	Targeted substitution
1	Lactic acid	1225	Polymer precusor	Lactic acid
2	Methyl lactate	1444	Solvent	N,N-dimethylformamide
3	Ethyl lactate	1329	Solvent	N,N-dimethylformamide
4	Heat, electricity	LHV: 17.9 MJ/kg	Utilitiy	District heating mix, Electricity from grid

impact in land use since the production of ethyl lactate from PC-PLA waste requires significant quantities of bioethanol, and is further assumed to replace only fossil-based solvents. Contrary to this, the hydrolysis of PC-PLA waste (scenario 1) leads to the lowest impact on land use since the virgin production of lactic acid from biogenic feedstock is substituted. Moreover, the hydrolysis of PLA does not require any biogenic inputs which further contributes to low impacts on land use. In addition to this, the alcoholysis of PC-PLA waste using methanol (scenario 2) as well as the incineration of PC-PLA waste (scenario 4) hardly influences the land use since neither biogenic inputs are required nor biogenic products are substituted. The high contribution of scenario 2 to ozone depletion should be particularly noted. This results from the use of chloroform in the process. The hydrolysis of PC-PLA waste contributes to the highest reduction in water scarcity since the production of virgin lactic acid from sugar is avoided.

Figs. 6-9 show a detailed contribution analysis of various process stages along the respective recycling scenarios. For scenarios 1-3, the investigated stages include (a) collection and pre-processing of the PC-PLA waste, (b) washing, drying and shredding, (c) recycling of the PC-PLA waste, (d) downstream processing to recover the product from other process residues, (e) credits for substituting equivalent reference

Table 10
Heat map of the LCIA results for all investigated scenarios (see Section 2.1.7 for impact categories).



Impact categories	Unit	Scenario 1	Scenario 2	Scenario 3	Scenario 4
A	[Mole of H+ eq.]	-3.0	-15.3	-11.2	-1.1
EU$_F$	[kg P eq.]	-0.26	-1.07	-0.97	-0.002
EU$_M$	[kg N eq.]	-0.005	-15.70	-12.60	-0.23
EU$_T$	[Mole of N eq.]	-5.31	-28.30	-12.20	-2.00
GWI, excl. biogenic carbon	[kg CO_2 eq.]	-59	-1387	-1230	-552
GWI, incl. biogenic carbon	[kg CO_2 eq.]	1670	-1389	-2910	1250
IR	[kBq U235 eq.]	-9.26	-518	-434	-69.90
LU	[Pt]	-84700	-12100	77900	-7110
OD	[kg CFC-11 eq.]	-2.12E-05	7.71E-02	-7.63E-04	-5.93E-12
POF	[kg NMVOC eq.]	-1	-7	-5	-1
RU$_E$	[MJ]	-331	-58200	-61737	-8140
RU$_M$	[kg Sb eq.]	-0.001	-0.059	-0.059	0.0001
RI	[Disease incidences]	-2.89E-05	-9.68E-05	-5.53E-05	-9.48E-06
WS	[Mole of H+ eq.]	-5800	-2200	-2203	112

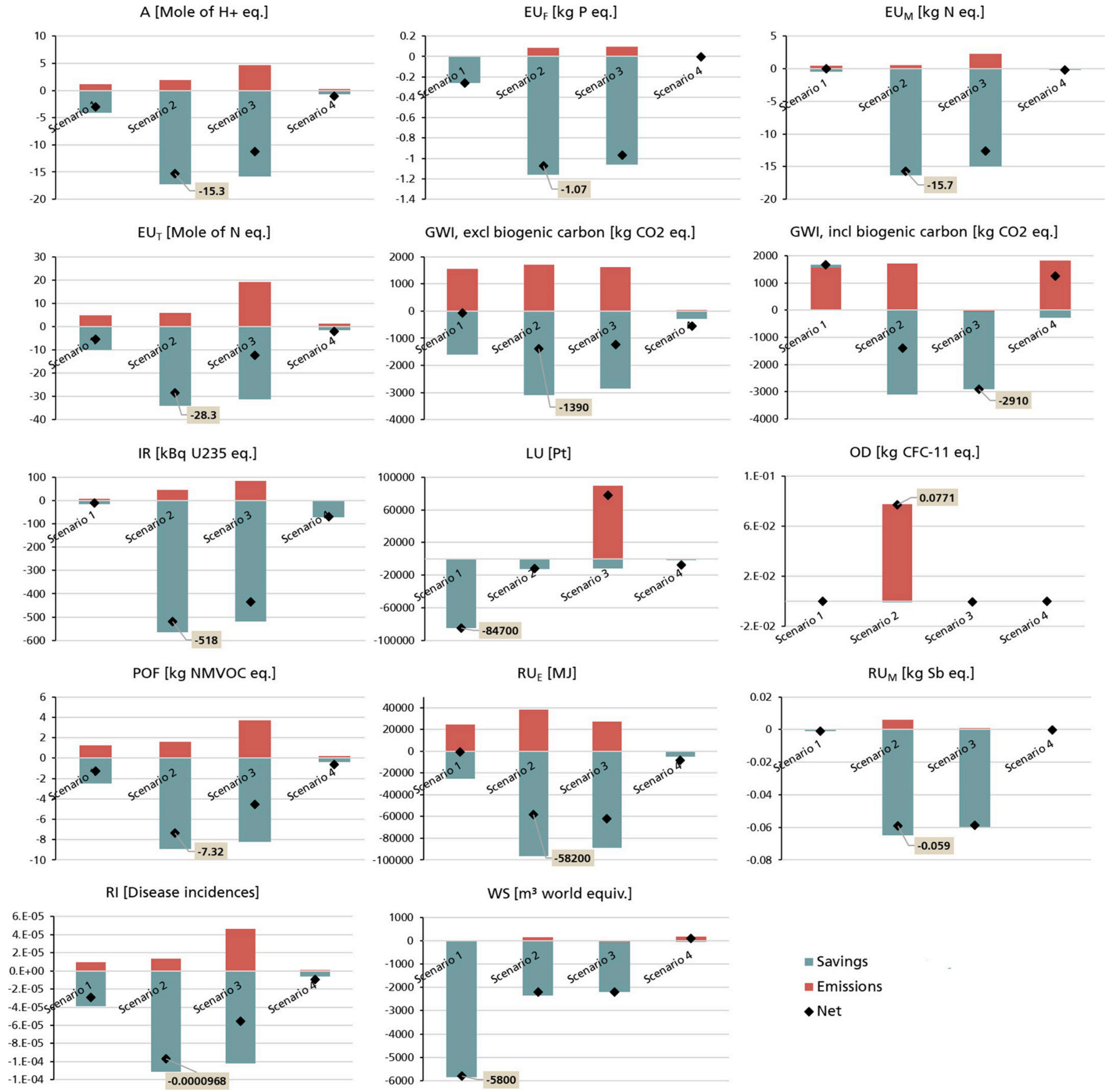


Fig. 5. Environmental impacts of the investigated scenarios (see Section 2.1.7 for impact categories).

products.

The contribution analysis quantifies the impact of each stage towards the chosen 14 impact categories. The negative values represent the savings obtained from each process or their respective credits gained from product substitution. In case of scenario 1, it can be concluded that the overall savings from the recycling of PC-PLA waste to produce lactic acid is considerably higher than the negative impacts associated with individual process stages. It can be observed that the highest net emission was recorded in the impact category GWI (including biogenic carbon) due to the high energy demand required for the production of lactic acid by hydrolysis of PC-PLA waste followed by subsequent product purification. In addition to this, considering a cradle-to-gate analysis, the production of virgin lactic acid from corn accounts for a negative carbon footprint (including biogenic carbon) due to a higher degree of CO_2 fixation as compared to the GHG emissions. Consequently, the substitution of virgin lactic acid by lactic acid obtained from recycling PC-PLA waste (scenario 1) further leads to additional GHG emissions.

For scenario 2, it is observed that the highest net emission was recorded in the impact category OD. This can be attributed to the use of chloroform as a solvent for the investigated strategy. In general, it can be concluded that recycling of PC-PLA waste to produce methyl lactate (scenario 2) has considerably higher savings as compared to penalties across all impact categories, except OD. This can be attributed to the fact that methyl lactate can serve as a potential substitute to fossil-based DMF.

The use of bioethanol as a nucleophile in scenario 3 contributes to a net negative impact in the category LU. However, in all the remaining impact categories, net savings are observed.

It is evident that the incineration of PC-PLA waste as investigated in scenario 4 leads to more GHG emissions (including biogenic carbon) than the savings resulting from energy recovery. Similarly, the impact category WS also shows net emissions, which can be attributed to the process itself (the chosen dataset employs a boiler operation with steam losses). Nonetheless, in the remaining impact categories, net savings can be observed by virtue of the credits acquired by electricity and thermal energy generated from incineration of PC-PLA waste in a MSWI plant.

4.3. Discussion on impacts of toxicity

The impact categories, human toxicity and ecotoxicity, were not a part of this investigation due to known methodological constraints owing to uncertainties in the characterisation factors (Hauschild and

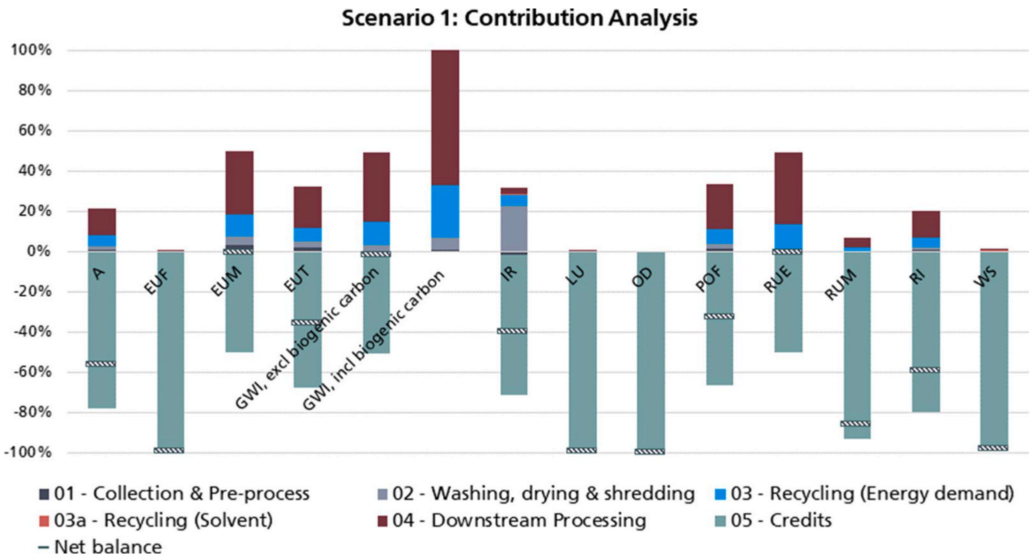


Fig. 6. Environmental impacts of the investigated scenarios (see Section 2.1.7 for impact categories).

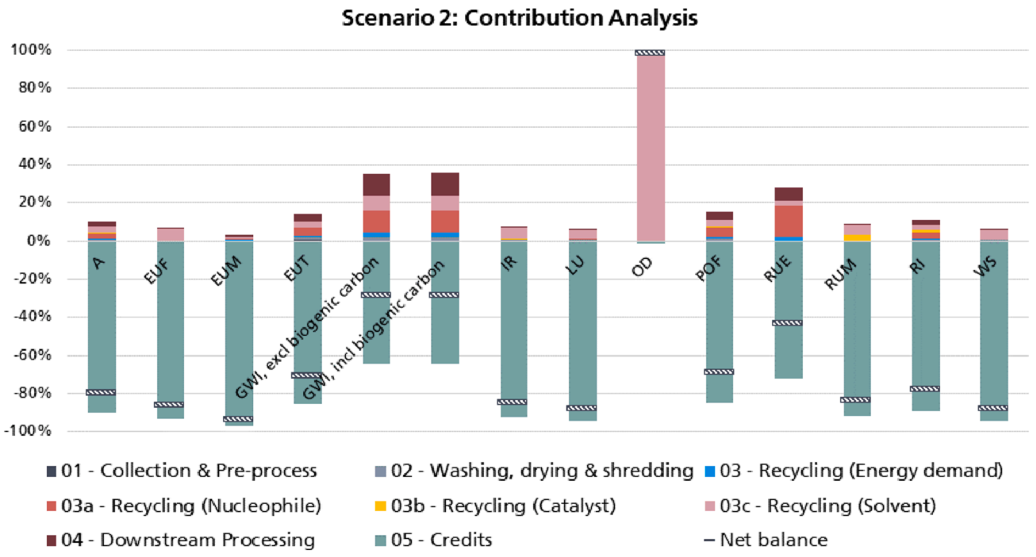


Fig. 7. Contribution analysis of scenario 2 (see Section 2.1.7 for impact categories)

Huijbregts, 2015). The recommended methodology to address toxicity in LCA, primarily the USEtox model (Rosenbaum et al., 2008), does not cover the impacts of toxicity at the work place. In addition to this, the LCI data regarding the release of possible toxic substances such as chloroform during the recycling process was not available. Nonetheless, it should be noted that, most of the solvents and the nucleophiles used in the investigated chemical recycling scenarios could have some general impacts on ecotoxicity and human toxicity, particularly in relation to the working environment. Table 11 summarizes the risk potential of the solvents, nucleophiles and the final products addressed in this study as a part of the investigated chemical recycling strategies.

Amongst the nucleophiles and solvents employed, chloroform, methanol and acetone raise concerns. The World Health Organisation (IARC) classifies chloroform as possibly carcinogenic to humans (IARC, 2020). The European regulation concerning the 'Registration, Evaluation, Authorisation and Restriction of Chemicals' (REACH), also classifies chloroform under the hazard code H351 and thus, has introduced restrictions on its use (ECHA, 2021). In addition to this, the nucleophile, methanol and the solvent, acetone employed in the recycling technologies investigated in this study are deemed problematic (Byrne et al., 2016). However, the final products, lactic acid, methyl lactate and ethyl lactate, are assessed to be non-problematic and can also be termed as green chemicals.

4.4. *Sensitivity analysis*

4.4.1. *Influence of the source of auxiliary materials*

In principal, methanol, acetone and ethanol can be produced either from fossil-based raw materials or biomass. Methanol can be produced from renewable raw materials either via thermochemical and biochemical conversion (Shamsul et al., 2014). Methanol can be also produced from CO_2 or steel mill gases (Schittkowski et al., 2018). However, both these pathways have not yet been realised at an industrial scale. The same applies for bio-based acetone (da Conceição Gomes et al., 2019). Therefore, methanol and acetone were excluded from this sensitivity analysis. In contrast, ethanol is produced worldwide from various renewable raw materials such as sugar cane, sugar beet, wheat, and corn. Additionally, it is industrially produced by the hydration of fossil-based ethylene. Hence, a sensitivity analysis based on the source of ethanol was conducted. Apart from ethanol from sugar beet (designated

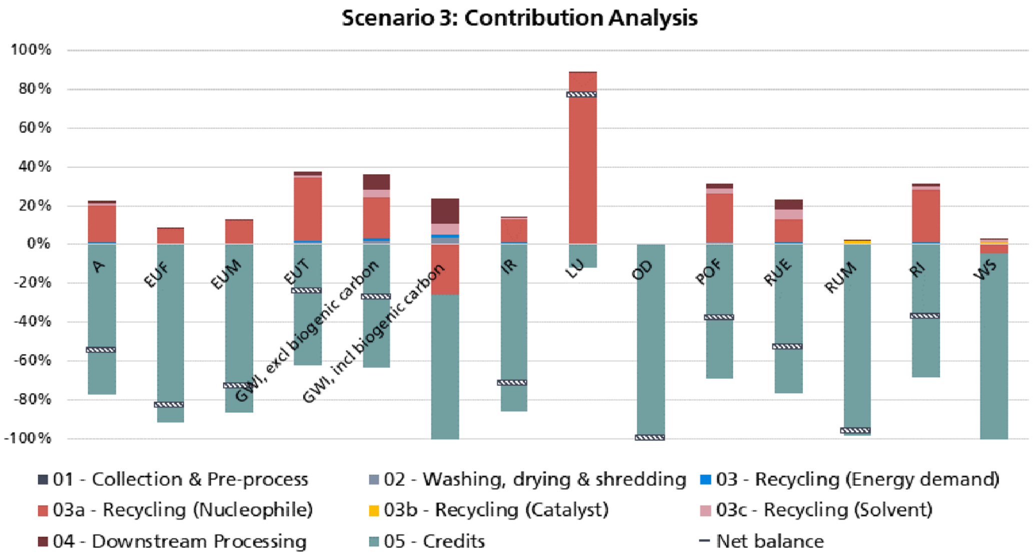


Fig. 8. Contribution analysis of scenario 3 (see Section 2.1.7 for impact categories).

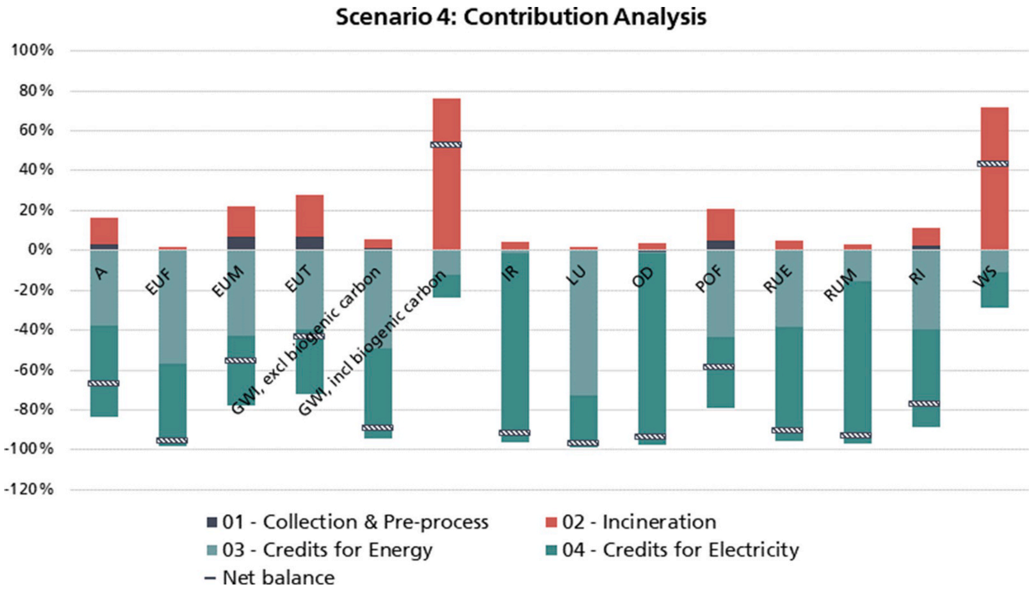


Fig. 9. Contribution analysis of scenario 4 (see Section 2.1.7 for impact categories).

Table 11

Solvents, nucleophiles and products from the three recycling scenarios (N: nucleophile; P: product; S: solvent) (Yang et al., 2012; Román-Ramírez et al., 2020).

Scenario	Compound	Type	Risk	Risk potential	Source
2	Methanol	N	🟧	problematic	(Byrne et al., 2016)
2	Chloroform	S	🟥	hazardous	(ECHA, 2021)
3	Ethanol	N	🟩	not problematic	(Byrne et al., 2016)
3	Acetone	S	🟧	problematic	(Byrne et al., 2016)
1	Lactic acid	P	🟩	not problematic	(Yang et al., 2012)
2	Methyl lactate	P	🟩	not problematic	(Román-Ramírez et al., 2020)
3	Ethyl lactate	P	🟩	not problematic	(Pereira et al., 2011)

as a base case for scenario 3), ethanol obtained from sugarcane, wheat, corn and other fossil-based raw materials was investigated in the sensitivity analysis. The following thinkstep datasets were chosen: “BR: Bioethanol from sugar cane at filling station”, “EU-28: Bioethanol from wheat, at filling station”, “EU-28: Bioethanol from corn, at filling station” and “DE: Ethanol”. The impacts pertaining to its transportation were neglected.

Table 12 shows the deviations in the net impact resulting from the substitution of ethanol obtained from sugar beet with that obtained from other sources. The savings observed under each impact category from the substitution of sugar beet based ethanol with other sources such as sugarcane, wheat, maize or fossil-based is shown in green. It can be observed that the use of ethanol derived from sugarcane instead of ethanol obtained from sugar beet has evident benefits in the impact categories, global warming and resource use (RU_E). In addition to this, impacts on ionising radiation are reduced by 16%. However, additional impacts are generated in the categories, acidification, eutrophication (marine and terrestrial), land use, photochemical ozone formation. In particular, its impact on the category, respiratory inorganics, is quite

Table 12
Heat map of the percentage deviations from net impact results of scenario 3 through different ethanol sources (see Section 2.1.7 for impact categories).

Impact categories	Unit	Sugarcane	Wheat	Maize	Fossil
A	[Mole of H+ eq.]	83%	43%	16%	-13%
EU_F	[kg P eq.]	-2%	3%	21%	-10%
EU_M	[kg N eq.]	140%	34%	7%	-11%
EU_T	[Mole of N eq.]	221%	183%	55%	-72%
GWI, incl. biogenic carbon	[kg CO_2 eq.]	-41%	4%	19%	82%
GWI, excl. biogenic carbon	[kg CO_2 eq.]	-18%	1%	8%	92%
IR	[kBq U235 eq.]	-16%	-5%	-5%	-14%
LU	[Pt]	72%	65%	204%	-114%
OD	[kg CFC-11 eq.]	0%	0%	0%	0%
POF	[kg NMVOC eq.]	238%	-12%	14%	-5%
RU_E	[MJ]	-20%	-1%	4%	66%
RU_M	[kg Sb eq.]	-1%	1%	5%	-1%
RI	[Disease incidences]	4065%	47%	46%	-43%
WS	[Mole of H+ eq.]	-1%	15%	165%	8%

high. Ethanol produced from wheat or corn performs worse in majority of the investigated impact categories as opposed to the base case involving ethanol obtained from sugar beet. In contrast to bio-based ethanol sources, the use of fossil-based ethanol shows clear benefits across impact categories except global warming, resource use for energy and water scarcity. This clearly demonstrates that the use of a bio-based nucleophile can reduce the impact on global warming and use of fossil resources at the expense of other impact categories. It can be observed that, the source of ethanol has negligible effect on the impact category concerning ozone depletion. This is because the results in this impact category primarily depend on the credits acquired by substituting the fossil-based solvent, DMF.

These environmental impacts particularly resulting from the cultivation of crops for the production of bio-based chemicals can be reduced further by using agricultural residues. (Maga et al., 2018) has demonstrated that the production of second generation bio-ethanol from agricultural residues (for e.g. bagasse) reduces the GHG emissions by 50%. In addition to this, the land use attributed to this production strategy is approximately 30 times smaller as compared to that associated with its production from sugarcane. (Maga et al., 2018).

4.4.2. *Influence on the substitution potential of the recyclates*

The recycling strategy illustrated in scenario 1 leads to the production of lactic acid while those instituted in scenarios 2 and 3 results in the production of methyl and ethyl lactate respectively. Fig. 10 shows the global warming impact of various fossil-based solvents that can be replaced by ethyl lactate.

Lactic acid is expected to replace virgin lactic acid derived from corn while methyl lactate is expected to replace DMF. The potential GHG savings resulting from these replacements have been already discussed in detail in previous sections (4.1 and 4.2). In contrast to this, ethyl lactate is a versatile solvent that can replace a wide spectra of fossil-based solvents.

While both methyl and ethyl lactate can substitute DMF as a solvent in the synthesis of cellulose-acetate membranes (Abdoul Raguime et al., 2007, Figoli et al., 2014, Medina-Gonzalez et al., 2011), ethyl lactate has promising prospects as compared to other low molecular weight lactate esters. For instance, it has the potential to replace toxic and carcinogenic solvents such as hexane or dichloromethane (DCM), which are traditionally used in the extraction of carotenoids and other phytonutrients (Ishida and Chapman, 2009, Kua et al., 2016). Furthermore, an experimental investigation aimed at replacing hazardous air-pollutants such as MEK, MIBK and toluene in magnetic tape-coatings demonstrated that, ethyl lactate can partially (up to an extent of 50%) replace the use of these solvents; thereby, leading to a safer work-place (Nikles et al., 2001). In addition to this, several efforts directed towards eco-friendly organic synthesis (Bennett et al., 2009, Paul et al., 2016) have illustrated the use of ethyl lactate as a promising reaction media. In an another attempt aimed at devising a greener chromatographic separation (Micăle et al., 2015), ethyl lactate was proposed as a potential non-toxic and bio-degradable alternative to acetonitrile for the separation of a mixture of 16 polycyclic aromatic hydrocarbons using reverse phase liquid chromatography. It has also been stated that, on account of its bio-degradability, low vapour pressure and surface tension and favourable solvating properties, ethyl lactate can completely replace several ozone-depleting halogenated solvents like DCM, chloroform and tetrachloroethylene in pertinent applications (Anastas and Hammond, 2016). Thus, on account of its desirable properties and a broad spectra of

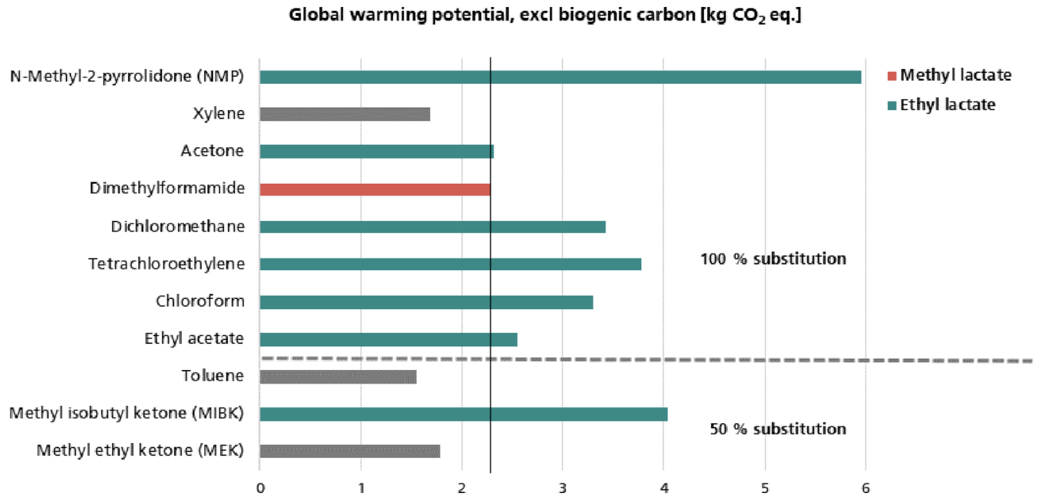


Fig. 10. Substitution potential of ethyl lactate. Please note: (i) solvents with an impact on the global warming potential higher than DMF are represented in green while those with a comparatively smaller carbon footprint are illustrated in grey; (ii) some solvents can be replaced on a 1:1 basis whereas some others can only be replaced partially (to an extent of 50%).

applicable sectors ranging from food and pharmaceuticals to paints and coatings (Pereira et al., 2011), ethyl lactate is undoubtedly the most valuable product amongst the family of lactate esters and is estimated to reach a market share of USD 92 million by 2024 (Lamberti et al., 2020).

5. Summary of results and conclusion

In this paper, the LCA methodology was utilised to investigate the environmental impacts generated from four different EoL handling and processing pathways for PLA waste. The functional unit of the study was the treatment of one tonne post-consumer PLA waste. The results reveal that all three chemical recycling technologies have a better environmental performance as compared to the reference scenario, which is the direct incineration of the PLA waste in a MSWI plant. Although the three chemical recycling scenarios are on a similar TRL, it must be noted that a one-to-one comparison between them is only possible to a limited extent, since the final products (recyclates) obtained from each of these scenarios are different.

Firstly, it must be understood that, chemical recycling is always beneficial as compared to incineration of PLA waste. Scenario 1 focusses on hydrolysis of PLA waste to produce lactic acid (LA) - a leading bio-based platform chemical, and is also used in the production of PLA. Thus, the hydrolysis of PLA reduces the environmental impact of PLA by substituting virgin LA, thereby avoiding associated agricultural activities. This results in lower impacts on land use and minimizes the possible impacts emanating from land use change. Hence, the hydrolysis of PLA can lower the impact of PLA since virgin LA is replaced and associated agricultural activities are avoided.

However, the use of PC-PLA waste for producing green solvents has higher environmental benefits. The alcoholysis of PLA (scenario 2 and 3) in particular showed clear environmental benefits in almost all impact categories except ozone depletion (scenario 2) and land use (scenario 3). Amongst these two scenarios, scenario 3 performs better from a global warming perspective, whereas scenario 2 performs slightly better in many other impact categories. One key reason for the better performance of scenario 2 is the higher product yield. However, there is no concrete information available about the tolerance of this recycling strategy to the presence of contaminants/impurities present in PC-PLA waste obtained from different value chains. In addition to this, if the substitution potential of the methyl lactate and ethyl lactate is considered, ethyl lactate shows evident benefits. In contrast to methyl lactate, ethyl lactate can substitute a spectrum of fossil-based products whereas methyl lactate has limited applications. Depending on the application, ethyl lactate can replace solvents with a bigger environmental footprint such as N-Methyl-2-pyrrolidone (NMP), leading to higher GHG savings. Furthermore, it should also be noted that, scenario 2 uses chloroform and methanol which have a negative impact on the eco-system and human health from a toxicological perspective. In contrast, scenario 3 uses acetone and ethanol which are associated with lower risks.

Another difference between scenario 2 and 3 is the possibility to use a bio-based nucleophile in the latter strategy. While scenario 2 depends on fossil-based methanol, scenario 3 can use ethanol from various sources, both bio-and fossil-based. In particular, the use of ethanol from sugarcane or alternative second generation feedstock allows further reduction in the impacts on global warming and the use of fossil resources. On the other hand, the use of bio-ethanol instead of fossil-based ethanol aggravates its environmental impacts in various impact categories such as acidification, eutrophication, respiratory inorganics and land use. This can be primarily attributed to the agricultural activities that are necessary to cultivate crops for the production of cellulosic sugar, which in turn serves as a feedstock for the production of bioethanol.

Thus, it is crucial to design plastics, especially bioplastics, with an aim to use them for as long as possible (concept of "eco-design"). In addition to this, they have to be up-cycled at the end of their life in order to conserve valuable biogenic resources for the future generations. However, this would require mutual co-operation between all the sectors along the value-chain.

Author statement

Venkat Aryan and Daniel Maga were mainly responsible for the development of methodology, application of LCA model, visualisation of results, and writing the manuscript. Pranav Majgaonkar and Ronny Hanich were responsible for the conceptualisation, technology development, provision of experimental and simulated data, and reviewing the manuscript. In addition to this, Pranav Majgaonkar and Venkat Aryan also undertook the project administration.

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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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2021.105670.

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