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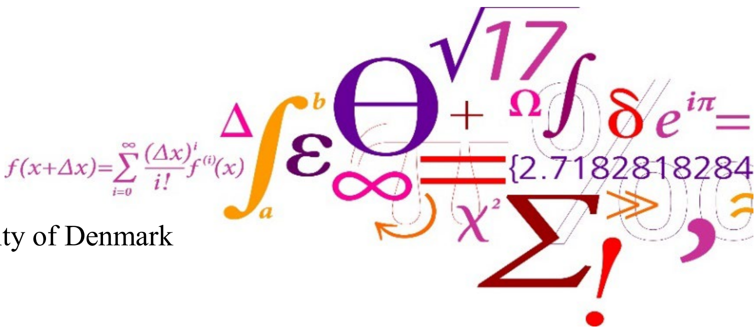

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PREFACE

This thesis, entitled "*Environmental Sustainability Assessment of Advanced Agricultural Waste Technologies and Agricultural Territories"* is a result of PhD research carried out at DTU Management from May 2017 to April 2020, under the supervision of two main supervisors Morten Birkved (now at Syddank Universitet) and Stig Irving Olsen in the group of Quantitative Sustainability Assessments at DTU. In addition the PhD project, hereafter referred to as "the project", was co-supervised by Michael Z. Hauschild, Sander Bruun (Københavns Universitet) and Hinrich Uellendahl (now at Hochschule Flensburg, Germany).

The project received funding and was carried out in coordination with the No Agricultural Waste (NoAW) Horizon 2020 project, grant agreement no. 688338, as well as receiving partial funding from DTU.

During the course of the project, the following manuscripts were produced, listed below and referred throughout this thesis by their roman numeral. Full copies of the articles are provided in the Appendix.

I. Croxatto Vega, G., Sohn, J., Bruun, S., Olsen, S. I., & Birkved, M. (2019). Maximizing environmental impact savings potential through innovative biorefinery alternatives: An application of the TM-LCA framework for regional scale impact assessment. *Sustainability*, *11*(14), 3836.

II. Croxatto Vega, G., Sohn, J., Voogt, J., Nilsson, A.E., Birkved, M., & Olsen, S. I. (2020) Insights from combining techno-economic and life cycle assessment – a case study of polyphenol extraction from red wine pomace. *Resources, Conservation & Recycling.* Submitted.

III. Croxatto Vega, G., Voogt, J., Sohn, J., Birkved, M., & Olsen, S. I. (2020). Assessing new biotechnologies with combined

TEA-TM-LCA for efficient use of biomass resources. *Sustainability* (2071-1050), 12(9).

IV. Croxatto Vega, G., Gross, A., Birkved, M. (2020) Accounting for priority impacts of plastic products – PlastLCI a simulation study for advanced Life Cycle Inventories of plastics. *Nature Sustainability*. Submitted.

In addition, I collaborated in the following co-authored publications, which are part of this thesis:

V. Sohn, J., Vega, G. C., & Birkved, M. (2018). A Methodology Concept for Territorial Metabolism-Life Cycle Assessment: Challenges and Opportunities in Scaling from Urban to Territorial Assessment. *Procedia CIRP*, *69*, 89-93.

VI. Corona, A., Ambye-Jensen, M., Vega, G. C., Hauschild, M. Z., & Birkved, M. (2018). Techno-environmental assessment of the green biorefinery concept: Combining process simulation and life cycle assessment at an early design stage. *Science of the Total Environment*, *635*, 100-111.

VII. David, G., Croxatto, G., Sohn, J., Nilsson, A. E., Helias, A., Gontard, N., Angellier-Coussy, H. (2020). Using Life Cycle Assessment to quantify the environmental benefit of up-cycling vine shoots as fillers in biocomposite packaging materials. *International Journal of Life Cycle Assessment.* Submitted.

VIII. Nilsson, A.E., Sohn, J., Croxatto Vega, G., Birkved, M., & Olsen, S. I. (2020). Testing the no agricultural waste concept – an environmental comparison of biorefinery value chains in various regions. Advanced draft.

Conference papers:

IX. Vega, G. C., Voogt, J., Nilsson, A. E., Sohn, J., Birkved, M., & Olsen, S. I. (2019) Lessons from combining techno-economic and life cycle assessment–a case study of polyphenol extraction from waste resources. *Publication in conference proceedings Heraklion 2019 – 7th International Conference on Sustainable Solid Waste Management.*

X. Sohn, J., Vega, G. C., Birkved, M., & Olsen, S. I. Incorporating Relative Importance: selecting a polyphenol production method for agro-waste treatment in an environmental and economic multi-criteria decision making context. (2019). *Publication in conference proceedings Heraklion 2019 – 7th International Conference on Sustainable Solid Waste Management.*

SUMMARY

The sustainability of emerging biotechnologies seeking to close the loop with circular economy alternatives is questionable. Biomass resources are finite and management of these resources has to improve dramatically if we are to ensure a sustainable future for coming generations. All things bio, such as bioproducts, bioenergy, biochemicals have a tendency to be perceived as environmentally superior to their fossil counterparts. However, research has shown this is not always the case and that the answer to the question, "is bio sustainable?" is always *it depends*.

During the course of this PhD project the sustainability of various biorefinery setups, which are a result of emerging biotechnological developments, and their products have been assessed with the life cycle assessment (LCA) methodology in specific regional contexts. The aim of this project is to increase our understanding of the variables and patterns that should be included in the assessment in order to succeed in the identification of sustainable bio-options. For this purpose, 3 perspectives of focus were introduced with regards to building appropriate assessments.

The territorial perspective, which includes considerations that must be made about the background system in LCA includes variables such as land, feedstock provisioning, and the energy grid supplying the biotechnologies. In this context, mass flow analysis of the regions of interest can be coupled with LCA i.e. material flows going in and out of the region, and dynamic inventories can be produced to account for changes in time i.e. changing background energy grid mix. The assessment of various systems throughout this project showed that the added information from a dynamic energy mix is a necessary component of future LCAs that either use biomass resources, or have energy intensive processing. On the other hand, the mass flow analysis is useful in drawing attention to potential pressures from the feedstock provisioning side,

though a more definitive analysis of land use change is necessary to avoid potential negative impacts from feedstock sourcing. This is a global issue and thus should be assessed taking the consequences at a global scale.

The foreground and early design perspective introduced in this project centers around process design, a.k.a. the foreground system. This refers to biotechnologies in early development phases, such as laboratory scale, which could benefit from a hot spot assessment that may point out process design improvement areas for more environmentally friendly technologies at industrial scale. Results showed that LCA is capable of pointing out design hot spots in biotechnologies at an early stage of development by the use of a quick carbon foot printing. The project exemplifies how to utilize process design software that is routinely used by the chemical/biotech industries, together with LCA to produce multi-angle assessments. The combination has the potential to become a powerful tool that would benefit from the level of standardization already available to LCA practitioners. Combining for example, techno-economic assessments (TEA) and LCA can lead to process design, which may be optimized from both an environmental and economic side. Furthermore, this project applied multi-criteria decision analysis methods in order to derive clear decision support from compound assessment such as combined TEA-LCA. The methods of MCDA tested during this project proved effective and were in agreement. However, further research is needed in order to decrease the subjectivity of weighting profiles and valuation of externalities.

The last perspective analyzed in this project is the products perspective. Novel products lack the level of coverage that conventional products have in LCA databases. Moreover, The functionality of novel products is, at times, poorly understood or contains value that is outside of the scope of the LCA methodology as it is today. Several improvement areas were identified in regards to increasing our understanding of novel

products and the methodological needs that are needed for a more complete assessment of these products. Most importantly, a framework was developed to include the impacts of plastic products in a more thorough way, which includes the contribution of microplastics to particulate matter formation. However, there is an urgent need for increasing our understanding of the microplastics cycle including, but not limited to: degradation rates of plastic in the natural environment, better understanding of degradation rates of conventional plastic in landfill and of littered plastic, increased understanding of the fate of macro and microplastic and finally, increased understanding of damage from this source of pollution to human health and ecosystem health. Additionally, the value of biodegradable materials should be carefully considered and might need to be redefined, as it is probable that the value of biodegradable materials extends beyond the scope of the LCA methodology into ecological perspectives poorly covered by LCA e.g. biodiversity, ecosystems services.

The three perspectives explored and findings produced during this project will facilitate the assessment of various biotechnologies in their regionally specific context. Depending on the goal and scope of future LCAs, the methodological elements identified here might be needed in their entirety or in a partial manner. Cross-disciplinary interactions will be key to ensure that the LCA methodology continues to develop and realizes its full potential, so that in the future when asked if bio is sustainable we can finally answer with a definitive statement.

DANSK SAMMENFATNING

Bæredygtigheden af nye bioteknologier, der søger at lukke kredsen med muligheder for cirkulære økonomier, er tvivlsom. Mængden af biomasse er begrænset, og forvaltningen af disse ressourcer skal forbedres dramatisk, hvis vi skal sikre en bæredygtig fremtid for kommende generationer. Alt ”bio”, såsom bioprodukter, bioenergi, biokemikalier har en tendens til at blive opfattet som miljømæssigt bedre end deres fossile kolleger. Imidlertid har forskning vist, at dette ikke altid er tilfældet, og at svaret på spørgsmålet, *er ”bio” bæredygtigt,* altid er: *det kommer an på*.

I løbet af dette ph.d.-projekt er bæredygtigheden af forskellige bioraffinaderiindstillinger, der er et resultat af den nye bioteknologiske udvikling, og deres produkter, blevet vurderet ved hjælp af livscyklusvurderings (LCA) -metodologien i specifikke regionale sammenhænge. Målet med dette projekt er at øge vores forståelse af de variationer og mønstre, der skal inkluderes i vurderingen for at få succes med at identificere bæredygtige bio-muligheder. Til dette formål er der introduceret 3 fokusperspektiver med hensyn til opbygning af passende vurderinger.

Det territoriale perspektiv, der inkluderer overvejelser, der skal tages med hensyn til baggrundssystemet i LCA, inkluderer variabler som jordbrug, forsyning af råvarer og energinettet der forsyner bioteknologierne. I denne sammenhæng kan massestrømningsanalyse dvs. materialestrømme der går ind og ud af regionen, sammenkobles med LCA. Samtidig kan der frembringes dynamiske opgørelser for at redegøre for ændringer i tiden, dvs. ændringer af energinettets kilder i baggrundssystemet. Evalueringen af forskellige systemer i hele dette projekt viste, at den tilføjede information fra en dynamisk energimix er en nødvendig komponent af fremtidige LCA'er, der enten bruger biomasseressourcer eller har energiintensiv behandling. På den anden side er massestrømningsanalysen velegnet til at henlede opmærksomheden på potentielt pres fra biomasseforsyningssiden, skønt en mere definitiv analyse af ændring af arealanvendelse er nødvendig for at undgå potentielle negative påvirkninger fra biomasseforsyningen. Dette er et globalt spørgsmål, og det bør derfor vurderes med hensyn til de globale konsekvenser.

Det tidlige designperspektiv, herunder forgrundsystemet, fokuserer på procesdesign. Dette henviser til bioteknologier i tidlige udviklingsfaser, f.eks. en laboratorieskala, som kunne drage fordel af en hot-spot vurdering, der kan påpege forbedringsområder for procesdesign til mere miljøvenlige teknologier i en industriel skala. Resultaterne viste, at LCA er i stand til at påpege design hot-spots i bioteknologier på et tidligt stadium af udviklingen ved hjælp af en hurtig carbon footprint analyse. Projektet illustrerer, hvordan man bruger procesdesignsoftware, der rutinemæssigt bruges af kemiske/bioteknologiske industrier, sammen med LCA, til at fremstille flervinkelsvurderinger. Kombinationen har potentialet til at blive et kraftfuldt værktøj, der vil kunne drage fordel af det standardiseringsniveau, der allerede er tilgængeligt for LCA-udøvere. Kombinationen af for eksempel teknologisk-økonomiske vurderinger (TEA) og LCA kan føre til procesdesign, som kan optimeres både fra en miljømæssig og økonomisk side. Desuden anvendte dette projekt beslutningsanalysemetoder med flere kriterier (Multi-Criteria Decision Analysis) for at opnå en klar beslutningsstøtte fra sammensatte vurderinger som for eksempel en kombineret TEA-LCA. MCDA metoderne testet under dette projekt viste sig at være effektive og i overensstemmelse med hinanden. Imidlertid er der behov for yderligere forskning for at reducere subjektiviteten af vægtningsprofiler og værdiansættelser af eksternaliteter.

Det sidste perspektiv, der analyseres i dette projekt, er produktperspektivet. Nye produkter mangler det dækningsniveau, som konventionelle produkter har i LCA-databaser. Desuden er funktionaliteten af nye produkter til tider dårligt forstået eller indeholder værdier, der ligger uden for rammerne af LCA-metodikken, som den er i dag. Der blev identificeret flere vigtige forbedringsområder med hensyn til at øge vores forståelse af nye produkter og de metodologiske behov, der er nødvendige for en mere fuldstændig vurdering af disse. Det vigtigste er, at der blev udviklet en ramme, der indbefatter påvirkningerne af plastprodukter på en mere grundig måde, som inkluderer bidraget fra mikroplastik til dannelse af atmosfærisk støv. Imidlertid er der et presserende behov for at øge vores forståelse af mikroplastik-cyklussen, herunder (men ikke begrænset til) nedbrydningshastigheder af plast i det naturlige miljø, bedre forståelse af nedbrydningshastighederne for konventionel plast i deponering og af spildt plast, øget forståelse af makro- og mikroplastiks skæbne og endelig en øget forståelse af hvordan denne type forurening skader menneskers og økosystemets sundhed. Derudover skal værdien af bionedbrydelige materialer overvejes nøje og der kan være nødvendig at omdefinere denne. Fordi det er sandsynligt, at værdien af bionedbrydeligt materiale strækker sig ud over LCA-metodens rækkevidde i økologiske perspektiver, bl.a. biodiversitet og økosystemtjenester, der er dårligt omfattet af LCA.

De tre perspektiver der blev undersøgt, og de resultater der blev produceret i løbet af dette projekt, vil lette vurderingen af forskellige bioteknologier i deres regions specifikke kontekst. Afhængigt af målet og omfanget af fremtidige LCA'er, kan de metodologiske elementer, der er identificeret her, være nødvendige i deres helhed eller delvist. Tværfaglige interaktioner vil være nøglen til at sikre, at LCA-metodologien fortsætter med at udvikle sig og realisere dens fulde potentiale, så vi i fremtiden, når vi bliver spurgt om ”bio” er bæredygtigt, endelig kan give et entydigt svar.

ACKNOWLEDGEMENTS

When I started this PhD, about 3 years ago, little did I know what a journey it would be. This journey has been full of personal and professional growth, which would not have been possible without the help and support from various people both at the office and at home.

First of all, I would like to thank all of my supervisors for the countless talks that helped clarify the path ahead. I would like to thank Morten for his reactivity, for always showing up. Many thanks to Stig for gracefully taking over the role of main supervisor mid-way through. To Sander for his hands on method of approaching my questions. To Hinrich for all the good talks about biomass conversion and for including me in your activities. And to Michael for the always welcomed insights. You all have no doubt helped me in many other ways, for all of it I thank you.

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Finally, I would like to thank my amazing husband, Martin, for putting up with me while getting through this PhD. I know it has not been easy to live with me, especially during these last few months. But also for being my sparring partner when Covid forced everyone to stay home. To my kids for being a constant reminder that there is more to life than working and for being so darn cute. Cuddling to reading you stories has been one of the best ways to relax after a tough day at the office. And to the rest of my family for being so excited about my work, even when I wasn't. There are many other ways in which you have helped get through this. I am eternally thankful for your consideration and love in this time.

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LIST OF ACRONYMS

AD	Anaerobic Digestion
ArgCW	Argumentation Context Weighted
BMP	Biomethane Potential
CapEx	Capital Expenditure
C.Cap	Carrying capacity
CE	Circular Economy
CF	Characterization factor
CFP	Carbon Foot-print
CHP	Combined heat and power
CWF	Context Weighted Factor
DALY	Disability-adjusted life year
DH	District Heating
DK	Denmark
DW	Dry Weight
EF	Emission Factor
EoL	End of Life
EU	European Union
EW	Equal weights
FU	Functional Unit
GDP	Gross domestic product
GWP	Global Warming Potential
IC	Impact Category
IPCC	Intergovernmental panel on climate change
iLUC	indirect Land Use Change
IRR	Interal Rate of Return
kW	kilo Watt
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
LDPE	Low density polyethylene
LUC	Land Use Change

iLUC	indirect Land Use Change
IT	Italy
MP	MicroPlastics
MW	Mega Watt
NPV	Net Present Value
OpEx	Operational Expenditure
PAH	Polycyclic aromatic hydrocarbon
PB	Planetary boundary
PE	Polyethylene
PET	Polyethylene terephthalate
PHA	Polyhydroxyalkanoates
PHB	Polyhydroxybutyrate
PHBV	Poly(hydroxybutyrate-co-hydroxyvalerate)
PLA	Polylactic Acid
PLE	Pressurized liquid extraction
PM10	Particulate matter with a size inferior to 10 μm
PM2.5	Particulate matter with a size inferior to 2.5 μm
PP	Polypropelene
PS	Polystyrene
PY	Person year
RIF	Relative Importance Factor
SDG	Sustainable Development Goal
TEA	Techno-Economic Assessment
TM	Territorial Metabolism
TOPSIS	Technique for Order of Preference by Similarity to Ideal Solution
TPS	Thermoplastic Starch
TRL	Technology Readiness Level
UN	United Nations
VFA	Volatile Fatty Acids
VOCs	Volatile Organic Compounds
VS	Volatile solids
TS	Total Solids

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READER'S GUIDE

This PhD thesis is divided into 6 chapters. In the Chapter 1, an overview of the existing literature relevant to this project and the methodological choices taken throughout is given. The introduction is subdivided into:

Section 1.1 – which presents the research objectives and approach. Section 1.2 – which presents current state of the art. This is followed by 3 subchapters, which represent 3 focus points of the thesis, in relation to determining the sustainability of biorefineries in a territorial context. The three subchapters refer to the background system (1.2.1.), the foreground system (1.2.2) and the outputs a.k.a. products of biorefineries (1.2.3) and relevant literature on these three topics.

After the introduction, chapters 2-5 are developed with the introductory literature in mind (but without repeating it). Each of these chapters begins with a research question, followed by the relevant methodology for the chapter. In each chapter, the method subsection is followed by a key results section. *It is advisable to read **Section 1.2.1**. followed by **chapter 2**, **section 1.2.3** followed by **chapters 3 and 4**, and **section 1.2.3 followed by chapter 5**, as these are complementary and together present the introduction, methods, results and discussion of the project.*

Finally, in Chapter 6, the project's achievements and future research needs are presented in relations to the project's findings. Recommendations for future work are made in this chapter.

1. INTRODUCTION

Waste in the agricultural sector is a resource. For centuries, farmers have used their waste to provide essential services such as fertilizer for their crops, soil amendment for their fields, and more. Now a day, with the advent of the circular economy (CE) (Kirchherr et al., 2017; Saidani et al., 2019), and the sustainable development goals (SDGs) (UNEP et al., 2012), valorizing waste resources has come more into focus. Various political targets have been proposed, in order to attain the aims of a circular economy (European Commission, 2015). Among other, the CE wishes to maintain the value of products, materials and resources in the economy for as long as possible, while at the same time minimizing the generation of waste (European Union, 2018). In agriculture, "waste" is often a by-product of primary production, and is rarely completely devoid of value. For production systems characterized by little change in production methods from year to year e.g. wine production, cereal production etc., a continuous, yet seasonal stream of biomass residue is produced. These streams may or may not be valorize by the farmers, and in either case, represent an opportunity under the CE aims.

Biotechnology is a field, which is rapidly expanding the portfolio of choices that allows society to valorize biomass residues. Recent advances in bioprocessing for the production of biosourced bioproducts are potentially valuable solutions to decarbonize the economy. Yet, the prefix bio is not akin sustainability (Jørgensen et al., 2012; Ögmundarson et al., 2018). For example, the sustainability of biofuels has long been debated, since growing of energy crops increases the pressure on the many services land must generate e.g. food and feed production, habitats, etc. Among other impacts, such as increased eutrophication or ecotoxicity due to increased use in pesticides and mineral fertilizers for growing bioenergy crops (Cherubini and Strømman, 2011; Dressler et al., 2012), there is the issue of indirect land use change (iLUC), which has the potential to release vast amounts of CO_2 due to expansion or conversion of e.g. intact forest to agricultural production

(European Commission, 2019; Tonini et al., 2016). These issues extend to other products in the bio spectrum, such as biochemicals. To exemplify, a review of the most prominent biochemicals in the market found that environmental impact varies widely, ranging from friendlier than fossil based chemicals to several orders of magnitude higher than fossil based options, in a wide range of environmental protection areas (Ögmundarson et al., 2020a). The final score for sustainability of bioproducts depends highly on the type of biomass resource used for productions, the area where it is produced, the impact of the production methods and the benefits of the product against other fossil based alternatives. The complexity of bio options makes the use of holistic quantitative tools imperative for determining the environmental performance of bio options. In this regard, life cycle assessment (LCA) is a well-equipped tool to handle the complexity of emerging technologies linked to biomass resources (Hauschild et al., 2018).

LCA is a standardized tool that can be used to quantify the environmental impacts of new production methods, products or services (Finkbeiner et al., 2006). The LCA methodology covers all relevant life cycle stages of the case study in question i.e. extraction of raw materials, processing/manufacturing, use phase and disposal (Hauschild et al., 2018). Furthermore, a set of rules governing the execution of LCAs (European Commission - Joint Research Centre, 2010) and various databases containing standardized data, such as the Ecoinvent database (Wernet et al., 2016), allow for a greater level of comparability between studies. An important strength of the LCA methodology, in regard to biomass valorization, is the fact that the various life cycle assessment impact assessment methods (LCIAs) cover a wide range of environmental areas of interest, a.k.a. impact categories (ICs), such as global warming, human and ecotoxicity, marine and freshwater eutrophication, acidification, fossil resource scarcity and many more. Impact categories, are an important aspect when assessing the sustainability of a system, since it has been shown in previous LCAs that it is possible to shift burdens from one IC to another, if for example, the assessment is based only on carbon foot-printing (Laurent et al., 2012; Weiss et al., 2012). Throughout this project, LCA was applied to assess the sustainability of biorefineries, bioprocessing and bioproducts with various different aims. In order to fulfill the aims of each study, various LCA methodological choices were taken. In the following subsections a brief overview of the literature supporting the methodological choices taken is given in relation to the various focus points of the work composing this PhD thesis.

1.1. Research Objectives and Thesis Structure

The main objective of this PhD project is to develop, improve and apply the LCA methodology in order to asses innovative, emerging biorefineries treating agricultural waste/residues and their products in a territorial context from an environmental impact perspective. In order to do so, various assessments were carried out with different approaches. Emerging biorefineries were assessed with 4 distinct areas of focus that were used concurrently in several of the articles produced. The areas of focus are:

- The potential for upscaling to a specific geography including future changes for the site's background system
- Optimization via process design from an environmental and economic perspective
- Clarity in the interpretation phase by combining environmental impacts and economic impacts into multi-criteria decision support.
- Challenges from emerging biorefinery products and their comparison to conventional products

In the course of the PhD project, sustainability criteria have emerged that may be used to assess biorefineries. These criteria or patterns can be framed under the following overarching research questions:

RQ1: *Which factors are potentially result-altering when deciding on the best technology to region pairing now and in the future? (the background system)*

RQ2: *How can LCA be used for early design optimization of emerging biotechnologies? (the foreground system)*

RQ3: *How can bilateral economic and environmental sustainability assessments be combined to increase clarity for decision support? (interpretation)*

RQ4: How *are key assessment parameters and LCA methodological challenges identified and overcome when assessing emerging biorefinery products against conventional products? (new products)*

An overview of how the questions are addressed throughout the thesis, as well as the research approach and thesis structure is shown in Figure 1-1, with regard to the various publications produced during the project.

Research Questions	Background	Method	Results, Perspectives, and Limitations
RQ1. *Which factors are potentially result-altering when deciding on the best technology to region pairing now and in the future?*	**Section 1.2.1.** Introduction to existing application of LCA in large geographical settings and use of dynamism in LCA in mostly the background system.	**Section 2.1.** Application of territorial metabolism in LCA. Empirical studies/LCAs with various regional contexts – Paper I, III, and VIII	**Section 2.2.** key findings from the background perspective and dynamic LCAs **Section 6.1.** formal answers to RQ1. Future research needs - **Section 6.2.**
RQ2. *How can LCA be used for early design optimization of emerging biotechnologies?*	**Section 1.2.2.** Introduction to status of optimization of the foreground system via LCA and combination of LCA with other methods.	**Section 3.1** Combining techno-economic assessment with LCA. Empirical LCA study, including process optimization – Paper II and Paper VI	**Section 3.2.** key findings for the foreground perspective and early design. **Section 6.1.** formal answers to RQ2. Future research needs - **Section 6.2.**
RQ3. *How can bilateral economic and environmental sustainability assessments be combined to increase clarity for decision support?*	**Section 1.2.2.** Introduction to the use of multi-criteria analysis to combine assessments into concise decision support.	**Section 4.1.** Application of MCDA. Extension and refinement of the method by progressively improving the application of TOPSIS for the LCAs from Paper I, II to III.	**Section 4.2.** key findings in regards to using multi-criteria decision analysis. **Section 6.1.** formal answers to RQ3. Future research needs - **Section 6.2.**
RQ4. *How are key assessment parameters and LCA methodological challenges identified and overcome when assessing emerging biorefinery products against conventional products?*	**Section 1.2.3.** Identification of knowledge gaps in relation to biodegradable biosourced materials and other emerging biorefinery products in comparison to conventional products.	**Section 5.1.** Empirical LCAs with products in focus, for protein extraction (VI), biocomposites (VII), filler and biodegradable biosourced polymers (VIII). Review of the literature (annex of IV) and conceptual framework for plastic impacts (IV).	**Section 5.2.** key findings in regards to the products perspective. **Section 6.1.** formal answers to RQ4. Future research needs - **Section 6.2.**

Figure 1-1 Thesis Structure and Research approach

1.2. SUSTAINABILITY OF BIOREFINERIES

A biorefinery, here defined as "a facility that integrates biomass conversion processes and equipment to produce fuels, power, and [or] chemicals from biomass" (Berntsson et al., 2012), works in a specific production context. Depending on the type of processing i.e. chemical, thermal or biological, various feedstock may apply to be used for the conversion e.g. forestry products, glucose rich feedstock, lignocellulosic crop residues etc., into value added products. Given the nature of a biorefinery, as a facility processing biomass, its sustainability may be defined or assessed, taking into consideration its interaction with surrounding systems. In LCA terms, a biorefinery system may be divided into the various parts that have to be included into the Life Cycle Inventory (LCI) i.e. the background, the foreground, and the outputs (Figure 1-2), where the background and outputs interact with external systems, while the foreground is unique to the case study biorefinery.

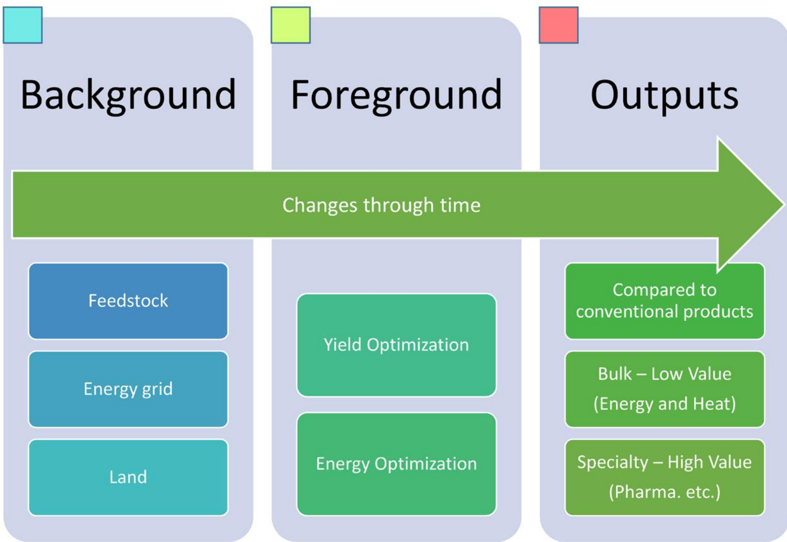


Figure 1-2 Sustainability nexus of a biorefinery

Components of each of these three parts are shown in Figure 1-2. The impacts generated by each of the three parts give a holistic representation of the potential environmental impacts of a biorefinery. However, depending on the goal of the assessment, more or less detail may be included into the LCI. For example, when making a political decision on subsidies for various biotechnologies, large scale implementation impacts may be important to consider in a regional context (Scarlat et al., 2015). Another factor of relevance may be the technology's performance in time (Ericsson et al., 2013). By contrast, if the decision entails choosing between two production pathways producing the same product from the same raw material, then it may be enough to assess the differences in processing parameters e.g. energy consumption, yields (Karka et al., 2017). These methodological choices and their relevance are discussed more in detail, starting with the background system.

1.2.1. THE BACKGROUND SYSTEM AND TERRITORIAL PERSPECTIVE

Raw material sourcing, geographically specific energy grids and land are all part of the background system that populates an LCI. Due to the capacity of some biorefineries to process very large amounts of biomass e.g. capacities from wood chips plants range from 2 MWe to >80MWe (BASIS, 2015; Bridgwater, 2003), constraints and impacts of sourcing of the raw material are relevant in determining sustainability. Mass flow analysis for large geographical areas has been applied to assess the impacts of cities under the concept of urban metabolism (UM) (Newman et al., 1996). In UM, an urban area is seen as a living organism requiring materials and energy to carry out basic functions, which in turn produces waste as a byproduct of living (Pincetl et al., 2012). Such mass flow analysis can give a comprehensive understanding of the available resources or needs of a city and the impacts attached to it. But rather than only looking at direct consumption, coupling of LCA with UM allows for including the impacts of upstream and downstream activities associated with the mass flows needed in the urban area (Goldstein et al., 2013). As demonstrated by the work of Goldstein et al. (2013) UM-LCA gives greater understanding of the flows carrying significant environmental loads, which allows for easy hot-spot identification of key metabolic contributors.

Similarly to UM-LCA, the concept of territorial LCA (T-LCA) seeks to apply LCA to meso- and macroscale objects/areas (Guinée et al., 2011; Hellweg and Canals, 2014). Two types of T-LCA have been identified, which differ in their system boundary application. The first contextualizes the LCA to a specific production or consumption activity, bound in a georeferenced territory. The second, takes into consideration the whole of consumption and production activities happening in a georeferenced territory, which is treated like a black box (Loiseau et al., 2018). Emphasis is placed on constructing spatially explicit inventory for explicit scenario development, which means data requirements are high.

A third option, the Territorial Metabolism LCA (TM-LCA)(Sohn et al., 2018) which is applied in this project (paper I, III, and VIII), is closely related to UM-LCA while incorporating some aspects from T-LCA. Like UM-LCA, TM-LCA is used to assess environmental impacts at a large scale i.e. city, region level, etc., but here a distinction is made and the territory is defined as the discontiguous area where the production/service in questions takes place, while unchanging background systems are excluded. Such a ‘producer territory’ also falls within the confines of a geopolitical entity and is certainly in a specific country. Data at the producer level in combination with data from the geopolitical region/country are used to build the LCI, with the advantage that a discontiguous area of production is less data intensive and thus more manageable without loosing its utility i.e. in relation to the other methods (UM-LCA and T-LCA). Thus, TM-LCA can be used to carry out large scale assessments with a systems perspective of the implications a change to a production type might have in a given region, linked to its location by regional data which provides information about potential improvement due to e.g. availability of raw material in the geopolitical region. Since the mass flow analysis inherently carries information about potential exploitation and also constraints, TM-LCA also lends itself well to finding potential unforeseen problems in the value chains assessed (logistics).

Another aspect that may be included into LCA and also TM-LCA is dynamic changes through time. As evidenced by the work of Sohn et al., (2020) in their review of dynamic LCA, the practice of including dynamic LCA, at least partially, is becoming more common among LCA practitioners. In their work, many types of dynamism are identified of which two have been applied in this project, namely dynamic systems inventory and dynamic process inventory. The advantage of including dynamic aspects is that it allows for prospective LCA and may provide added information about how sensitive the system is to changes in e.g. the background system due to changing energy grids. Though dynamic LCA is not necessary in every case, Pinsonnault et al., (2014) showed the biomass and biofuels sectors to be highly sensitive to changes in the background system, after varying the background for 4,034 product systems, though it is worth noting that the changes were mild for 95% of the cases studied. Still, other ways to handle dynamism, as for example with different temporal resolutions (month vs. years) in a case of domestic hot water, exhibited rank reversal for the best environmental performer (Beloin-Saint-Pierre et al., 2017) and timing of GHG emissions has in more than one occasion been demonstrated as an important aspect with varying potential for global warming potential outcomes (Ericsson et al., 2013; Levasseur et al., 2013; Schmidt et al., 2015). Thus, the added information from dynamics into the system may be of benefit for the decision making process and for ensuring that LCA results obtained today hold in the future.

1.2.2. The Foreground and Early Design Perspective

In an effort to move the industry towards more sustainable ways, several methods under the umbrella of "industrial ecology", have been applied in industry (Jacquemin et al., 2012). These include the establishment of the Green Chemistry principles (Anastas and Eghbali, 2010), risk assessment, environmental impact assessment and of course LCA. When applied to industry special focus is placed on the development of clean technologies by optimization of process design. This also applies to biorefineries, which have a foreground made up of several unit processes that transform biomass into valuable products. Since the 1990s application of LCA for multi-objective optimization of industrial technologies began its journey (Furuholt, 1995) when it was first applied as a tool for process optimization (Kniel et al., 1996; Stefanis et al., 1995). Since then, LCA has been applied many times in a "cradle-to-grave" or "cradle-to-gate" approach, to find environmental "hotspots" in the value chain (Parajuli et al., 2017), for the selection of operating conditions (Gerber et al., 2011) and in combination with other optimization tools (Levasseur et al., 2017; Sukumara et al., 2014). Among promising combinations, is the combination of techno-economic assessment TEA and LCA, which covers economic and environmental spheres well (Patel et al., 2016).

There are many reasons to combine TEA and LCA. To carry out a TEA, an inventory of, among other, the foreground (unit processes) is carried out, with the objective to obtain an overview of capital and operational expenditure for the specified project. The TEA inventory is thorough and contains the information needed for an LCI e.g. electricity and heat consumption, yields, biomass demand, some environmental emissions, etc. These information allow for easy identification of hotspots from both an environmental and economic perspective and gives the chance to carry out process optimization (Barlow et al., 2016; Cai et al., 2018). Furthermore, the data produced may then be summarized into valuable indicators such as production costs, net present value (NPV) or internal rate of return (IRR).

Recently, TEA and LCA have been combined into concrete decision support through the monetization of externalities (Ögmundarson et al., in press). This method has the advantage of presenting the results from the LCA and TEA as one single indicator result, in monetary terms, which is easy to understand. As per the authors, special considerations should be made so that the functional unit and technological system boundaries for both TEA and LCA are aligned (Ögmundarson et al., in press). The drawbacks of this method are the uncertainties of the monetary amounts assigned for the impact indicators from the LCA, which as some authors point out, need further refining of the framework for monetization of impacts bias (Kalbar et al., 2012; Ögmundarson et al., in press; Sohn et al., 2017). Another option for deriving clearer interpretation of combined LCA and TEA results is applying multi-criteria decision analysis (MCDA), such as the Technique for Order of Preference by Similarity to Ideal Solution (TOPSIS) method (Hwang and Yoon, 1981). This is a versatile method that can be used on LCA results alone, or may be used to combine economic and environmental information. Furthermore, the method has the added advantage of reproducibility, thereby it has recently been applied to attain transparent and repeatable decision support (Kalbar and Das, 2020; Köksalan et al., 2011).

1.2.3. The Product Perspective

The outputs of a biorefinery vary widely between low value bulk products, such as energy, to niche products, such as drop-in biochemicals and pharmaceuticals (de Jong and Jungmeier, 2015). To account for the benefits of biorefinery products in LCA, credits are assigned to biorefinery outputs based on either system expansion or allocation, or a mix, depending on the type of LCA and methodological choices taken (Ahlgren et al., 2013). Product substitution for emerging innovative products has to be considered carefully, for various reasons (Civancik-

Uslu et al., 2018). Missing information on functionally equivalent products on the market, lack of information on the products material properties or formulation, incomplete understanding of the product's performance at full scale and missing standardized data in the available LCA databases, are some of the issues making the choice of substituting product or allocation of product credits complex (Civancik-Uslu et al., 2018). Furthermore, when comparing biorefinery products to conventional products that are functionally equivalent, emerging products often lose out, due to an often more burdensome processing life cycle stage and lower TRL (Yates and Barlow, 2013).

In relation to the products assessed in this project, which include biodegradable biosourced polymers, biocomposites, biogas/energy, digestate and protein, several methodology gaps and challenges have been identified.

A common pattern observed in LCAs of biodegradable materials is higher impacts during the end of life stage (EoL). Biodegradable products, such as polylactic acid (PLA), thermoplastic starch (TPS), and the family of polymers polyhydroxyalkanoates (PHA), including polyhydroxybutyrate (PHB) and poly(hydroxybutyrate-co-hydroxyvalerate) (PHBV) require new EoL options for appropriate disposal that can exploit their inherent value of degradability. When assessed through LCA, generally, the preferred order for disposal of these materials from lower to higher impacts is recycling followed by composting, incineration and finally landfilling (Beigbeder et al., 2019; Lazarevic et al., 2010; Rajendran et al., 2012; Wäger and Hischier, 2015). These studies agree with the waste hierarchy established by the EU under the Waste framework Directive (European Parliament and Council, 2008). However, studies do not always align with the waste hierarchy and composting can at times be the worst EoL option (Hottle et al., 2013; Rossi et al., 2015). A few of the key parameters causing this misalignment between LCAs originate from variability of composting and landfilling conditions, the treatment of biogenic emissions in the polymer and different assumptions regarding the degradable fraction of each polymers under different disposal conditions. These same considerations apply to bio-composites, which exhibit similar properties as biodegradable polymers during EoL. Nevertheless, for recycling of biodegradable materials to become a viable option, separate collection of the materials must be justified and infrastructure must be in place, which is not yet the case (Rossi et al., 2015).

On the other hand, inclusion of impacts for conventional plastics is badly covered in LCAs. For example, polyethylene (PE) and other conventional plastics, are also susceptible to biodegradation and though their degradation in the environment may be slow, the production of methane and volatile organic compounds (VOCs) from this source should not be disregarded (Royer et al., 2018). Adding to this is the unaccounted microplastic fraction and the effect of micro and macro-plastic litter on ecosystems (Anbumani and Kakkar, 2018; Woods et al., 2019). Though these are beginning to appear in the LCA literature, efforts are necessary so that it becomes common practice (Boucher et al., 2019; Ryberg et al., 2019).

Thus, an early development and more burdensome production phase for emerging biodegradable materials, has to be weighed against the incompletely covered burdens of conventional fossil-based plastics in order to give a clear picture of the competitive potential of biobased plastic and biocomposites. An expansion of the LCA methodology to include plastic impacts, together with creative ways of including benefits of biodegradable materials may be necessary. As for example, in the case of bioactive packaging i.e. packaging delaying rotting of the contents. In a few of these cases, an increase of 30% in shelf-life (Lorite et al., 2017) and a 6% reduction is food waste (Dilkes-Hoffman et al., 2018) would make the packaging more competitive against their fossil-based counterparts. For innovative bioactive packaging, advantages may be represented by an extension of the system boundary, in order to include the impacts of delaying food waste creation.

Another product coming out of biorefineries that must be carefully considered when assessing impacts is digestate and other products containing nutrients, and being of little value for anything else than as a fertilizer. As a living product, emissions from field application of digestate and its effect on plant yields can vary widely depending on: 1) digestate characteristics, such as nutrient content and plant availability, volatile solids (VS) and total solids (TS) content, etc., 2) soil characteristics, such as fertility level, soil texture, C/N ratio, bulk density, in combination with 3) climatic conditions such as soil temperature, moisture and finally 4) management practices such as time of incorporation, use of other fertilizers alongside digestate, ploughing, etc. Thus, the comparison of the application of a conventional fertilizer, or undigested manures to the application of soil and the subsequent emissions formed is a complex task with extreme site specificity, invalidating the possibility to make generalizations. When one considers the wide variability of factors governing the emissions, it is no surprise that there is still no consensus on whether the application of digestate leads to lower or higher environmentally relevant emissions (N_2O, NO_3^-, among other) and crop yields than mineral fertilizers or undigested manures (Möller and Müller, 2012), though some consensus exist when it comes to NH_3 emissions generally thought to be higher for digestates (Nkoa, 2014). Needless to say, an error in the N_2O emission factor, which has an impact factor of 298 kg CO_2 equivalents for global warming potential (GWP) is capable of introducing large errors to any LCA.

The three perspectives are discussed further in the following chapters, in relation to the project's findings.

2. Territorial Application and Temporally Dynamic LCA

This chapter addresses the first research question by first presenting an overview of the method used in paper I, III and VIII. The methodology concept is outlined step by step in (Sohn et al., 2018) (Paper V). Here, focus is placed on giving a brief overview of the method and discussing the application of it, while synthesizing the most relevant results for the project. An overview of the key methodological choices, systems, feedstock, and products analyzed in the three papers is provided in Table 2-A.

RQ1: *Which factors are potentially result-altering when deciding on the best technology to region pairing now and in the future?*

2.1. Method in Brief

From Paper I "assessing large systems, […] can be approached by defining the geographical boundaries in terms of a "producer territory" (Sohn et al., 2018) so that the LCA can be applied for assessment of a delimited "territory", e.g., wine-producing areas, within a broadly defined region, e.g., Southern France. The producer territory is thus defined as the area of interaction between the aggregated producers and other systems within the region. The TM-LCA framework reduces data demand by aggregating individual areas of the production of, for example, a specific product, supply chain or waste treatment technology, while ignoring unchanging background systems, i.e., only changes to the region interacting with the producer territory are assessed. At the same time, representativeness is increased by merging local inventory data from individual producers with regional and nation-wide data in order to fill in data gaps. In this way, an environmental performance improvement in the territory, due to, e.g., the implementation of a new technology or new management technique, can be quantified in the non-contiguous production area and is reflected in the results for the region. When combined with dynamic and prospective LCA (Sohn et al., 2020b), this approach offers a comprehensive assessment that gives temporally and geographically resolved results. Moreover, it has the added utility of providing prospective insights that can more accurately support decision makers, production owners, and technology developers (Sohn et al., 2018)."(Croxatto Vega et al., 2019)

"A point of departure for the application of the TM-LCA framework is the functional unit. The functional unit, the treatment of one ton of feedstock of specific composition, is treated by two different technology alternatives, described in more detail below. From here, the following steps are applied:

a) Alternative technology is defined.

b) The producer territory is defined and limited to systems interacting with the technological options being assessed within a geographical region.

c) Temporal dynamics are incorporated into the systems, e.g., in dynamic background electricity energy provision and technological efficiency improvements [in the foreground system].

d) The assessment is scaled to encompass the whole region so that all feedstock available that may fulfill the functional unit is treated by the technological alternatives being assessed. However, only changes in systems and in the region are assessed" (Croxatto Vega et al., 2019).

In Paper I, two technology systems were assessed; a biogas only scenario produces biogas and digestate, while a PHA-biogas scenario produces PHA, biogas and digestate. The multi-product output is included in the LCA through system expansion and biogas is valorized in a combined heat and power engine (CHP). In Paper III, an additional AD-based technology scenario is added, the AD-Booster, which is based on wet explosion (Aps et al., 2017), which, “increases the conversion yield of cellulose to biogas from 52% to 88% and the conversion yield of hemicellulose to biogas from 75% to 98%, [in comparison to AD]” (G. Croxatto Vega et al., 2020).

The functional unit is the treatment of 1 ton of agricultural residues (Paper I, III, VIII). In Paper I the residues have a composition of 50% liquid cow manure, 15% solid cow manure, and 35% wine pomace. For the territorial assessment, the functional unit is scaled up i.e. all available feedstock as described in the FU contained in the region is used for the assessment. The two wine producing regions are the Languedoc-Roussillon region in southeast France and the Willamette, Umpqua, Rogue, and Columbia valleys of Oregon State in the USA. The feedstocks come into the system burden free, since they are by-products and thereby the burdens are allocated to the primary production i.e. meat production and wine production. This is the procedure followed for scale up of the functional unit in all three papers, with some differences arising due to the types of feedstock considered (see Table 2-A).

The foreground systems for Paper I and III were modelled using the process design software Superpro Designer ® (Intelligen Inc, 2018). The OpenLCA software (GreenDelta, 2019) was used for the LCAs (all Papers in the thesis), along with the Ecoinvent v3 database (Wernet et al., 2016). The ReCiPE Hierarchist (H) (Huijbregts et al., 2017), method was used for impact characterization. All impact categories were included in the assessments and analyzed at midpoint (Paper I, III, VIII) and endpoints (Paper III, and VIII).

Dynamics were included as follows in Paper I. “Dynamic inventories of the electricity mix for the two locations, modelled for a period of 20 years from 2015–2035, were used in the analysis. Four different dynamic energy futures, developed by the French government, with yearly shifting percentages of contributing sources of energy, were used for all electricity provision in the scenarios for Languedoc-Roussillon (Réseau de Transport d'Électricité, 2014). Likewise, three different dynamic energy futures were developed based on the legislation for Oregon State, which regulates the share of renewables in Oregon's future energy grid (Oregon State, 2017). Qualifying renewables, i.e., renewable energy sources accepted by Oregon legislation on renewables, were introduced in varying amounts. Thus, (1) a scenario where biomass was increased more than other qualifying renewables, (2) a scenario where wind and solar were increased more than other qualifying renewables, and (3) a scenario where all qualifying renewables were increased evenly were developed. Static electricity mix scenarios were also included for both locations." Dynamics in the foreground system, for the PHA-biogas technology, "was modelled as becoming more energy efficient, improving by 1% annually for the 20-year period, based on similar technology learning curves (Bugge et al., 2006)"(Croxatto Vega et al., 2019). This was included since the PHA-biogas technology assessed is expected to improve over the next years due to its early current TRL. Instead, in Paper III a dynamic sensitivity analysis was performed for the background energy system by switching the energy mix of each location for a theoretical green energy mix.

For Paper III, the FU is the treatment of 1 ton of feedstock again and it is scaled up as described above for Paper I. However, an assessment of sustainably available feedstocks, as well as installed anaerobic digestion (AD) capacity was carried out for the two regions; Veneto, Italy and Bavaria, Germany. In this case, the scaled up FU represents agricultural co-products normally grown in these regions and compatible with AD, as well as feedstock used in two real farms that provided data for their AD operations. These two farms, with two different plant scales, i.e. 200 kWe and 1 MWe, were included in the assessment.

Table 2-A. LCA methodological aspects in regard to the territorial perspective.

Parameter	Paper I	Paper III		Paper VIII
Biotechnologies assessed	Biogas plant PHA-Biogas plant	Biogas plant PHA-Biogas plant Biogas+AD-Booster		Biogas plant PHA-Biogas plant Polyphenol extraction Filler/Biocomposites All in various compatible biorefinery setups
Regions	Willamette, Umpqua, Rogue, and Columbia valleys of Oregon State, USA Languedoc—Roussillon, FR	Bavaria, DE Veneto, IT		Bavaria, DE Languedoc—Roussillon, FR Oregon State, USA Skåne, SE Veneto, IT
Functional Unit	1 ton of agricultural residues	1 ton of agricultural residues And all biogas compatible feedstock in the region		1 ton of agricultural residues (phase 1) All straw, wine pomace, animal manures, and vine shoots in the regions (phase 2)
Inputs % wet weight	50% liquid manure 15% solid manure 35% wine pomace	Veneto 59% cow manure 7% pig manure 2% chicken manure 8% energy crop 3% straw 4% maize straw 8% sugar beet pulp 6% soybean straw 3% wine pomace 1% vine shoots	Bavaria 53% cow manure 7% pig manure 27% energy crop 11% straw 1% maize straw 1% sugar beet pulp	For the regional assessment, the percentages of feedstock change so that the regional feedstock is used by the biorefinery setups in order of best environmental performance to least.

Input allocation	No impacts allocated to the inputs	No impacts allocated to the manures, wine pomace, vine shoots. 100% of impacts of energy crop production are included Ecoinvent allocated straw is used. Economic allocation for the rest.	No impacts allocated to the manures, wine pomace, vine shoots. Ecoinvent allocated straw is used.
Output and allocation	Biogas - Valorized in CHP, Electricity substitutes average energy mix, heat is not valorized in these regions PHA - substitutes either PLA or PET in different ratios based on material properties Digestate - substitutes ammonium nitrate at a 67.5% ratio. Field emissions of digestate are modelled against field emissions of the mineral fertilizer replaced.	Biogas - Valorized in CHP, Electricity substitutes average energy mix, heat is utilized for District Heating (DH) only for DE, otherwise it is not valorized. PHA - substitutes average global thermoplastic production 1:1 ratio as granules. Digestate - substitutes manure and manure application. Field emissions are not included. Emission from reduced storage of manure are included.	Biogas – average utilization which includes: Valorized in CHP, electricity substitutes average energy mix, heat is utilized for District Heating (DH) only for DE and SE. Upgraded to biomethane of natural gas grade (IT, DE, SE), for utilization in transport (SE). PHA and biocomposites- substitutes average global thermoplastic production 1:1, and 0.3:1 ratio as granules, respectively. Polyphenols – substitute ascorbic acid, ratio of 1:1. Digestate - substitutes manure and manure application. Field emissions are not included. Emission from reduced storage of manure are included. (Details in Table 2, Paper VIII)
System Boundary	Cradle to gate	Cradle to gate	Cradle to grave
Dynamic	Background and foreground systems	Background for sensitivity analysis	None
Type of Data	Average data	Average Data	Average Data
LCIA	ReCiPE Hierarchist midpoints	ReCiPE Hierarchist midpoints and endpoints	ReCiPE (H) midpoints and endpoints

Only feedstock not typically valorized, other than for biogas production, came into the system burden free, while for dedicated crops, the full burden of their production was accounted. “For agricultural residues currently valorized in the market, such as sugar beet pulp, corn stover and soybean straw, the burden of production was distributed by economic allocation, while for wheat straw an existing Ecoinvent process was used”(G. Croxatto Vega et al., 2020). A TEA was performed on the two plant scales and at regional scale to clarify the economic potential of the technologies. For the territorial assessment only the 1MWe plant size was considered due to guidance provided from the TEA, which put into evidence that installing equipment required for PHA and AD-Booster production at a small scale (200 kWe) was not economically feasible (discussed further in Ch. 5).

A few modifications were necessary in Paper VIII, in order to answer the overarching research question. For this study, the technologies were analyzed in a modular way, using the results generated by Paper II, III, and VII on the following biotechnologies: polyphenol extraction, AD and AD+PHA, and filler, respectively in addition to many synergistic combinations between the aforementioned technologies (Figure 1, in Paper VIII). This results in 16 mini-LCAs per region with a FU as the treatment of 1 ton of agricultural residues (100% of the same residue), though the residues considered were limited to animal manures, straw, wine pomace and vine shoots. Regional feedstock is then scaled up as the functional unit, which is determined with guidance from the mini-LCAs. “ In order to select the most environmentally preferable [biorefinery value chain] for a given region, an order of preference metric was used. This metric was carried out through a sequence of logic consisting of a series of binary questions. The first of these questions is ‘is this technology shown to be the most environmentally preferable (based on the given environmental impact measurement, technology, and feedstock pairing)’. If the answer is yes, then it is assumed that as much of the feedstock as technically possible should be used in said technology, barring an overriding factor. If the answer is no, then the next available technology for the feedstock is queried in the manner previously described. Once the most preferable technology for a given feedstock-region pairing is selected, it is determined if the maximal use of said feedstock would preclude the most preferable use of any other assessed feedstock available in said region. If no preclusion is found, then it is assumed that a maximum technically feasible amount of the given feedstock should be allocated to said feedstock's most environmentally preferable technology. If preclusion of another technology that is most preferable for another feedstock is found, then the two competing technologies are compared. This is carried out as follows: if, based on a given region and its feedstock availability, there are two competing feedstocks, F_a and F_b, with two technologies, T_a and T_b, the potential environmental impact of utilization of a technically maximal amount of F_a and F_b in T_a is compared to the potential environmental impact of utilization of a technically maximal amount of F_a and F_b in T_b. Thus, if the lesser environmentally valued utilization of a feedstock-technology pairing results in an overall system benefit, the lesser environmentally valued technology is still selected. This results in a given regional system utilizing the overall most environmentally beneficial mix of technology-feedstock pairings as possible" (Nilsson et al., n.d.).

2.2. KEY RESULTS AND DISCUSSION

In regard to RQ1, when assessing the impact a biotechnology may have in a specific regional context, whether it is for a delimited producer territory or a geopolitical region, the influence of the background energy system is important. Paper I showed that evolving energy grids i.e. the increase of renewables in the average energy mix of a location, has consequences for the LCA results. For systems producing large amounts of energy such as the biogas and PHA-biogas plants shown in Figure 2-1, increasing renewables mean lower credits from substitution of fossil energy production. In Figure 2-1 this is observed more clearly in the Oregon State scenarios, which have a sharp decrease in the share of fossil-based energy due to political targets. The sharp decrease (solid blue lines) means that the difference between the biogas and the PHA-biogas plant increases by 50% from 2015 to 2035, where the negative values indicate that the PHA-biogas plant incurs higher savings to GWP than biogas only. On the other hand, the static scenarios show that the difference between the biogas only scenario and the PHA-biogas scenario stays the same in time. But, this is in a way misleading once one compares with the dynamic scenarios, where one can make a more definitive choice for the PHA-biogas scenarios due to the large difference between the two. Material production in the form of the bioplastic PHA, which in this case replaces either PLA or PET, keeps the PHA-biogas savings more stable through time in terms of GWP, since this factor is not affected to a high degree due to changes in the background mix.

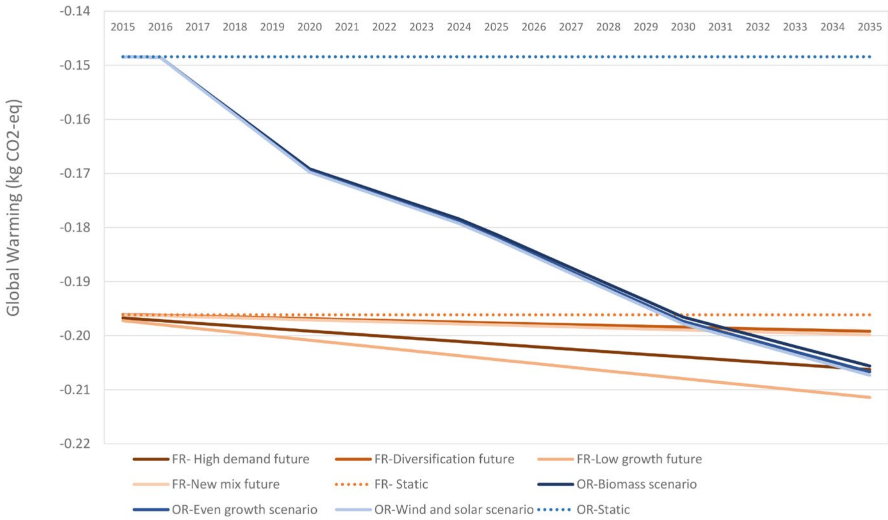


Figure 2-1 Yearly difference of global warming potential (GWP) impacts, i.e., PHA-biogas minus biogas-only scenarios. Figure reflects the evolution of the energy mixes in the two locations. Negative values mean PHA-biogas has higher savings than biogas-only. Source:(Croxatto et al., 2019).

Similarly, the variation of present energy grid to a future theoretical energy grid in Paper III, has large consequences for GWP. With present energy grids, (Figure 2-2 and 2-3), the scenarios producing the highest amounts of electricity, namely the AD+Booster technology followed by regular AD, are the best performing scenarios in terms of GWP.

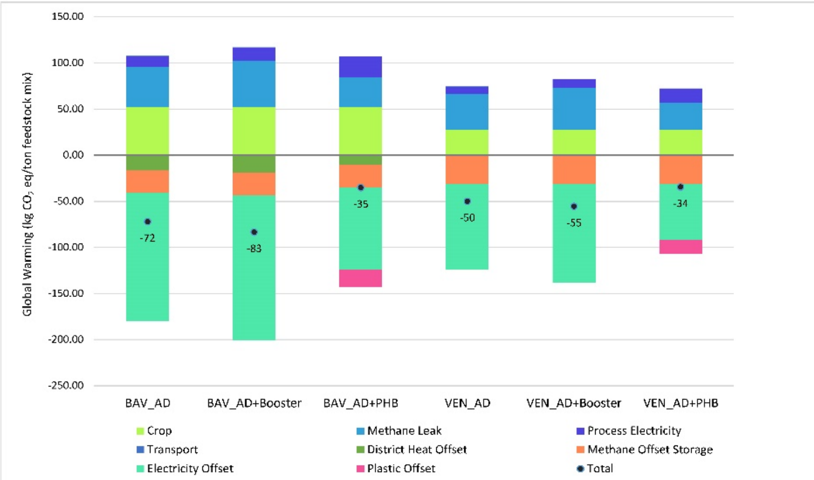


Figure 2-2 GWP contribution per ton of feedstock mix for the two regions, BAV for Bavaria and VEN for Veneto, for the three technology options i.e. AD, AD+Booster and AD+PHB. Source: (G. Croxatto Vega et al., 2020)

Furthermore, an important aspect regarding feedstock composition can be appreciated in these two figures, namely that the feedstocks for which burden of production is included fully or partially i.e. energy crops, straw, corn stover, sugar beet pulp, soybean straw etc., have a higher biomethane potential (BMP) and the energy savings attained out-weigh the production burdens that are accounted. This is true for both regions assessed, as well as both scales assessed (Figure 2-3). However, with a clean energy future this ceases to be the case, and the scenarios with the highest energy production have the highest impacts for GWP (Figure 2-4).

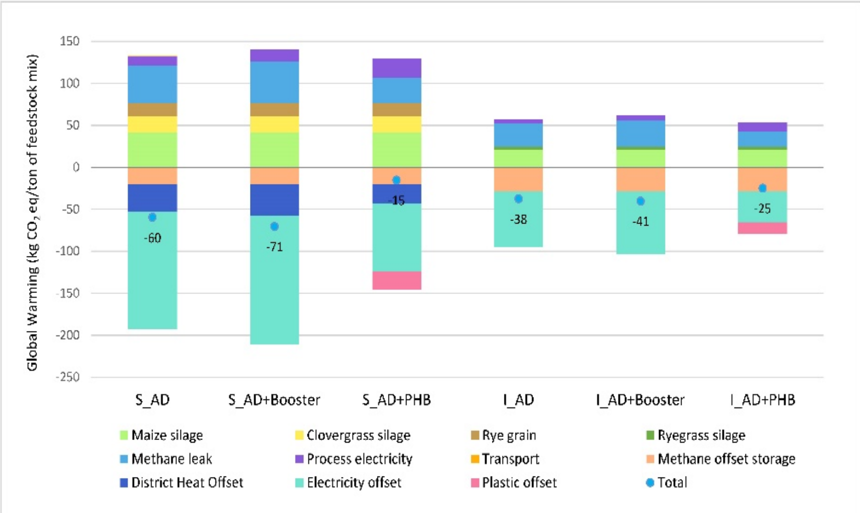


Figure 2-3 Global warming potential results for the small scale (200 kW) and Industrial scale (1000 kW) cases, per ton of feedstock, as well as contribution to GW by each stage. Scenarios are named as S for small scale and I for industrial scale followed by each technology scenario (AD, AD+Booster, AD+PHB). Source: *(G. Croxatto Vega et al., 2020).*

All else equal, the savings from replacing electricity shrink with the theoretical green future. This has consequences for GWP but not for the remaining impact categories, which highlights the possibility to shift burdens from one IC to others, if the decision was to be solely based on GWP (Laurent et al., 2012). This is the reason for applying a refined method for weighting the results and producing single score indicators (presented in Ch. 4).

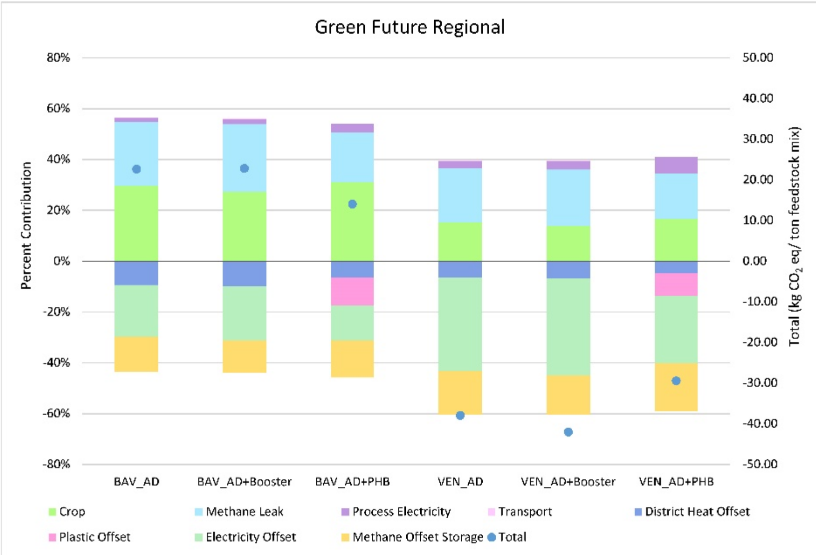
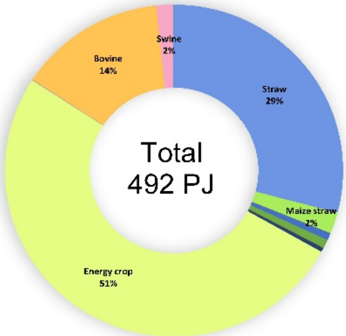


Figure 2-4 Global warming potential for a future with a theoretical green energy mix. Scenarios are named by the first three letters of the region (VEN or BAV) followed by each technology scenario (AD, AD+Booster, AD+PHB). Source: *(G. Croxatto Vega et al., 2020)*.

On the contrary, the inclusion of a dynamic foreground system in Paper I, for the PHA-biogas technology, did not result in large variations in the results. The changes to GWP induced by reducing the process energy consumption by 1% per year, produced an improvement in GWP ranging from 0.1% to 1.5% of total GWP. Predictive technology development is a challenge for the LCA practitioner, and can be approached in various ways. The approach taken here, which is to emulate similar technology learning curves is considered appropriate (Arvidsson et al., 2018) and has been used in LCA before (Bergesen and Suh, 2016). It has the advantage that it avoids a potential mismatch between a dynamic background system and the foreground system. However, the improvement modelled in this case, which is for energy efficiency of the PHA-biogas technology overall, is not the only factor that might improve for this technology in the future. Yield improvements for the polymer production might indeed be a more important factor of improvement however, correct determination of a future yields for the technology are more difficult to determine. In this case it would be advisable that future research includes possible yield improvements which could be included as scenario ranges, which test extremes (Arvidsson et al., 2014).

The territorial assessment allows to put absolute savings into perspective. In Paper I, for example, even though not all the AD compatible feedstock was analyzed, it was shown that "GWP impacts [could be] normalized using planetary boundary carrying capacity-based normalization factors (Bjørn and Hauschild, 2015). Assuming a 985 kg CO_2 eq. per person year (PY) carrying capacity (C.Cap) (Bjørn and Hauschild, 2015), and assuming that PHA replaces PET with a 93% replacement ratio (RR) and that the PHA process improves in terms of energy efficiency at 1% annually, the production of PHA induces an average reduction in GWP impacts relative to biogas-only equating to nearly 1400 PY of C.Cap. When broken down by region, the French scenarios indicate an average relative maximum potential GWP saving of over 2400 PY of C.Cap, with Oregon exhibiting just over 80 PY of C.Cap in average relative maximum potential GWP savings" (Croxatto Vega et al., 2019). Similarly, in Paper III, a comparison between the two regions showed that "the Bavarian region is capable of obtaining GWP savings 7.4, 7.7 and 5.4 times higher than in the Veneto region for AD, AD+Booster, and AD+PHB, respectively, on an annual basis. This is explained in part by the scale of the regions, feedstock density of the regions, as well as the energy density of each feedstock employed in the mix. While Veneto is also the smaller of the two regions, the lower GWP savings are partly due to an average 25% lower feedstock mass production per area relative to Bavaria. Moreover, the regional feedstock mix in Bavaria contains ca. 7% more crops and crop residues, among which maize silage is a prominent one, whilst Veneto contains ca. 7% more animal manures, which have a low methane/VFA productivity" (G. Croxatto Vega et al., 2020). This is similar to findings by other authors, who report that agricultural yields and farming intensity has indeed a positive influence in environmental GHGs savings in other regionalized LCAs (Dressler et al., 2012; O'Keeffe and Thrän, 2020).

Furthermore, by closer inspection of the two regions, by the full analysis of sustainable feedstock availability it was also possible to appreciate the level of utilization of the feedstocks in each regions, in comparison to their total potential calculated based on electrical capacity (Figure 2-5).



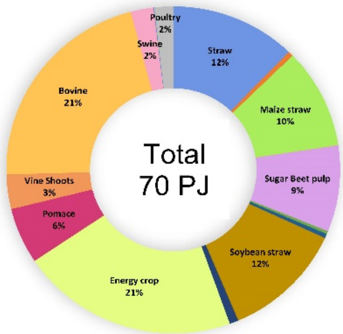


Figure 2-5 Complete energetic potential from agricultural residues for Bavaria (left) and Veneto (right) as % energy basis (without the removal of built AD capacity). Source: *(G. Croxatto Vega et al., 2020)*.

Considering the built AD capacity of each region, it was estimated that "153 PJ and 38 PJ remain as unexploited feedstock" (G. Croxatto Vega et al., 2020), meaning that roughly 69% and 46% of the feedstocks assessed are currently being used in Bavaria and Veneto, respectively. Upon closer inspection of the literature, it was revealed that pressures on land use in Germany have been high due to feed in tariff assigned to maize, ryegrass silage, among other, previously subsidized by the German government (Thrän et al., 2020). Though the Renewable Energy Act, has also put a cap on the use of land use sensitive feedstock, small scale plant are exempt from sustainability criteria (European

Commission, 2018). Nevertheless, the feedstock availability assessment highlighted that many small plants exert considerable pressure on biomass resources and land resources. Though this is not a definitive land use study, it has the potential to highlight pressures of biomass resources. Compared to other studies that have included land use change (LUC) and indirect land use change (iLUC), it is clear that not including these is a disadvantage. For example, (Tamburini et al., 2020) demonstrated that including LUC and iLUC in regards to maize silage for biogas production results in a failure to attain the sustainability threshold of 60% GHGs savings in comparison to the fossil reference for these plants (European Commission, 2018). Similarly, (Styles et al., 2015) found that the inclusion of iLUC increased GWP burdens by a factor of 3-8 for various biogas and biofuel scenarios, and altered the results from a savings status to burdens. Thus, including LUC and iLUC is recommended when assessing the performance of biorefineries.

Many factors are important when attempting to determine biotechnology to region pairing. Of the factors addressed though this project, the background energy mix seems to have the highest potential to induce changes in the overall performance of biotechnologies, and its change should be carefully considered in LCAs. Results from Paper VIII support this stance once more and highlight the possibility of the technologies to perform differently depending on feedstock and region of implementation. Figure 2-6 shows the variability in performance of pomace, straw and vine shoots, in up to 6 technology setups, in each of the assessed regions. It is, for example, evident that filler performs differently across regions, sometimes providing benefit for GWP, while other times inducing burdens to GWP e.g. filler from pomace and vine shoots in Bavaria. When compared to monetized environmental damages (MED) in US dollars, the same filler producing technology shows disagreement with GWP results, most notably in Skåne, Oregon and Bavaria (refer to section 4.1 for the method used to monetize impact damages).

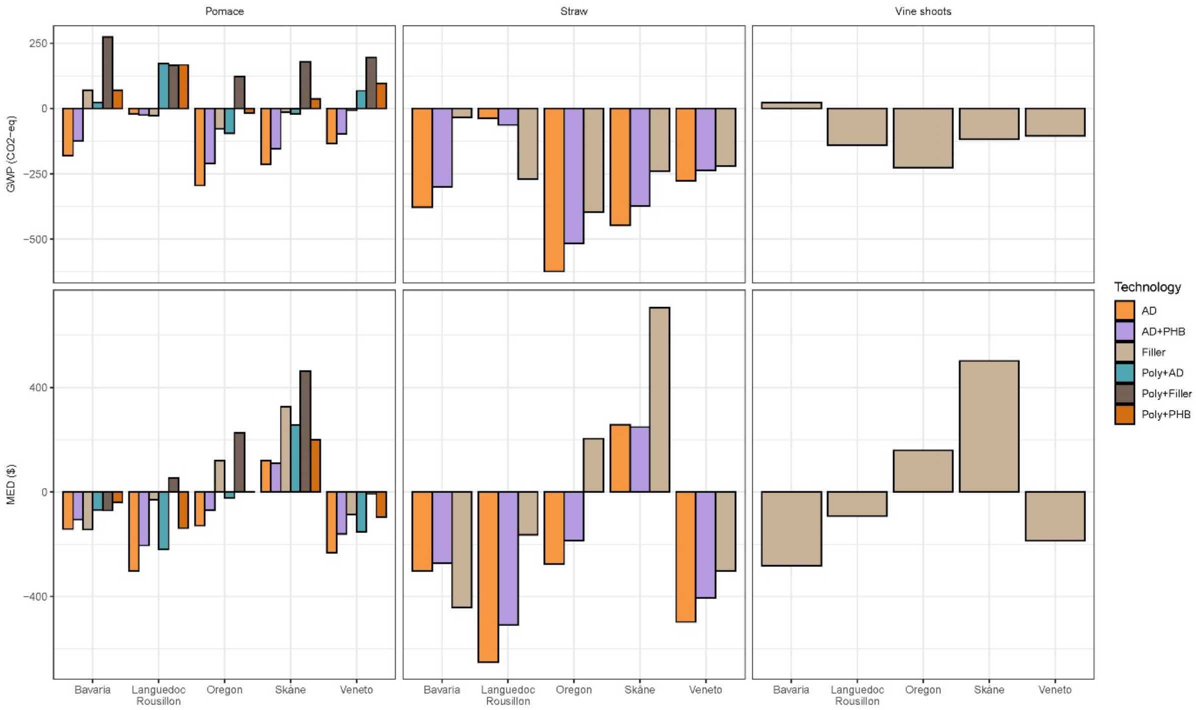


Figure 2-6 Preference according to region, feedstock and biorefinery setup for pomace, straw and vine shoots. Global warming potential (GWP) impacts (top) and monetized environmental damages (MED) (bottom) in U.S. dollars. Source: (Nilsson et al., n.d.)

The differences can partly be explained by variations in the background energy mix of the regions, as well as in utilization of the co-products (biodiesel in Skåne, electricity and natural gas in Oregon). As can be observed in Figure 2-7, electricity needed to process manure into biogas or biogas plus PHA, or to upgrade biogas into biomethane, induced large MED for Skåne, while causing insignificant burdens to GWP, for the same region. Skåne's energy mix, which contains around 50% re-newables, show a high degree of burden shifting when comparing GWP to MED. This is due to impacts coming from impact categories other than GWP, from the activities that require electricity.

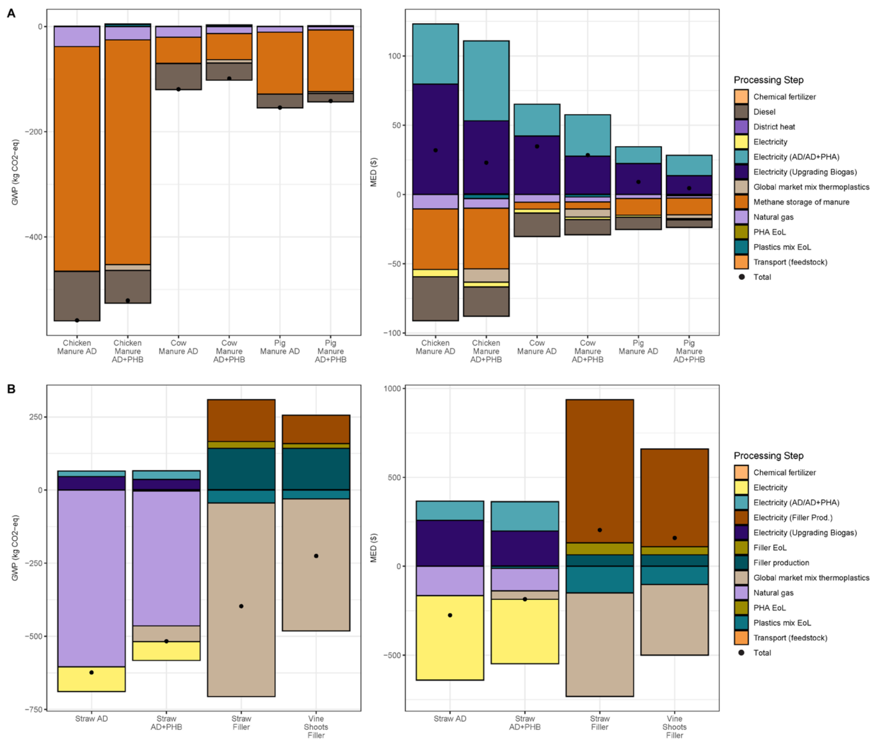


Figure 2-7 Contribution analysis for impacts (GWP) and damages (MED) from biorefining A) manures in the Skåne region and B) biorefining straw and vine shoots in Oregon. Source: (Nilsson et al., n.d.)

Moreover, biorefinery setups show varied environmental profiles for each region due to the potential in each region to scale up depending on feedstock availability, background energy mix, and utilization of co-products. It is, thus, useful to preset biorefinery setups as such (Figure 2-8), so that the most efficient feedstocks in terms of providing high environmental benefits for the region in question can be prioritized for a compatible biotechnology. On the other hand, this highlights the need for context specific assessments and the danger of generalizations.

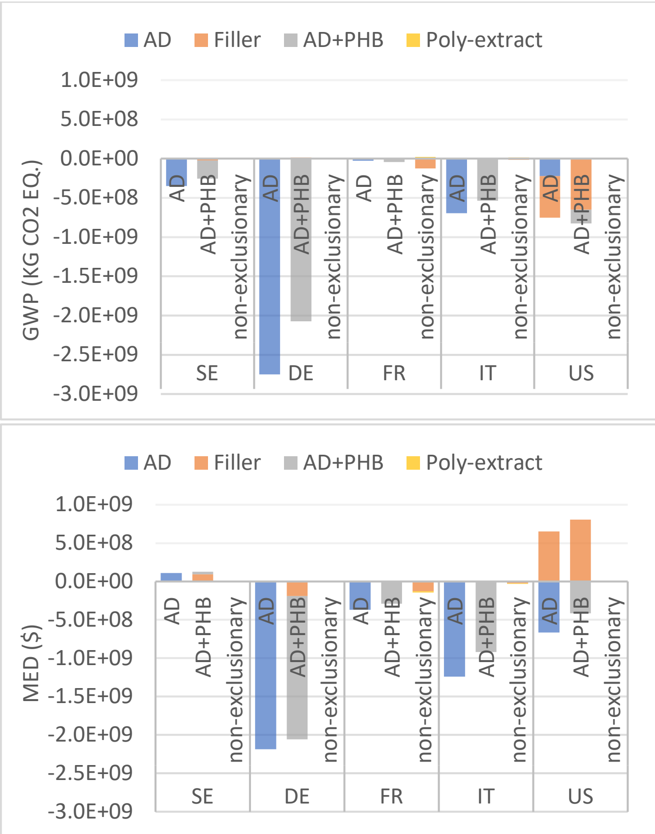


Figure 2-8 Regional value chain implementation environmental impacts for Bavaria (DE), Veneto (IT), Languedoc Roussillon (FR), Skåne (SE), and Oregon (US) for both global warming potential (GWP) and monetized environmental damages (MED), non-exclusionary technologies utilize feedstock that do not preclude the utilization of any other technology in a potential value chain. Source: (Nilsson et al., n.d.).

Though there are many ways to include regionalization and dynamism, whether it is by following political targets as presented in this work, testing energy grids known for their carbon intensity or relative greenness (Ögmundarson et al., 2020b), or following predictions of well respected institutions such as the International Energy Agency or the IPCC (Arvidsson et al., 2018), it is clear that the added level of information is valuable for the LCA. While quantifying the resources available in a region can help give an overall picture of resource use or limitations, adding changes to the background system will increase the likelihood that the results will hold true in the future, and considering the lifetime of these technologies (20-30 years) this factor is of high importance. This is in agreement, with other studies that have considered the biomass sector (Pinsonnault et al., 2014).

3. Combining TEA and LCA for Process Optimization

Process optimization was the focus of Paper II, where various assessment types were applied in order to improve process design. The lessons learned from this paper are described below in relation to RQ2.

RQ2: *How can LCA be used for early design optimization of emerging biotechnologies? (the foreground system)*

3.1. Method in Brief

In order to answer RQ2, LCA was applied at different stages of technology development to two polyphenol extraction methods being developed at low TRL. Polyphenols are bioactive compounds that "have been shown to have excellent health promoting qualities, such as anti-diabetic, anti-inflammatory, anti-bacterial and anti-cancer properties (Nowshehri et al., 2015)." They are commonly used in the food industry as antioxidant rich additives and they can be the key component in new materials, allowing designers to make bioactive packaging i.e. food packaging that extends the shelf life of its contents. Polyphenols are found in large quantities in grapes, and in this case the starting material was residues from wine production i.e. wine pomace.

The aim of Paper II was two-fold: 1) to pinpoint hot spots of two different polyphenol extraction methods in development in the laboratory, and 2) to propose improvements to the process design that would then be scaled up within the NoAW project. In order to reach these goals the following methodology was followed:

- ❖ Data on the two extraction methods, solvent extraction (SE) and pressurized liquid extraction (PLE) was collected from the technology developers.

- A gate to gate carbon footprint (CFP) was carried out, using the operational parameters provided by the two labs. The model was scaled up using data from project partners and implemented using SuperPro Designer, with industrial equipment and scale, without altering key parameters such as yield, or solvent to dry weight (DW) ratios.

- CFP hotspots were identified and the industrial scale design was then modified using information from literature (Dávila et al., 2017a, 2017b; Fiori, 2010; Todd and Baroutian, 2017; Viganó et al., 2017), the CFP and by completing a TEA of the newly designed process.

- The new industrial design was then analyzed with LCA using all impact categories with the ReCiPe (H) LCIA method.

- Finally, results from the TEA and LCA were combined to produce single indicator results (discussed in detail in Ch. 5).

For Paper II, an accounting LCA is enough to fulfil the goals of the study. The functional unit is the production of 1 kg of polyphenols, expressed as Gallic Acid Equivalents (GAE). The assessment was done from gate to gate, meaning that the only life cycle of interest was the production stage. Average data was used for the LCA and CFP. The functional unit and inventories of the LCA and TEA were aligned. The TEA provided information about capital and operational expenditure of each extraction method (CapEx and OpEx). In total, 6 industrial setups were assessed, each having a different solvent to DW ratio.

3.2. KEY RESULTS AND DISCUSSION

In this case, the CFP was useful in pointing out the hotspots of the polyphenol extraction methods. The high solvent to DW ratios used in the laboratory result in increased energy consumption due to high demand for heating and pressurizing of the systems. Thereby, from a quick CFP assessment it could be stated that a high degree of optimization would need to be done to these methods before they would be feasible at industrial scale. The difference between laboratory scale parameters and designed operational conditions can be seen in Figures 3-1 and 3-2.

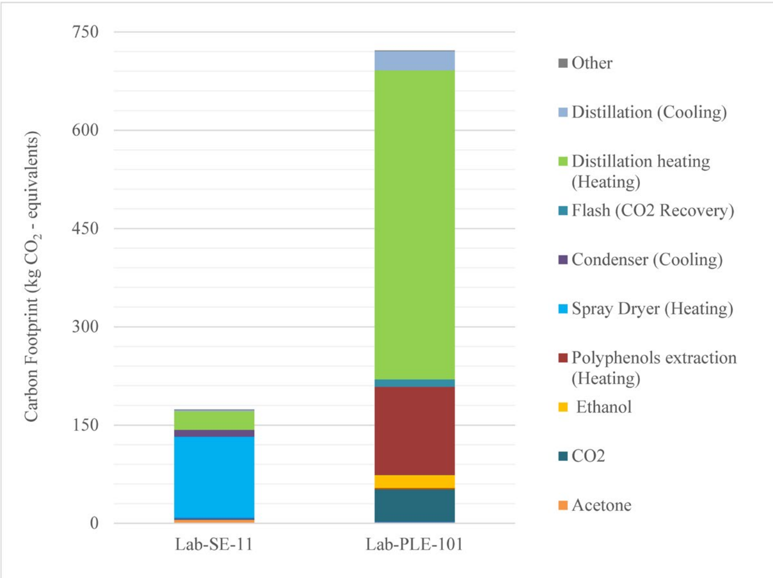


Figure 3-1 Global warming potential results per kg GAE of polyphenol extraction scenarios at laboratory scale. SE is solvent extraction, while PLE is pressurized liquid extraction. The number at the end of each scenario indicates the solvent to DW ratio for the extraction process. Source: (G. C. Croxatto Vega et al., n.d.)

The differences in processing performance were mainly attained by improving the solvent to DW ratios, by adding countercurrent extraction steps in the system. The placement of drying units was also changed, so that concentration steps (filtration) were performed before drying the liquid phase containing the polyphenols. The TEA of the improved design calculated productions costs for each scenario. The LCA of the improved designs aligned well with TEA results, suggesting that at least in this case, expensive extraction steps are a heavy load from both monetary and environmental perspectives.

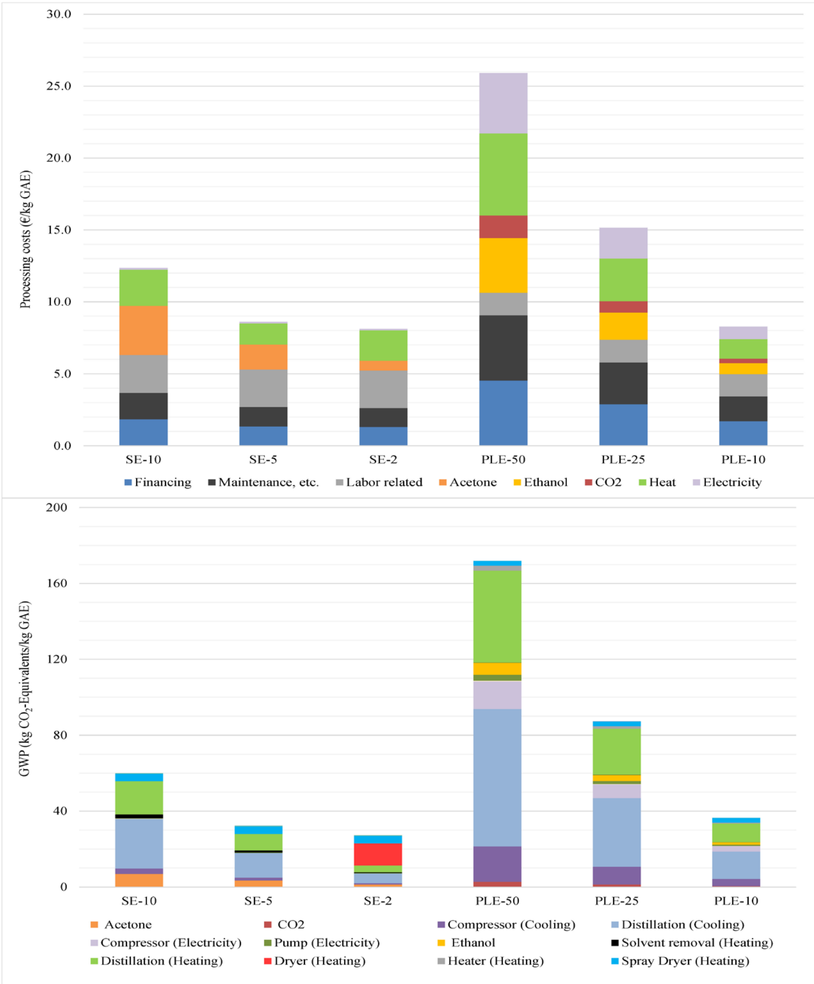


Figure 3-2 Contribution per processing step to Capex and Opex (top) and GWP (bottom) of the designed process at industrial scale, cut-off 1% of overall impact. SE is solvent extraction, while PLE is pressurized liquid extraction. The number at the end of each scenario indicates the solvent to DW ratio for the extraction process. Source: G. C. Croxatto Vega et al., n.d.

In the context of the NoAW project, these assessments were used to provide decision support for the project, which had to choose one method to scale up. For the most part, midpoint impact categories exhibited the same ranking as GWP, but considering the consequences of providing this recommendation, further steps were taken to ensure the robustness of the results. In order to do this, and so that no burden shifting was overlooked, further interpretation was done on these results, explained in detail in Ch. 5. With this increase interpretation level, and iterative meetings with all involved stakeholders in the decision, which is to say the technology developers, the institute carrying out scale up experiments, and those performing the assessments, solvent extraction was chosen as the method to scale up. Technical considerations made it clear that PLE-10 was unlikely to be attainable with the equipment in place.

In regard to RQ2, it was shown that a simple CFP can be used as a useful tool that, without building an overtly detailed inventory, may be able to point out important design hot-spots to new methods of production. That said, it is important that such an assessment is followed by more refined assessments, as done in this work, including but not limited to an LCA with all ICs and TEA. Quantitative process design, using software such as Superpro Designer is advantageous, in a sense that it is a tool that respects thermodynamic laws and thereby produces a more reliable inventory of production, in terms of energy consumption and equipment limitations. Process design has also been singled out in the literature as a way to bridge LCA and the field of biotechnology, since for example, the chemical industry routinely uses process design to test up and coming production methods (Ögmundarson et al., 2018). Thus, the opportunity to combine approaches to assess emerging production methods via process design, with TEA and LCA should be explored in future research. In the absence of process design software, scaling laws can be used to predict industrial settings, though uncertainty would need to be evaluated (Caduff et al., 2012). Together with scenario analysis and testing of extreme ranges of the process parameters having a large influence on the performance of the system, this approach can be a good way to test options at a more mature stage (Arvidsson et al., 2018). The advantage of including TEA, together with LCA, is that it is possible to optimize environmental performance without completely compromising economic viability.

4. INCREASING CLARITY OF LCA RESULTS

Interpreting LCA results is not always a straightforward matter. Considering, for example, the 18 midpoint results for the ReCiPe method for 18 different impact categories is a serious issue for the LCA practitioner, especially when these vary widely in impact. If conflicting economic information is also added into this mix, it make matters even more confounding. Thus, this chapter addresses the methods that have been applied in order to ease the interpretation phase of the combined assessments that have been used through this project, in order to answer the following RQ. Table 4-A shows an overview of the methods applied.

RQ3: *How can bilateral economic and environmental sustainability assessments be combined to increase clarity for decision support? (interpretation)*

4.1. METHOD IN BRIEF

Two methods were applied to the interpretation phase of LCA throughout this project. 1) monetization of environmental impacts based on endpoint damages (Ögmundarson et al., 2018), and 2) the MCDA method called TOPSIS (Hwang and Yoon, 1981), with different weighting profiles.

Monetization, used in Paper III, takes "ReCiPe endpoint damages (Huijbregts et al., 2016) to calculate the external costs of the implementation of a given technology at a given scale or region. This was done through two methods. The first, for ecosystem damages, is based on budget constrained ability to pay, which is used to derive a valuation for species years (Species.Yr) gained or lost (Weidema, 2009), as this is suggested as the least uncertain method for this valuation (Pizzol et al., 2015). For that valuation, 65,000 USD2003 per Species.Yr was utilized. In order to valuate the disability adjusted life year (DALY) loss or gain, a value from Dong et al. ,who assessed a number of different methods, was utilized (Dong et al., 2019). The valuation derived in these different methods varies significantly, on the range of 1 to 2 orders of magnitude. So, here […] the value, 110,000 USD2003 per DALY, [is used], which is also in line with the value derived from budget constraint monetization (Weidema, 2009), which again should have the least uncertainty. Since resource scarcity endpoint damages are already expressed in monetary terms, no further interpretation is necessary" (G. Croxatto Vega et al., 2020).

The second method, TOPSIS, was used in Paper I, II, and III with a few differences in the weighting profile (see Table 4-A).

In Paper I, TOPSIS was used merely to check if there was burden shifting between the GWP IC and the other ICs. TOPSIS was applied with equal weights for all ICs and was compared to GWP to see if preference varied between the two single score indicators.

In Paper II, the information derived from the TEA, i.e. production cost, was used as criteria for applying TOPSIS, as well as the LCA results of the 18 midpoint ICs for each of the polyphenol extraction options tested. Then normalization factors (PRé, 2019) per impact category (i) were used to derive a relative importance factor (RIF), which is essentially the average value of each midpoint IC for each extraction method divided by the normalized impact (per IC) of an average European's annual environmental impact.

In Paper III, for "deriving a single score, based on the ArgCW-LCA method (Sohn et al., 2020a), ReCiPe midpoint environmental impacts along with a valuation of required subsidy for profitability to represent the economic impacts [derived from the TEA] were used as the input criteria for TOPSIS utilizing weighting based on what Sohn et al. describe as a context weighting factor (CWF) (Sohn et al., 2020a). […] For this application, normalization for an average European person year emissions was used (PRé, 2019). Thus, weighting of the environmental impacts is derived, as described in the ArgCW-LCA method (Sohn et al., 2020a), by taking an average of two values: the average of the normalized midpoint impacts for impact category 'i' amongst all assessed scenarios, and the difference of the minimum and maximum normalized impacts for impact category 'i' amongst all assessed scenarios. This accomplishes two things. The first, taking the average of the normalized impacts, scales the importance of emissions of the system to status quo emissions. And, the second, taking the difference between the maximum and minimum normalized impacts, is to scale relative to the ability for choosing amongst the available alternatives to cause significant change in status quo emissions. This was completed for all impact categories resulting in the CWF for the environmental impacts. Economic impacts were ascribed a range of weights relative to the sum of weighting given to environmental impacts ranging from 10%-90%. The system was also run using equal weights for all criteria as a point of comparison to the context weighted and the other single score results" (G. Croxatto Vega et al., 2020).

Table 4-A. Methods for integration of TEA and LCA results into single scores.

Parameter	Paper I	Paper II	Paper III
Method applied	MCDA - TOPSIS	MCDA – TOPSIS	- MCDA – TOPSIS - Monetization of end-point damages
TOPSIS weighting	Equal weights	Equal weights, Relative importance factor	Equal weights, Context weighting factor
Economic weighting	None	10%-90% given to production costs	10%-90% given to subsidy requirement
Single scores compared	GWP compared to TOPSIS with equal weights	TOPSIS with Equal weights and RIF	TOPSIS with equal weights and CWF, and Monetization

4.2. Key Results and Discussion

Here emphasis is placed in results from Paper II and III, since the main purpose of the application of TOPSIS in Paper I was to check for burden shifting and not to combine economic and environmental results. Though it is important to note that checking for burden shifting is an important step that should be followed in LCA. As the results throughout this thesis, but also in the work of other authors, has shown that burden shifting is common, leading to optimization of one impact category e.g. GWP, while other ICs are sub-optimized (Corona et al., 2018; Laurent et al., 2012; Ögmundarson et al., 2018).

The application of TOPSIS is helpful in deriving single score indicators that are easy to understand and communicate. In Paper II, the work done with this method was used to provide decision support for the NoAW project. The results, shown in Figure 4-1, were discussed in several iterative meetings with technology developers and project stakeholders.

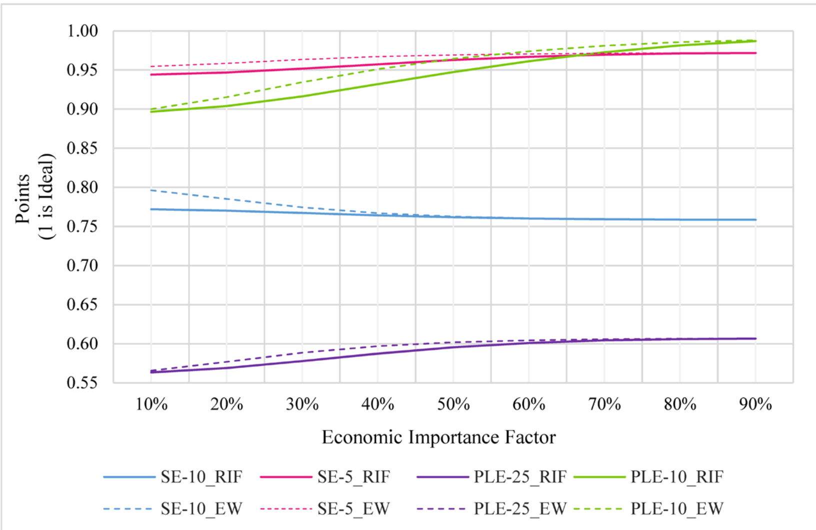


Figure 4-1 TOPSIS derived single score indicator of idealness (most ideal=1) for both Relative Importance Factor (RIF) derived environmental weighting and Equal Weights

(EW) environmental weighting amongst a range of weights given to economic performance. SE is solvent extraction, while PLE is pressurized liquid extraction. The number in each scenario indicates the solvent to DW ratio for the extraction process. Source: (G. C. Croxatto Vega et al., n.d.)

From figure 4-1 it is clear that the best environmental performer is the solvent extraction option with a solvent to DW ratio of 5 (SE-5), and that a closely competitive option is the pressurized liquid extraction option with a solvent to DW ratio of 10 (PLE-10). It is also interesting to see that there is a shift in preference with increasing economic weight, where these two options change i.e. PLE-10 is the best performing above 55% economic weight (with equal weights) and above 65% economic weight (with RIF). Furthermore, it is clear that the other options assessed perform far worse than SE-5 and PLE-10. However, as mentioned in Paper II, caution must be taken to understand what the weighting means. As highlighted by other authors, equal weighting is a rather arbitrary choice, however, no more arbitrary than basing decision making on CFP (Pizzol et al., 2017). On the other hand, " there is a level of uncertainty in the normalization factors used to derive the RIF, and the decision to use current emissions as a reference point, i.e. by using a European's environmental impact as normalization factor, does not necessarily have a relationship to the severity or consequences of environmental impacts. However, it does provide an indication of the relative importance of an emission, or reduction thereof, to the status quo. If absolute sustainability related factors were available for all relevant impact categories, the application of these instead of normalization factors would be preferable, as they would provide a stronger link to environmental impact. Ideally, this process would be completed relative to planetary boundaries (Steffen et al., 2015) using an absolute relationship to impacts from LCA (Bjørn et al., 2015). However, this cannot be done because this absolute relationship is not yet well enough understood/developed, nor has it been developed to include all impact categories covered in LCA" (G. C. Croxatto Vega et al., 2020). The same consideration is valid for the integrated interpretation method applied in Paper III.

The comparison of various weighting profiles derived in Paper III, is a useful organization of the results, which allows one to see that the bio-technology preference is the same for all regions and plant scales assessed, whether the results are expressed through CWF TOPSIS, monetized endpoint damages, or EW TOPSIS (Figure 4-2). Some patterns are clearly visible, for example, there is an economic preference for the options producing more energy i.e. AD+Booster, followed by AD, which approach the ideal (1) with higher economic weight. Nevertheless, it is possible to observe how the type of feedstock used for production influences the results. With a feedstock high in animal manures (80% for the Industrial (I) case), the technologies converge with increasing economic weight, whereas the opposite is true when the feedstock includes more crop residues (VEN scenarios), i.e. residues that have a cost. Furthermore, the single indicators are an important aspect in Paper III. As mentioned in section 2.2, a sensitivity analysis with a green energy future showed that the results flip for the Bavarian region for GWP. Yet, the overall performance of the region remains the same as with today’s energy mix (Figure 4-2), due to impacts in ICs other than GWP, which weigh more than GWP impacts under the CWF applied to TOPSIS.

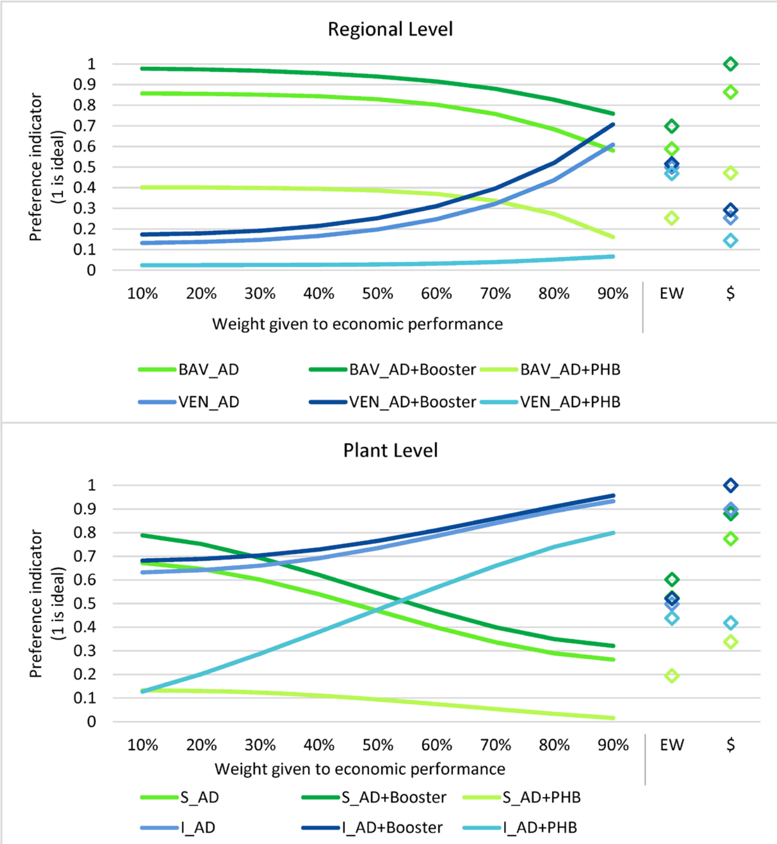


Figure 4-2 TOPSIS results for the regions (top) and scales (bottom), with varying economic importance (10% to 90%), equal weights (EW), and internally normalized monetization ($) of endpoint damages. Scenarios are named by the first three letters of the region (VEN or BAV) or scale size S for small and I for Industrial, followed by each technology scenario (AD, AD+Booster, AD+PHB). Source: (G. Croxatto Vega et al., 2020).

Thus, there is little change other than GWP with the green energy future, a change which is not reflected in the single score indicators. The combination of different derivations of single score ease the interpretation of conflicting results for large systems. As long as these are used in combination with midpoint impact results and the weighting is presented clearly and transparently they can be used to make decisions with a clear decision context in mind. Additionally, the inclusion of several single score indicators that agree on the technology preference of the assessment speaks strongly to the robustness of the results. Though TOPSIS eases the interpretation phase, it has to be recognized that normalization adds an extra level of subjectivity. As mentioned before, relating the emissions of the systems assessed in this chapter to the average emissions of a European person year does not necessarily give a good frame of reference to the environmental profiles of the systems, considering the environmental footprint of a European in comparison to other regions (Wiedmann and Lenzen, 2018). Thus, further research is needed in this regard. Improvements could encompass normalization factor derived from absolute sustainability carrying capacities or possibly focus on refining the LCIA methods by going beyond impacts to consequences arising from a highly globalized world, such as in the newly developed LC-impact method (Verones et al., 2017).

5. ASSESSING EMERGING BIO-PRODUCTS

This chapter discusses the special considerations that must be taken when assessing emerging products, whether these are in the development phase or because the information we have on them is in some ways incomplete or conflicting, as exemplified in section 1.1.3. The chapter aims to answer RQ4 and covers results from Paper IV, VII, VIII, VI, with more focus given to Paper IV.

RQ4: *How are key assessment parameters and LCA methodological challenges identified and overcome when assessing emerging biorefinery products against conventional products?*

5.1. METHOD IN BRIEF

This methodology section is split up into 3 sections. The first dealing with the efforts that have been made during this project to increase our understanding and capabilities to quantify impacts from the life cycle of plastics. The second deals with various challenges for the LCA methodology in regards to degradable materials and lastly there is a method section describing challenges regarding the quantification of emissions from digestate application. The various biorefinery products are discussed in regards to their competing conventional products.

5.1.1. FOR ADDITIONAL PLASTIC IMPACTS

The aim of Paper IV is to extend and apply the LCA methodology to long-term effects of plastic products. Focus was placed in the LCI part of the assessment, by exploring how plastic losses can increase the PM_{10} and $PM_{2.5}$ elementary flows. The method followed consisted of:

- A mini-review was carried out to establish variables and relationships needed to clarify the impacts of conventional and biodegradable plastics in the environment and to identify the items necessary to build a complete inventory representing the life cycle of various plastic products (Supplementary Information to Paper IV).

- Based on the concepts discovered in the mini-review, a conceptual framework was established for inventory building which can be related to specific regional contexts.

- The conceptual framework was tested on a case study of LDPE mulch film for agricultural uses. An accounting cradle-to-grave LCA was performed and focus was placed on the ICs affected by microplastic (MP) development.

The conceptual framework is made up of 3 main pillars 1) a dynamic plastic degradation module, 2) an emission and redistribution into various compartments module and finally 3) and an impacts module. The degradation model predicts polymer surface degradation due to UV exposure, as well as subsequent degradation of the particles formed (the 1st set of particles). For littered plastics the removal of particles from the surface of the polymer allows other layers of the polymer to be exposed to UV degradation. The model compiles subsequent sets of particles generated each year and summarizes the final amount (in kg) of particles that reach the $<PM_{10}$ range in the time horizon of the LCA, throughout all life cycle stages of the product. These are further subdivided into the integrated ranges of PM_{10} and $PM_{2.5}$. Furthermore, it quantifies the degradation of the plastic into decomposition gases, based on values from the literature. A complete inventory of MP should include losses during manufacturing, the use phase and end of life. The latter varies according to location and is dependent on local waste treatment infrastructure. EoL is grouped into 4 different categories: recycling, landfill, incineration and littering. Average EoL data for the location determines, to some degree, the potential for secondary MP formation i.e. the potential is higher for plastic in landfills since they are

more exposed to the elements, while it is much lower for incinerated plastic.

The emissions module links land cover data for the specific location and local wind data to the final emission of plastic particles, grouping them in an air and ground compartment. Local 10 m wind data is converted to 2 m wind data and transformed via the wind shear formula to account for surface roughness (Holton, 2004). A two dimensional micro-physical model was developed (Hansen et al., 2020). It considers both wind speed and MP shape to determine if the MPs will lift off the ground. A fate factor can be adjusted to account for MP capture in soils and sediments. A conceptual figure of the conceptual framework is available in the Papers annex, under Paper IV, figure 1.

Once the dynamic inventory is in place the impacts of plastic litter and microplastics follow a traditional LCIA characterization, where substances are subdivided by their contribution into the relevant impact categories and are characterized using existing characterization models. On the other hand, ecosystem damages are estimated by applying the characterization factor for species loss in Ryberg et al., (2019) to the developed inventory. Regionalized characterization values were used for impact damages at endpoints (Verones et al., 2016). Lastly, impact damages were monetized using the method described in section 4.1.

5.1.2. Biocomposites, Polyphenols and Biodegradable Polymers

In Paper VII, a standard LCA was performed for filler material, which can be mixed with various polymers to produce biocomposites. The filler material is made from vine shoots, which is a currently an undervalorized residue from wine production in the Languedoc-Roussillon region of France. The residues are typically burned in this region, but changing legislation requires farmers to find other ways to “dispose” of vine shoots. The LCA compared first rigid virgin polymer trays made out of PHBV, polypropylene (PP), and PLA and then the same polymer matrices with the addition of vine shoots filler material. The assessment was performed from cradle to grave with a cut-off system.

Paper VIII tests the premise of the No Agricultural Waste Concept, by arranging the various biorefinery setups assessed through this project into combinations that aim to extract the highest possible environmental benefit out the agricultural residues present in a region. The assessments, composed of two phases, 1) mini-LCAs of the treatment of 1 ton of feedstock in each region per biorefinery setup, and 2) a scale up phase utilizing the sustainable/technical available feedstock of the region, for 5 different regions. The products arising from the various combinations were biogas, digestate, PHB, filler, and polyphenols. The assessment was done from cradle to grave, taking into consideration average EoL options in each of the regions in question. Product substitution was handled via system expansion, so for example, in the case of production of PHB, while composting, incineration and landfilling are the induced EoL options for the PHB, the substituted product i.e. global thermoplastic production, is avoided and so is the EoL of the global thermoplastic, a.k.a recycling, landfilling, and incineration.

5.1.3. DIGESTATE

In Paper I "the field application of the digestate was modelled, and conventional ammonium nitrate fertilizer was assumed to be replaced. It is well known that digestates mineralize at a slower rate [than mineral fertilizers] so a share of the organic nitrogen present in digestate will be bound and will thereby not be available for crop uptake or emissions [immediately after digestate application]. Thus, an average mineral fertilizer equivalency value of 67.5%, calculated from a review of values that are commonly used in this type of assessment, was used for the substitution of mineral N fertilizer (Brockmann et al., 2018). Emissions resulting from the field application of digestate were modeled based on the approach in Bruun et al., (2016), which applied the agronomic model Daisy (S. Hansen et al., 2012) to estimate long-term emissions from different types of soils with different histories of management, i.e., high or low inputs of organic matter in the form of organic fertilizers, such as digestate and compost. As shown in this work, the crop's response to nutrient inputs is highly dependent on the previous fertilization history of the field. Emission factors (EFs) for [losses of N through the drains and leaching to ground water] for high and low crop response after digestate application were taken from Yoshida et al., (2016), which follows the same approach described by Bruun et al., (2016) and had soils and overall conditions which more or less match the soils in the geographical areas assessed here. However, for N_2O emissions, the Intergovernmental Panel on Climate Change (IPCC) methodology (Eggleston et al., 2006) and EFs were used" (Croxatto Vega et al., 2019). The robustness of the conclusions was tested by also applying the N_2O EFs found in Yoshida et al. (2016), which are higher than the ones derived from the IPCC. The emission factor for N_2O is very important because of its high GWP. Furthermore, it has been shown in the past that small changes in N_2O emissions factor can have a large influence on GWP results (Croxatto Vega et al., 2014).

Additionally, a multi-product green biorefinery was analyzed in Paper VI, with various options for press-pulp utilization. The products were protein of different qualities, animal feed, lysine, energy in the form of electricity and heat, insulation material and digestate. The products were handled via system expansion in the LCA. The implications of the utilization of the products by substituting conventional products is discussed briefly in the following section.

5.2. Key Results and Discussion

When emerging bioplastics such as PLA and PHA are compared to conventional polymers they often perform worse in terms of processing (Dietrich et al., 2017; Madeleine R Yates and Barlow, 2013). However, a growing body of literature is bringing to light several different health and ecosystem concerns associated with conventional plastics, which have not been included in the LCA methodology (at the time of writing).

The list of problems includes for example entanglement of marine fauna (Woods et al., 2019), ingestion increasing mortality or ability of birds to feed (Browne et al., 2015), habitat destruction, while for humans it has been shown that we are breathing microplastics (Vianello et al., 2019).

Efforts are needed by the LCA community to include the emerging impacts from plastic products in a standardized manner, to avoid conflicting information from assessments including these new impacts. Thus, Paper IV proposed a framework to standardize the inventory collection of plastic losses with focus on the formation of secondary microplastics. Some key findings from applying this framework to a case study of mulch film, typically used in agriculture to avoid weeds, increase soil temperature (Steinmetz et al., 2016), among other are discussed.

The inventory compiled from the deterioration of LDPE included particulate matter from microplastic formation during the use phase, landfilling, and littering. Additionally, decomposition gases including methane, ethylene, ethane and propene were inventoried and contribute to photochemical ozone formation, as well as climate change. The inclusion of these emissions can be seen in Figure 5-1, where it is evident that the contribution to these three impact categories is small, at least with the baseline shown here which corresponds to Danish conditions of degradation and a 10% littering rate, after the use phase. It is important to note the difference between the two impact assessment methods used, namely ReCiPE Hierarchist midpoint and ILCD midpoint+. The difference arises from the characterization factors assigned to PM_{10} and $PM_{2.5}$, most notably ReCipe’s CF for PM_{10} is 0, so the impact of PM formation is underestimated by the latter. On the other hand, ILCD’s GWP CF for methane is outdated, having the old CF of 26 kg CO_2 eq/kg CH_4.

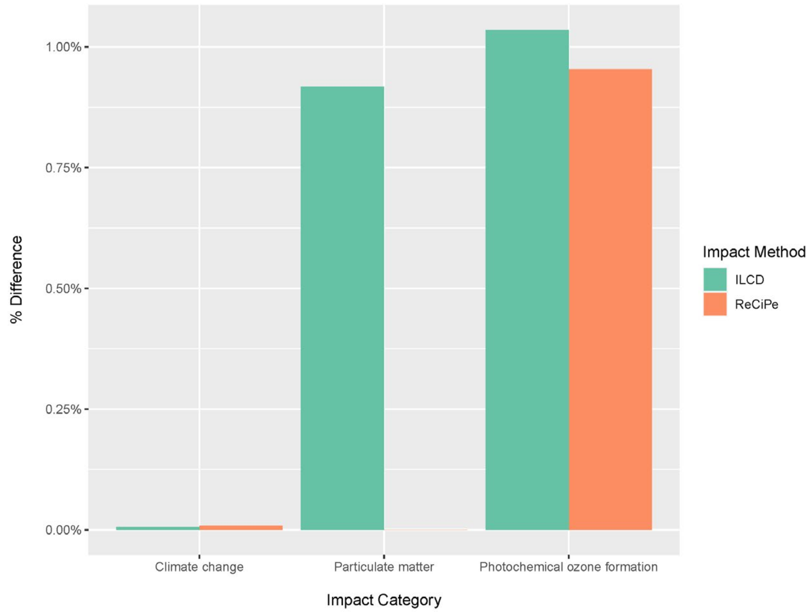


Figure 5-1. Increase of potential particulate matter, climate change, and photochemical ozone formation per kg of LDPE from deterioration throughout its life cycle, including production, use, and EoL. Comparison between the ILCD Midpoint+ and ReCiPe midpoint (H) impact assessment methodologies. Source: (Croxatto Vega et al., n.d.).

The degradation rates obtained from the degradation module in Paper IV were compared to recently published results by (Chamas et al., 2020) and were found to be 2 orders of magnitude lower. This is logical, since Paper IV only includes the effect from UV radiation and not the subsequent photo-oxidation and other polymer decomposition accelerating factors reported in literature. Thus, to test the model, the values for degradation in Chamas et al (2020) were applied as sensitivity. The degradation parameter was found to be sensitive, since a faster degradation of the LDPE film signifies an increase of 40% in particulate matter potential compared to a situation were no MPs are included in the system. Thus, future research would benefit from clarifying degradation rates to which littered plastics, as well as plastics decomposing in landfill and during use, are subjected. Furthermore, a large increase in both PM fractions is observed when accounting for the subsequent degradation of the particles generated in year 1 at the end of the 100 year time horizon used for the case study. The mass of PM_{10} increases by 592% and $PM_{2.5}$ increases by 376% by the end of the 100 years (Italian scenario). However, the mass of these particles is very small and the PM impact categories of both LCIA methods used are rather unsusceptible to the increases. At the same time, part of the particle size distribution is constantly eliminated due to particle masses approaching zero. This is rather similar to experimental results from accelerated weathering of plastics, were the authors were not able to account for ca. 75% of the initial mass lost, which they suggest is because particles reach the submicron level, becoming undetectable (Song et al., 2017).

Regional differences were less important in regards to the final impact of the LDPE's life cycle. The case study was tested for Italy (IT) and Denmark (DK), to which results mainly differed depending on the types of EoL treatment present in each country. For example, a higher contribution from incineration is seen for DK, while a higher contribution from landfill is seen for IT. Monetized particulate matter damages are shown in Figure 5-2, calculated with the same method as in Paper III (refer to section 4.1). The values were obtained by applying the regionalized endpoint CF for particulate matter impacts developed in (Verones et al., 2016) and applying (Ryberg et al., 2018) for characterization of species loss, though only damages to human health are shown in the figure since these are the more visible damages.

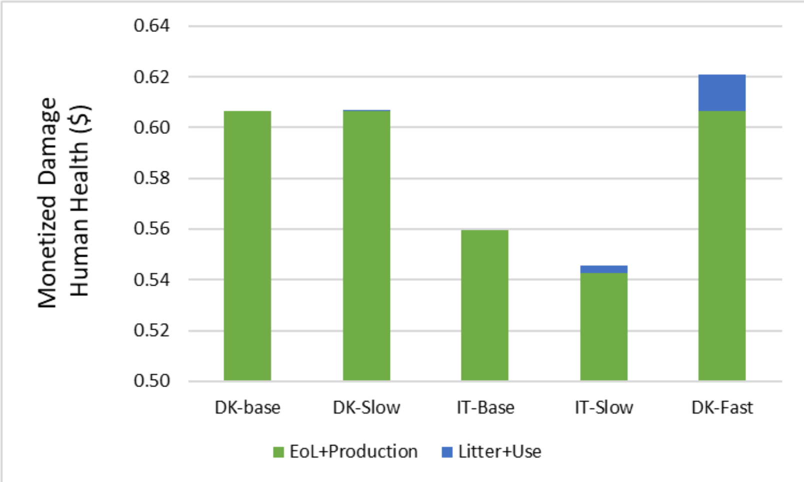


Figure 5-2 Monetized damage to human health per kg of mulch film. Derived from DALY valuated as 110,000 USD$_{2003}$ per DALY. Source: (Croxatto Vega et al., n.d.). *Scenarios with –base at the end do not include additional impacts from plastic litter and MPs. Slow refers to a slow degradation of plastic in the environment (only UV), and fast corresponds to a faster degradation rate, including various degradation factors as per the work of* (Chamas et al., 2020).

"When expressed in monetary terms (USD), the per kg contribution of monetized damage is low, only a few cents per kilo of LDPE, though more noticeable for IT and also for a faster degradation rate. Though these amounts may seems small when observed on a per kilo basis, it is informative to think about them in terms of, for example the EU's consumption of LDPE. Scaled up to European level, the additional impacts of plastic considered in Paper IV cost the EU between 3.5 million USD and 133 million USD per year in human health damages and around 800 USD in damages to species. If similar damages are considered for the whole of the EUs consumption of plastic (though this is better assessed on a per type of polymer basis), then the damage to human health could potentially cost from 20 million to 755 million USD, and ca. 4250 USD in damage to species. These values though highly uncertain are worth considering carefully"(Croxatto Vega et al., n.d.). Furthermore, the sensitivity results for photochemical ozone formation suggest that plastics degrading in the environment may be an important diffuse source of NMVOCs and thereby of photochemical ozone formation, especially if the degradation rate is fast. Further work is needed in this regard, to determine gas production rates for various polymer types in various degradation conditions.

Paper IV is only a beginning step to include the impacts of plastic losses. Additional work is needed to include the possibility of MP as vector for disease and also for toxic substances (Plastic Soup Foundation, 2019; Prata, 2018). The degradation module is an important component of the model, which shows the potential evolution of particles into sizes that are dangerous to human health, posing health risks of particulate matter (Raaschou-Nielsen et al., 2013) (see figure 3 of Paper IV), with mounting evidence that microplastics may contribute to reduce function in the lungs (Plastic Soup Foundation, 2019). Degradability is a methodological Achilles heel, not only when considering MP formation, but also when assessing other biorefinery products.

Various biodegradable products have been assessed throughout this project. As in other publications (Madeleine R Yates and Barlow, 2013), in Paper VII the life cycle of the biodegradable polymers PLA and PHB has been found to be more burdensome than the life cycle of PP, which is a highly optimized fossil-based polymer. Adding filler material lowers the GWP of the biodegradable polymer matrices and also of the PP matrix (Fig 5 in Paper VII), however PP continues to perform better. With the tools we have today, it is however, not possible to capture the advantages of truly biodegradable products. A biocomposite made from PHB and filler material from vine shoots, that leaves no trace on earth once it has undergone complete decomposition sounds at the very least desirable (for many applications). Are there possibly ecosystem services that we are not accounting for? Is it the reduction of the Oceanic garbage patch and a lower risk of entanglement for various species, or is it a mere aesthetic value? There are undoubtedly different directions to go and much needed work for accounting for value in these materials.

Another area of incongruence for the LCA methodology is evident when exploring the EoL options of the global plastic mix substituted in Paper VIII, and also during the assessment with PlastLCI (Paper IV). Lack of representativeness in the Ecoinvent database for specific materials in the various end of life processes is an issue needing urgent attention. For example, a 1% degradability is assigned to conventional polymers, such as Polyethylene terephthalate (PET), PE, PP and polystyrene (PS) in landfill, however from the literature reviewed during this project it is evident that degradation of conventional polymers does happen to a larger extent and is faster than initially thought under various conditions (Castro-Aguirre et al., 2017; Chamas et al., 2020; Restrepo-Flórez et al., 2014; Royer et al., 2018; Shah et al., 2008). Even for biodegradable polymers, the level of degradation experienced will depend on degradation conditions, e.g. type of compost setup, moisture content to name a few, and subsequently, the decomposition products (gas emissions) will also vary according to conditions during degradation (Arcos-Hernandez et al., 2012; Emadian et al., 2017; Itävaara et al., 2002; Wang et al., 2018).

Much like degradation of materials, biological activity in agricultural fields after the application of digestate is a source of high uncertainty for LCAs. Particularly, an erroneous assessment of N_2O emissions from field application of mineral fertilizers, animal slurries or digestate can alter the results of an LCA. Figure 5-3 shows the influence a change of EFs for N_2O had on overall results in Paper I. For the LCA practitioner a common approach is to adopt the IPCC guidelines for national GHG inventories, Tier 1 approach (Eggleston et al., 2006), which is what was used for the baseline in Paper I. However, the IPCC encourages the use of more representative approaches, as for example a Tier 2 approach, which would base the calculation on country-specific EFs or a Tier 3 approach based on rigorous models that include climatic conditions, and field management, among other, or based on field measurements (Brocks et al., 2014).

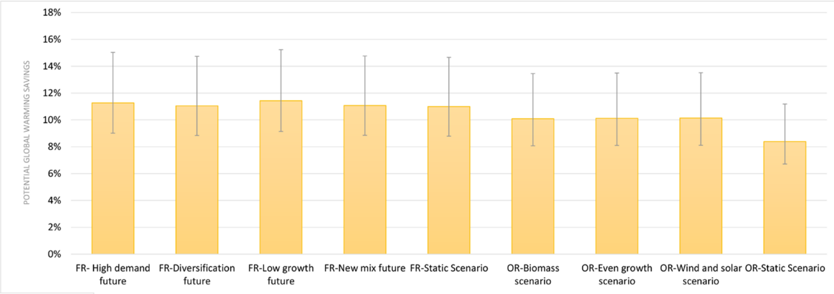


Figure 5-3 Sensitivity of field emissions to a change in N_2O emission factor

To demonstrate the importance of N_2O, it is worth noting that with the Tier 1 approach, all scenarios assessed in Paper I induce savings, while with a Tier 3 approach, as shown in Figure 5-3, all scenarios induce burdens to GWP, though ranking between scenarios does not change. The potential for error is high if N_2O is misrepresented, and thus it is recommended to adopt an advanced Tier approach, as for example by the use of regionalized disaggregated N_2O emission factors representative of the region (Brocks et al., 2014), or a modelling approach as with for example the Daisy model (S. Hansen et al., 2012) were the current status of the soil as well as climatic conditions, and field management are taken into account (Bruun et al., 2016).

Finally, of the products assessed through this project, the polyphenols case assessed in Paper II and VIII would benefit of an increase understanding of their relationship to reducing food spoilage, which has not been taken into account yet in the present work. Polyphenols can be used in bioactive packaging formulations, where their antioxidant properties will delay food spoilage (Piccolella et al., 2019). In the LCAs where this facet of polyphenols has been taken into account, the savings in the system increased dramatically (Dilkes-Hoffman et al., 2018; Lorite et al., 2017). However, the functional unit of the assessment will then need to be re-formulated to include the consequences on food production, as for example "providing customers with 100 000 kg fresh fruit in one year" (Lorite et al., 2017). This presents a challenge for assessing the full potential of polyphenols in the biorefinery setups as assessed in Paper VIII, increasing the complexity of the system and potentially introducing an unbalanced view. However, careful scoping of the functional unit could be done to include the benefits from avoiding food waste.

6. CONCLUSIONS

6.1. ACHIEVEMENTS OF THE PHD PROJECT

The main goal of this project was to develop and expand the LCA methodology in order to improve assessments and determine good LCA practice when related to emerging biorefineries and their products in a regional or territorial context. In order to define appropriate methodological steps, four research questions were presented.

RQ1: *Which factors are potentially result-altering when deciding on the best technology to region pairing now and in the future? (the background system)*

In Paper I, focus was placed in adding dynamics to the foreground, and background systems. In Paper III, sustainable and technical feedstock potentials were quantified for the inventory, giving a more complete overview of the status of exploitation in the assessed region. While in Paper VIII, region/feedstock/technology combinations were explored. The results gathered show that dynamism, especially in the background system, is of high importance for systems producing bioenergy and processing setups that are energy intensive. Given today's focus on moving towards greener energy mixes it is recommended that regional predictions or political targets are used at least as a sensitivity analysis to check the influence of these on the general environmental profile of the biotechnology, as it is a result-altering factor. Using regionalized data gives additional context to the LCA and increases our understanding of the level of some of the technologies' proliferation and exploitation of available feedstocks. It also provides a good overview of regional potentials for improvements and/or impacts related to the various mass flows interacting within the region. However, in paper III an important limitation of the method is observed in the need to more accurately quantify the pressures of land resources, whether by intensification or transformation. This limitation could be overcome by adding a more thorough assessment of land use change and indirect land use change, which should be assessed at a global scale. Finally, a step-by-step method to determine biorefinery setups suitable for the biotechnology using a specific feedstock in the explicit regional context was tested. Paper VIII puts into evidence the need for region-to-feedstock-to-biotechnology assessments and shows how various biotechnologies fail to bring environmental improvement in some regions, while being successful in others. This approach shows that when it comes to different residue treatment methods, even when these are 2nd generation biomass, their sustainability is not a given and decision to implement a technology have to be made on an individual case basis.

RQ2: *How can LCA be used for early design optimization of emerging biotechnologies? (the foreground system)*

Chapter 3 showed that it is possible to pinpoint hot-spots in new production methods by doing a simple assessment of laboratory scale methods, by using process design software to scale up the process. The simple assessment can be used to guide and inform process designers on potential improvements they may be able to attain with a few changes. However, this type of simple assessment should always be followed by a more rounded and complete assessment, such as a complete LCA and TEA supported by process design software. The latter allows LCA practitioners to build more reliable inventories in terms of energy consumption and equipment limitations. The opportunity to standardize the use of TEA-LCA in the chemical/biotech industries should be explored in future research, since the industry already uses TEA coupled with process design software. As requirements for sustainability increase for industry combined TEA-LCA will be a useful tool, especially if it attains/is subjected to standardization.

RQ3: *How can bilateral economic and environmental sustainability assessments be combined to increase clarity for decision support? (interpretation)*

Throughout this project effort has been placed into synthesizing the often many and conflicting results that are obtained when all IC results from an LCA are observed. In this regard, a few options are available to LCA practitioners when combining results into single scores, whether results are from several ICs alone, or from ICs and TEA results. One of the options is TOPSIS, which is a type of MCDA that measures distance to an ideal solution. The advantages of this option include a more nuanced assessment of value assigned to either economic or environmental impacts, which gives a clear decision context. On the other hand, weighting is necessary and will always introduce an extra layer of subjectivity, while it also requires extra time investment for the TOPSIS calculations. Another possible method, which is easier to implement is monetization of impact damages. While this method is the easiest, determination of the cost of externalities is not and carries with it a large level of uncertainty (Pizzol et al., 2015). However, in terms of communication, representing impacts as monetary values is highly relatable and can ease the dissemination of results. Moreover, the comparison between monetized values and TOPSIS results showed a high level of agreement, suggesting that both of these methods are correctly summarizing overall LCA impacts.

RQ4: How *are key assessment parameters and LCA methodological challenges identified and overcome when assessing emerging biorefinery products against conventional products? (new products)*

The products assessed throughout this project range from biodegradable polymers (Paper I, III, VIII), to polyphenols (Paper II and VIII), to biocomposites or filler material (Paper VII, VIII), to protein (VI) and digestate (Paper I, VIII). Paper IV focused on expanding the LCA methodology to include impacts that have not previously been included in LCAs to date, such as the impact of particulate matter formation from conventional fossil-based plastic products. The impacts from PM have the potential to be large depending on the degradation rate of the fossil-based polymer in the environment. Several methodological challenges will need to be addressed in the future to correctly account for the impacts of plastic products with LCA. Other challenges related to biodegradable products have been identified through this project. The question of value of biodegradable materials, or multifunctional products such as bioactive packaging pose challenges for the LCA community. Efforts should be made to clarify the value of biodegradable materials, which might extend beyond the general LCIA methodology into the social or ecosystem services disciplines and might require cross-disciplinary development. Furthermore, data paucity and lack of representative data in the databases used by LCA practitioners can only be overcome by a continued development of the field and other disciplines, which contribute valuable knowledge that can be applied to database building.

6.2. Future Perspectives

"The more I learn, the more I realize how much I don't know" – Albert Einstein (or Socrates, no one knows)

Though this project has suggested many methodological recommendations in relation to assessing emerging biorefineries and their products in a regional/territorial context, there are many unanswered questions that require future collaboration on the topics covered in this project. Future research necessary for the improvement of the LCA methodology is not limited to work done by LCA practitioners. Establishing of cross-disciplinary teams is a key aspect that will lead to enriching the methods used in LCA, as we often rely on real world data to make meaningful assessments. Though this is true for all weak methodological elements that have been identified throughout this project it is pointedly true when related to the assessment of the impacts of plastic, which must improve in the future. In particular, effort should be placed in exploring the possibility of microplastics as vectors for toxic substances, as well as the effect (damage) of the abstruse nano-fraction. Determination of a possible need for microplastic specific characterization factors is imperative for the development of this area of study. Future research will benefit from knowledge development in this area and from connecting new knowledge to the standardized LCA methodology.

Standardization is another area where major efforts should be devoted. A continued growth of the databases used by LCA practitioners facilitates better quality in LCA assessments. This is very relevant for products such as digestates, which need both standardization of the methods for emissions calculations and site/geographic differentiation. Overall, regional specificity is an important component in the effort to improve the quality of LCAs and I hope the clarifications of methodological choices attained here will continue to aid the development of quality LCA assessments.

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PAPERS

PAPER I

Maximizing Environmental Impact Savings Potential Through Innovative Biorefinery Alternatives: An Application of the TM-LCA Framework for Regional Scale Impact Assessment

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Maximizing Environmental Impact Savings Potential through Innovative Biorefinery Alternatives: An Application of the TM-LCA Framework for Regional Scale Impact Assessment

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Abstract: In order to compare the maximum potential environmental impact savings that may result from the implementation of innovative biorefinery alternatives at a regional scale, the Territorial Metabolism-Life Cycle Assessment (TM-LCA) framework is implemented. With the goal of examining environmental impacts arising from technology-to-region (territory) compatibility, the framework is applied to two biorefinery alternatives, treating a mixture of cow manure and grape marc. The biorefineries produce either biogas alone or biogas and polyhydroxyalkanoates (PHA), a naturally occurring polymer. The production of PHA substitutes either polyethylene terephthalate (PET) or biosourced polylactide (PLA) production. The assessment is performed for two regions, one in Southern France and the other in Oregon, USA. Changing energy systems are taken into account via multiple dynamic energy provision scenarios. Territorial scale impacts are quantified using both LCA midpoint impact categories and single score indicators derived through multi-criteria decision assessment (MCDA). It is determined that in all probable future scenarios, a biorefinery with PHA-biogas co-production is preferable to a biorefinery only producing biogas. The TM-LCA framework facilitates the capture of technology and regionally specific impacts, such as impacts caused by local energy provision and potential impacts due to limitations in the availability of the defined feedstock leading to additional transport.

Keywords: biorefinery; territorial metabolism; life cycle assessment; biogas; multi-criteria decision assessment; bioplastic; polyhydroxyalkanoates; agricultural residues

1. Introduction

Life cycle assessment (LCA) is a tool designed to quantify the environmental impact potential of products and services [1]. Recent advances in the field of LCA, such as the inclusion of temporal dynamism [2] and the coupling of LCA to urban metabolism [3] increase the applicability of the LCA methodology. Dynamism in LCA allows for the quantification of impacts while taking into consideration changing background and foreground systems, e.g., amounts of renewable and fossil energy sources in the electrical energy mix of a specific location in the background, and improvement to processing technologies in the foreground. On the other hand, coupling urban metabolism to LCA allows for large-scale assessments that better predict large-scale consequences of implementing a change at regional scale. These advances are an especially important input that can help guide the transition into a sustainable bioeconomy, as they allow for prospective studies. LCA of production systems/technologies, such as various agricultural productions, e.g., wine, cereal, and meat, can benefit from applying some of the new developments, since the large inputs and outputs to these systems, most likely, will have great environmental implications when changes to the production are implemented.

By applying the TM-LCA framework, as used in this study, it is possible to assess said systems in the specific context of the region, i.e., taking into consideration the region's infrastructure, feedstock availability and accessibility, and the technical feasibility of technology implementation. Assessing large systems, as mentioned above, can be approached by defining the geographical boundaries in terms of a "producer territory" [4] so that the LCA can be applied for assessment of a delimited "territory", e.g., wine-producing areas, within a broadly defined region, e.g., Southern France. The producer territory is thus defined as the area of interaction between the aggregated producers and other systems within the region. The TM-LCA framework reduces data demand by aggregating individual areas of the production of, for example, a specific product, supply chain or waste treatment technology, while ignoring unchanging background systems, i.e., only changes to the region interacting with the producer territory are assessed. At the same time, representativeness is increased by merging local inventory data from individual producers with regional and nation-wide data in order to fill in data gaps. In this way, an environmental performance improvement in the territory, due to, e.g., the implementation of a new technology or new management technique, can be quantified in the non-contiguous production area and is reflected in the results for the region. When combined with dynamic and prospective LCA [2], this approach offers a comprehensive assessment that gives temporally and geographically resolved results. Moreover, it has the added utility of providing prospective insights that can more accurately support decision makers, production owners, and technology developers [4].

A point of departure for many LCAs is a static product system, where, for example, technology A might be assessed against technology B for the making of a product. The static nature of LCA is problematic when applied to products or systems with long service lives [5], due to inconsistencies in time horizons and changes in background systems [6,7]. Previous work has demonstrated the importance of incorporating various types of dynamism into LCA, as this can significantly affect the results of the study [6]. In this regard, it is possible to add dynamism to the various stages of the LCA in a consistent, systematic, and transparent manner, as outlined in [2] and shown in various other publications [7–9]. Following the TM-LCA framework, dynamism can be added in a consistent manner from the start, which provides added information regarding the sensitivity of the system to background changes. Real production systems are rarely static, and results based on static systems can sometimes exhibit rank reversal when compared to dynamic results [10]. Thus, basing future decisions on static LCAs can result in building significant error into the models and associated results. Adding dynamic aspects to LCAs can increase the analytical accuracy of results [11].

The added layers of information to the TM-LCA mean that the interpretation phase becomes more resource demanding. This can be eased by the use of extra tools, such as multi-criteria decision assessment (MCDA). Midpoint results for 18 different impact categories of an LCA are often difficult and time consuming to synthesize into clear and readily applicable decision support. When adding dynamism, this translates into temporally specific results for, e.g., each year of the time horizon, for each of the 18 impact categories. Out of the many MCDA methods that exist, one that has shown great capability in dealing with LCA results is Technique for Order of Preference by Similarity to Ideal Solution (TOPSIS) [12,13]. The output from TOPSIS is given in the form of a single score performance index, which is used to derive preference between the scenarios being assessed. By checking a multiple criteria decision support tool used with equal weightings for all midpoint impact categories, it is easy to realize and visualize burden shifting amongst the midpoint impact categories, when used in conjunction with a visual inspection of internally normalized results. The MCDA approach is considered preferable, as using carbon footprint alone has been shown to give potentially misleading results [14].

The present study's goal is to implement an assessment based on the TM-LCA approach [4] in order to provide a comparison of potential biorefinery choices for the treatment of agricultural residues. For the demonstration of TM-LCA, a biogas production scenario is compared to a scenario of combined biogas and Polyhydroxyalkanoates (PHA) production, which is currently being developed at pilot scale. Polyhydroxyalkanoates are naturally occurring polymers produced by a consortium of bacteria, which can feed on the volatile fatty acid (VFA) stream generated by the acidogenic phase of anaerobic digestion (AD) [15]. PHA, which is also found as polyhydroxybutyrate (PHB), can be used to produce biodegradable plastic products. In this case, PHB production substitutes the production of polyethylene terephthalate (PET) or polylactide (PLA). The two biorefinery scenarios are modeled with dynamics built into both foreground and background systems. In the foreground system, dynamics are included as a yearly decrease in the amount of energy consumption needed to produce PHA. In the background system, the electrical energy mix, hereafter referred to as energy mix or energy grid, of both locations is varied yearly for a period of 20 years with four possible provision mixes for Oregon, and five possible choices of provision for the energy mix futures of France. The scenarios are then tested at a territorial scale as described above, i.e., processing all the feedstock in the region in the two geographically dissimilar production territories, to observe the effects of regional differences on territorial performance. Since the use of global warming potential (GWP) as a single indicator has been shown to provide potentially misleading results [14], MCDA is applied in the interpretation phase to help ease the interpretation of results.

2. Materials and Methods

2.1. TM-LCA Framework Application

The application of the TM-LCA framework is described in general terms here. A point of departure for the application of the TM-LCA framework is the functional unit. The functional unit, the treatment of one ton of feedstock of specific composition, is treated by two different technology alternatives, described in more detail below. From here, the following steps are applied and described through the methodology:

(a) Alternative technology is defined.
(b) The producer territory is defined and limited to systems interacting with the technological options being assessed within a geographical region.
(c) Temporal dynamics are incorporated into the systems, e.g., in dynamic background electricity energy provision and technological efficiency improvement.
(d) The assessment is scaled to encompass the whole region so that all feedstock available that may fulfill the functional unit is treated by the technological alternatives being assessed. However, only changes in systems and in the region are assessed.

2.2. Goal and Scope

In order to implement the TM-LCA framework, two options for the treatment of agricultural residues were modelled and compared in two geographic locations, the Languedoc-Roussillon region in southeast France and the Willamette, Umpqua, Rogue, and Columbia valleys of Oregon State in the USA. Advancements in biogas technology make it possible to treat a plethora of agricultural residues, and recent innovation allows for the production of value-added products, in this case, the family of biopolymers known as polyhydroxyalkanoates (PHA). This innovative technology, which effectively creates a biogas platform for new biorefineries, is a contender to conventional biogas production where the only products are biogas and digestate. The proliferation of biogas plants makes this new addition to anaerobic digestion a highly transferable technology, which can be implemented wherever agricultural residues are available. Since biorefineries, in general, have a long service life (decades) and draw from large discontiguous areas, both territorial and dynamic aspects of this assessment are an advantage for decision makers considering biorefinery options for their region. However, it should be emphasized that the study only compares two different biorefinery types. It cannot be used to decide whether to increase the total use of residues for biorefineries.

Functional Unit

The basis for the comparison of the scenarios is the treatment of 1000 kg of feedstock. The feedstock is assumed to be agricultural residues of the following composition: 50% liquid cow manure, 15% solid cow manure, and 35% wine pomace or wine marc, hereafter used interchangeably. Feedstock characterization is based on laboratory tests performed onsite at an Italian biogas plant for the liquid and solid manure, while for wine pomace it is based on literature values. While other types of feedstock can be treated by the biorefineries being considered, the choice of feedstock was limited to the above in order to better appreciate the difference between biorefineries rather than differences arising from choice of feedstock. The feedstock physiochemical properties are presented in the supplementary information (SI).

2.3. Scenarios

Two baseline scenarios were assessed with the OpenLCA [16] software and the Ecoinvent 3.4 database [17]. The two alternative technological pathways possible for treating the functional unit are:

2.3.1. Biogas Only

Conventional biogas production was modelled as the anaerobic digestion step of biogas production, which produces biogas and digestate. The biogas was assumed to be burned in a combined heat and power (CHP) engine, producing electricity and heat based on the energy content of the biogas. Process energy consumption was calculated to be 7% of the electricity output, based on data received from an industrial scale biogas plant in Northern Italy, while the co-generated heat is assumed to be wasted. This is due to the geographical areas of implementation of the scenarios, where the excess heat is not used. Furthermore, adding the produced heat to this study would only change the magnitude of the savings from displaced energy production, and not the ranking of the scenarios, as seen in [18], as the magnitude of heat production is similar across scenarios. All other operational parameters were also based on the data acquired from the abovementioned biogas plant and are available in the supplementary information (SI).

Processing steps that are equal for both scenarios and emissions occurring therein, e.g., feedstock storage, animal housing and digestate storage, were excluded from the assessment, as they would result in no relative difference. Similarly, phosphorus fertilizer replacement was left out because the starting content of P is the same, and processing is not expected to change this. Adding replacement of P fertilizer to the assessment would only elucidate differences between digestate and mineral fertilizers, which is not the focus of this study.

2.3.2. Field Application of Digestate for All Scenarios

The field application of the digestate was modelled, and conventional ammonium nitrate fertilizer was assumed to be replaced. It is well known that digestates mineralize at a slower rate so that a share of the organic nitrogen present in digestate will be bound and will thereby not be available for crop uptake or emissions. Thus, an average mineral fertilizer equivalency value of 67.5%, calculated from a review of values that are commonly used in this type of assessment, was used for the substitution of mineral N fertilizer [19]. Emissions resulting from the field application of digestate were modeled based on the approach in [20], which applied the agronomic model Daisy [21] to estimate long-term emissions from different types of soils with different histories of management, i.e., high or low inputs of organic matter in the form of organic fertilizers, such as digestate and compost. As shown in this work, the crop's response to nutrient inputs is highly dependent on the previous fertilization history of the field. Emission factors (EFs) for high and low crop response after digestate application were taken from [22], which follows the same approach described by [20] and had soils and overall conditions which more or less match the soils in the geographical areas assessed here. For N_2O emissions, the Intergovernmental Panel on Climate Change (IPCC) methodology [23] and EFs were used. The sensitivity of N_2O EFs was tested in the sensitivity analysis due to the multiple models available for deriving EFs. The nutrient content of the digestates, as well as emission factors for all N-related emissions, for digestates and mineral N fertilizer are presented in Supplementary Tables S1, S2 and S4.

2.3.3. PHA-Biogas

The second scenario represents a tweaking to the AD process, where AD is split so that the VFA production that occurs during the first days of digestion is diverted and used to produce and feed biomass capable of producing PHA. Operational data from a PHA-producing pilot plant run by Innoven Srl were obtained and used to create an industrial scale model of PHA production. The co-production of biogas and PHA is executed, albeit with a lower biogas yield. Just as above, digestate continues to be produced and replaces mineral N fertilizer. Additionally, the extraction of polyhydroxybutyrate (PHB), a polymer in the family of polyhydroxyalkanoates, i.e., PHAs, is included as the addition of process energy consumption for the extraction, and hydrogen peroxide is included as an extraction agent. All other model parameters are equal to the biogas scenario.

PHA production is here assumed to be 100% PHB and replaces the production of petroleum or bio-based polymers, referred to as the replacement polymers (RP). In the first run of the model, PHB replaces PET at the factory gate, with a replacement ratio of 0.93:1 PHB to PET. In terms of material properties, several performance indices (PI) based on yield strength (σ), tensile strength, and density (ρ) were used to derive the replacement ratios (RR) (Equation (1)). The ratio of replacement is tested in the sensitivity analysis so as to represent different applications of the polymer more accurately. The choice of polymer substitution is also tested; since PHA is a bio-sourced biopolymer, a sub-scenario with replacement of biobased polylactide (PLA) is also presented. The RR is 0.64 for PHB substitution of PLA, based on Equation (1).

$$RR = \frac{PI_{PHB}}{PI_{RP}}, \text{ and } PI = \frac{\sigma}{\rho} \tag{1}$$

Equation (1) Polymer replacement ratio, where RR = replacement ratio, PI = performance index, σ = yield strength, RP = replacement polymer and ρ = density.

The addition of PHA production in this scenario is not burden-free, inducing impacts from energy consumption and via the production of the extraction agent. However, due to missing data from the pilot plant, the additional energy consumption was calculated using the process design software Superpro Designer® [24]. This yields an additional 7 kwh/functional unit (FU). It was assumed that process energy consumption for PHA could improve over time, so a 1% decrease in energy demand per year for PHA production was modeled for the assessed period. This represents the maturation of PHA extraction technology, which is a likely scenario as the implementation of PHA extraction in biorefineries becomes more widespread and further optimization of the technology takes place. This efficiency improvement rate is tested in the sensitivity analysis to explore the possibility of faster and slower improvements to the process. Key parameters for the production of PHB are presented in Supplementary Table S3.

2.3.4. System Boundaries

The system boundary of the two scenarios extends from when the feedstock enters AD to the application of digestate onto the field (see Figure 1). End of life was not included in the assessment, as the LCA methodology lacks an appropriate characterization of the effects from plastic degradation in the environment, such as microplastic formation and the production of methane among other decomposition gases [25,26].

Applying a dynamic approach, all background and foreground processes were modified so that the two geographical areas are accurately represented with likely different future energy production scenarios in accordance with the national and state-specific energy legislations and policies.

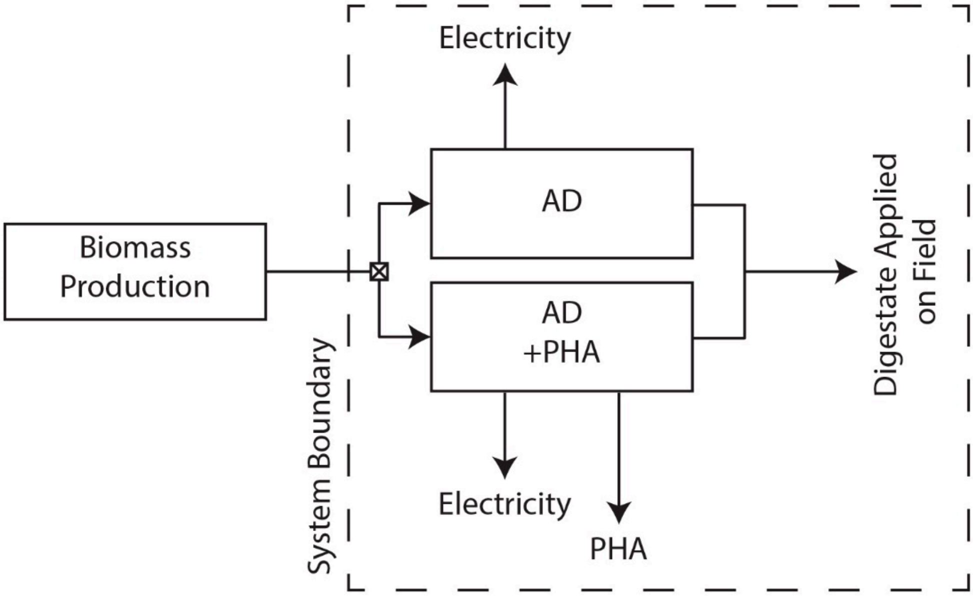

Figure 1. System boundary definition.

2.4. Dynamics

Dynamic inventories of the electricity mix for the two locations, modelled for a period of 20 years from 2015–2035, were used in the analysis. Four different dynamic energy futures, developed by the French government, with yearly shifting percentages of contributing sources of energy (Figure 2), were used for all electricity provision in the scenarios for Languedoc-Roussillon [27]. Likewise, three different dynamic energy futures were developed based on the legislation for Oregon State (Figure 3), which regulates the share of renewables in Oregon's future energy grid [28]. Qualifying renewables, i.e., renewable energy sources accepted by Oregon legislation on renewables, were introduced in varying amounts. Thus, (1) a scenario where biomass was increased more than other qualifying renewables, (2) a scenario where wind and solar were increased more than other qualifying renewables, and (3) a scenario where all qualifying renewables were increased evenly were developed. Static electricity mix scenarios were also included for both locations.

To maintain consistency in the foreground and background systems, the electricity provision component of all Ecoinvent processes used in the assessment was exchanged with the dynamic mixes developed. This included the electricity for fertilizer production, conventional polymer production, and the electricity replaced in the grid. This use of the local grid mix in the commodity production may not be a 100% accurate representation of a market reaction for the background systems, but it is deemed a better representation than the static processes. Further discussion on this subject can be found in Section 4.

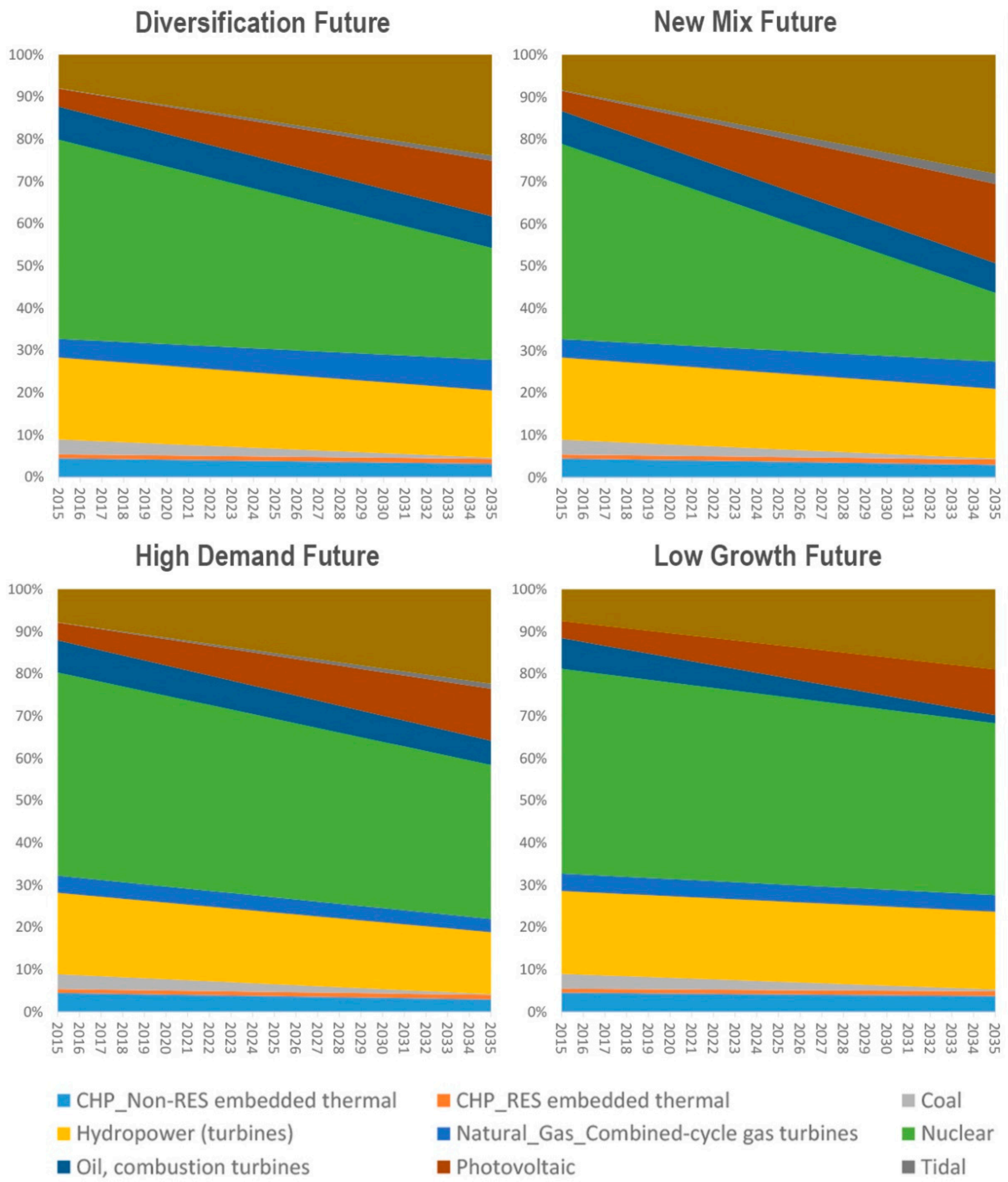

Figure 2. Evolution of the French electricity grid based on future scenarios defined by [27].

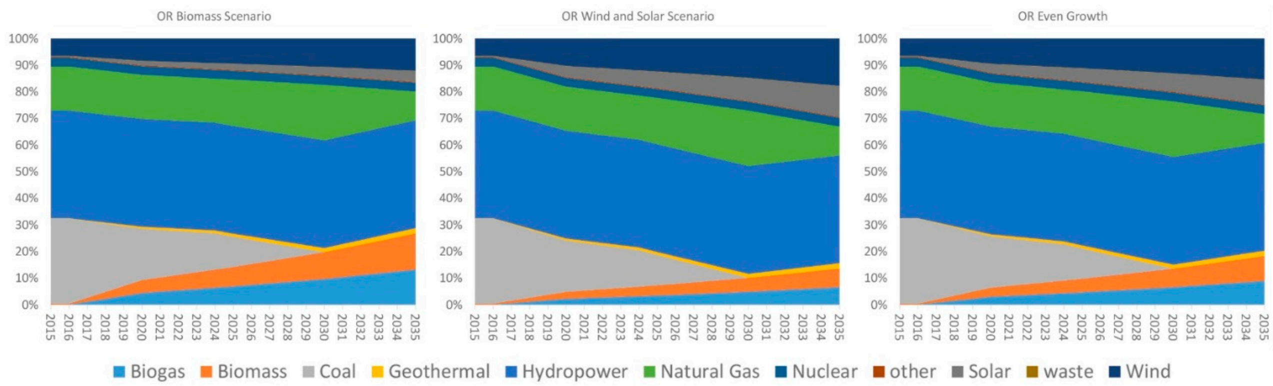

Figure 3. Evolution of the Oregon electricity grid based on three possible future scenarios for the fulfillment of legal requirements for decommissioning fossil-based production facilities.

PHA Process Energy Consumption

PHA production, which has been around since the 1980s, is already practiced at industrial level with first generation feedstock such as sugars from corn and sugarcane. Plants already exist with capacities ranging from 2000 to 50,000 tons of annual production [29]. Furthermore, PHA production has been introduced to the waste water treatment sector [30,31] and is also possible from second generation biomass. Due to important experience in the market with regards to PHA production, the PHA production for second generation biomass, as in the present study, will likely attain vast improvements in the future, eventually reaching a maturity level comparable to current industrial PHA production. To reflect this, dynamics in the PHA inventory were included in terms of electricity consumption (i.e., energy efficiency), in addition to the dynamic electricity provision. Hence, while PHA production was modelled starting as 7 kwh/FU more burdensome than the biogas-only scenario, thereafter the process was modelled as becoming more energy efficient, improving by 1% annually for the 20-year period, based on similar technology learning curves [32]. This improvement rate was also tested in terms of influence on total impacts (see Section 2.7).

2.5. Implementation of Territorial Scale Assessment

In order to assess the implications of implementing PHA technology at a territorial scale, the two study regions, in France and Oregon respectively, were analyzed regarding ability to provide feedstock for application in the two assessed biorefinery scenarios, i.e., impacts arising from treating all feedstock available in the region by biogas-only or combined PHA-biogas. The territories were defined as the interacting areas of residue production and the treatment plants. However, as defined in the TM-LCA method [4], only the areas undergoing change are included in the assessment. In this case, the change is an average change reflected in the residue treatment centers. Therefore, it is not expected that this change will affect the production of the residues in any way, ergo feedstock producers are left out of the assessment in terms of environmental impact. Likewise, transport from producers to treatment centers is not expected to change, as the volume of residues produced will not change as a consequence of implementing PHA technology. Where there is potential for transport that would deviate from the status quo, namely in the transport of grape marc which is the lighter of the two feedstock, impacts from transport were assessed (see 0, Sensitivity Analysis). These impacts were not included in the main results, as the induced impacts from transport would be equal in both the PHA-biogas and the biogas-only scenarios.

Feedstock Provision

Several assumptions were made in relation to determining the amounts of residue produced in each region for input into the regional scale assessment (Table 1). For wineries, it is assumed that grape marc is produced at a rate of 0.13 tons per ton of processed wine grapes [33]. It is further assumed that in France, where production data are reported in hectoliters of wine instead of mass of grapes at crush, 140 kg of grapes are used to produce 1 hectoliter of wine [34]. For feedstock coming from cattle, it is assumed that all waste comes from dairy cattle and that dairy cattle produce waste at a rate of 54.5 kg per head per day [35].

Due to the relative scale of wine production and the cattle industry in Oregon, the production capacity of the biorefinery systems in Oregon is limited by the production of grape marc, assuming that the co-digestion of cow waste and grape marc is not augmented with alternative feedstock. With nearly 2.4 million tons of waste produced by dairy cattle annually [35] and only 8010 tons of grape marc produced annually, the treatment of all grape marc (at 35% of total treated biomass) would require appx. 1% of the dairy cattle manure provision capability of Oregon. However, the total production of this system might not be enough to provision a fully industrial scale biogas plant, though it would be enough to provision a smaller scale plant, and implications of this are discussed in Section 2.7.4.

Conversely, in relation to Oregon, the capacity of the biorefinery systems in Languedoc-Roussillon is limited by the production of manure. With only 18,700 dairy cattle [36], the region would only be able to supply appx. 0.37 million tons of the 0.39 million tons manure needed for co-digestion with the 0.21 million tons of grape marc produced in the region annually (CIVL—Conseil Interprofessionnel des vin AOC du Languedoc et des IGP Sud de France—Languedoc Wines). This relationship, unlike that in Oregon, is fairly well balanced. However, unlike in Oregon, there are well-established uses for grape marc, so the ability to provide grape marc as feedstock would therefore compete with existing demand (see Section 4).

Table 1. Feedstock provision for Languedoc-Roussillon and Oregon.

	Languedoc-Roussillon	**Oregon**
Annual Grape Marc Production (tons at crush)	212,940	8,009
Annual Cow Waste Production (tons)	372,300	2,389,091
Max. Co-digestion Feedstock Availability at 35% Grape Marc (tons/day)	1569	62
Cow Waste Demand at 100% Grape Marc Utilization (tons)	395,460	14,875
Grape Marc Demand at 100% Cow Waste Utilization (tons)	200,469	1,286,433
Cow Waste Demand at 100% Grape Marc Utilization (% of available cow waste)	106%	0.62%
Grape Marc Demand at 100% Cow Waste Utilization (% of available grape marc)	94%	16,061%

2.6. Impact Assessment Method

The ReCiPe 2016 Hierarchist method was used for impact assessment [37]. Impacts were assessed at the midpoint level with a time horizon of 100 years from the time of emission. All impact categories were included in the assessment of the dynamic system model and in all scenarios.

While all impact categories were modelled, using all indicators creates difficulty in relation to the interpretation of the results. To avoid this obstacle, GWP was chosen as a single indicator for impacts. In order to check for potential burden shifting when solely using GWP as an indicator impact, TOPSIS was applied with equal weighting to all impact categories. Ranking of the scenario results was then performed in a pairwise fashion, i.e., within each energy mix future, for the two scenarios, biogas-only and PHA-biogas, using both GWP as a single score indicator and TOPSIS.

2.7. Sensitivity Analysis

Important modelling parameters and assumptions were tested through a sensitivity analysis. These include:

2.7.1. Process Energy Consumption Related to PHA Production

Energy consumption related to PHB production was calculated using process design software, and it was subsequently tested to see if the overall results were sensitive to this parameter. Thus, a scenario where the energy consumption of PHB production does not improve over time was tested. For contrast, a scenario where processing improves by 5% per year was also explored.

2.7.2. Replacement Ratio Conventional Polymers

Replacement ratios of PHB to PET and PLA were estimated using the following material property indices: tensile strength, yield strength (σ), and the average between tensile strength and yield strength. RRs in the first model run were based on yield strength (σ), which applies to brittle polymers that are loaded in tension. This is done in order to relate the polymer matrix to its final application, which is unknown and is most likely several different applications for this case study. Thus, by choosing a handful of material properties, it is possible to estimate more realistic RRs that apply to desired properties. The values used of the RR estimation are presented in Table 2.

Table 2. Material properties, performance indices of polyethylene terephthalate (PET), polylactide (PLA) and polyhydroxybutyrate (PHB). Replacement ratios are derived from material properties using Equation (1).

	PET [38]	***PLA*** [39]	***PHB*** [40]
Yield strength, σ (Mpa)	2410.0	3830.0	2200.0
Tensile strength (Mpa)	38.8	48.0	32.0
Density (kg/m^3)	1.3	1.2	1.2
Performance index (YS)	1882.8	3088.7	1833.3
Performance index (TS)	30.3	38.7	26.7
Average performance	956.6	1563.7	930.0
Replacement Ratio (RR), YS	0.97	0.59	
RR, TS	0.88	0.69	
RR, AVG	0.93	0.64	

2.7.3. Mineralization of N in Digestate

An important source of uncertainty comes from the application of digestate to the field. In the first model run, EFs for N_2O emissions were based on IPCC values. To test the possible range of impact arising from N_2O emissions in the field, a powerful greenhouse gas, a second model run was performed using the N_2O emission factors published by [22]. Though these are not local EFs, they are used to portray the potential variation of greenhouse gas emissions after digestate application. The values used are found in Supplementary Table S5.

2.7.4. Feedstock Provisioning Scenarios

In both regions, there is potential for increased ground transportation induced by transport of grape marc for PHB production. Transport for grape marc is, in most cases, non-existent in Oregon whereas transport is used to distribute grape marc amongst various end-users in France. This means that implementing a PHA-producing biorefinery would either route or re-route the grape marc needed as feedstock to the biorefinery. To account for this, the system was modelled with ground transport of the grape marc by lorry. This was done for various potential transport distances ranging from 50–500 km for the PET replacement scenario.

3. Results

Results showed that the PHA scenarios outperformed the biogas-only scenarios in almost every impact category with a few exceptions (Figure 4). Exceptions included the French energy scenarios for the Ionizing Radiation (IR) impact category and almost all scenarios for Land Use (LU), except in one instance, the Oregon Static scenario, where PHA-biogas performed better than biogas-only in terms of LU.

It is worth noting that in some of the impact categories the difference between the two scenarios is so small that, keeping in mind the considerable uncertainty of LCA results in general, it is fair to say that both PHA-biogas and biogas-only are essentially equal in terms of environmental impact. This is true for the Particulate Matter (PM), Fresh Water Ecotoxicity (FWE), Land Use (LU), Marine Ecotoxicity (MEtox), Marine Eutrophication (ME), Mineral Resource Scarcity (MRC), both Ozone Formation categories, Terrestrial Acidification (TA), and Stratospheric Ozone Depletion (SOD) impact categories. The remaining impact categories show a greater degree of difference, where it is clear that the PHA scenarios are generally preferable. Midpoint impact category results are presented as percent reduction in environmental impact from the implementation of PHA production in relation to biogas-only scenarios, for all energy provision scenarios. These are shown both for scenarios replacing PET with a ca. 93% RR and a 30% RR, to show the influence of RR in impact results (Figure 5).

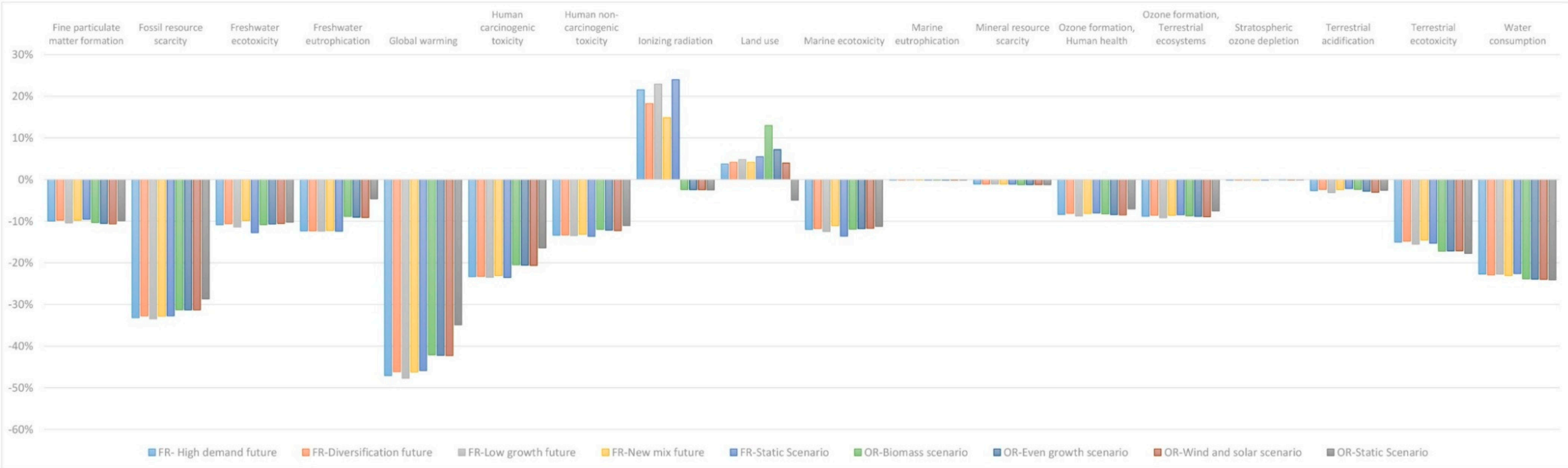

Figure 4. Relative difference for the PHA-biogas scenario. Negative values indicate that PHA-biogas outperforms Biogas-only scenarios.

	Fine particulate matter formation	Fossil resource scarcity	Freshwater ecotoxicity	Freshwater eutrophication	Global warming	Human carcinogenic toxicity	Human non-carcinogenic toxicity	Ionizing radiation	Land use	Marine ecotoxicity	Marine eutrophication	Mineral resource scarcity	Ozone formation, Human health	Ozone formation, Terrestrial ecosystems	Stratospheric ozone depletion	Terrestrial acidification	Terrestrial ecotoxicity	Water consumption
FR- High demand future	10.09%	36.88%	11.10%	14.86%	64.33%	27.53%	14.08%	-30.54%	-5.62%	12.23%	-0.13%	1.08%	8.62%	9.09%	-0.15%	2.70%	15.19%	22.78%
FR-Diversification future	9.78%	36.02%	10.79%	14.82%	61.39%	27.41%	14.00%	-28.86%	-6.19%	11.94%	-0.13%	1.09%	8.31%	8.78%	-0.14%	2.39%	14.89%	23.08%
FR-Low growth future	10.63%	37.46%	11.82%	14.94%	66.58%	27.80%	14.22%	-31.14%	-6.99%	12.89%	-0.13%	1.08%	9.08%	9.55%	-0.16%	3.24%	15.90%	22.80%
FR-New mix future	9.80%	36.13%	9.83%	14.68%	61.66%	27.04%	13.77%	-26.68%	-6.18%	11.10%	-0.13%	1.08%	8.36%	8.83%	-0.14%	2.42%	14.53%	23.32%
FR-Static Scenario	9.55%	35.99%	13.64%	14.96%	60.60%	27.91%	14.45%	-31.55%	-7.81%	14.40%	-0.13%	1.11%	8.17%	8.64%	-0.14%	2.17%	15.55%	22.60%
OR-Biomass scenario	10.52%	33.21%	11.07%	9.78%	50.11%	22.54%	12.19%	13.64%	-14.91%	12.13%	-0.15%	1.23%	8.47%	8.92%	-0.03%	2.39%	18.26%	24.50%
OR-Even growth scenario	10.77%	33.25%	10.89%	10.01%	50.44%	22.68%	12.46%	13.86%	-9.70%	11.99%	-0.15%	1.22%	8.68%	9.14%	-0.07%	2.86%	18.18%	24.64%
OR-Wind and solar scenario	10.92%	33.27%	10.73%	10.13%	50.63%	22.77%	12.61%	13.99%	-5.91%	11.86%	-0.15%	1.22%	8.79%	9.25%	-0.10%	3.12%	18.14%	24.72%
OR-Static Scenario	10.01%	28.67%	10.35%	4.65%	34.92%	16.41%	11.06%	14.30%	10.55%	11.29%	-0.15%	1.24%	7.08%	7.55%	-0.10%	2.60%	19.09%	24.93%

(A)

	Fine particulate matter formation	Fossil resource scarcity	Freshwater ecotoxicity	Freshwater eutrophication	Global warming	Human carcinogenic toxicity	Human non-carcinogenic toxicity	Ionizing radiation	Land use	Marine ecotoxicity	Marine eutrophication	Mineral resource scarcity	Ozone formation, Human health	Ozone formation, Terrestrial ecosystems	Stratospheric ozone depletion	Terrestrial acidification	Terrestrial ecotoxicity	Water consumption
FR- High demand future	-0.26%	13.73%	-0.31%	4.36%	29.86%	7.80%	3.55%	-36.84%	-14.44%	0.54%	-0.12%	0.14%	1.36%	1.53%	0.03%	-6.54%	2.63%	4.72%
FR-Diversification future	-0.55%	12.89%	-0.59%	4.33%	26.00%	7.69%	3.47%	-35.83%	-14.93%	0.29%	-0.12%	0.15%	1.07%	1.23%	0.04%	-6.82%	2.35%	4.98%
FR-Low growth future	0.23%	14.30%	0.34%	4.44%	32.95%	8.05%	3.67%	-37.19%	-15.59%	1.14%	-0.12%	0.14%	1.79%	1.95%	0.02%	-6.07%	3.27%	4.73%
FR-New mix future	-0.52%	13.00%	-1.45%	4.20%	26.35%	7.34%	3.26%	-34.52%	-14.91%	-0.47%	-0.12%	0.14%	1.12%	1.28%	0.04%	-6.79%	2.02%	5.20%
FR-Static Scenario	-0.75%	12.86%	1.99%	4.46%	24.99%	8.16%	3.89%	-37.42%	-16.27%	2.51%	-0.12%	0.16%	0.94%	1.10%	0.04%	-7.01%	2.95%	4.55%
OR-Biomass scenario	0.13%	10.18%	-0.34%	-0.30%	12.79%	3.24%	1.81%	-5.76%	-22.08%	0.45%	-0.14%	0.29%	1.22%	1.37%	0.15%	-6.82%	5.43%	6.28%
OR-Even growth scenario	0.36%	10.22%	-0.50%	-0.10%	13.13%	3.37%	2.05%	-5.58%	-17.82%	0.33%	-0.14%	0.28%	1.41%	1.56%	0.11%	-6.40%	5.36%	6.41%
OR-Wind and solar scenario	0.49%	10.24%	-0.65%	0.01%	13.34%	3.45%	2.19%	-5.47%	-14.66%	0.21%	-0.14%	0.28%	1.52%	1.67%	0.08%	-6.17%	5.33%	6.49%
OR-Static Scenario	-0.33%	5.96%	-0.99%	-4.91%	-1.62%	-2.10%	0.77%	-5.22%	-0.34%	-0.30%	-0.14%	0.30%	-0.07%	0.09%	0.08%	-6.63%	6.20%	6.68%

(B)

Figure 5. Percent reduction in environmental impact for all midpoint impact categories for the implementation of PHA production relative to biogas-only, for all energy provision scenarios with a PET RR of (**A**) 93% and (**B**) 30%.

The first model run shown in Figure 4 has PET as the conventional polymer to be replaced by PHB. The model was checked to see if a different polymer substitution material would alter the results. It was found that a change to PLA as the polymer substitution material did not change the general ranking, but the magnitude of the difference between PHA-biogas and biogas-only, i.e., the advantage that PHA-biogas has over biogas-only, decreased. Figures and tables for the PHA-biogas results for PLA are shown in the SI (Supplementary Figure S4 and Table S7).

Figure 6 shows the difference between the PHA-biogas and biogas-only scenarios, i.e., PHA-biogas CO_2-eq *minus* biogas-only, in CO_2-eq. For all 20 years, the PHA-biogas scenario induces greater savings than the biogas-only scenarios, which is why the results are always negative. Furthermore, the general negative slope of all scenario lines shows that as time progresses PHA-biogas becomes more attractive, inducing higher savings in comparison to biogas-only. More interestingly, it is possible to observe the difference between plans for energy grid development in the two locations. Hence, Oregon scenarios show a steeper slope, i.e., a drastic pull back from the use of fossil fuels and, more specifically, the use of coal. In contrast, the French slopes are less pronounced, as improvements to the grid are subtler because there is already a large share of non-fossil-based energy production in use in France. The difference between the two regions is larger at the beginning of the period, getting smaller in time as the grids progressively increase their share of renewable energy.

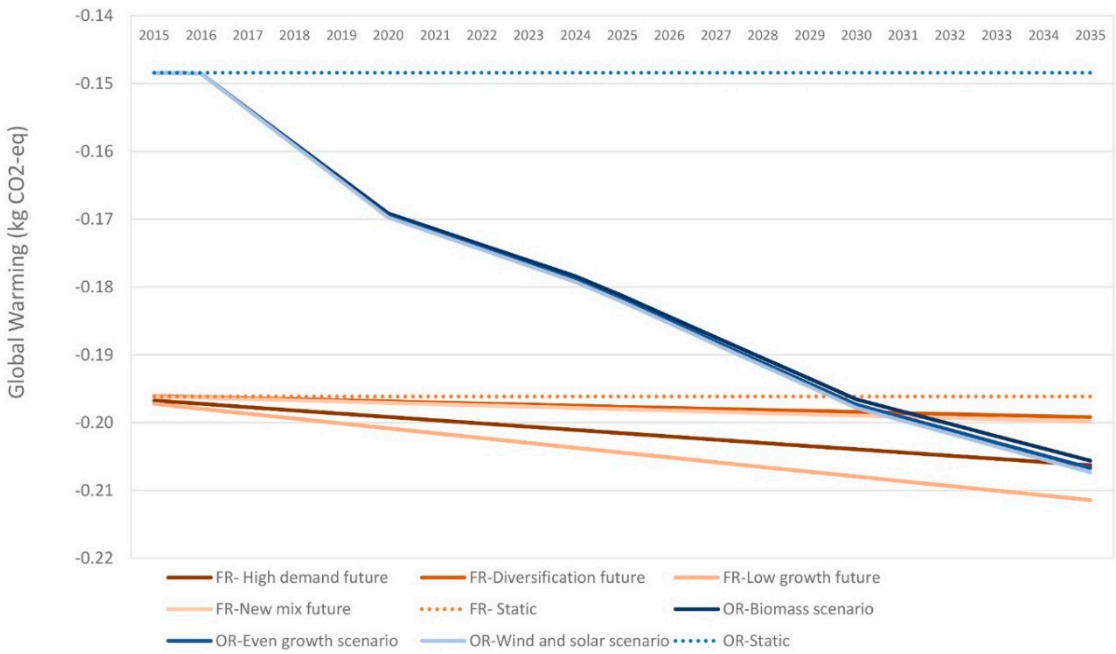

Figure 6. Yearly difference of global warming potential (GWP) impacts, i.e., PHA-biogas minus biogas-only scenarios. Figure reflects the evolution of the energy mixes in the two locations. Negative values mean PHA-biogas has higher savings than biogas-only.

3.1. Sensitivity Results

The robustness of model results was checked by varying different parameters, as described in the methodology, Section 2.7. After each change, indicators were checked with the TOPSIS and GWP single indicators, but for the most part, there was no change to the preference ranking of the scenarios, and combined PHA-biogas production continued to perform better. Thus, it can be said that the model results are robust in regards to the most influential parameters analyzed.

In more detail, changes to the replacement ratio (RR), i.e., the PHB: PET mass ratio that is allowed by different material properties, as discussed in Section 2.7.2, was shown to be a moderately sensitive parameter. A 5% change in the replacement ratio lead to a 3–4% change in results for PHA-biogas with PET (Figure 7), and a 2.5–4% change in results for PHA-biogas with PLA. Thus, it can be said that a general trend is observed of lower savings with lower RR (or higher savings with higher RR), while the effect of the change is nearly proportional to the change seen in the results.

The sensitivity to efficiency improvements for PHA-producing technology was also tested and it is shown in the SI, Figure S3. This parameter was showed to have very little effect on overall model results, with GWP changing in the range of 0.1–1.5%.

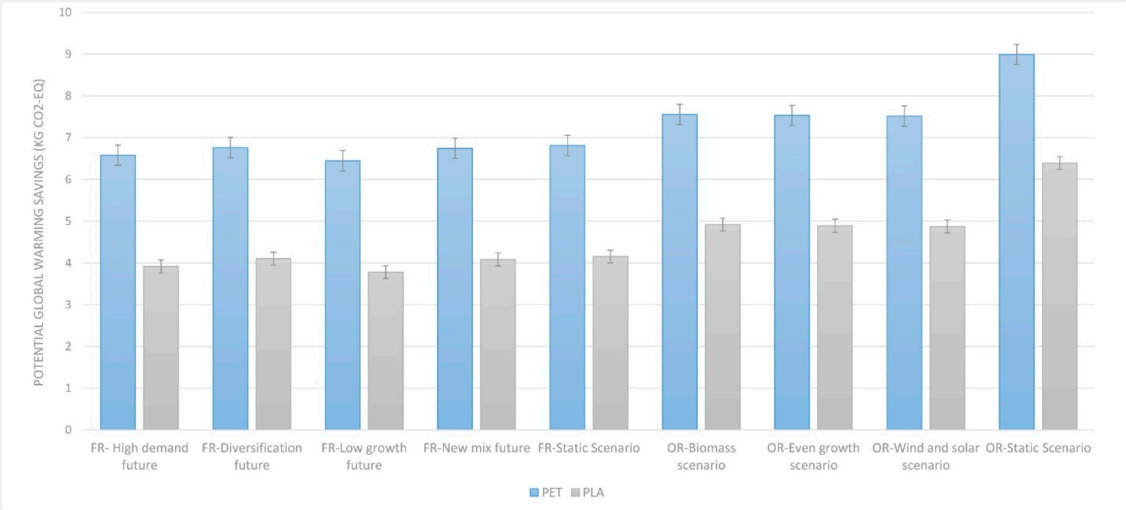

Figure 7. Sensitivity analysis of replacement ratio of conventional polymers by PHB. Cumulative GWP savings for substitution of PET, in blue, or PLA, in gray, by PHB. Bars represent savings in relation to the biogas-only scenario and the upper and lower error bars represent the range of potential savings, which depends on material properties' performance indices. PHA scenarios only.

Sensitivity of N_2O Emission Factor

Cumulative global warming impacts switch from a savings inducing status to a burden inducing status when N_2O emission factors for the field application of digestate from [22] are applied (Supplementary Figure S4). However, the ranking between PHA-biogas and biogas-only stays the same, with combined PHA-biogas scenarios continuing to perform better than biogas-only scenarios. The results show that N_2O emissions play an important role, and considering the strong dependency on local conditions, they should as much as possible be spatially differentiated. The variability of N_2O emissions for the EFs employed can be seen in Figure 8.

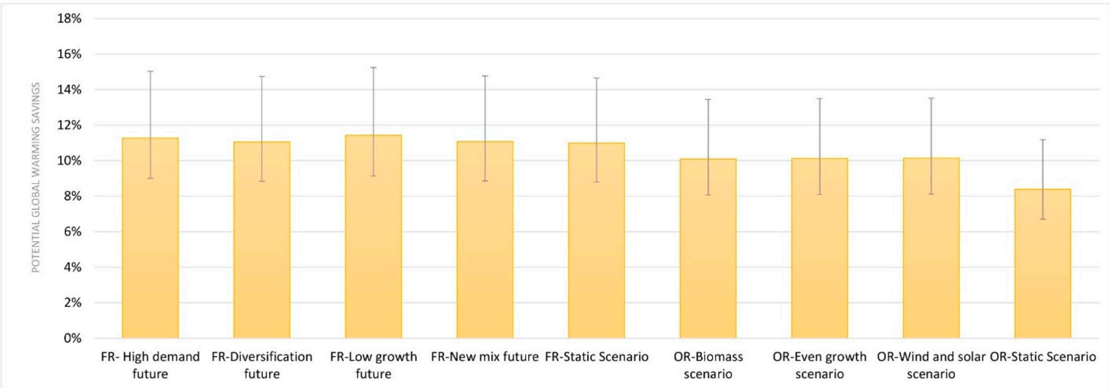

Figure 8. Cumulative PHA-biogas GWP minus cumulative biogas-only GWP. Yellow bars indicate relative savings of PHA-biogas scenarios in relation to biogas-only for each energy mix future. Error bars indicate variation in the savings induced by PHA-biogas due to N_2O emissions after application of digestate. Upper error bars correspond to the high crop response case, while lower error bars correspond to the low crop response case, as explained in Section 2.3.2.

3.2. Territorial Scale Application

Application of the biorefinery alternatives at a territorial scale would lead to potential reductions in regional environmental impact. In order to give a measure of scale to the potential savings induced by the implementation of maximum (limited by feedstock availability) PHA-biogas production relative to biogas-only, the GWP impacts were normalized using planetary boundary carrying capacity-based normalization factors [41]. Assuming a 985 kg CO_2 eq. per person year (PY) carrying capacity (C.Cap) [41], and assuming that PHA replaces PET with a 93% RR and that the PHA process improves in terms of energy efficiency at 1% annually, the production of PHA induces an average reduction in GWP impacts relative to biogas-only equating to nearly 1400 PY of C.Cap. When broken down by region, the French scenarios indicate an average relative maximum potential GWP saving of over 2400 PY of C.Cap, with Oregon exhibiting just over 80 PY of C.Cap in average relative maximum potential GWP savings. Using the same assumptions, except exchanging the replacement polymer with PLA production at a 64% RR, then the maximum implementation in France and Oregon of the PHA-biogas scenario induces an average annual potential relative GWP impact reduction of 493 PY of C.Cap when compared to production of biogas-only, with 871 and 21 PY of C.Cap in France and Oregon, respectively, see Table 3.

Table 3. Carrying capacity normalized GWP reduction for maximum application of the PHA-biogas relative to the biogas-only biorefinery alternative in France and Oregon based on replacement of PET with 93% RR and a 1% annual energy efficiency improvement for PHA production. Reduction per functional unit (FU).

	GWP (Kg CO2e) Reduction/Fu	Person Years (PY) of Carrying Capacity (C.Cap) Reduction Daily	PY of C.Cap Reduction Annually
FR-HIGH DEMAND FUTURE	4.23	6.74	2460.75
FR-DIVERSIFICATION FUTURE	4.15	6.61	2413.15
FR-LOW GROWTH FUTURE	4.29	6.84	2495.46
FR-NEW MIX FUTURE	4.16	6.62	2417.67
FR-STATIC SCENARIO	4.13	6.57	2399.86
OR-BIOMASS SCENARIO	3.79	0.24	86.98
OR-EVEN GROWTH SCENARIO	3.80	0.24	87.25
OR-WIND AND SOLAR SCENARIO	3.80	0.24	87.41
OR-STATIC SCENARIO	3.14	0.20	72.14

Sensitivity Analysis of Transport at Territorial Scale

The importance of transport was tested via sensitivity analysis of different theoretical grape marc transport distances for both the biogas-only and PHA-biogas scenarios (Table 4). For all scenarios, a 500 km transport distance results in overall elimination of environmental benefits, and at 200 km, transport of grape marc reduces average impact savings from the various biorefinery-region scenarios by 42.5% for all midpoint indicators. In terms of GWP, a 200 km transport distance induces impacts of a maximum of appx. 284% and a minimum of 68% of the magnitude of GWP savings without transport. At 50 km, all scenarios show reductions in GWP. At 100 km, all PHA production scenarios and France biogas-only scenarios induce GWP savings, while the Oregon biogas-only production scenarios eliminate the GWP benefit of implementing the biorefinery. Furthermore, if the introduction of centralized PHA-Biogas biorefineries were to induce transport of grape marc, relative to existing decentralized biogas production, then GWP savings are overwhelmed by the induced impact from transportation at any distance greater than appx. 125 km.

Table 4. Sensitivity to inclusion of transport of grape marc in percentage change to midpoint impacts without transport.

	50 km	100 km	200 km	500 km
AVERAGE CHANGE AMONGST ALL IMPACT CATEGORIES	11%	21%	43%	106%
AVERAGE CHANGE IN GWP	36%	73%	145%	363%
MAX. CHANGE IN GWP	71%	142%	284%	710%

4. Discussion

Overall, the model results obtained were robust and indicate that implementing PHA production technology is preferable to conventional anaerobic digestion, when the functional unit (FU) equals 1 ton of feedstock treated. Combined PHA-biogas scenarios, whether with PET or PLA as the replaced polymer, performed better across almost every impact category. This is largely due to the added benefit of replacing conventional polymers, which are associated with significant impacts. As evidenced by the replacement ratio (RR) sensitivity analysis, decreasing or increasing the amount of PHB needed to equate the function of PET or PLA resulted in an almost proportional effect in the outcome. RR of PET would have to decrease by around 80% and be as low as 20% before there is rank reversal between the two options in some of the impact categories. This was confirmed by both single score indicators, which prefer combined PHA-biogas scenarios until reaching values close to 20% RR (Table 5). However, the GWP single indicator still preferred PHA-biogas, even at a 20% RR, except for the OR-Static Scenario. On the contrary, the TOPSIS single indicator, which is equally weighted between impact categories, starts preferring biogas-only scenarios earlier, with a 35% RR. In this regard, there was less operating space for the GWP indicator, when PLA is the replacement polymer, which starts signaling biogas-only as the preferred choice already at 30% RR. On the contrary, TOPSIS selects biogas-only at low RR of 9–16%. Thus, there is disagreement between the GWP and TOPSIS single indicators, which is, furthermore, replacement polymer-dependent. This points to two issues to consider: (1) choosing GWP as the only impact category for the assessment can potentially result in burden shifting to other environmental impact categories and (2) the choice of polymer substitution affects impact categories other than GWP, here exemplified by the difference in the TOPSIS results when choosing PET or PLA as polymer replacement. To elaborate, the difference lies in PET's production being more burdensome for impact categories other than GWP in comparison to PLA's production. However, the single score indicators employed generally indicated a similar scenario prioritization, i.e., combined PHA-biogas production being the preferred choice across all future energy scenarios, as long as RRs were higher than 20% for PET and 30% for PLA. It is worth noting that such a low replacement ratio is considered unrealistic, as the material properties of PHB allow for various applications [40].

Table 5. Single indicator preference, by TOPSIS with equal weights or GWP. Sensitivity values shown. For energy demand of calculated PHA production, values start with 10 times the calculated energy needed. For RR, values are shown for a replacement rate lower than 42%; above this value, PHA-biogas is always preferred.

		FR-High Demand Future	FR-Diversification Future	FR-Low Growth Future	FR-New Mix Future	FR-Static Scenario	OR-Biomass Scenario	OR-Even Growth Scenario	OR-Wind and Solar Scenario	OR-Static Scenario
Energy Demand for PHA Production (kWh/FU)										
70.70	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
	TOPSIS Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
77.70	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
	TOPSIS Preference	PHA	PHA	PHA	PHA	Biogas	PHA	PHA	PHA	PHA
84.84	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
	TOPSIS Preference	Biogas	Biogas	Biogas	PHA	Biogas	Biogas	PHA	PHA	PHA
98.98	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	PHA	PHA	PHA
106.10	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	PHA	PHA
113.12	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	Biogas
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	PHA	PHA
127.26	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	Biogas
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	PHA
226.34	GWP Preference	PHA	PHA	PHA	PHA	PHA	Biogas	Biogas	Biogas	Biogas
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas
388.85	GWP Preference	PHA	PHA	PHA	PHA	Biogas	Biogas	Biogas	Biogas	Biogas
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas
537.32	GWP Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas
Polymer replacement ratio (PHB:PET)										
42%	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
	TOPSIS Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
32%	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA
	TOPSIS Preference	PHA	PHA	PHA	PHA	Biogas	Biogas	Biogas	Biogas	PHA
22%	GWP Preference	PHA	PHA	PHA	PHA	PHA	PHA	PHA	PHA	Biogas
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas
12%	GWP Preference	PHA	Biogas	PHA	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas
	TOPSIS Preference	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas	Biogas

Much like with polymer replacement ratios, TOPSIS and GWP do not always agree when the limits of process energy consumption are tested. If process energy consumption reaches 134 kWh per FU of added energy demand for PHA production, then TOPSIS (unlike GWP) indicates preference for biogas-only, for all energy scenarios, which indicates there is a potential for burden shifting if GWP is chosen as the only indicator. However, unlike the replacement ratio, improvements in process energy consumption for the production of PHA lead to very small changes in results. If there is no improvement in process energy consumption, meaning production of PHA consumes 7 kWh more per FU than the biogas-only scenario, results still stay the same. The break-even point of energy consumption for PHA production is high, i.e., it takes 12 times this value, 85 kWh of added process energy consumption of PHA per ton feedstock, before the TOPSIS-derived single indicator shows preference for biogas-only over combined PHA-biogas production for several of the French energy scenarios and one Oregon scenario. Moreover, it takes 16 times this value, or 113 kWh/FU more, before it is possible to observe prioritization change for the GWP single indicator for one Oregon scenario, the OR-Static Scenario, and 32 times the initial value, 226kWh/FU, before all Oregon energy scenarios show a preference for biogas-only. As for France, it is not until PHA production consumes 55 times this value, 389 kWh/FU, before there is a change in the GWP single indicator in preference of one of the energy future scenarios; the FR-Static Scenario. Thus, it is possible to conclude that there is large leeway in process energy consumption for PHA production before the decision support will change, in terms of GWP. As exemplified here, this is also dependent on the share of renewable energy sources in the future energy grid, which is why results are more robust for France in terms of GWP, i.e., requiring 55 times, 7 kWh/FU, more energy consumption before seeing a change in GWP impact category. The energy prediction mix is thereby an important factor when deriving the impacts of the system, which are heavily affected by energy mix usage.

In this regard, using dynamic energy grids for the background is a powerful tool. Many nuances are highlighted and originate from the predicted/expected changes in the share of renewable energy for the different locations. The most obvious of these subtleties can be observed in the Ionizing Radiation category (Figure 4), where it is evident that there is a higher share of nuclear energy in the French background system than in that of Oregon. As seen in Figure 6, the evolution of the energy grid reveals a sharp decrease for Oregon, while France's energy grid remains somewhat unaltered. This is due to legal requirements in Oregon, which are intended to increase the share of renewables from 15% to 50% by 2040 [28]. Greening of the energy grids increases the difference between biogas-only and PHA-biogas in the future, as is exhibited by the negative slopes of the lines in Figure 6. Despite the increasing environmental importance of plastic replacement as opposed to electricity replacement, it is worth restating that PHA-biogas is consistently preferable in terms of GWP, i.e., negative values throughout the assessment period. One major area discussion regarding the dynamic inventory is the use of local energy mix scenarios in commodity replacement. It is likely that the increased production of PHA would have no direct effect on the production of PET or PLA in Oregon or France. However, by using a local instead of global process, it is possible to develop processes that are treated equally, in terms of system dynamism, for their inventory development. Furthermore, this is seen as a cautious choice, as the localized dynamic processes for the replaced polymers exhibit lower impacts than the global average. Thus, it is possible that this inclusion slightly under-represents the potential impact reduction gains from increased PHA production and is hence considered unlikely to over-state impact reduction gains.

As shown in the sensitivity analyses, biogas-only scenarios are preferred only in extreme cases where polymer replacement ratio or consumption of energy during PHA production are set to extreme values, i.e., very low RR and very high process energy consumption for PHA. Another area of uncertainty is N_2O emissions after digestate application, which have also been shown to be highly uncertain in several LCAs [42–44]. N_2O emissions were shown to have the potential to induce impacts for all scenarios, though the ranking of PHA-biogas in relation to biogas-only was not affected. Due to the closeness in results from the field application of digestates generated from the model for biogas and PHA scenarios, it can be concluded that both digestates act more or less in the same way during field application. Results were also tested without the field emissions, leading to the same technology prioritization. Nevertheless, it is important to highlight the large impact that N_2O emissions have in assessing agricultural product systems, and the necessity to improve inventories of these emissions in LCA assessments. Incidentally, the TM-LCA framework advocates for the use of local inventory data as much as possible.

One area that is made evident by including the territorial assessment, where there is potential for inducing impacts that would eliminate the environmental benefits of the system, is transport. Due to the relatively low energy and chemical value density in grape marc, increases in present transport of grape marc greater than 200 km cause induced impacts in all biogas-only scenarios. When transporting grape marc 250 km, both PHA-biogas with PET replacement and biogas-only induce impacts, except for the PHA-biogas scenario with static energy grid in Oregon, i.e., a dirtier energy mix than impacts from transport. Furthermore, if the PHA-biogas scenario induces transport relative to the biogas-only scenario (no added transport for biogas-only), then 150 km of grape marc transport eliminates the GWP benefit of the PHA-biogas scenario. While the PHA production scenario remains clearly preferable to biogas-only in all transport scenarios, this result does underline the need to assess potential re-routing of the feedstock if a new biorefinery technology were to be implemented.

It is also notable that the present use of feedstock, omitted in the results of this study as the impacts would be equal in both the PHA-biogas and the biogas-only scenarios, varies significantly between the two assessed territories. In France, there is a well-established market for distillation of wine residues, and in Oregon the wine residues are often used as compost. This said, it is also important to highlight that the feedstock mix used in this assessment can also be changed, as the PHA-producing technology is compatible with all types of organic waste, e.g., the organic fraction of household waste, waste-water treatment sludge, other animal slurries, other crop residues etc. The option to change the feedstock mix was not investigated in this study, as it would change the functional unit and was thus omitted from the present work. However, it is quite possible that there is further exploitable feedstock in both assessed regions. A good indication of feasibility is if there is an industrial sized biogas plant already in operation in the region; this would indicate that there is already feedstock enough to run PHA production. However, it is important to keep in mind that the use of crops has not been investigated in this report and so this study's conclusions do not apply if the feedstock is food crops.

5. Conclusions

Based on the results of this study, it can be concluded that when a biorefinery is installed in Oregon or Languedoc-Roussillon to handle a mix of grape marc and cow waste, it is very likely that it would be environmentally beneficial to include PHA production in addition to energy and digestate production. When relating the impact reductions between PHA-biogas and biogas-only, based on the maximum potential implementation capacity of the specific region, to planetary boundaries-based carrying capacity, it is shown that the impact reductions correspond to up to nearly 2500 person years in France and up to nearly 90 person years in Oregon. This corresponds to 1.59 and 1.40 person years of avoided GWP per ton of treated feedstock per day in France and Oregon, respectively. However, based on the results of the sensitivity analysis regarding transportation, special care needs to be taken in regards to assessing the potential increase in biomass transport; otherwise, it is likely that all environmental benefit from the biorefinery will be offset by the induced impacts of transportation. Likewise, the induced environmental impact reductions cannot be ensured if the feedstock for the biorefinery is to be rerouted from another use. Thus, it is concluded that PHA production should be seen as a potentially valuable add-on for biogas platforms.

The TM-LCA framework has the added benefit of elucidating the influence of potential future energy provision and the impact this has on potential environmental benefits. As indicated by the results, the benefit of including co-production of PHA in biogas plants increases as energy grids become greener, an element that can have significance in terms of decision support for its implementation from the regional planning or governance perspective. The framework also provides perspective on the scale of potential benefits (in person years) and added emphasis on single score indicators that point out possible burden shifting to environmental problems other than global warming.

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Paper II

Insights from combining techno-economic and life cycle assessment – a case study of polyphenol extraction from red wine pomace.

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Abstract: To determine the environmental and economic performance of emerging processes for the valorization of red wine pomace, a techno-economic assessment (TEA) and a life cycle assessment (LCA) of two polyphenol extraction methods, solvent extraction (SE) and pressurized liquid extraction (PLE), were combined into concise decision support, using Multiple Criteria Decision Analysis (MCDA), at an early design stage. SE performs better than PLE, due to a lower solvent to DW ratio and a less expensive processing setup. LCA at laboratory scale aided in showing potential environmental hotspots and highlighted the need to reduce solvent use. The MCDA showed a shift in decision support depending on how strongly economic or environmental benefits are valued. Both SE and PLE with a solvent to DW ratio of 5 and 10, respectively, perform competitively while SE with a solvent to DW ratio of 10 outperforms PLE with a solvent to DW ratio of 25.

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Title: Insights from combining techno-economic and life cycle assessment – a case study of polyphenol extraction from red wine pomace

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Insights from combining techno-economic and life cycle assessment – a case study of polyphenol extraction from red wine pomace

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Abstract

To determine the environmental and economic performance of emerging processes for the valorization of red wine pomace, a techno-economic assessment (TEA) and a life cycle assessment (LCA) of two polyphenol extraction methods, solvent extraction (SE) and pressurized liquid extraction (PLE), were combined into concise decision support, using Multiple Criteria Decision Analysis (MCDA), at an early design stage. SE performs better than PLE, due to a lower solvent to DW ratio and a less expensive processing setup. LCA at laboratory scale aided in showing potential environmental hotspots and highlighted the need to reduce solvent use. The MCDA showed a shift in decision support depending on how strongly economic or environmental benefits are valued. Both SE and PLE with a solvent to DW ratio of 5 and 10, respectively, perform competitively while SE with a solvent to DW ratio of 10 outperforms PLE with a solvent to DW ratio of 25.

Key words: Techno-economic assessment, Life cycle assessment, polyphenol extraction, solvent extraction, pressurized liquid extraction, process design optimization

1. Introduction

Biomass demand for the production of bioenergy, biomaterials and biochemicals is estimated to increase by 70-110 % by 2050 compared to 2005 levels (Mauser et al., 2015). A paradigm shift to renewable sources of production has long been discussed, in the context of circular economy and valorization of biomass waste resources produced through the agricultural value chain. The bioeconomy today is estimated to have a €2.4 billion annual turnover, and it is only expected to increase in the future (Scarlat et al., 2015). Yet, the prefix bio does not guarantee sustainability. For example, growing biomass for biofuels has long been debated (Haberl et al., 2010; Murphy et al., 2011; Popp et al., 2014), prompting the Renewable Energy Directive (The European Commission, 2018) at an international (pan-European) level to ensure valid quantification of greenhouse gas reductions claims. In this regard, integration of methods such as life cycle assessment (LCA) and techno-economic assessment (TEA) are valuable input for quantitative sustainability assessments.

Combined TEA-LCA has been applied in many occasions to assess the environmental and economic ramifications of implementing new technologies (Cai et al., 2018; Hise et al., 2016; Vaskan et al., 2018). More interestingly, TEA-LCA has been used to quantify and monetize externalities, namely environmental damages, to provide a more complete picture of the financial burdens arising from environmental problems (Ögmundarson et al., 2018; Pizzol et al., 2015). Recently, combined TEA and LCA has been used to optimize new production routes from an early design phase, as in the case of integrated wastewater treatment and microalgae production for biodiesel production (Barlow et al., 2016), or the integration of power-to-gas technology of methane and photovoltaics (Collet et al., 2017). Combined TEA and LCA lends itself well to finding production hot spots and opportunities for optimization. This is even more relevant when applied to renewable resources such as biomass, which have to be managed sustainably.

New materials like biodegradable bio-sourced biopolymers and bioactive molecules such as, polyphenols obtained from agricultural residues can be combined to create new and innovative products (Vannini et al., 2019). Polyphenols present interesting possibilities as they can be utilized by various industries, such as in the pharmaceutical, nutraceutical and cosmetic industries (Pérez-López et al., 2014). Among other, polyphenols have been shown to have excellent health promoting qualities, such as anti-diabetic, anti-inflammatory, anti-bacterial and anti-cancer properties (Nowshehri et al., 2015). This versatility means that polyphenols may be used in niche markets as well as in mass markets, with various uses that may be of importance to the bioeconomy e.g. active packaging, coloring, food supplements, etc. Wine pomace is a residue rich in polyphenols, with a global production of 68 million tons of wine pomace annually (Nowshehri et al., 2015). To ensure a sustainable exploitation of polyphenol rich biomass, innovative polyphenol extraction methods at the laboratory scale were analyzed using TEA-LCA in order to identify hotspots and potentially environmentally problematic production steps.

On the other hand, results from the application of TEA-LCA can sometimes be confounding if, for example, one option performs better environmentally while incurring financial loss. The multitude of factors that must be taken into account remains an issue, when policy makers, corporations, or any other actor is faced with the need to decisively and definitively choose between alternative solutions to a given problem. In order to handle this, the decision-making context surrounding such a choice can be handled in many ways, from community-based decision making to round table discussions or even executive fiat. But, without a tool for interpreting fundamentally conflicting information, the results of the decision making process can vary wildly and may depend on happenstance and or subjective factors. Multiple Criteria Decision Analysis (MCDA) has been applied to aid in alleviating these problems by introducing a transparent and repeatable form of decision support (Kalbar and Das, 2020; Köksalan et al., 2011).

When assessing environmental issues in an LCA perspective, oftentimes practitioners turn to single indicators such as global warming potential (carbon foot-printing), but this poses potential downfalls such as burden shifting e.g. shifting environmental burdens from carbon emissions to environmental or human toxicity (Laurent et al., 2012). In other cases, practitioners turn to endpoint damage modeling, but these have high levels of uncertainty, can lead to unintentional bias (Kalbar et al., 2012a; Sohn et al., 2017), and still leave the decision maker with several categories of environmental damages e.g. ecosystem health, human heath, and resource availability. Furthermore, neither of these methods can be directly combined with economic indicators. In some cases, LCA practitioners have monetized impacts in order to combine environmental and economic indicators, however these suffer from issues, among others, involving the relationship of internalized and externalized costs (Reap et al., 2008). These issues have lead some LCA practitioners to turn to MCDA for providing decision support (Kalbar et al., 2016, 2012a; Sohn et al., 2017), as applying MCDA with a defined decision context to results from TEA-LCA is advantageous when a final decision must be taken.

Therefore, in this study LCA is applied at an early design stage to obtain a preliminary carbon footprint (CFP) of the polyphenol extraction methods. Subsequently, TEA is applied to optimize and improve the design of the extraction processes and LCA is applied again with all environmental indicators in simulated industrial conditions, optimized with guidance from literature, the TEA and the preliminary CFP. This is done with the goal of obtaining a holistic picture of the economic feasibility and possible environmental impacts of each polyphenol extraction method. Lastly, MCDA is applied to the decision context of choosing between the polyphenol extraction methods and a weighting-profile derivation method (ArgCW-LCA) is applied (Sohn et al., 2020). The criteria from the LCA and TEA are then incorporated to provide concise decision support for selecting one of the methods for scale-up.

2. Material and Methods

Results of laboratory scale experiments of different methods for the extraction of polyphenols from red wine pomace were evaluated using a combination of TEA and LCA. Two different labs, one located at the University of Bologna, Italy, and a second located at the Research Institute of Sweden (RISE), provided operational parameters for their laboratory setups. Yields, solvent amounts, temperature and time were then used to complete the inventory to carry out a preliminary carbon foot-printing (CFP) LCA of the laboratory scale experiments. The parameters of the most successful setups i.e. those producing the highest polyphenol yields, were used for the CFP and are described in detail in Table S1 of the supplementary information. The laboratory methods are described briefly in section 2.1. Following this step, industrial scale processes of the laboratory methods were designed and optimized for key parameters using TEA (described in section 2.3). An LCA of the optimized industrial scale processes including all environmental indicators was then carried out. Lastly, a multiple criteria framework for decision support where the economic and environmental indicators are combined was applied to the results from the TEA-LCA.

2.1. Polyphenol extraction methods and laboratory experiments

The CFP of two different extraction methods, solvent extraction (SE) and pressurized liquid extraction (PLE), was determined. One SE setup and 3 different PLE setups, where the main difference is the solvent amounts used, were assessed for this step, of which the most relevant setups are briefly described below, and the remainder can be found in the SI, since they did not become relevant for the industrial case. The laboratory extraction methods are also described in detail in (Ferri et al., in press).

2.1.1. Solvent extraction with acetone – Lab-SE-11

Batch extraction was performed in the laboratory with 61% acetone, and 39% water as solvent on a per mass basis, with a solvent to DW ratio of 11:1. Extraction was performed in an air-tight vessel at 50°C at atmospheric pressure. The solvent and pomace were kept in contact for 2 hours, after which polyphenol content of the extracts was analyzed. Polyphenol content is expressed in kg gallic acid equivalents (kg GAE).

2.1.2. Pressurized liquid extraction with ethanol – Lab-PLE-101

PLE was performed with 37% ethanol, 39% water and 25% supercritical CO_2 on a per mass basis. The extraction is performed at 80°C and 100 bar. As this is a continuous set up, it leads to high solvent to DW ratio of 101.

2.2. Carbon foot-printing of laboratory scale experiments

A CFP was performed on the extraction methods described above, using only the Global Warming potential (GWP) impact category as the environmental indicator. The ReCiPe 2016 Midpoint Hierarchist (H) method (Huijbregts et al., 2017), which has a 100 year time horizon from point of emission, was used as impact assessment method, supplied by the Ecoinvent 3.4 Database (Wernet et al., 2016) and processed with the open source software OpenLCA (GreenDelta, 2019). The functional unit for the CFP is the production of 1 kg of polyphenols in GAE, assuming equal functionality. The process design software, Superpro Designer v.10 (Intelligen Inc, 2018), was used to simulate the polyphenol extraction methods with industrial scale equipment, while keeping all other operational parameters from the laboratory experiments (see Table S1, in the SI). The polyphenol producing plant is assumed to be placed in Italy and thereby, background processes for Italy from the Ecoinvent database were used as much as possible, e.g. the electricity grid.

2.3. Design of industrial scale processes

The process design focused on optimizing the operational parameters of the laboratory extraction methods so that it would be economically feasible to implement a polyphenol extraction at industrial scale. In order to achieve this, solvent recovery is essential i.e. several process steps are required such as distillation, pressing and desolventizing (Figure 1 and Figure 2). The solvent loss and the energy required for solvent recovery should be reduced as much as possible. The solvent to DW ratio is an important parameter in solvent recovery. To reduce the solvent to DW ratio, industrial scale processes usually have multiple extraction stages in a counter current flow setup to maintain a driving force (Berk, 2018).

Based on literature (Dávila et al., 2017a, 2017b; Fiori, 2010; Todd and Baroutian, 2017; Viganó et al., 2017), process setup was designed for both SE (Figure 1) and PLE (Figure 2). Both designs assume multiple extraction stages in counter current flow. Compared to the laboratory scale experiments the residence times were adjusted as well as, flow and equipment sizes. The total extraction time is assumed to be 60 minutes for all processes. By this set up, the solvent to DW ratio used in the laboratory scale experiments can be reduced, while the extraction yield, i.e. the amount of polyphenols extracted per kg DW, is maintained. As mentioned previously the solvent to DW ratio is an important parameter. The reduction of the solvent to DW ratio in the industrial scale processes is difficult to estimate precisely, therefore, based on Dávila et al., 2017a, 2017b; Fiori, 2010; Todd and Baroutian, 2017; Viganó et al., 2017 and expert knowledge, three feasible solvent to DW ratios were used in the TEA and LCA for each extraction method. The parameters of these scenarios are shown in Table 1. In all scenarios, the amount of polyphenols extracted is assumed to be equal to the laboratory scale experiments, though this will have to be validated by further experiments. The solvent to DW ratios and the solvent compositions were corrected for the amount of water in the pomace. The number in each scenario name refers to the solvent to DW ratio.

Table 1 Design parameters for industrial scale processes used in TEA and LCA.

	SE-10	SE-5	SE-2	PLE-50	PLE-25	PLE-10
Solvent to DW ratio (kg/kg DW)	10	5	2	50	25	10
Extraction stages	2	2	5		2	
Residence time (min/stage)	30	30	12		30	
Polyphenols extracted (g GAE/kg DW)		47			79	
Temperature (°C)		50			80	
Pressure (bar)		1			100	
Composition solvent						
- Water		33.3%			37.5%	
- Acetone		66.7%			-	
- Ethanol		-			37.5%	
- CO_2		-			25.0%	

The designs of both extraction processes include grinding of pomace to increase contact with the solvent, multiple extraction stages, distillation for solvent separation and recovery, nano filtration to reduce liquid volume and finally spray drying for recovery of the polyphenols in powder form (Figure 1 and Figure 2). The solvent to DW ratio determines the concentration of polyphenols after extraction and distillation i.e. the higher the solvent use the lower the polyphenol concentration in the liquid. The extracted polyphenols after distillation are concentrated i.e. water is removed, by nano filtration to 25% DW and then to 95% DW by spray drying.

For SE, the solvent is recovered from the pomace by first pressing i.e. separating the majority of the solvent from the pomace and distilling the liquid fraction, while the pomace is sent to desolventizing (drying). The composition of the solvent in the recycle is 95% acetone and 5% water. For scenario SE-2, it is necessary to dry the pomace prior to extraction, because otherwise the required solvent composition cannot be obtained. This dryer is not shown in Figure 1, but is taken into account in the TEA and LCA.

For PLE, the solvent is recovered from the pomace by flashing the CO_2 and distilling the extract. The composition of the solvent in the recycle is assumed to be 90% ethanol and 10% water.

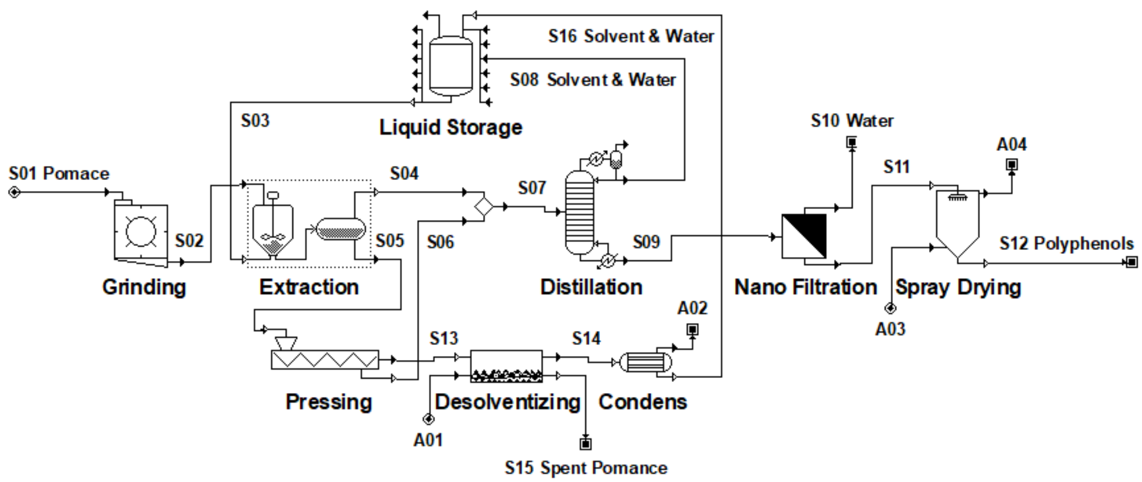

Figure 1 Process flow diagram for solvent extraction with acetone and water, for polyphenol recovery from grape pomace. Process includes input of wine pomace (S01), grinding, addition of solvents (S03) from liquid storage, extraction of polyphenols, distillation for solvent recovery and recycle (S08), nano filtration and spray drying for concentration and final recovery of polyphenols (S12), pressing and desolventizing of the wet pomace, condensation for additional recovery of solvent from the soaked pomace (S16).

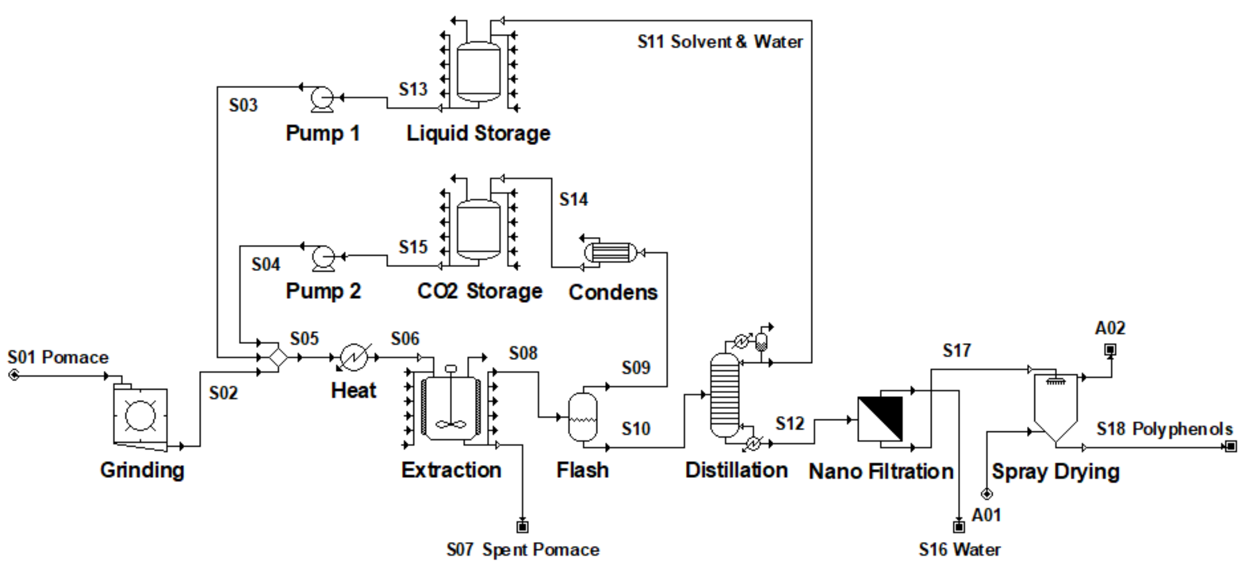

Figure 2 Process flow diagram for pressurized liquid extraction with ethanol, water, and supercritical CO_2 for the extraction of polyphenols from grape pomace. Process includes input of wine pomace (S01), grinding, pressurization by pump 1 and 2, addition of liquid solvents (S13) from liquid storage and supercritical CO_2 (S15) from CO_2 storage, extraction of polyphenols, flashing for CO2 recovery (S09) and distillation for liquid solvent recovery and recycle (S11), nano filtration and spray drying for concentration and final recovery of polyphenols (S18). Spent pomace (S07) is not desolventized.

2.4. Techno-economic assessment of industrial scale processes

TEA of the designed industrial scale processes was carried out in order to investigate the economic repercussions of installing a polyphenol extracting plant. The TEA includes Capital Expenditure (CapEx) and Operating Expenditure (OpEx). Assumptions and simplifications were made in order to fill in data gaps. The most important assumptions considering the TEA are reported in Table 2. Assumptions of economic parameters were based on Intelligen Inc, (2018); Maroulis and Saravacos, (2007); Peters et al., (2003); and Sinnott and Towler, (2009).

Based on the flow sizes of the designed processes, equipment were scaled. Purchased equipment cost and CapEx were based on the literature used for the process designs (Dávila et al., 2017b, 2017a; Fiori, 2010; Todd and Baroutian, 2017; Viganó et al., 2017) and the references mentioned above. Scaling rules were taken into account and the CapEx of the extraction vessels was corrected for the pressure.

In several wine growing areas wine pomaces and other residues are currently processed on industrial scale in centralized processing plants, so called distilleries. It is assumed that the polyphenol extraction will be performed in a setting similar to that of existing distilleries e.g. as in Italy and France, where 100% and 90% of wine pomace is sent to distilleries for treatment, respectively (Galanakis, 2017). The raw material costs for the polyphenol extraction are assumed to be negligible, since pomace is already part the current residue processing system.

The labor related costs were assumed to be the same for all scenarios and were based on: 2 shift positions, 4.8 operators per shift position, and an operator salary of k€ 30/y. Costs for supervision, direct salary overhead, and general plant overhead are added to the costs for operating labor.

Maintenance, including tax, insurance, rent, plant overhead, environmental charges, and royalties are assumed to be 10% of the CapEx per year. The financing costs are based on an amortization of the CapEx over 10 years with no interest (Peters et al., 2003; Sinnott and Towler, 2009).

Table 2 Parameters for the techno-economic assessment.

Production hours	8000	h/y
Red wine pomace	20	kton wet/y
	2500	kg wet/h
	36%	DW
Polyphenols extracted		
- with SE	340	ton GAE/y
- with PLE	572	ton GAE/y
Labor related costs	891	k€/y
Maintenance, etc.	10%	of CapEx/y
Financing costs	10%	of CapEx/y

The heat required in the dryer for SE-2 and in the spray dryer, as well as the energy required for solvent recycle is assumed to be two times the heat of evaporation of the concerning stream, based on the flow size of the scaled up version. For SE, this energy is distributed as follows: 90% for distillation (heat) and 10% for desolventizing (heat). For PLE, this energy is distributed as follows: 90% for distillation (heat), 5% for pumping (electricity), and 5% for heating prior to extraction. The electricity usage of the processing units is assumed to be: 10 kWh/ton input for grinding, 5 kWh/ton input for pressing, 5 kWh/ton permeate for nano filtration, 10 kWh/ton input for spray drying (atomization). Cooling water is used for cooling, for which the costs are assumed to be negligible. Despite all measures in the designed processes to recover the solvent, solvent loss is inevitable.

Therefore, for all scenarios, a solvent loss of 2% of the solvent in the recycle is assumed. Prices, CO_2-equivalents, and heat of evaporation of relevant utilities and solvents are given in Table 3.

Table 3 Parameters for utilities and solvents

		Price €/kWh	GWP CO_2-eq/kWh
Electricity		0.10	0.43
Heat		0.04	0.37
Cooling		0.00	0.56
	ΔH vap kJ/kg	Price €/kg	GWP CO_2-eq/kg
Water	2260	0.00	0.0002
Ethanol	841	0.80	1.34
Acetone	539	1.20	2.87
CO_2	380	0.50	0.85

2.5. Life cycle assessment of industrial scale processes

Following the TEA, an accounting LCA was performed on the newly designed industrial systems as modelled by the TEA. The functional unit is the production of 1 kg of polyphenols expressed as 1 kg GAE. The assessment is a "gate-to-gate" LCA and includes all actions carried out in order to obtain polyphenols from red wine pomace. This includes all steps from when the pomace enters the production system to the product, the polyphenols, leaving the production facility, e.g. all processing steps, such as grinding, drying, adding solvents, filtering, distillation and more (Figure 1 and Figure 2). The assessment does not include the end of life of the polyphenols or any transport throughout the life cycle, since this is deemed equal for all processing methods. Also, any potential burden of the raw material, the red wine pomace, is not accounted for, since the wine pomace is waste from wine production. Likewise, no credits are assigned for the production of polyphenols potentially replacing similar products in the market. The LCA includes all 18 impact categories in ReCiPe 2016 Midpoint (H) methodology (Huijbregts et al., 2017). The geographical location of the polyphenol plant is Italy.

2.6. Development of weighting for Multi-Criteria Decision Analysis

In order to incorporate the various environmental, as well as the economic criteria derived from the previous assessments, the Technique for Order of Preference by Similarity to Ideal Solution (TOPSIS) method of MCDA (Hwang and Yoon, 1981) is used. This is chosen due to its previous application in the context of LCA and because it is one of the most widely applied compensatory methods of MCDA when cardinal indicators are available for all alternatives (Kalbar et al., 2012b; Kalbar and Das, 2020).

All midpoint indicators from LCA and production prices of the various polyphenol production methods from the TEA (Table S3) are used as criteria in the application of TOPSIS.

When applying TOPSIS, there is an inherent application of weighting, even in its default mode, equal weights are applied (Pizzol et al., 2017). This presents a problem because the selection of the ideal alternative is directly related to weighting, which is further discussed in section 4.1.1. In this case, following the ArgCW-LCA method (Sohn et al., 2020), normalization factors (NF) (PRé, 2019) per impact category (i) are used to derive a relative importance factor (RIF), relating the average value, amongst all of the alternative extraction methods, of each of the midpoint impacts (MI) to the average European's annual environmental impact such that $RIF_i = \overline{MI_i} \,/\, NF_i$ represents the relationship between environmental and other criteria (Equation 1). For example, for calculating the RIF_{GW}, if the average GW impact amongst all assessed technologies ($\overline{MI_{GW}}$), were 80 kg CO_2 eq., then because the NF_{GW} for GW is 7990 kg CO_2 eq., the RIF_{GW} will be approximately equal to 0.01. In this case, production cost is then normalized such that production cost is allocated the desired weight and the sum of all weights is equal to 1000. The resultant weighting is then displayed in tabular form to promote full transparency in the assessment (Table S4, and Table S5).

3. Results and Discussion

3.1. Carbon foot-printing of laboratory scale experiments

The CFP analysis clearly shows that if laboratory conditions are maintained at industrial scale, then the acetone based solvent extraction method outperforms all other scenarios by a large margin, in terms of global warming potential (GWP), Figure 3. This is largely due to the amounts of solvent used in each scenario, which are lowest for the Lab-SE-11 scenario. The large amount of solvent used in the continuous setup for all Lab-PLE scenarios results in a very high heating demand in, for example, heating during polyphenol extraction, and heating during distillation to recover the solvents.

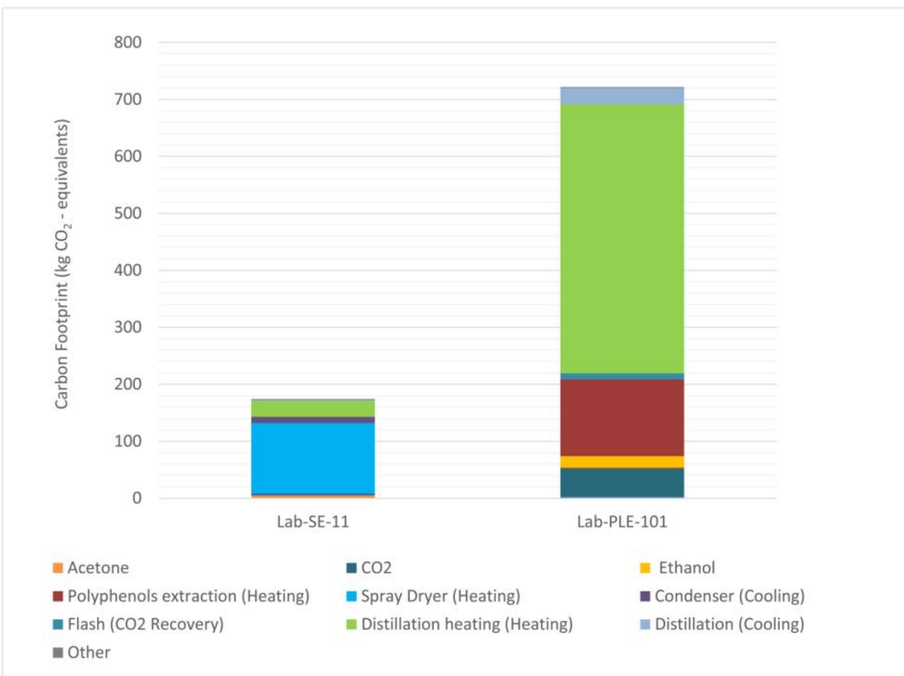

Figure 3 Global warming potential results per kg GAE of polyphenol extraction scenarios at laboratory scale. SE is solvent extraction, while PLE is pressurized liquid extraction. The number at the end of each scenario indicates the solvent to DW ratio for the extraction process.

From the CFP, the importance of keeping the solvent ratio as low as possible is evident. This has a trickledown effect on the energy demand of the whole system. The results can be used in the early design phase, in order to avoid excessive environmental burden later on. By identifying hot spots early on, it is possible to envision adjustments to the production setup, so that the identified hot spots are addressed. Measures, such as increasing the time of contact between solvent and pomace were identified after the CFP. Systems with multiple extraction stages and lower solvent to DW ratios were considered in the TEA.

3.2. Techno-economic assessment of industrial scale processes

The estimated CapEx for the different scenarios are: M€ 6.3 for SE-10, M€ 4.6 for SE-5, M€ 4.5 for SE-2, M€ 25.9 for PLE-50, M€ 16.6 for PLE-25, and M€ 9.8 for PLE-10. For the assessed solvent to DW ratios, the estimated CapEx are significantly higher for PLE compared to SE. Higher solvent ratios require larger equipment and a higher pressure results in more expensive equipment. Due to higher required solvent to DW ratios, the costs related to solvent recovery (i.e. electricity and heat) and solvent supplement are also higher for PLE compared to SE. On the other hand, PLE has a higher extraction yield compared to SE. By looking at processing costs expressed in €/kg GAE (Figure 4), it is clear that the higher extraction yield for PLE does not compensate the higher costs. Only labor related costs are lower for PLE. Scenario SE-2, which has the advantage of a low solvent to DW ratio, has the lowest processing costs. However, because of the required drying step and the low solvent to DW ratio, the assumed extraction yield was considered to be uncertain. As a result, the most feasible options, from a techno-economic perspective, are SE-5 and PLE-10. In the technically feasible range of solvent ratios, SE performs techno-economically better compared to PLE. Details on estimated CapEx, solvent loss, and utility usage for all assessed scenarios is shown in Table S2 of the SI.

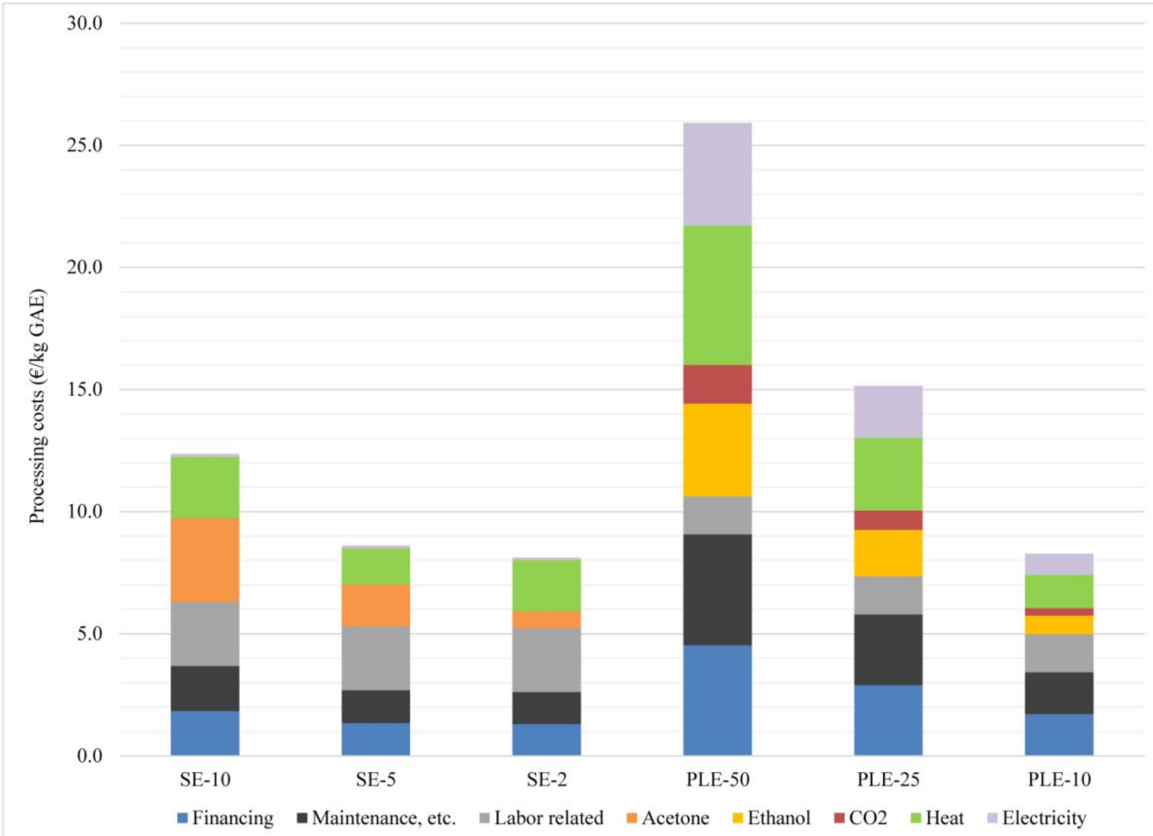

Figure 4 TEA results of polyphenol extraction at industrial scale. SE is solvent extraction, while PLE is pressurized liquid extraction. The number at the end of each scenario indicates the solvent to DW ratio for the extraction process.

3.3. Life cycle assessment of industrial scale design

The LCA of optimized operational conditions showed that if seeking to alleviate GWP it would be preferable to choose SE-2, that is to say, a solvent extraction using acetone with a solvent ratio of 2, Figure 5. However, as mentioned previously, the extraction yield of SE-2 was considered to be uncertain and therefore SE-2 was not considered to be a competitive option. Moreover, PLE-50 and PLE-25 perform far worse than the other options in terms of GWP and all other impact categories (Figure S2, SI), so these are also not deemed competitive options.

From Figure 5 it is possible to see the effect of the optimization performed via process design. The hotspot analysis still point towards solvent quantities as a key parameter for environmental outcomes, e.g. energy used for cooling and heating for distillation dominate the CO_2 burden, and energy for compressing the system. However, through process optimization it is possible to drastically reduce some impacts that were large in the laboratory scale CFP, as for example the impact from the spray dryer for the SE options, by adding a concentration (filtration) step before the drying. On the other hand, it is possible to see that adding a drying step for the pomace in option SE-2, does not pay off in comparison to not drying in SE-5, as the dryer plus distillation heating and cooling, are on the same range of impact as just distillation heating and cooling in SE-5. The overall GWP is lower for all options due to the reduction in solvent use and addition of extraction steps.

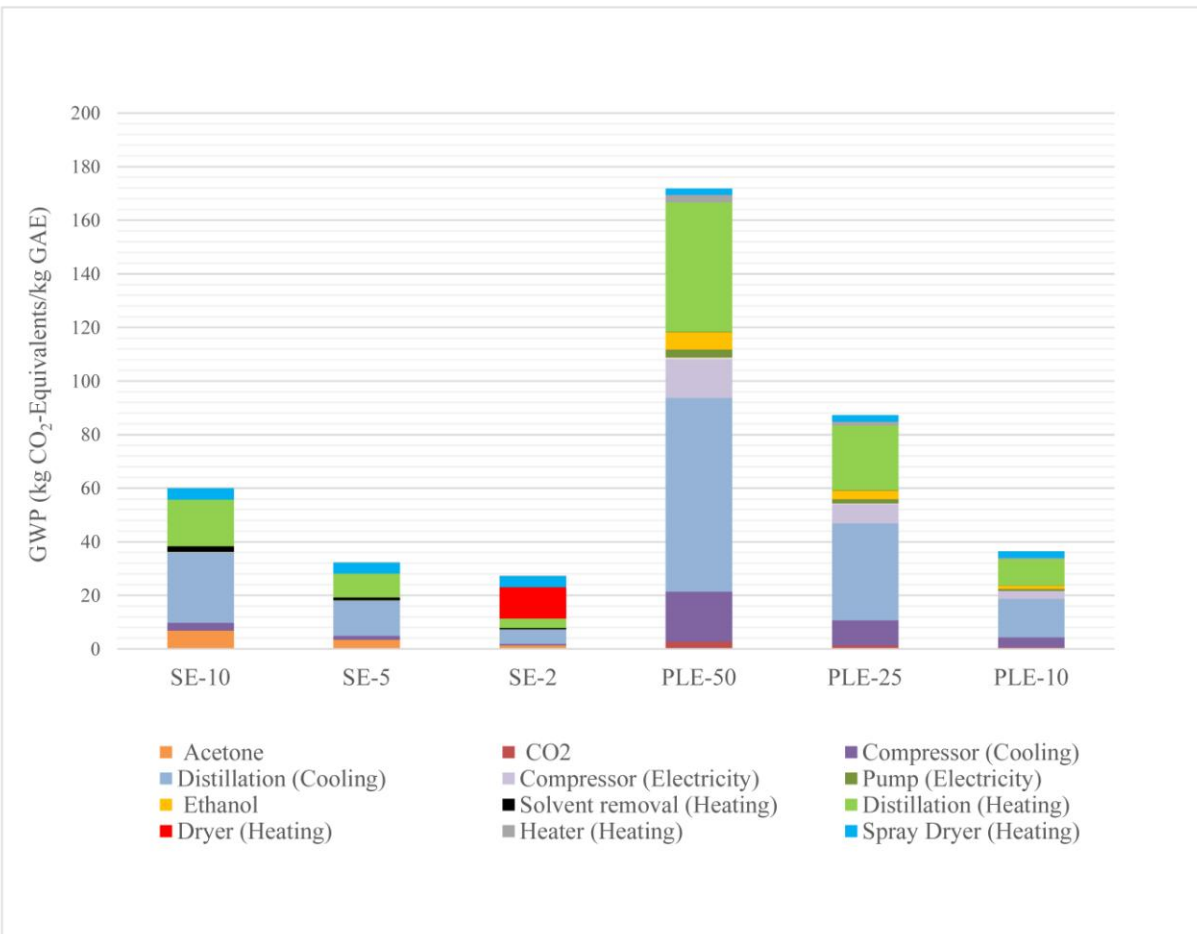

Figure 5 Global warming potential for scenarios tested in kg of CO2-equivalents. Contribution per processing step, cut-

off 1% of overall impact. SE is solvent extraction, while PLE is pressurized liquid extraction. The number at the end of each scenario indicates the solvent to DW ratio for the extraction process.

Results of the TEA show the importance of the solvent to DW ratio for the feasibility of extraction processes. High use of solvent leads to high operational costs and increased demand for electricity and heat, which affect the results of both TEA and LCA. On the other hand, higher yields allow more leeway for higher energy consumption, though not always fully compensating for all impacts. A lower solvent to DW ratio results in lower costs for solvent recovery, lower solvent loss, and lower CapEx. These results are mirrored in the LCA, where results benefit from lower solvent use, while impacts are increased due to the extra heating demand from large solvent volumes. In this regard though, it was clear in the LCA that solvent use, especially if the solvent is acetone, comes with higher impacts than electricity or heat use. This is easily illustrated when looking at the CO_2-Equivalents per 1 kg of acetone compared to 1 kg of ethanol or 1 kWh of electricity, as shown in (Table 3). From Table 3 it is possible to visualize that, in terms of the overall LCA assessment, added acetone or ethanol weigh more than added heat or electricity, with acetone being two times more burdensome than ethanol. Nevertheless, the use of solvent in the PLE options is high enough that even though ethanol is less burdensome the total impact outweighs the acetone use in the SE options.

In this regard, it is also worth mentioning that the ethanol used for this assessment is of petrochemical origin. However, since the waste being treated is wine pomace, it is quite possible that a biorefinery treating this waste would also produce bioethanol. This is true for distilleries placed in Italy and France, which currently treat wine pomace in order to produce bioethanol, bioenergy and food additives, among others (Lempereur and Penavayre, 2014). Bio-sourced ethanol will incur different environmental impacts, which were not investigated in this study.

Furthermore, the TEA in this study considers the processing costs including the financing costs. The market price of the product, the extracted polyphenols, and the market volume are yet to be explored. Once a market price or price range is known, then CapEx and processing costs can be compared to the benefits, and profitability indicators, such as net present value and internal rate of return. A larger investment for more complex technology (PLE instead of SE) might be justified if the benefits are significantly larger e.g. a higher yield for PLE than in the present study.

The most competitive options based on all midpoint impacts (Fig S2) and TEA; SE-10, SE-5 and PLE-10, were analyzed further to see if there is burden shifting between environmental indicators and to derive single scores for the options.

3.4. Single score results

After applying RIF, weighting strings can be derived for the application of TOPSIS with a range of importance given to economic impact from 0-1000, of a sum of 1000 available points distributed in the weighting profile between economic weight and environmental weight (Table S4). The relative importance amongst environmental impacts can also be shown in a single string to improve transparency of the weighting (Table 4).

Table 4 Weighting strings for RIF of environmental impacts used in this study, developed as described in section 2.6.

Impact category	RIF	Impact category	RIF
Fine particulate matter formation	12.14	Marine ecotoxicity	171.22
Fossil resource scarcity	256.66	Marine eutrophication	0.94
Freshwater ecotoxicity	197.95	Mineral resource scarcity	0.004
Freshwater eutrophication	90.31	Ozone formation, Human health	22.35
Global warming	54.5	Ozone formation, Terrestrial ecosystems	26.86
Human carcinogenic toxicity	60.66	Stratospheric ozone depletion	2.06
Human non-carcinogenic toxicity	4.21	Terrestrial acidification	19.86
Ionizing radiation	31.02	Terrestrial ecotoxicity	39.87
Land use	0.6	Water consumption	8.79

This is also done for equal weights (EW) amongst environmental impacts and the same range of importance of economic impact (Table S5). Applying these weightings to the criteria derived from LCA and TEA using TOPSIS, it is possible to provide decision support in the form of a single score indicator of idealness of the various technological alternatives (Figure 6 A and B). Furthermore, based on the results of the application of TOPSIS, a preference ranking can be made, with PLE-25 ranked fourth, SE-10 ranked third, and either PLE-10 or SE-5 ranked first and second. The ranking for first and second is based on the weight given to economics in the decision making process.

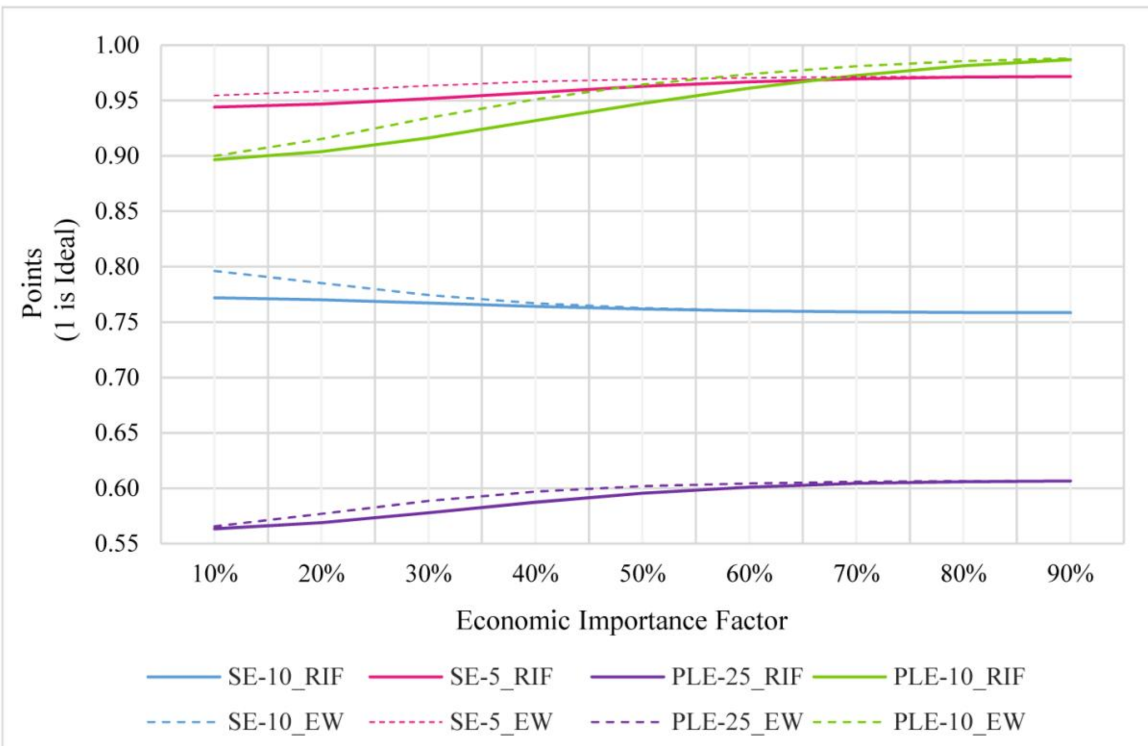

Figure 6 TOPSIS derived single score indicator of idealness (most ideal=1) for both Relative Importance Factor (RIF) derived environmental weighting and Equal Weights (EW) environmental weighting amongst a range of weights given to economic performance. SE is solvent extraction, while PLE is pressurized liquid extraction. The number in each scenario indicates the solvent to DW ratio for the extraction process.

Based on the application of TOPSIS, it can be easily concluded that the PLE-10 and SE-5 methods outperform all other alternative extraction methods. While PLE-10 is the best economic performer, SE-5 proved to be the best environmental performer. This results in a shift in decision support depending on the weight given to economic factors. In addition, SE-10 consistently performs better than PLE-25 both environmentally and economically. This results in a preference of SE-10 over PLE-25 regardless of weight given to economics.

As can be seen in Table 4 and Table 5, there is significant range in the importance of specific environmental impacts in RIF for the assessed methods. For example, some impacts such as human non-carcinogenic toxicity, marine eutrophication, and land use are insignificant in relative importance, and mineral resource scarcity is almost entirely irrelevant. On the other hand, fossil resource scarcity and freshwater ecotoxicity make up nearly half of weighting applied to environmental impacts due to the scale of their impact compared to the other environmental criteria relative to the average European's environmental impact.

One other element of note is the difference of decision support between EW and RIF in terms of the importance given to economic impact when PLE-10 is preferred over SE-5. When applying the RIF, this switch in preference occurs at appx. 65% weight to economic factors while for EW, the switch occurs at 55%. This is primarily due to the effective removal of environmental impact categories where the two alternatives are relatively equal that were compensating for other impact categories where the technologies were less equal in terms of performance. This occurs through the application of the ArgCW-LCA RIF weighting (Table 5) because some impact categories do not present much relevance to the decision context. This can be because there is either very little variation of the particular impact category amongst the assessed alternatives or because the given impact is smaller relative to status quo per capita emissions in relation to the other impacts of the assessed system.

Table 5 Relative weight (RW) of environmental impacts between RIF and EW weighting ($RW = W_{RIF}/W_{EW}$)

Impact category	RW	Impact category	RW
Fine particulate matter formation	21.85%	Marine ecotoxicity	308.19%

Fossil resource scarcity	461.99%	Marine eutrophication	1.69%
Freshwater ecotoxicity	356.31%	Mineral resource scarcity	0.01%
Freshwater eutrophication	162.56%	Ozone formation, Human health	40.23%
Global warming	98.10%	Ozone formation, Terrestrial ecosystems	48.35%
Human carcinogenic toxicity	109.18%	Stratospheric ozone depletion	3.70%
Human non-carcinogenic toxicity	7.57%	Terrestrial acidification	35.76%
Ionizing radiation	55.84%	Terrestrial ecotoxicity	71.77%
Land use	1.07%	Water consumption	15.83%

Another important element in interpreting the results from RIF weighting is understanding that there is a level of uncertainty in the normalization factors used to derive the RIF, and that the decision to use current emissions as a reference point, i.e. by using a European's environmental impact as NF, does not necessarily have a relationship to the severity or consequences of environmental impacts. However, it does provide an indication of the relative importance of an emission, or reduction thereof, to the status quo. If absolute sustainability related factors were available for all relevant impact categories, the application of these instead of normalization factors would be preferable, as they would provide a stronger link to environmental impact. Ideally, this process would be completed relative to planetary boundaries (Steffen *et al.*, 2015) using an absolute relationship to impacts from LCA (Bjørn *et al.*, 2015). However, this cannot be done because this absolute relationship is not yet well enough understood/developed, nor has it been developed to include all impact categories covered in LCA.

An alternative to either of these methods would be to derive a RIF weighting from endpoints using e.g. monetization. While this might seem appealing, as there is a stronger connection with environmental damages when using endpoint indicators in LCA, the challenge comes in determining the relative importance of the different damage categories. This relative importance is purely subjective, and as such a specific cultural perspective would be applied to the derivation of the weighting profile. While this could be carried out in a scientific fashion to be representative of a decision maker group, the results would already contain some bias toward certain impacts introduced in the endpoint calculation (Kalbar et al., 2016; Sohn et al., 2017). This would make the results more challenging to interpret and potentially lead to decision support that in the end does not reflect the true preferences of the decision maker. And, though midpoint impacts are not devoid of subjectivity, utilizing RIFs based on midpoint impacts effectively reduces the layers of interpretation applied in the interpretation phase of the impact assessment relative to endpoint derived single scores. Thus, making the elements driving decision support easier to track and understand.

4. Conclusions

Out of the solvent to DW ratio ranges of the TEA-LCA, SE options have potential to perform better than PLE. Despite higher yields for PLE, higher economic and environmental burdens outweigh the benefit of higher yield for this option. The same is highlighted by the single score indicator which concluded the potential performance is better when utilizing SE-5 than PLE-10, though a shift in preference is seen for higher economic weight. The addition of a transparent and reproducible decision assessment process aided in the understanding of the holistic impacts of the alternatives i.e. the introduction of RIF for deriving weighting.

Acknowledgements

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Supplementary Material to Insights from combining techno-economic and life cycle assessment a case study of polyphenol extraction from red wine pomace

Lab-PLE-101b is performed without liquid CO2 and instead there is 100% co-solvent composed of equal parts ethanol and water. While the third PLE option, Lab-PLE-586-oil, is divided into two extraction steps. One with 100% supercritical CO2 at 350 bar and 80°C for one hour, with a flow of CO2 of 30g per minute, leading to the production an oily phenolic extract. A second extraction step with the same EtOH:H2O:CO2 ratio as applied for Lab-PLE-101 is performed to obtain polyphenols as dry extract. The solvent flow for the second step is 8g per minute.

Table S 1 Operational parameters used as inventory for the preliminary LCA, for four options of polyphenol extraction.

Scenario Name	**Lab-SE-11**	**Lab-PLE-101**	**Lab-PLE-101b**	**Lab-PLE-583-oil**
Yield (g polyphenol/kg DW)	47	48	44	49
Solvents (% per mass)				
- Water	39%	39%	51%	39%**
- Ethanol		37%	49%	37%**
- Acetone	61%			
- CO_2		25%		100%*, 25%**
Solvent to DW ratio	11	101	101	583
Stages (no.)	1	1	1	2
Total extraction time (min)	120	60	60	90
Temperature (°C)	50	80	80	80
Pressure (bar)	1	100	100	350*, 100**

*first stage
** second stage

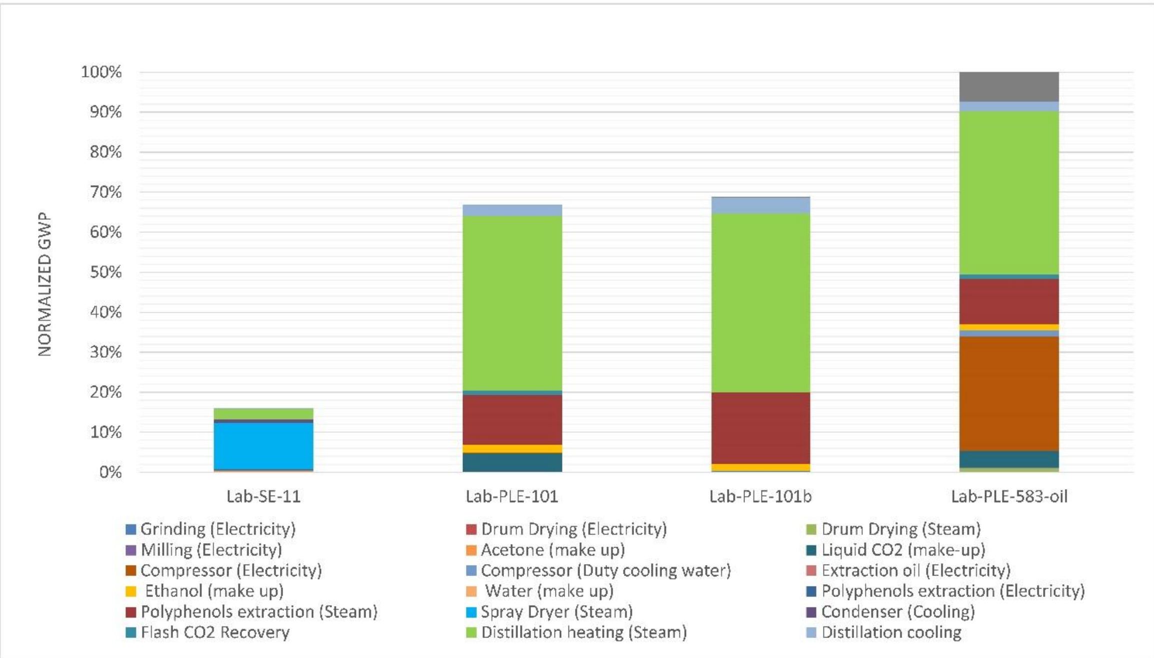


Figure S 1 Laboratory scale LCA of four extraction methods. Global warming potential is shown relative to worse performing scenario (Lab-PLE-583-oil)

Table S 2 Estimated CapEx, solvent loss, and utility usage for all scenarios assessed.

		Solvent Extraction Acetone & Water			Press. Liquid Extraction Ethanol, Water & SCCO2		
Solvent ratio	kg/kg DW	10	5	2	50	25	10
Pomace + Solvent	ton/h	10	5.4	2.7	46	24	10
Extraction vessel volume (of a single vessel)	m3	4	2	1	17	9	4
Fixed capital (CapEx)	M€	6.3	4.6	4.5	25.9	16.6	9.8
- Drying	M€			1.1			
- Grinding, Extraction, Solvent recovery	M€	3.3	2.3	1.5	16.8	11.2	6.7
- Nano filtration	M€	1.1	0.4	0.0	7.3	3.5	1.3
- Spray drying	M€	1.8	1.8	1.8	1.8	1.8	1.8
Solvent loss							
- Acetone	kg/h	121	61	24			
- Ethanol	kg/h				339	170	68
- CO2	kg/h				226	113	45
Utilities							
Electr. - Grinder	kW	25	25	25	25	25	25
Heat - Dryer	kW			1333			
Heat - Distillation	kW	1995	997	399	9266	4633	1853
Cool - Distillation	kW	1995	997	399	9266	4633	1853
Electr. - Pressing (SE)	kW	13	13	13			
Heat - Desolventizing (SE)	kW	222	111	44			
Cool - Condensor (SE)	kW	222	111	44			
Electr. - Compressor (PLE)	kW				2388	1194	478
Cool - Condensor (PLE)	kW				2388	1194	478
Electr. - Pump (PLE)	kW				515	257	103
Heat - Heater (PLE)	kW				515	257	103
Electr. - Nano Filtration	kW	11	4	0	73	35	13
Electr.- Spray Dryer	kW	5	5	5	5	5	5
Heat - Spray Dryer	kW	463	463	463	463	463	463
Heat - Total	kW	2679	1571	2239	10243	5353	2419
Cool - Total	kW	2216	1108	443	11654	5827	2331
Electicity - Total	kW	53	47	43	3006	1517	623

Table S 3 Midpoint results ReCiPe 2016 (H) for all scenarios assessed.

	Solvent Extraction			Pressurized Liquid Extraction			
	Acetone & Water			Ethanol, Water & $SCCO_2$			
	340 ton GA/y			572 ton GA/y			
	Solvent ratio: 10	Solvent ratio: 5	Solvent ratio: 2	Solvent ratio: 50	Solvent ratio: 25	Solvent ratio: 10	
Impact Category	**S-Acn-10**	**S-Acn-5**	**S-Acn-2**	**PLE-EthOH-50**	**PLE-EthOH-25**	**PLE-EthOH-10**	Unit
Fine particulate matter formation	0.0418	0.0226	0.0193	0.1234	0.0627	0.03	kg PM2.5 eq
Fossil resource scarcity	21.1886	11.2888	8.9655	57.0390	28.9122	12.04	kg oil eq
Freshwater ecotoxicity	0.5986	0.3092	0.1772	2.1480	1.0793	0.44	kg 1,4-DCB
Freshwater eutrophication	0.0065	0.0035	0.0026	0.0254	0.0128	0.01	kg P eq
Global warming	60.0484	32.2763	27.2810	171.8774	87.2234	36.44	kg CO2 eq
Human carcinogenic toxicity	0.8053	0.4235	0.2890	2.5925	1.3076	0.54	kg 1,4-DCB
Human non-carcinogenic toxicity	15.5926	8.0736	4.7722	56.7402	28.5185	11.59	kg 1,4-DCB
Ionizing radiation	1.3004	0.7365	0.7004	6.6696	3.3797	1.41	kBq Co-60 eq
Land use	0.3407	0.1971	0.2229	1.5936	0.8113	0.34	m2a crop eq
Marine ecotoxicity	0.9062	0.4702	0.2855	3.1791	1.5988	0.65	kg 1,4-DCB
Marine eutrophication	0.0004	0.0002	0.0002	0.0019	0.0010	0.00	kg N eq
Mineral resource scarcity	0.0552	0.0282	0.0149	0.1983	0.0995	0.04	kg Cu eq
Ozone formation, Human health	0.0650	0.0350	0.0294	0.1795	0.0912	0.04	kg NOx eq
Ozone formation, Terrestrial ecosystems	0.0676	0.0364	0.0303	0.1861	0.0945	0.04	kg NOx eq
Stratospheric ozone depletion	0.0000	0.0000	0.0000	0.0001	0.0000	0.00	kg CFC11 eq
Terrestrial acidification	0.1109	0.0605	0.0543	0.3194	0.1625	0.07	kg SO2 eq
Terrestrial ecotoxicity	75.0523	40.4591	35.6819	248.2986	125.8477	52.39	kg 1,4-DCB
Water consumption	0.2918	0.1530	0.0865	0.9974	0.5020	0.20	m3
product production cost	12.4	8.6	8.1	25.9	15.1	8.3	€

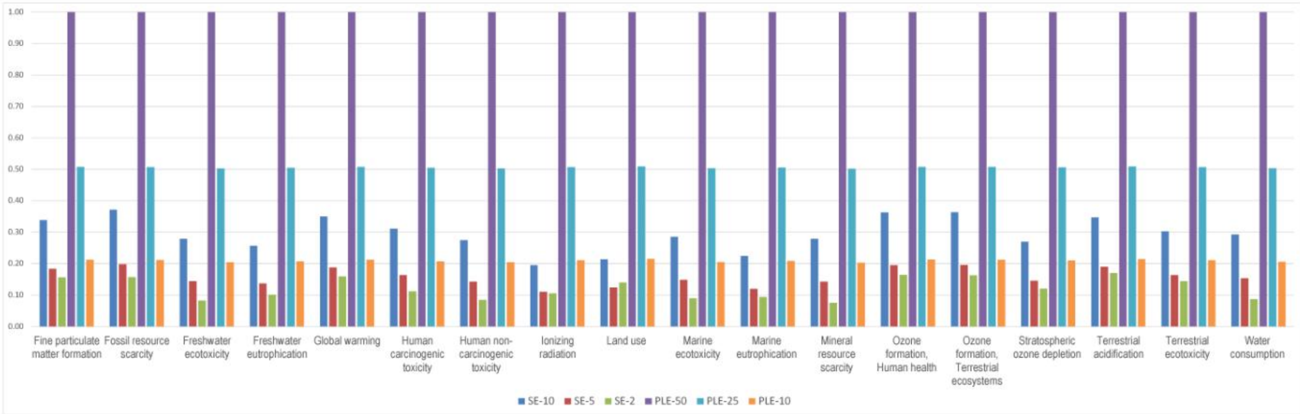


Figure S 2 Bar graph of midpoint results of assessed scenarios, internally normalized to worse performing scenario PLE-50.

Table S 4 Weighting strings including RIF for environmental impacts and a range of importance of economics

product production cost	Fine particulate matter formation	Fossil resource scarcity	Freshwater ecotoxicity	Freshwater eutrophication	Global warming	Human carcinogenic toxicity	Human non-carcinogenic toxicity	Ionizing radiation	Land use	Marine ecotoxicity	Marine eutrophication	Mineral resource scarcity	Ozone formation, Human health	Ozone formation, Terrestrial ecosystems	Stratospheric ozone depletion	Terrestrial acidification	Terrestrial ecotoxicity	Water consumptio n
0	12.14	256.66	197.95	90.31	54.50	60.66	4.21	31.02	0.60	171.22	0.94	0.004	22.35	26.86	2.06	19.86	39.87	8.79
100	10.93	231.00	178.15	81.28	49.05	54.59	3.78	27.92	0.54	154.09	0.84	0.003	20.12	24.17	1.85	17.88	35.88	7.92
200	9.71	205.33	158.36	72.25	43.60	48.53	3.36	24.82	0.48	136.97	0.75	0.003	17.88	21.49	1.65	15.89	31.90	7.04
300	8.50	179.66	138.56	63.22	38.15	42.46	2.94	21.72	0.42	119.85	0.66	0.003	15.65	18.80	1.44	13.91	27.91	6.16
400	7.28	154.00	118.77	54.19	32.70	36.39	2.52	18.61	0.36	102.73	0.56	0.002	13.41	16.12	1.23	11.92	23.92	5.28
500	6.07	128.33	98.97	45.16	27.25	30.33	2.10	15.51	0.30	85.61	0.47	0.002	11.18	13.43	1.03	9.93	19.94	4.40
600	4.86	102.66	79.18	36.12	21.80	24.26	1.68	12.41	0.24	68.49	0.38	0.002	8.94	10.74	0.82	7.95	15.95	3.52
700	3.64	77.00	59.38	27.09	16.35	18.20	1.26	9.31	0.18	51.36	0.28	0.001	6.71	8.06	0.62	5.96	11.96	2.64
800	2.43	51.33	39.59	18.06	10.90	12.13	0.84	6.20	0.12	34.24	0.19	0.001	4.47	5.37	0.41	3.97	7.97	1.76
900	1.21	25.67	19.79	9.03	5.45	6.07	0.42	3.10	0.06	17.12	0.09	0.000	2.24	2.69	0.21	1.99	3.99	0.88
1000	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.000	0.00	0.00	0.00	0.00	0.00	0.00

Table S 5 Weighting strings including equal weighting for environmental impacts and a range of importance of economics

product production cost	Fine particulate matter formation	Fossil resource scarcity	Freshwater ecotoxicity	Freshwater eutrophication	Global warming	Human carcinogenic toxicity	Human non-carcinogenic toxicity	Ionizing radiation	Land use	Marine ecotoxicity	Marine eutrophication	Mineral resource scarcity	Ozone formation, Human health	Ozone formation, Terrestrial ecosystems	Stratospheric ozone depletion	Terrestrial acidification	Terrestrial ecotoxicity	Water consumption
0	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6
100	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0
200	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4
300	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9
400	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3
500	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8
600	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2
700	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7
800	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1
900	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6
1000	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0

***conflict of Interest Statement**

Declaration of interests

☒ 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.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Highlights

Polyphenol extraction laboratory methods were optimized through process design

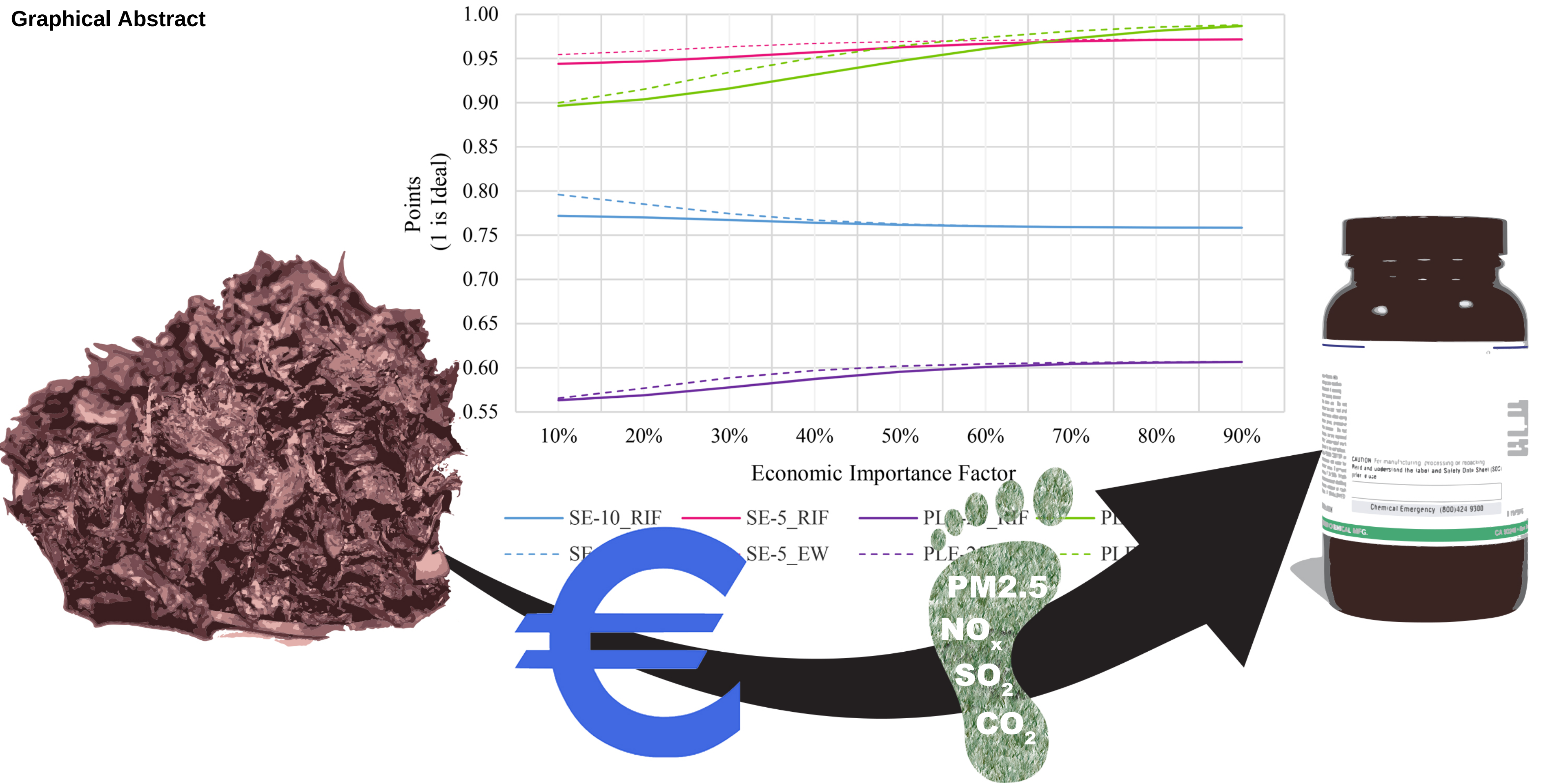
TEA and LCA were used to assess the designed industrial scale extraction

TOPSIS-based MCDA was used to choose the best polyphenol extraction option

Within feasible solvent ratios, SE exhibits better eco/enviro performance than PLE

Either SE or PLE is preferred depending on level of importance assigned to economics

Graphical Abstract



Credit Author Statement

Giovanna Croxatto Vega: Conceptualization, formal analysis, investigation, methodology, project administration, writing original draft, writing- review & editing.

Joshua Sohn: Conceptualization, formal analysis, Investigation, methodology, writing original draft, writing- review & editing.

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PAPER III

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Assessing New Biotechnologies by Combining TEA and TM-LCA for an Efficient Use of Biomass Resources

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Abstract: An efficient use of biomass resources is a key element of the bioeconomy. Ideally, options leading to the highest environmental and economic gains can be singled out for any given region. In this study, to achieve this goal of singling out an ideal technology for a given region, biotechnologies are assessed by a combination of techno-economic assessment (TEA) and territorial metabolism life cycle assessment (TM-LCA). Three technology variations for anaerobic digestion (AD) were assessed at two different scales (200 kW and 1 MW) and for two different regions. First, sustainable feedstock availability for two European regions was quantified. Then, the environmental impact and economic potential of each technology when scaled up to the regional level, considering all of the region's unique sustainably available feedstock, was investigated. Multiple criteria decision analysis and internalized damage monetization were used to generate single scores for the assessments. Preference for the technology scenario producing the most energy was shown for all regions and scales, while producing bioplastic was less preferable since the value of the produced bioplastic plastic was not great enough to offset the resultant reduction in energy production. Assessing alternatives in a regional context provided valuable information about the influence of different types of feedstock on environmental performance.

Keywords: anaerobic digestion; polyhydroxyalkanoates; life cycle assessment; techno-economic assessment; territorial metabolism; regional assessment; wet oxidation; biogas; biomass valorization

1. Introduction

One of the goals of the European Union (EU) is to stimulate the creation of a competitive low carbon economy that is able to provide a reduction of 80%–95% greenhouse gas (GHG) in Europe by 2050 [1]. Energy production is an important sector where changes can be made in order to reach this ambitious target. Shifting from fossil-based energy to renewable sources of energy can lead to GHG reductions, provided the value chains for the renewable energy sources can lead to better overall environmental performance. A careful evaluation of new renewable energy pathways has previously been recommended [2] and various studies have shown wide ranges for GHG emissions of renewable energy systems [3,4]. Moreover, and particularly relevant to biomass based renewable energy, in some cases lower GHGs are not accompanied by lower emissions of other environmentally concerning emissions, such as those contributing to eutrophication, acidification, and human/ecosystem toxicity [5,6]. Within the various renewable energy sources, biomass is important as in 2015 it already supplied 10% of the global demand for primary energy consumption [7]. In Europe, demand for electricity biomass, heating, and transport was around 5010 PJ in 2012 and it is estimated to rise to 7437 PJ in 2020 in order to meet renewable energy targets in the EU. Thereby, it is important to consider additional renewable energy with holistic perspectives that can quantify the environmental performance of renewable energy from biomass resources. Life cycle assessment (LCA) is an internationally recognized, standardized tool with a mature methodology capable of assessing large systems and giving a complete assessment of environmental impacts [8]. As such, LCA has been used widely and is aligned with the sustainable development goals (SDGs) developed by the United Nations [9], which incorporate life cycle thinking into, for example, goal number 12 (sustainable production and consumption patterns) [10]. Under the umbrella of SDGs, decoupling economic growth from the unsustainable use of resources is of prime importance so that future generations may enjoy precious natural resources. Thus, measuring progress towards these goals is necessary from both an economic and environmental perspective, which makes the use of mixed assessments necessary.

Out of the estimated 7437 PJ demand for biomass energy in 2020, 887 PJ are expected to come from biogas [11]. Biogas production has increased significantly in the EU, from 92 PJ in 2000 to 654 PJ of primary energy in 2015, with a total of 17,400 installed biogas plants [7]. Anaerobic digestion (AD) is a versatile technology for many reasons, one being that it is possible to install decentralized plants near agricultural sources of feedstock. In terms of biomass resources, AD can utilize various types of organic waste aside from agricultural residues, including industrial wastes such as slaughterhouse wastes and residues from food production, sewage sludge and the organic fraction of municipal solid waste. The produced biogas can be valorized in several ways, such as for heat and electricity production in combined heat and power engines (CHP); injection into the natural gas grid after an upgrade to biomethane; or use in the transport sector. It is at least in part due to this versatility that AD can serve as a successful platform for the bioeconomy. In addition, the latest developments in biogas technology expand the platform beyond energy into materials production [12]. While some of the advances focus on optimizing energy extraction, such as wet explosion pretreatment aimed at unlocking the lignocellulosic fraction of waste [13], or adding a separate dark fermentation step before methanization so as to increase hydrogen content of the biogas [14], other innovation allows for the production of biopolymers via the modification of the AD process [15]. By isolating the volatile fatty acids (VFAs) produced during the AD process and feeding them to microbes in a multi-stage process, intracellular polymer, such as polyhydroxybutyrate (PHB) of the polyhydroxyalkanoates (PHAs) family of biodegradable polymers can be produced and later extracted from the bacteria. In this way, it is possible to turn biogas plants into chemical platforms, which can expand the acting field of AD to new utilization and valorization opportunities.

Needless to say, biogas relies on available biomass and by definition is constrained to these finite resources. Various studies have focused on mapping out the availability of biomass in Europe for the production of energy and biogas [16–21]. Though the quantified potentials vary widely due to methodological selections and database choice, it is generally acknowledged that the extraction of biomass must be done with care to avoid competition with food resources and unwanted market effects, such as increases on land and maize prices [22,23]. Still, Scarlat et al. [11] warns that even though domestic biomass supply in the EU is enough to satisfy the demand required to accomplish national renewable targets, as much as a quarter of the biomass demand may be sourced from third countries (outside of the EU) in 2020. Since this is due to market effects, it is imperative to take economics as well as environmental aspects into account so that the appropriate support systems are in place for the development of a sustainable renewable energies market and thereby a sustainable biogas sector. In this regard, it is important to determine if the emerging biogas innovations mentioned are environmentally sound and lead to environmental performance improvements in comparison to the status quo. As has been pointed out before, the prefix bio does not guarantee sustainability [24]. Biogas capacity already built in Europe is an important aspect when analyzing any additional capacity that may be built in an area, e.g., considering that 50% of the EU's biogas capacity is in Germany [7]. As has been pointed out by Bojesen [25] and colleagues, who estimated service areas for existing and future biogas plants in Denmark, the availability of feedstock in relation to plant location is an important aspect. An inadequate assessment of a plant's sourcing ability may lead to high operation costs from increased transport demand or inadequate sourcing of feedstock [25]. In turn, high transport distances may negate the environmental benefits brought about by biorefineries, as shown in Croxatto Vega et al. [26] which applied the territorial metabolism-LCA approach (TM-LCA) [27] and found distances of 50 km to be the upper limit.

This study performs a step-wise assessment starting from individual plant level and investigates the implementation potential of the PHB and AD-Booster technologies in two different plant scales. A techno-economic assessment (TEA) and LCA are carried out for this aim. The results from the TEA-LCA are used to structure implementation of the technologies at the regional level. The TEA relates the plant scale and processing capacity to capital expenditure (CapEx) and operational expenditure (OpEx) of the plant, and to the break-even prices of products. In the LCA, the environmental aspects of different technologies are quantified. The implementation of the two technologies is analyzed for two regions defined by the nomenclature of territorial units for statistics (NUTS) from Eurostat's definition of regions (NUTS2 regions): Bavaria, Germany and Veneto, Italy. We analyze the potential impacts of the two innovative technologies (PHB and AD-Booster) against the current level of biogas implementation for the regions. First, we use TEA to analyze the effect of scale on the economic potential considering relevant plant sizes. Concurrently, we provide a mass flow analysis for the regions to better understand the energetic potential of agricultural residues produced within the regions (i.e., both the residues already in use for biogas and not yet exploited) as well as the level of development of the biogas sector (i.e., installed capacity). Finally, we use the results from the TEA of each technology to perform a TM-LCA, which will be able to tell us the possible environmental improvements (or deterioration) potentials for the whole region, if all of the residues are processed with the new technologies. We place special attention on the repercussions for the farmer, especially from installation of large biogas plants, which can potentially monopolize biomass resources over a large area. Vice versa, we explore the possible needs and constraints for biogas developers in the two regions. In this way, we seek to explore new biotechnological implementation potentials from a stakeholder's perspective.

2. Method

2.1. Plant Level Assessment

The potential of implementing new AD technology was analyzed at two different scales. Data was collected from two biogas plants: a 1 MW installed electric capacity plant in Veneto, Italy and a smaller 200 kW plant in Bavaria, Germany, hereafter referred to as "the farms". Both plants operate on a mixture of cow manure, crop residues, and maize silage (Table 1). Both plants valorize biogas in CHP units, which produce heat as a waste product. Both plants utilize the co-produced heat in the plant's operation and additionally, in the Bavarian case, the surplus heat produced is utilized in the district heating network for a nearby village [28].

Table 1. Feedstock mix employed in the farms.

	200 kW		1 MW	
	% ww [1]	ton/day	% ww [1]	ton/day
Cow manure	57%	11.3	82%	131.4
Maize Silage	27%	5.5	14%	23.0
Grass silage	14%	2.7	3%	5.4
Grain	2%	0.4	0%	0.0

[1] Percent on a wet weight basis.

2.2. Technology Description

Three technology scenarios were assessed. Conventional AD was chosen as the baseline and two emerging treatment processes that can be added to existing AD were assessed for the comparison. All technology scenarios are modelled with a biogas leak of 3% of the produced biogas [22]. The technology set ups are: AD, AD + Booster, and AD + PHB.

2.2.1. Anaerobic Digestion

Conventional AD was modelled using SuperPro Designer, following the details received from the farms (Figure 1). The feedstock is grinded before it enters the anaerobic digester. The anaerobic digester produces biogas and digestate. The AD model was populated with the most common stoichiometric equations governing anaerobic digestion in [29]. Internal electricity consumption for the whole process was 7.5% of produced electricity based on data obtained from the farms operating biogas plants. A methane content of 50% for the biogas and an electrical efficiency of 38% for the CHP unit was used, based on the received data, yielding a 1.9 kWh/m^3 of biogas. Internal thermal energy usage was assumed to be 40%. The methane content, electrical efficiency, energy content of the biogas, and internal heat use was equal in all technology scenarios.

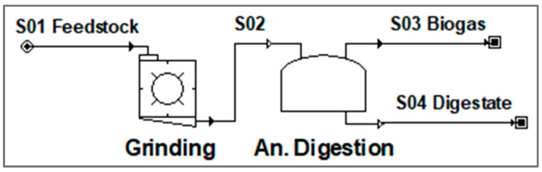

Figure 1. Simplified flow chart of anaerobic digestion.

2.2.2. AD + Booster

The AD + Booster technology consists of an extra tank where the wet explosion technology is applied under high heat and pressure conditions [13]. The AD + Booster scenario (Figure 2) was designed with information obtained from the technology developers [30]. In comparison to AD, the AD + Booster technology increases the conversion yield of cellulose to biogas from 52% to 88% and the conversion yield of hemicellulose to biomass from 75% to 98%. This scenario has an internal electricity consumption of 9.5% of produced electricity. On the other hand, the biogas yield is 12% to 16% higher than the AD scenario.

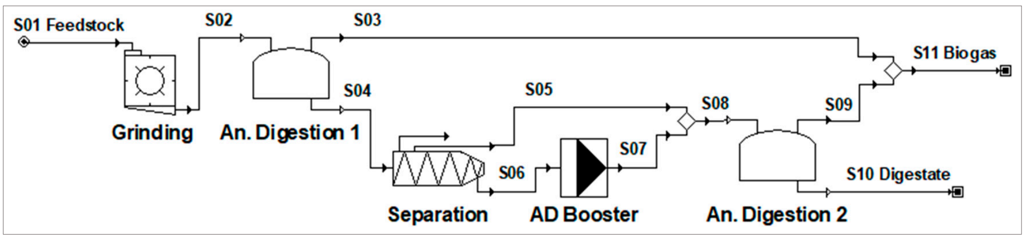

Figure 2. Simplified flow chart of the anaerobic digestion (AD) + Booster technology.

2.2.3. AD + PHB

In order to include a PHB producing section into an existing AD plant, a few extra pieces of equipment are necessary (Figure 3). AD is split into two tanks, the first is of short retention time and is where the VFA are produced and rerouted for PHB production. After this step, a screw press and a filtration unit separate solid from liquid. The solid fraction is fed to the AD tank where it continues the regular AD process, while the liquid fraction goes into a series of bio-oxidation units where selection and accumulation occurs via the feast and famine method [15]. The bio-oxidation equipment, in SuperPro Designer, was populated with stoichiometric equations obtained from the technology developers. Finally, PHB can be extracted using sodium hypochlorite and a final filtration step recovers a crude PHB. In comparison to AD, this scenario has an internal electricity consumption of 15% of produced electricity and a biogas yield from 24% to 30% lower than AD.

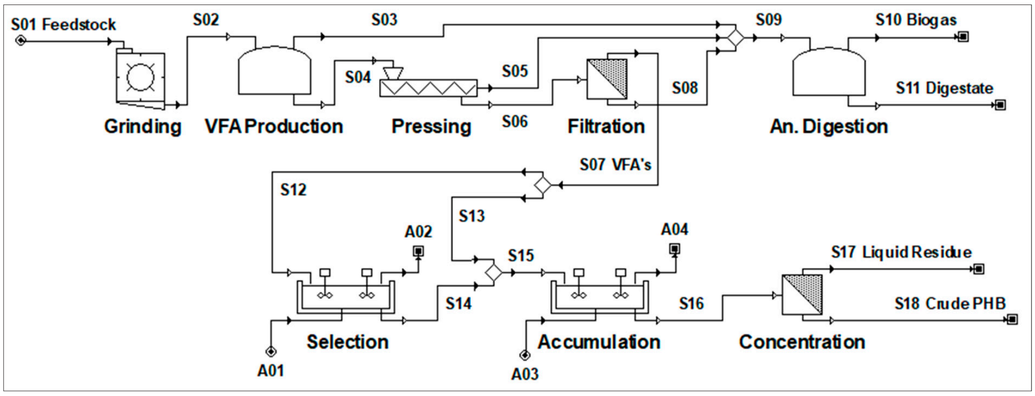

Figure 3. Simplified flow chart of the AD + polyhydroxybutyrate (PHB) technology.

2.3. *Regional Feedstock Availability*

2.3.1. Crops

Primary production amounts and land cover of individual crops was obtained from the Eurostat database (apro_cpnhr, [31]) for the two NUTS2 regions. As much as possible, the most recent data on production statistics was used, for the period of 2008–2018. For Veneto, data coverage on crops by Eurostat was very incomplete. Thus, it was preferable to use data from the National Italian Institute of Statistics (ISTAT) [32], where data is available for the whole period at the NUTS2 level. The production yield (production amounts divided by area of production) was then averaged over the period to derive an average production per year for the regions. Residue:crop ratios where then applied to the production yield to derive a total annual amount of residues for each crop. A list of the residue:crop ratios (Table 2), as well as grouping of Eurostat categories used are provided (Table A1, in Appendix A). For the most part, it was assumed that only residues are used for AD (or AD + innovation), with the exception of energy crops where the whole plant is assumed to enter digestion. Energy crops included are maize silage, green maize, and sorghum. Only crops which are most commonly considered for biogas operation were included in the study; we excluded horticultural crops that do not typically serve a purpose in AD.

Because wine grapes are an important crop in the Veneto region, they are also present, though to a smaller extent in the crop mix of Bavaria. Amounts of winery pomace were also taken into account as potential AD feedstock. Data on wine production was obtained from Eurostat (vit_t1, vit_an5, vit_an7, [33,34]) for both regions and the data for Veneto was checked against ISTAT data. The period for Eurostat wine data differed slightly and included the years 2001–2009, and 2015. The amount of pomace produced was estimated based on [35], which reports a 25% conversion rate from grapes to pomace.

After obtaining total crop residue amounts for the region, it is necessary to estimate a technical and sustainable potential for collection of the residues. The technical potential may potentially exclude the share of residues which is too difficult to collect, as well as the share that has known competing applications (Table 3). For example, this is the case for straw from cereal production, which is typically used for bedding and feed for cattle and other animals [36]. Sustainable potential collection, on the other hand, takes into consideration soil fertility. Residues may, for example, be left on agricultural fields to uphold the organic matter content of the soil and protect it from excessive erosion. Sustainable potential collection factors typically used in the literature vary from around 10% to 50% of the most common types of residues, i.e., excluding pomace and energy crop [37], and it has been shown that residue removals above 50% may negatively affect soil organic carbon storage [38]. Nearly all studies [11,16,19–21,36,39] that evaluate biomass potential for bioenergy purposes apply some sort of technical/sustainable collection factor, yet many of these studies do not report the actual values used or leave values out. We report all the values used in Table 2, since this is one of the most important determinants of potential for biomass utilization.

Table 2. Sustainable removal factor of various crops.

	Fraction of Total Residues	References
Cereal straw	0.3	[40]
Rice straw	0.5	[19,21]
Maize	0.5	[19,21]
Leguminous	0.1	[36,41]
Sugar neet	0.5	[36,41]
Rape	0.5	[19,21]
Sunflower	0.5	[19,21]
Soya	0.4	[17]
Oily	0.1	[36,41]
Industrial	0.4	[17]
Forage	0.1	[36,41]
Energy crop	0.9	[19,21]
Pomace	0.99	[42]

Table 3. Competing application factors for cereal straw.

	Feed [1]	Bedding [1]
Straw for bovine [2]	0.1	0.8
Straw for swine [2]		0.6
Straw for sheep [3]	0.025	0.2
Straw for goat [3]	0.025	0.2
Straw for poultry [4]	0.0125	0.1

[1] unit is ton per livestock unit*yr. [2] [40]. [3] Estimated value. Sheep and goat use a fourth of bovine. [4] Estimated value. Poultry uses a half of sheep and goat.

2.3.2. Manure

Animal production data was obtained from the Eurostat database (agr_r_animal) for bovine, swine, sheep, and goats for the period of 2008–2018. At the NUTS2 level, it is possible to obtain data for the number of animals in thousand heads. It is then necessary to estimate the amounts of manure excreted by the different types of animal, which varies also with their age (dairy cows, calves, sows, piglets, etc.). Values of manure production are calculated using the methodology detailed in [43] following the definitions for the various animals in [44]. The values are reported in Appendix A, in Table A2. Poultry production is not reported in the above-mentioned database, thus it was necessary to use the ef_lsk_main Eurostat database, which reports livestock units (LSU) for poultry for the years 2005, 2007, 2010, 2013, and 2016 at the appropriate regional level. This was the best available data for poultry at the NUTS2 level. LSU values were converted to poultry heads, following the methodology outlined in [43].

Similarly to crop residues, a technical potential was considered for animal manures. Here, for cattle, the potential collectable manure was estimated based on the type of housing and rearing. Since European regulation on organic production of agricultural products specifies that organic “livestock should have permanent access to open air areas” in most cases [45] and that there shall be a connection between land management by the use of manure, i.e., meaning that organic production must maintain the fertility of soil by applying cover crops, green manures or organic livestock manure, it was assumed that manure could only be collected in the harsh winter months (at most) from organic cattle farms [46]. The estimate for housing types was derived from the Farm Structure Survey (FSS) [47] carried out in 2010, since more recent FSS could not be located. The types of housing were assumed to stay proportionally equal to the values in 2010, though after taking into consideration the growth in the organic farming sector for cattle rearing. Data on the share of organic livestock was obtained from statistical data summarized by Eurostat at the national level [48]. For animals other than cattle, the share of organic production was disregarded since the share is very low (<1% of animals) [48]. Manure collection factors are given in Table A3 of Appendix A, for all animals and various types of housing.

2.3.3. Installed AD Capacity

Already installed AD capacity has to be considered when assessing additional potential implementation in the regions. Regional data on biogas installation was collected from various sources. In Veneto, a total of 220 biogas plants were in operation by 2018, of which 89% were considered agricultural plants, i.e., treating crop residues, energy crops and animal manures [49]. By contrast, 2566 plants were installed in Bavaria by 2019 [50], of which 93% were considered agricultural AD [7], while the rest were landfill gas and sewage gas. A breakdown of types of installed capacity (scale) was obtained from a census of installed plants [51] in 2011 in the Veneto region. It was assumed that installation continued in the same fashion through to 2018, with a preference for plants of capacity slightly lower than 1MW, due to an all-encompassing subsidy [52]. For Bavaria, data obtained was detailed down to city/rural district level, which made it possible to use average capacity to determine the scale breakdown of installed capacity. The types of capacity installed estimated for Veneto and Bavaria are shown in Table 4.

Table 4. Scale of installed biogas plants in Veneto and Bavaria.

Type of Capacity	Veneto 2018		Bavaria 2019	
(kWe)	n	%	n.	%
<100	23	12%	9	0%
101–500	43	22%	1352	56%
501–1000	118	60%	1010	42%
>1000	3	1%	11	0%
Biogas in broiler	0	0%	0	0%
No data	10	5%	15	1%

2.3.4. Regional Energetic Potential

The methane potential of various feedstocks (Table A4, Appendix A) was used to derive the quantities of feedstock currently being processed by the already installed AD capacity in each region. Since it was not possible to obtain specific data on precisely what types of feedstock are used at the NUTS2 level, statistics on the manure to crop share processed in AD were scaled down from national to regional level. For Germany, feedstock inputs for agricultural biogas plants are on average 45% manures and 55% crop material [28], while in Veneto the mix is on average 55% manures and 45% crop material [53]. A CHP electrical efficiency of 38% and a value of 9.97 kWh per liter of methane (CH_4) were assumed. The capacity installed in each region corresponds to 137 MW in Veneto and 1237 MW in Bavaria. Taking account for the installed capacity, the average mix of manure and crop material present in each region is then used to estimate more precisely the feedstock already used in AD. The final available potential can then be calculated by taking the total agricultural feedstock produced and subtracting competing applications for animals, soil organic matter and already installed capacity.

2.4. TEA Method

TEA of the different technologies, utilizing different feedstock mixes was carried out. Financing costs, maintenance and plant overhead costs, labor related costs, and feedstock costs were aspects considered for the TEA. For all scenarios, it was assumed that the AD plant has a productivity of 8760 hours per year.

The CapEx of the AD plants were estimated using a CapEx of M€ 4 for a 1 MW plant complete with AD, H_2S washer, and generator as a reference, which scales with a power of 2/3 to the electricity output [28,54]. The AD + Booster technology requires extra equipment for the separation and heat treatment, but it also reduces the required hydraulic retention time and therefore the required equipment size of the digester. Based on expert knowledge, it was assumed that regarding the CapEx these aspects equalize and therefore the CapEx of the AD + Booster scenarios is equal to that of the AD plants. The PHB production requires extra equipment for separation, filtration, selection, accumulation, and concentration. Based on expert knowledge, the CapEx for the AD + PHB scenarios was estimated to be 25% higher compared to that of the AD plant.

The financing costs were based on an amortization of the CapEx over 10 years with no interest. Maintenance, tax, insurance, rent, plant overhead, environmental charges, and royalties were assumed to be 10% of the CapEx per year [55,56].

The AD plants were assumed to have a high level of automation, thus, the labor related costs for a 1 MW plant are based on a 1 shift position. Assuming that an operator earns a salary of €18/h and including costs for supervision (+25%), direct salary overhead (+63%), and general plant overhead (+122%) [55], resulted in total labor related costs of k€ 500/y. For the 200 kW plant the labor related costs were divided by five, assuming farm personnel are available part-time. As the PHB production requires a number of extra unit operations and produces an extra product, the labor related costs were assumed to be 50% higher.

The feedstock costs including raw material, and handling and transportation costs are shown in Table 5. The costs for the different types of manure were estimated based on short distance transport costs of manure of €1/ton wet weight (WW) and thereby depend on the dry weight (DW) content of each feedstock. Grass and corn silage were assumed to be produced close to the AD plant and costs were estimated based on [57] and [58]. The costs for wheat straw, corn stover, and soybean straw were based on baling and transportation costs. The costs for vine shoots were based on harvesting and transportation costs. The costs for grape pomace, sugar beet pulp, and grain were based on [58].

Table 5. Feedstock costs in euro per dry weight.

Feedstock	Costs
Chicken manure	€5/ton DW
Cow manure	€9/ton DW
Pig manure	€18/ton DW
Grass silage	€100/ton DW
Corn silage	€120/ton DW
Wheat straw	€40/ton DW
Corn stover	€40/ton DW
Soybean straw	€40/ton DW
Vine shoots	€60/ton DW
Grape pomace	€150/ton DW
Sugar beet pulp	€150/ton DW
Grain	€200/ton DW

Based on the total costs, the break-even prices for electricity and crude PHB were calculated. In the scenarios in which crude PHB is produced, the break-even price of electricity is equal to the regular AD scenario. The break-even prices were compared to selling prices of electricity and PHB (Table 6). As in the AD + PHB scenarios a concentrated crude PHB is produced, extra required purification costs were included. For comparison between the economic performance of each scenario, the required subsidy, i.e., the difference between the selling prices and the break-even prices was calculated.

Table 6. Product selling prices.

Product	Specification	Price	Reference
Electricity	Germany	€0.042/kWh	[59–61]
	Italy	€0.058/kWh	[59–61]
Thermal energy	Germany	€0.025/kWh	[28]
PHB	Purified PHB	€3.6/kg	[15,62]
	Purification costs	€1.8/kg	[62]

2.5. LCA Method

LCA is a standardized methodology governed by international standards and guidelines [8]. Among these, the ILCD handbook offers detailed guidance on how to carry out LCAs in accordance with the definitions set out by the European guidelines [63]. Using this guidance, the study at hand is considered a situation A “micro-level decision support”, since structural changes are not foreseen to occur in the background system, due to the small share of biogas in the overall context of renewable energy. Thus, average mixes were used for the background system and replacement of substituted products. Where co-products are produced, such as in the case of AD + PHB, system expansion is used. The same was done for heat, which is produced as a by-product when biogas is burned in a CHP unit. Though in the latter no credits were awarded in the Veneto region for the produced heat, since this is not yet valorized in Italy [51], apart from what is used for own consumption from operation of the plant. In Germany, the situation is slightly different, and thus, a credit was given to the co-produced heat at a rate of 0.52 kwh heat/kwh electricity, based on the amount of heat utilized at national level [28].

Residue feedstocks that are presently not typically valorized, apart from biogas production, come into the system burden free, since the burden of production was allocated solely to the main product. This is the case for animal manures, pomace and vine shoots. However, for energy crops, the full burden of production was taken into account, i.e., maize silage, grain and grass silages. For agricultural residues currently valorized in the market, such as sugar beet pulp, corn stover, and soybean straw, the burden of production was distributed by economic allocation, while for wheat straw an existing Ecoinvent process was used. The allocation key is shown in Table 7.

Table 7. Economic allocation key for crop by-products.

	%	% of	Reference
Corn stover	47	maize production	[64,65]
Sugar beet pulp	6	sugar production	[66]
Soybean straw	12	soybean production	[67]

In order to visualize the benefit of digesting manure, emissions from storing manure have been included in the assessment. A period of 50 days of manure storage, minus two weeks of unavoidable storing to account for losses and manure in housing units, is avoided by instead treating the manure with the technology scenarios. The quantity of avoided methane is directly proportional to the quantity of manure available in the region or the amount of manure is the feedstock mix. Values used for the calculation are included in Table A5 of Appendix A.

The product system modelling software OpenLCA [68] was used for the modelling and subsequent analysis, utilizing the Ecoinvent v3 database [69]. ReCiPE Hierarchist (H) [70] was chosen as the impact assessment method, and results were generated at midpoint and endpoint. The time horizon for calculation of impacts is 100 years from point of emission.

2.5.1. Plant Level

The functional unit (FU) at plant level is the treatment of 1 ton of feedstock of local characteristics, defined in Table 1 for each plant. Biogas is burned in a CHP, producing heat and electricity. Electricity substitutes the production mix corresponding to the geographical location of each biogas plant. Heat utilization was modeled as substituted district heat for the 200 kW Bavarian plant based on their data, while there is no heat utilization for the industrial size plant in Veneto. PHB production offsets average global thermoplastic production (Table A6, Appendix A).

2.5.2. Regional Level

The FU at NUTS2 level is the treatment of all the AD compatible feedstock defined through the mass flow analysis of available potential for each region (see Section 3.1). An energetic cut off of 1% was applied, so that feedstocks contributing less than 1% of total energetic potential of all feedstock in the region were left out. To simplify matters further, partly due to results from the TEA, the regional assessment was done for plants of industrial size, i.e., 1000 kW for both locations, processing a feedstock mix corresponding to the regional availability, which is defined in the regional feedstock availability assessment. Transport for the regional and plant level assessments was included as 1 km of feedstock transport, and other distances were tested in a sensitivity analysis.

A second sensitivity analysis was also included. The energy grid of each location was replaced with a theoretical future green energy mix, in order to observe the effect of changing energy grids through time. This follows best practices for including partially dynamic LCA in systems with a long service life [71].

2.6. Interpretation of Environmental Impacts

In order to interpret the results, several methods were used. Because of political importance as well as ease of understanding, GHG emissions were used as a proxy for environmental impacts in some discussion, though due to the potential issues with only using GHG emissions, e.g., burden shifting [72], other interpretation methods are were also used. In particular, two methods were used: the first is a monetization of environmental impacts based on endpoint damages [73] and the second uses a form of multiple criteria decision assessment called technique for order of preference by similarity to ideal solution (TOPSIS) [74], utilizing the implementation method ArgCW-LCA [75].

In the first of these two methods, monetization and ReCiPe endpoint damages [76,77] are used to calculate the external costs of the implementation of a given technology at a given scale or region. This was done through two methods. The first, for ecosystem damages, is based on budget constrained ability to pay, which is used to derive a valuation for species years (Species.Yr) gained or lost [78], as this is suggested as the least uncertain method for this valuation [79]. For that valuation, 65,000 USD_{2003} per Species.Yr was utilized. In order to evaluate the disability adjusted life year (DALY) loss or gain, a value from Dong et al., who assessed a number of different methods, was utilized [80]. The valuation derived in these different methods varies significantly, on the range of 1 to 2 orders of magnitude. Therefore, here we used the average of these values, 110,000 USD_{2003} per DALY, which is also in line with the value derived from budget constraint monetization [78], which again should have the least uncertainty. Since resource scarcity endpoint damages are already expressed in monetary terms, no further interpretation is necessary.

In the second method used for deriving a single score, based on the ArgCW-LCA method [75], ReCiPe midpoint environmental impacts [76] along with a valuation of required subsidy for profitability to represent the economic impacts were used as the input criteria for TOPSIS utilizing weighting based on what Sohn et al., describe as a context weighting factor (CWF) [75]. Per a suggestion from the ArgCW-LCA method, as there was no specific stakeholder group present, the stakeholder perspective element was omitted from the method application. For this application, normalization for an average European person year emission was used [81]. Thus, weighting of the environmental impacts is derived, as described in the ArgCW-LCA method, by taking an average of two values: the average of the normalized midpoint impacts for impact category 'i' amongst all assessed scenarios, and the difference of the minimum and maximum normalized impacts for impact category 'i' amongst all assessed scenarios. This accomplishes two things: (1) taking the average of the normalized impacts scales the importance of emissions of the system to status quo emissions and (2) taking the difference between the maximum and minimum normalized impacts is to scale relative to the ability for choosing amongst the available alternatives to cause significant change in status quo emissions. This was completed for all impact categories resulting in the CWF for the environmental impacts. Economic impacts were ascribed a range of weights relative to the sum of weighting given to environmental impacts ranging from 10%–90%. The system was also run using equal weights for all criteria as a point of comparison to the context weighted and the other single score results.

3. Results and Discussion

3.1. Regional Feedstock Availability and Potential Bioenergy Production

A complete table of the sustainable/technical feedstock potential is presented in Appendix B, Table A7, for Bavaria and Veneto. These amounts were used for the TEA-LCA as the regional feedstock mix, though with a 1% cut-off based on the energetic potential of the feedstock.

When graphed on a % wt basis (Figure 4), a relatively large proportion of production of energy crops is evident in Bavaria. Both regions are rich in cattle manure and have a noteworthy amount of swine manure. After energy crops, the most abundant residues are cereal straw for Bavaria and sugar beet straw and soybean residues for Veneto. The regions notably differ from each other, in particular with regard to the production of certain crops, for example sugar beet, soya and grapes. The grapes, represented by pomace, are much more prominent in the Veneto region.

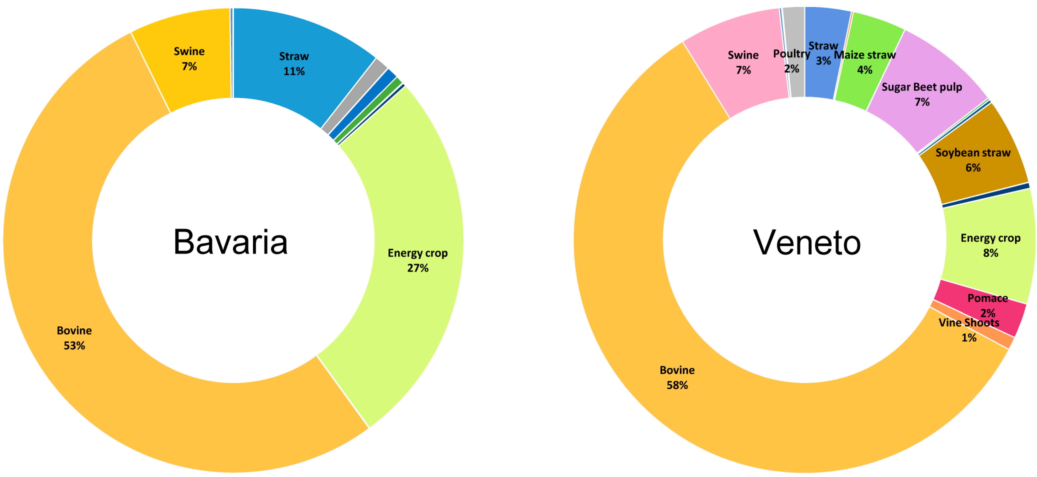

Figure 4. Share of total mix of agricultural residues on a % wt basis for Veneto and Bavaria.

In comparison to Veneto, Bavaria has a much larger share of energy crops, mainly maize silage. This greater share of energy crops is explained by feed incentives given to biogas plants using energy crops in Germany during 2004–2008 [82]. Although maize production has been capped by several German rulings, from 60% by mass input in 2014 lowered to 50% in 2018 and 44% by 2021, the combination of a high animal density and fodder production means that growing of maize has increased exponentially with unintended consequences, such as increasing land prices [22,82]. In Veneto, the feedstock mix exhibits more variability and the expansion of energy crops has not been as dramatic. This may likely be due to the Biogasdoneright™ concept promoted by the Italian Biogas Association, which originates in northern Italy, under which sequential crop cultivation is practiced, where the primary food crop goes to its intended purpose, and a secondary cover crop serves as feedstock for biogas plants [83].

Nevertheless, in energy terms, the potential of the feedstock mix is different than the availability based on mass, mostly due to the poor methane potential of some of the feedstocks. Without subtracting the feedstock that is already being used in the installed capacity of these regions, the energetic potential (based on electrical power) is seen in Figure 5. The largest share of potential is dominated by different feedstocks in the two regions. In Bavaria, the largest share can be obtained from energy crops, while in Veneto the largest share can come equally from cattle manure or energy crops. As a rough estimate, 153 PJ and 38 PJ remain as unexploited feedstock. This represents 31% and 54% of the total available feedstock potential, in Bavaria and Veneto, respectively, which is estimated as described in Section 2.3.4. However, for the LCA, all of the feedstock in the region was assumed to be utilized by the technologies, since in theory biogas plants can be retrofitted with the additional equipment needed for implementation of the AD + Booster technology and PHB production.

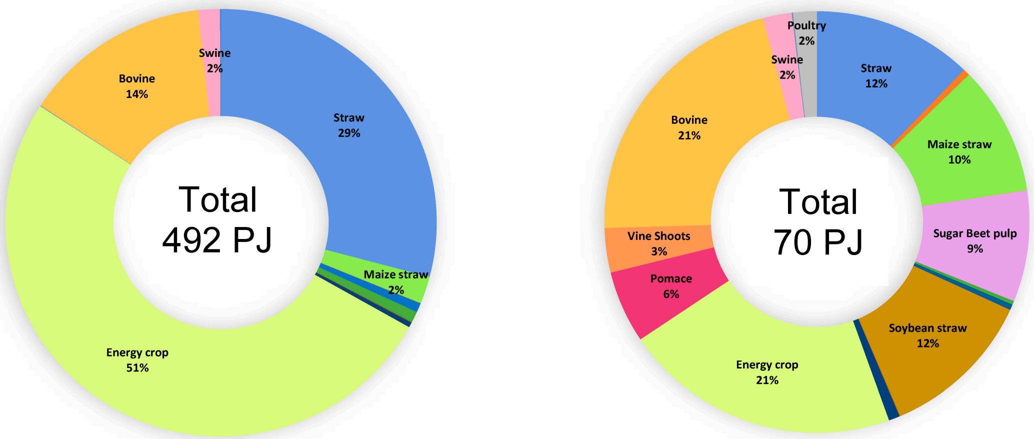

Figure 5. Energetic potential from agricultural residues for Bavaria (left) and Veneto (right) as % energy basis.

3.2. TEA Results

Based on the technical description of the different technologies and the different feedstock compositions, the flow sizes, flow compositions, production of electricity, heat, and crude PHB were estimated. Linking these process parameters to the economic parameters results in the TEA in Table 8.

In all scenarios, the financing, maintenance, labor-related, and feedstock costs are in the same order of magnitude. The contributions of these cost aspects to the total cost vary between 19% and 34%. The small-scale scenarios have, relative to annual production, a larger CapEx compared to the industrial scale, therefore financing and maintenance costs increase the break-even prices for the small-scale scenarios. This results in a break-even price are 34% higher for electricity and 27% higher for crude PHB for the small-scale scenarios, compared to the industrial scale scenarios. As all cost aspects are in the same order of magnitude, the extra required labor in the AD + PHB scenarios results in a significant contribution to the total costs. Logically, the extra labor related costs increase the break-even price of crude PHB. Compared to the feedstock costs of the studied plants, the regional level feedstock in both Bavaria and Veneto have a slightly higher contribution to the costs and to the break-even prices. In the Bavaria scenarios, the revenues of the thermal energy cause a reduction to the break-even prices of 8% for the small scale and 6% for the industrial scale, relative to scenarios that do not utilize the thermal energy.

For the 1 MW AD plant scenarios the average estimated break-even price for electricity is €0.22/kWh. For the AD + Booster scenarios, the average estimated break-even price for electricity is €0.19/kWh, a reduction of 12% in comparison to AD alone. Using the break-even for electricity of regular AD in the AD + PHB scenarios results in an estimated break-even price for crude PHB in the range €4.3/kg to €4.7/kg. When the purification costs of €1.8/kg are included, the break-even price range for PHB is in the range €6.1/kg to €6.5/kg. Due to the difference between market price and the break-even prices, as outlined in Section 2.4 (Table 6), it is clear that both electricity and PHB require large subsidy contributions to be profitably produced in AD plants. Relative to their respective market prices, the required amount of subsidy for the production of PHB is smaller compared to the subsidy for the production of electricity. Nevertheless, the production of PHB requires the co-production of electricity (Table 8).

Table 8. Techno-economic assessment (TEA) results of different scenarios.

Plant Level								
			Small scale (200 kW)			Industrial scale (1 MW)		
			AD	AD + Booster	AD + PHB	AD	AD + Booster	AD + PHB
CapEx		M€	1.4	1.4	1.4	4.0	4.0	4.0
Electricity	Produced	kW	200	224	138	1000	1124	662
	Internal use	kW	15	19	30	75	95	150
	Offset	kW	185	205	108	925	1029	512
Thermal energy	Produced	kW	326	365	224	1632	1834	1080
	Internal use	kW	131	146	90	653	734	432
	Offset	kW	196	219	135			
Crude PHB	Offset	ton/y			58			287
Costs	Financing	k€/y	137	137	171	400	400	500
	Maintenance, etc.	k€/y	137	137	171	400	400	500
	Labor-related	k€/y	100	100	150	500	500	750
	Feedstock	k€/y	142	142	142	440	440	440
	Total	k€/y	516	516	634	1740	1740	2190
Break-even price	Electricity	€/kWh	0.29	0.26	0.29	0.21	0.19	0.21
	Crude PHB	€/kg			5.7			4.3
Subsidy	Electricity	k€/y	405	393	236	1274	1222	705
	Crude PHB	k€/y			225			711
	Total	k€/y	405	393	460	1274	1222	1416
Regional Level								
			Bavaria region (1 MW)			Veneto region (1 MW)		
			AD	AD + Booster	AD + PHB	AD	AD + Booster	AD + PHB
CapEx		M€	4.0	4.0	5.0	4.0	4.0	5.0
Electricity	Produced	kW	1000	1144	742	1000	1155	755
	Internal use	kW	75	95	150	75	95	150
	Offset	kW	925	1049	592	925	1060	605
Thermal energy	Produced	kW	1632	1866	1211	1632	1885	1232
	Internal use	kW	653	746	485	653	754	493
	Offset	kW	481	545	308			
Crude PHB	Offset	ton/y			255			227
Costs	Financing	k€/y	400	400	500	400	400	500
	Maintenance, etc.	k€/y	400	400	500	400	400	500
	Labor-related	k€/y	500	500	750	500	500	750
	Feedstock	k€/y	558	558	558	509	509	509
	Total	k€/y	1858	1858	2308	1809	1809	2259
Break-even price	Electricity	€/kWh	0.22	0.19	0.22	0.22	0.19	0.22
	Crude PHB	€/kg			4.4			4.7
Subsidy	Electricity	k€/y	1415	1356	906	1343	1275	879
	Crude PHB	k€/y			660			666
	Total	k€/y	1415	1356	1567	1343	1275	1545

3.3. LCA Results

3.3.1. Midpoint Results

Results were obtained both at midpoint and endpoint level, using the ReCiPE 2016 (H) LCIA methodology. The results were internally normalized and ranked relative to the best-performing technology scenario. Midpoint level results for both regions and scales showed, for the most part, the same technology preference, pointing to AD + Booster as the best performer across impact categories (ICs), followed by AD and lastly AD + PHB. In the Veneto region, slightly more variation is observed across impact categories (Figure 6) and AD + PHB can at times be the best performer, as seen in the Ionizing Radiation, Land Use, and the Mineral Resource Scarcity ICs. The importance of this variation was tested with TOPSIS and is discussed further in Section 3.4.

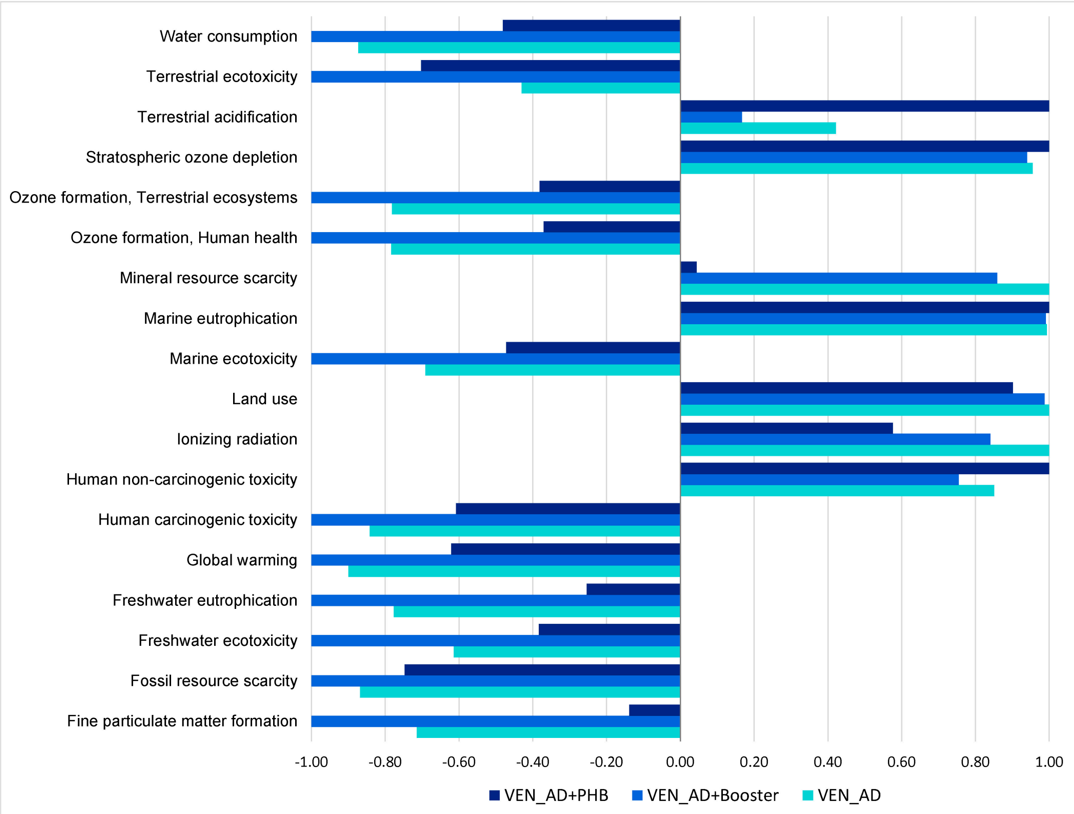

Figure 6. ReCiPE (H) Midpoint results for the region of Veneto for the three technology options i.e., AD, AD + Booster, and AD + PHB. Results are normalized per impact category to the worst or best performing scenario. Negative values show impact savings while positive values show burdens.

Midpoint results for the two farms assessed the small scale 200 kW farm in Bavaria and the 1000 kW farm in the Veneto region showed identical preference to the regional assessment when ranked within geographical location. However, more rank switching is observed when ranking is done across scales; this is explored further and discussed in Section 3.4., where rank reversal is checked thoroughly for both regional and scale assessments. Figures of normalized midpoint impacts for the Bavarian region, small and industrial scale are shown in Appendix B (Figures A1–A3), as well as tables of raw midpoint/endpoint results (Tables A8–A11).

3.3.2. Global Warming

As mentioned previously, global warming potential (GWP) shows the same technology preference as other ICs, with AD + Booster performing better than AD, which in turn performs better than AD + PHB. Looking at the contribution to GWP from the various elements that make up the system, it is possible to understand this preference. As can be seen in Figure 4, the higher energy production of the AD + Booster induces a higher electricity offset, which is largely responsible for the technology preference exhibited by the results. It is also evident that the offset for substituting plastic in the market for the AD + PHB options is very moderate and occurs on account of lower energy production, resulting overall in the lowest GWP savings out of all technology options. Figure 4 also shows the difference between the two regions on a per ton feedstock mix basis. An important difference can be observed in the crop mixture used for each region, where it is evident that Bavaria uses a more burdensome mix than Veneto. Other than crop differences, methane leaks from the facilities, here assumed to be 3% of the biogas produced, is an important source of GHGs. This is worth noting, as it can diminish the savings intended by these technologies. On the other hand, an important savings is attained by degassing animal manures, which would otherwise sit in storage facilities for a longer period producing methane that would be released to the atmosphere. This benefit can be seen in Figure 7 as the "methane offset storage" and is higher for Veneto due to the higher availability of animal manures on a %wt basis in this region.

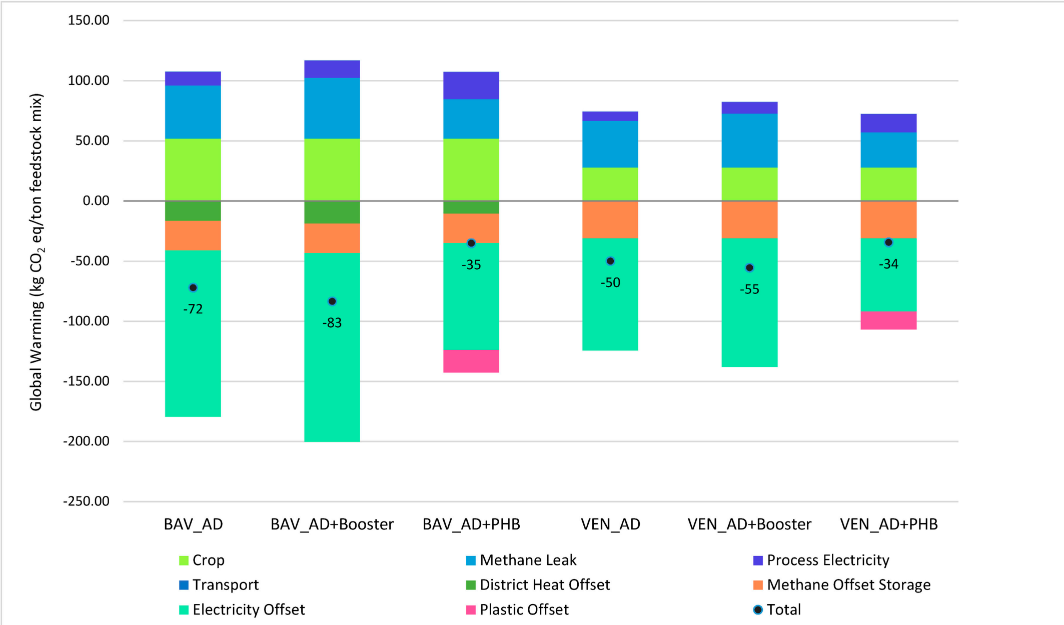

Figure 7. Global warming potential (GWP) contribution per ton of feedstock mix for the two regions, BAV for Bavaria and VEN for Veneto, for the three technology options, i.e., AD, AD + Booster, and AD + PHB.

Figure 8A,B, shows total GWP savings for both Veneto and Bavaria, respectively. As a total, the Bavaria region is capable of obtaining GWP savings 7.4, 7.7, and 5.4 times higher than in the Veneto region for AD, AD + Booster, and AD + PHB, respectively, on an annual basis. This is explained in part by the scale of the regions, feedstock density of the regions, as well as the energy density of each feedstock employed in the mix. While Veneto is also the smaller of the two regions, the lower GWP savings are partly due to an average 25% lower feedstock mass production per area relative to Bavaria. Moreover, the regional feedstock mix in Bavaria contains ca. 7% more crops and crop residues, among which maize silage is a prominent one, whilst Veneto contains ca. 7% more animal manures, which have a low methane/VFA productivity. The feedstock mix of Bavaria results in a higher electricity offsets, even though its feedstock mix contains a higher share of primary production (1st Generation) feedstock, i.e., maize silage, rather than secondary production such as straw. In addition, the utilization of waste heat in the Bavarian system for district heating gives an extra considerable impact offset to the region. If the heat were to be utilized in Veneto, then an extra 23%–25% savings in GWP could be attained there.

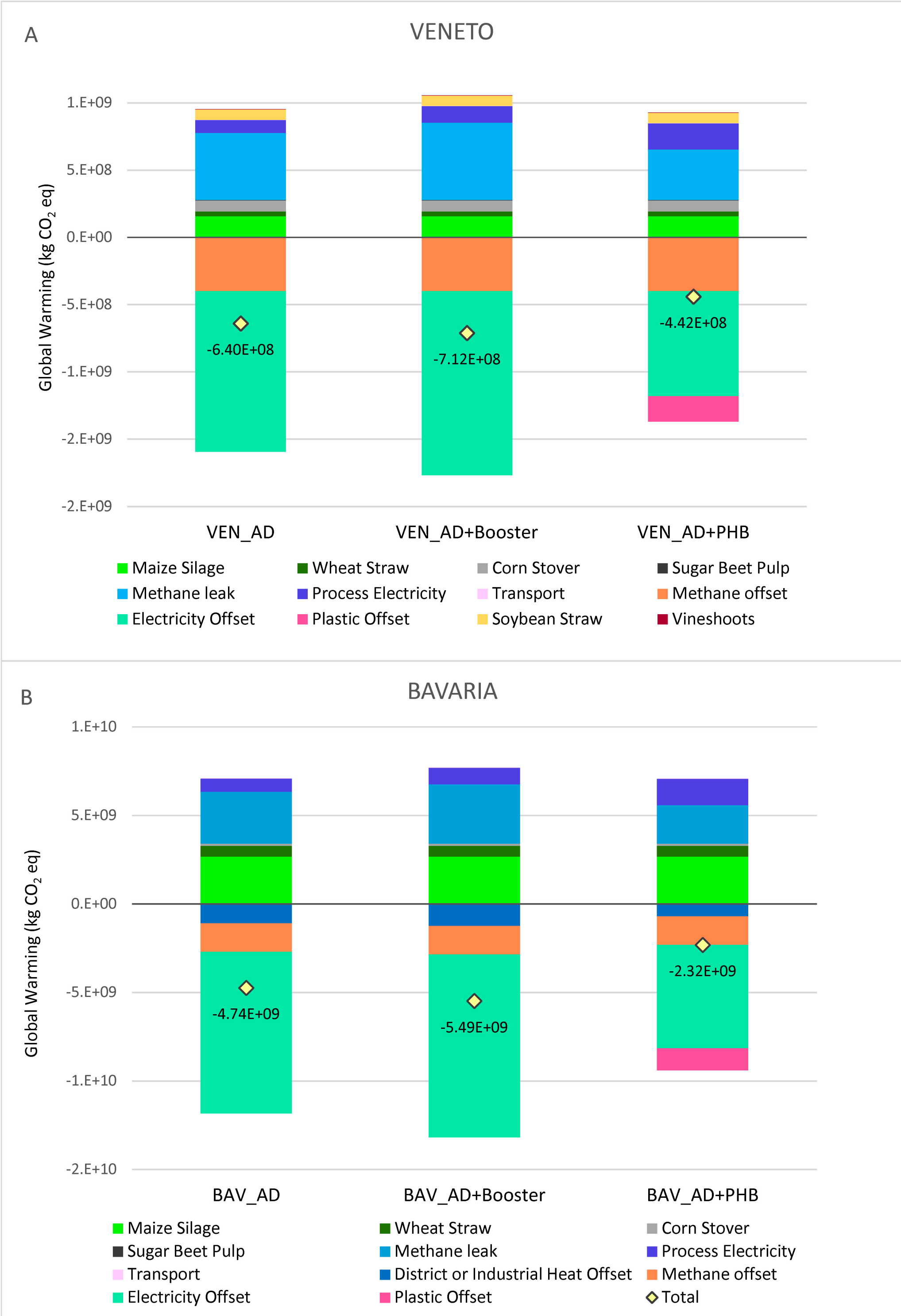

Figure 8. Global warming potential if all of the regional feedstock is treated on an annual basis for (**A**) Veneto and (**B**) Bavaria, as well as GWP contribution by the various system phases. Scenarios are named by the first three letters of the region (VEN or BAV) followed by each technology scenario (AD, AD + Booster, AD + PHB).

The pattern of feedstock efficiency is repeated when comparing the technologies on a scale basis. In fact, using more energy dense feedstock, i.e., feedstock that has a higher methane potential, leads to higher GWP savings for the small-scale facility (S + technology scenario), on a per ton feedstock basis, than for the industrial scale (I + technology scenario) (Figure 9). This is true even though the feedstock mix used in the small scale is more burdensome in terms of GWP, due to the cultivation phase of the feedstocks. The industrial scale facility still incurs savings to GWP, albeit lower, due to the poor characteristics of the feedstock utilized, which in this case is ca. 80% cow manure. Technology preference largely stays the same for both scales, though it is worth mentioning that a friendlier feedstock mix, i.e., with less first generation feedstocks, such as the one in the industrial scale is more important for the AD + PHB option, as can be observed when comparing S_AD + PHB and I_AD + PHB, which have savings of −15 and −25 kg CO_2 eq/ton, respectively.

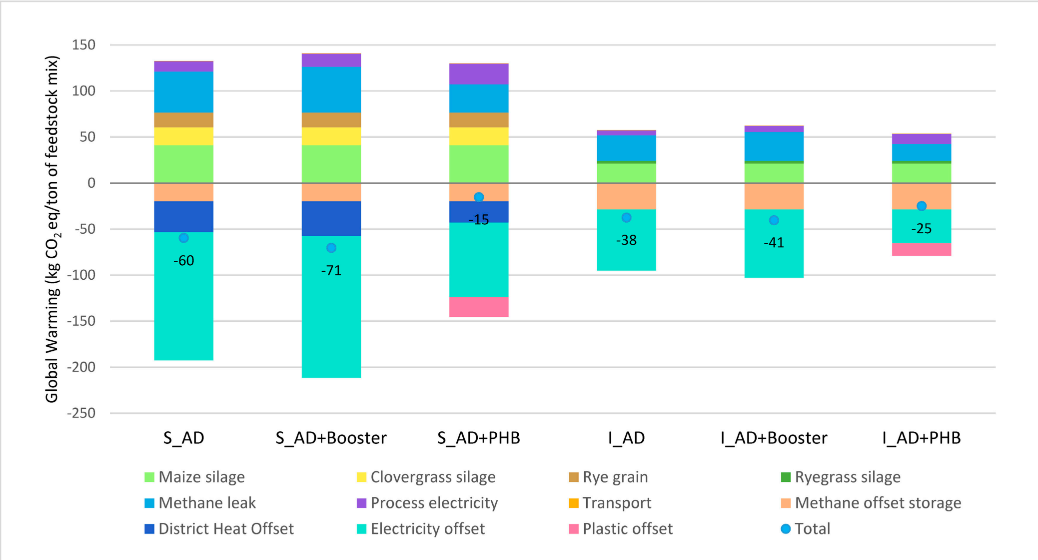

Figure 9. Global warming potential results for the small scale (200 kW) and industrial scale (1000 kW) cases, per ton of feedstock, as well as contribution to GW by each stage. Scenarios are named as S for small scale and I for industrial scale followed by each technology scenario (AD, AD + Booster, AD + PHB).

3.3.3. Sensitivity

Two parameters were tested to assess the sensitivity of the results: transport distance of the feedstock and the effect of a theoretical future green energy mix in the system.

The effect of transport varies depending on how well the technologies perform. The initial results, which include a 1 km transport distance were varied and transport was added up to 100 km. The result can be observed in Figure 10, where it is evident that a further transport distance can be allowed for the AD + Booster technology in both regions since this is the best performing technology. The point at which each technology scenario goes from GWP saving to GWP burden can also be seen in the graph. This point (the y-intercept) is 86, 99, and 42 kilometers respectively for BAV_AD, BAV_AD + Booster and BAV_AD + PHB, in Bavaria. In Veneto these distances are lower, because of the lower performance of the technologies in this region, where a transport distance below 59, 65, and 41 kilometers for VEN_AD, VEN_AD + Booster, and VEN_AD + PHB respectively, would ensure that the technologies continue to induce GWP savings. Needless to say, the lower the transport distances for the feedstock, the better the technologies perform.

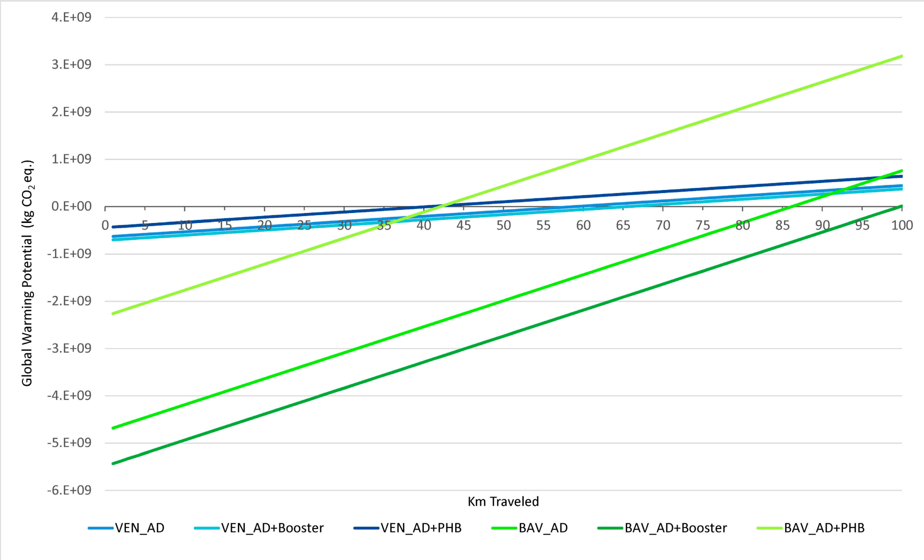

Figure 10. Effect of transport of feedstock on GWP savings. Scenarios are named by the first three letters of the region (VEN or BAV) followed by each technology scenario (AD, AD + Booster, AD + PHB).

The effect of switching the current production mix for the provisioning of process electricity and electricity offset with a future energy mix mainly composed of renewable sources is substantial for GWP results. For the regional assessment in Bavaria, all technology options result in impact burdens for GWP, while they continue to be impact savings for Veneto (Figure 11). This is due to the feedstock mix emissions in Bavaria, which are no longer counterbalanced by high emissions savings from offsetting of electricity. As has been shown before [26,84], offsets from replacing GHG intensive sources of electricity production such as coal, diminish as 'green' energy sources are implemented in the energy grid. The implications of this are very important for technologies producing renewable fuels, as their potential to produce savings will be bound to this future component.

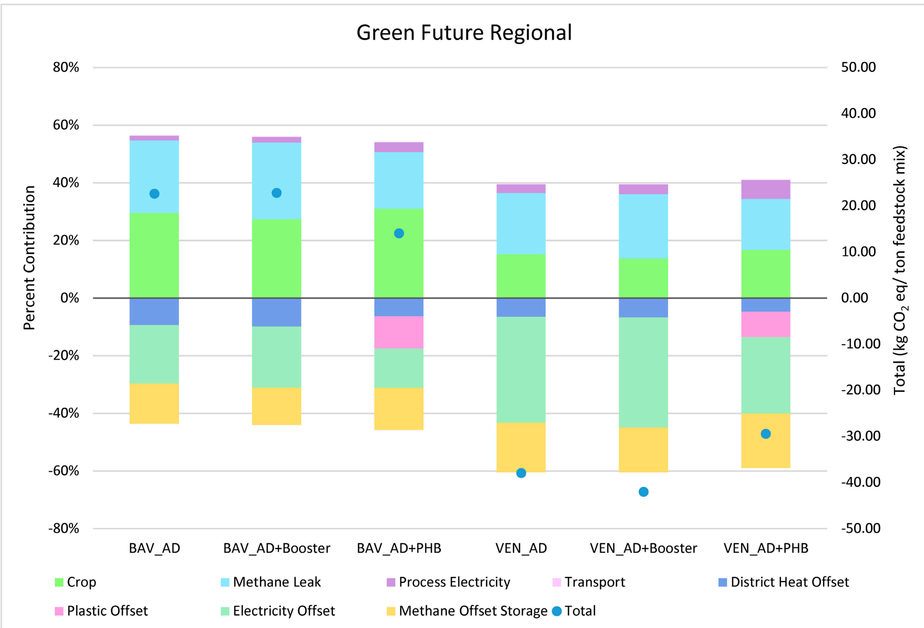

Figure 11. Global warming result for a future with a theoretical green energy mix. Scenarios are named by the first three letters of the region (VEN or BAV) followed by each technology scenario (AD, AD + Booster, AD + PHB).

On the other hand, BAV_AD + Booster, which is the worst performing scenario in terms of GWP continues to be the best performing scenario for most other impact categories in the green energy future SA (normalized midpoint results in Appendix B, Figures A4 and A5). As this clearly points to burden shifting the results were subjected to two single indicator interpretation methods to clarify the results.

3.4. Single Score Interpretations

Single score results, via TOPSIS developed by applying the ArgCW-LCA methodology [75], with environmental weights relating the results to a European's consumption patterns, and an economic weight derived from the TEA are discussed in this section. When assessed through TOPSIS (Figure 12), the initial regional results are very clear. Technology preference does not change within each region no matter which weighting is given to the results. AD + Booster is always the preferred choice, whether there are equal weights and high or low weight is given to economics. Furthermore, when impacts are monetized ($) so that the costs of environmental protection are visualized, these results also agree with the ArgCW-LCA and equal weights (EW) TOPSIS results. From the figure it is clear that the AD + Booster is also the best performer in terms of economic preference in Veneto (going up to 90% econ level), and on the contrary AD + PHB appears to be the worst. However, it is worth noting that in the Veneto region, if environmental concerns are weighed more heavily (<55% econ level), it is not easy to single out one of the technologies as unequivocally the best performing option, since the results perform close to equally well. This is not the case for Bavaria where the more burdensome feedstocks result in a more indisputable preference for the AD + Booster option, which produces the most energy. The implication of these results, namely that the more burdensome the energy production is, the more important the energy offsets become, is even more obvious for the plant level assessment. Here we see that though the technology preference is always the same (AD + Booster > AD > AD + PHB), the relative difference between the options becomes smaller the higher the economic weight (approaching 90%) for the Industrial plant in Veneto. This is a different pattern than the one observed for the regional level, where the distance between options, with and without PHB, increases with economic weight, and as supported by the assessment of midpoint results, the technology scenarios are closer to each other when the feedstock mix contains more animal manures than crop residues (see Figure 9). The same trend is seen for the small-scale plant in Bavaria, where the distance between the AD + Booster and AD + PHB option decreases with increasing economic weight. Though in this case, the plant's economic performance, which is very low in comparison to the industrial plant, is an important factor pulling all technology options further from the ideal.

The green energy future sensitivity was also checked with the single indicator methodology. The results again showed to be robust in terms of technology preference for the assessment (Figures A6 and A7). It is important to point out, however, that if the decision was based solely on GWP, then when looking at the green energy future one would choose AD + PHB in Bavaria, but continue to choose the AD + Booster in Veneto (Figure A6, Appendix B).

Overall the results are robust, though some clear patterns emerge. The single indicator results clearly highlight the dependency on the energy extraction efficiency of the options, which have increasing importance for regions with a more burdensome production, i.e., in the cultivation of energy crop for biogas production (the BAV and S scenarios). In this case, the electricity offsets are very important, not only for GWP, but all impact categories considered in an LCA, as evidenced by the single indicator preference. There are trade-offs when production utilizes a higher share of energy crops. On the one hand, electricity production is higher and with today's electricity mixes offsetting this type of production is highly valuable. On the other hand, it is worth noting that sustainability criteria for biofuels and biomass fuels might limit this type of production even more in the future. As it stands today, the renewable energy directive II sets out a cap on energy crops for renewable fuels and national caps are also present in various member states. The EC has also singled out feedstock of high potential for indirect land use change (iLUC), so that renewable fuels do provide the GHG reductions they are meant to bring. Though small plants are exempt from this cap (ca. <500 kW electric), one needs only to look at the German case, where around 50% of plants are small, as an example of how many small biogas plants can in fact have large consequences for how agricultural land is used.

The assessment also shows that varied production, i.e., not only energy, can be a viable option for plants with a high content of manures in the mix. In a future with an optimized PHB production this might be even more beneficial, also if we are to avoid the impacts of microplastic pollution, which are yet to be included in LCA studies. For now, strong subsidies are needed to increase technology penetration in the market with constant revision on sustainability targets. Continuing to green the energy grid should be a top priority by making as much energy as possible and fomenting technologies that increase the energy that can be obtained from biomass (like the AD booster). Future research on the possible synergies between technologies such as the AD-Booster + PHB could be interesting to explore.

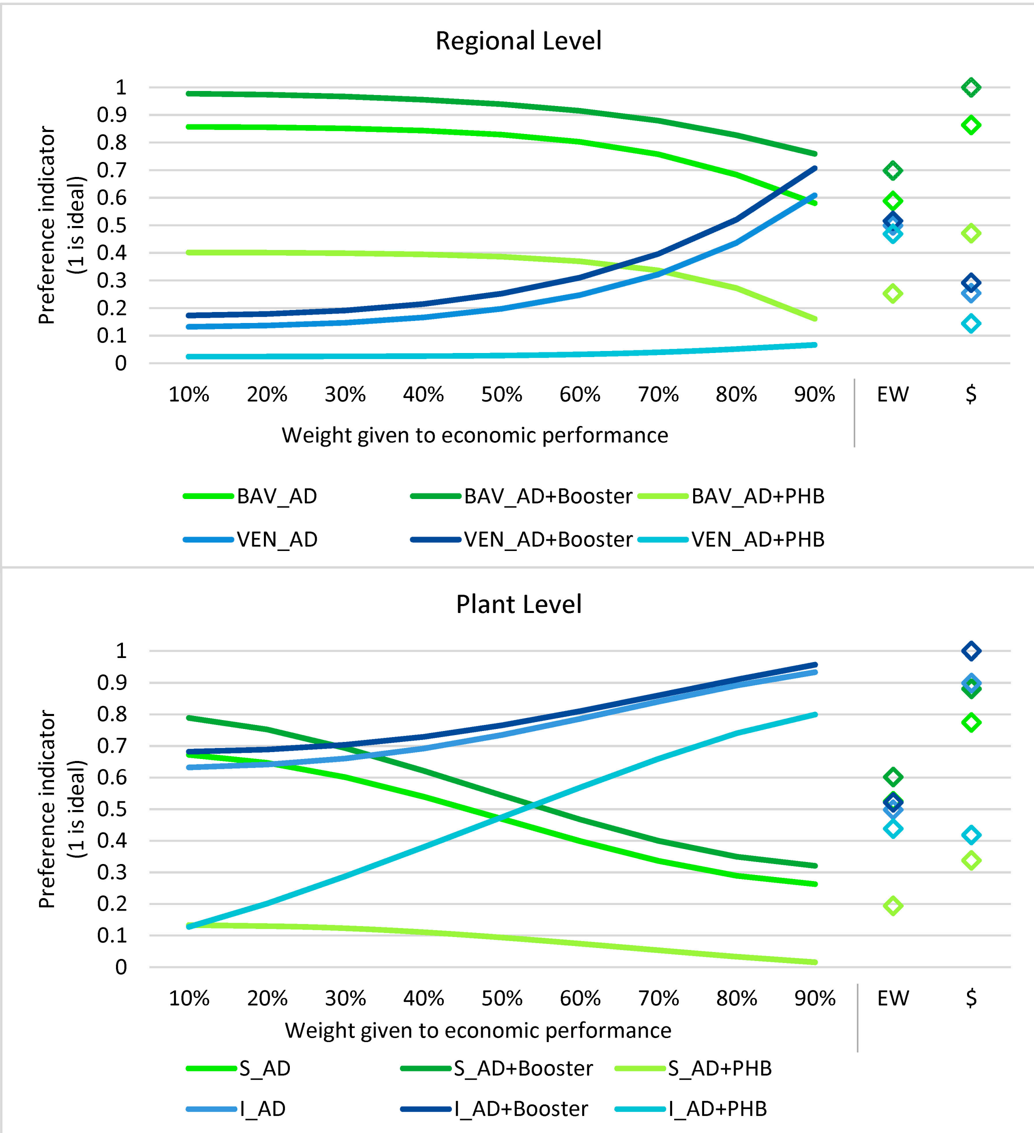

Figure 12. TOPSIS results for the regions (top) and scales (bottom), with varying economic importance (10% to 90%), equal weights (EW), and internally normalized monetization ($) of endpoint damages. Scenarios are named by the first three letters of the region (VEN or BAV) or scale size S for small and I for industrial, followed by each technology scenario (AD, AD + Booster, AD + PHB).

4. Conclusions

The production scale of the industrial set up assessed, with electricity ca. 1 MW and crude PHB production at ca. 300 ton/y, is small compared to their fossil and non-fossil alternatives. As a result, the financing, maintenance, and labor related costs increase the break-even prices significantly. Crude PHB production in AD plants requires the co-production of electricity in order to be adequately valorized, though benefits from avoided plastic particle pollution, which could be important, have not been included in the TEA and LCA. With today's energy mixes in the regions in question, it is highly valuable to offset electricity production and thereby options such as the AD + Booster are preferred for all environmental areas of protection. Material production in scenarios such as the AD + PHB perform equally well to more energy efficient scenarios for plants with a feedstock mix high in animal manures. Future caps on certain types of feedstock are worth considering when deciding on technology options to be implemented and/or subsidized.

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Conflicts of Interest: The authors declare no conflict of interest.

Appendix A

Table A1. Grouping of crops, Eurostat names, and codes for crops and residue crop ratios.

Grouping	Eurostat Code and Name	Residue:Crop Ratio	Reference/Assumption for Residue:Crop Ratio
Cereal Straw	C1110-Common wheat and spelt	1.00	[19,21,85]
	C1111-Common winter wheat and spelt	1.00	assumed same as wheat
	C1120-Durum wheat	0.95	Assumed as triticale, [19,21,85]
	C1200 - Rye and winter cereal mixtures (maslin)	1.10	[19,21,85]
	C1300-Barley	0.93	[19,21,85]
	C1410-Oats	1.13	[19,21,85]
	C1420-Spring cereal mixtures (mixed grain other than maslin)	1.00	Average of common wheat, durum wheat, barley and rye
	C1600-Triticale	0.95	[19,21,85]
Rice Straw	C2000-Rice	1.70	[19,21,85]
Maize	C1500 - Grain maize and corn-cob-mix	1.13	[19,21,85]
Leguminous	P0000 - Dry pulses and protein crops for the production of grain (including seed and mixtures of cereals and pulses)	1.50	Assumed as soy
	P1100-Field peas	1.50	Assumed as soy
Oil-bearing	I1140-Linseed (oilflax)	1.42	[19,21,85]
Rape	I1110-Rape and turnip rape seeds	1.70	[19,21,85]
Sunflower	I1120-Sunflower seed	2.70	[19,21,85]
Soya	I1130-Soya	1.50	[19,21,85]
Industrial	I3000-Tobacco		Not relevant for regions
Energy Crop	C1700-Sorghum	1.30	[19,21,85]
	G3000-Green maize	1.00	Whole plant [21]
Forage	G1000-Temporary grasses and grazing	1.00	Whole plant [21]
	G2000-Leguminous plants harvested green	1.00	Whole plant [21]
	G9100-Other cereals harvested green (excluding green maize)	1.00	Whole plant [21]
Sugar Beet	R2000-Sugar beet (excluding seed)	0.23	[19,21,85]

Table A2. Livestock unit conversion factors and manure production per animal type [7].

	Livestock Unit	Manure	Manure
	LSU	kg/head/day	t/head/year
calves	0.40	8.00	2.90
bovine	0.70	20.00	7.30
male bovine	1.00	25.00	9.10
dairy cows	1.00	53.00	19.30
other cows	0.80	25.00	9.10
piglets	0.03	0.50	0.20
other pigs	0.30	4.50	1.60
sows	0.50	11.00	4.00
sheep	0.10	1.50	0.50
goat	0.10	1.50	0.50
broilers	0.01	0.10	0.04
laying hens	0.01	0.20	0.07
other poultry	0.03	0.30	0.11
Live poultry average	0.02	0.20	0.07

Table A3. Manure collectability factors based on different types of housing and type of production [47,48].

	Collectability
	factor
Stanchion	0.98
Loose housing	0.95
Organic	0.25
Poultry	0.98
Swine	0.98
Sheep	0.5
Goat	0.1

Table A4. Methane potentials of various feedstocks [7].

	DM	VS	Methane Yield	Methane Yield
	%	%	L CH4/kg VS	L CH4/kg fresh
Pig slurry	5.5	75	300	14
Cattle slurry	9	77.5	225	16.5
Poultry manure	20	75	325	52.5
Sheep [1]				16.5
Goat [1]				16.5
Maize silage [2]	35	92.5	350	119
Grass [3]	25	92.5	375	91.5
Alfalfa [4]	22.5	92.5	400	87.5
Sugar beet	17.5	92.5	305	51.5
Straw [5]	87.5	85	225	169
Pomace	35	92.5	600	194.5

[1] Assumed same as cattle slurry. [2] Used for energy crops. [3] Used for forage crops. [4] Used for leguminous crops.
[5] Used for rice straw, rape straw, sunflower straw, soya straw, oil-bearing straw, industrial crop straw, and vine shoot.

Table A5. Parameters used for methane emission from manure storage [86].

		Cattle	Pig	Poultry
Dry matter content	kg DM/kg WW	10.8	5.5	20
Volatile solids	kg VS/kg DM	0.714	0.638	0.638
Methane production in storage (50 days)	g CH4/kg VS	19	98.5	98.5
Inevitable storage and losses (15 days)	g CH4/kg VS	5.7	29.55	29.55

Table A6. Composition of global average plastic production, including low density polyethylene (LDPE), high density polyethylene (HDPE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), polyethylene terephthalate (PET) and polylactic acid (PLA) [87].

Polymer Type	
LDPE	22.8%
HDPE	18.6%
PP	24.3%
PS	8.9%
PVC	13.6%
PET	11.8%
PLA	0.1%

Appendix B

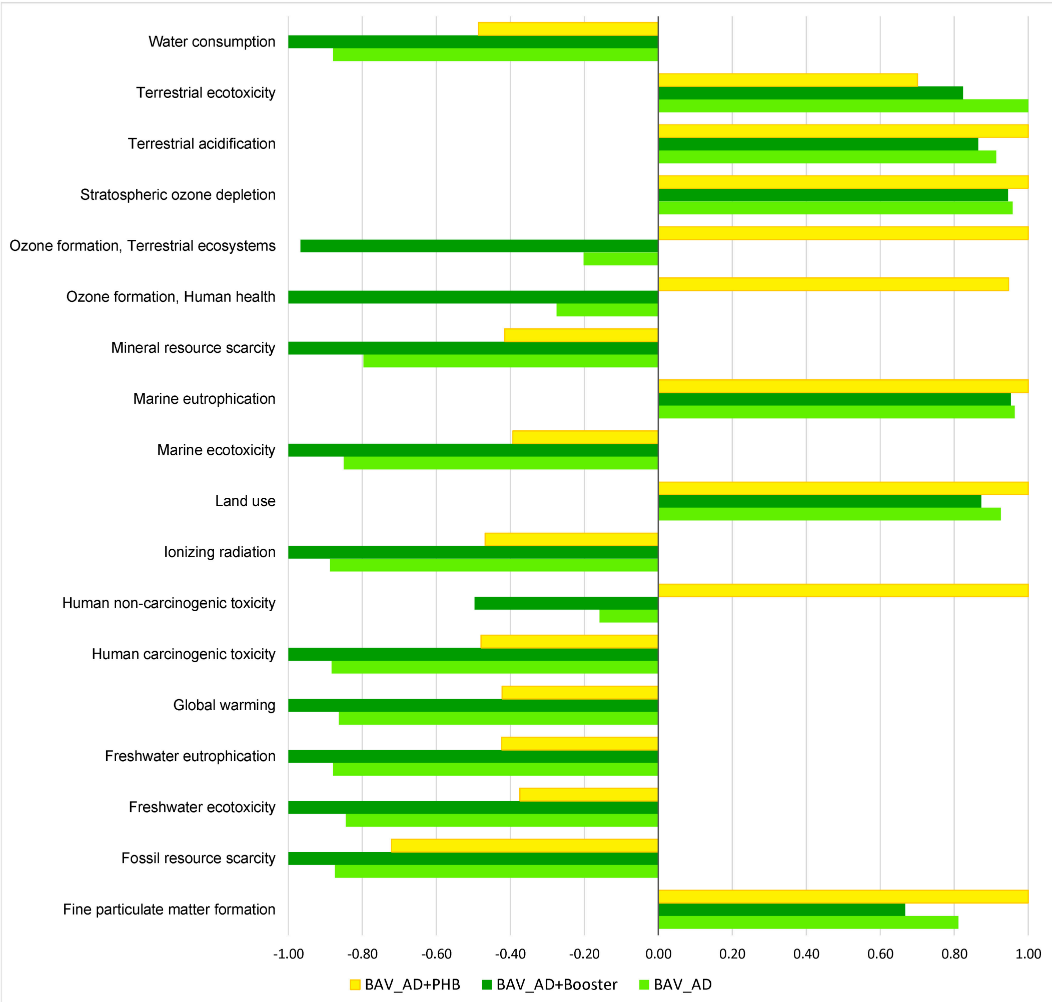

Figure A1. ReCiPE 2016 (H) midpoint results for the region of Bavaria. Results are normalized per impact category to the worst or best performing scenario. Negative values show impact savings while positive values show burdens.

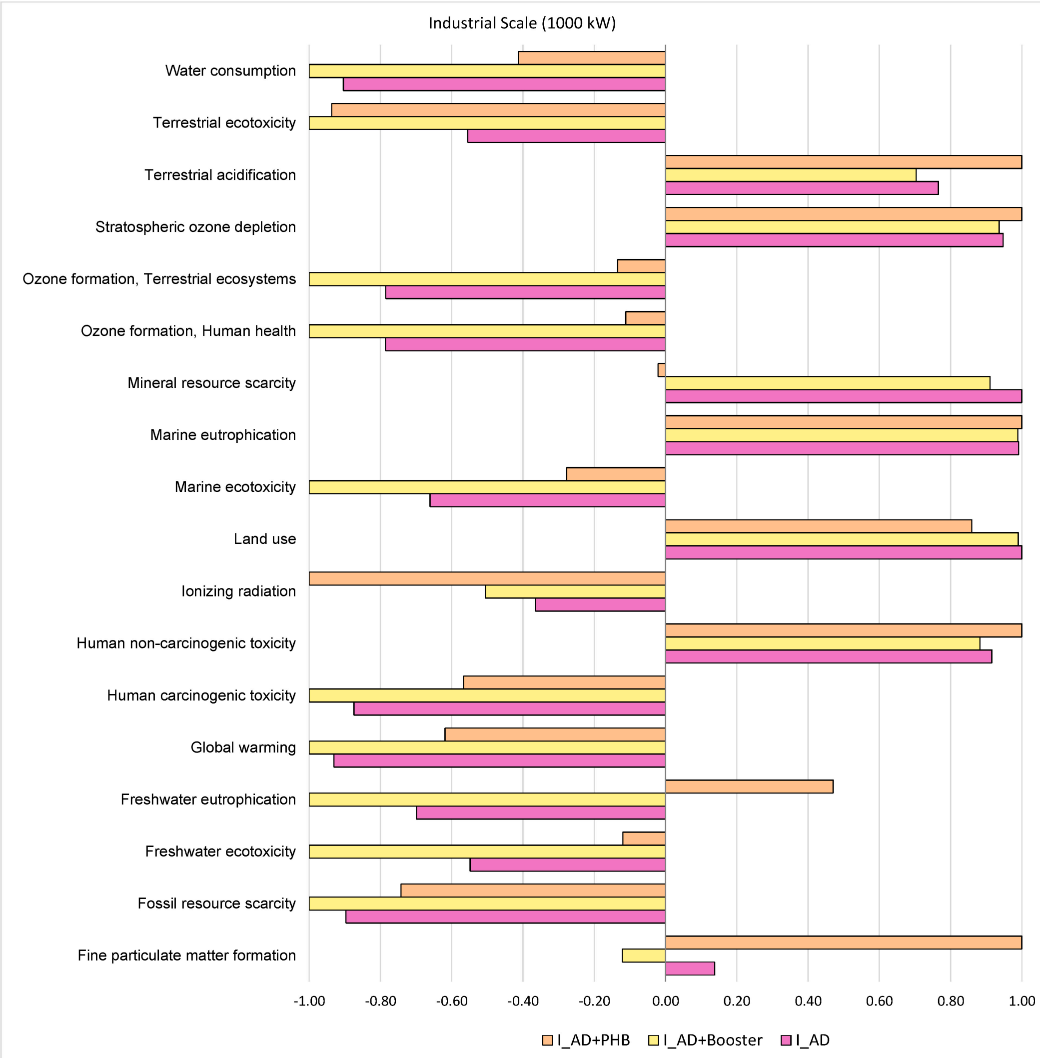

Figure A2. ReCiPE 2016 (H) midpoint results for the region of the industrial scale plant. Results are normalized per impact category to the worst or best performing scenario. Negative values show impact savings while positive values show burdens.

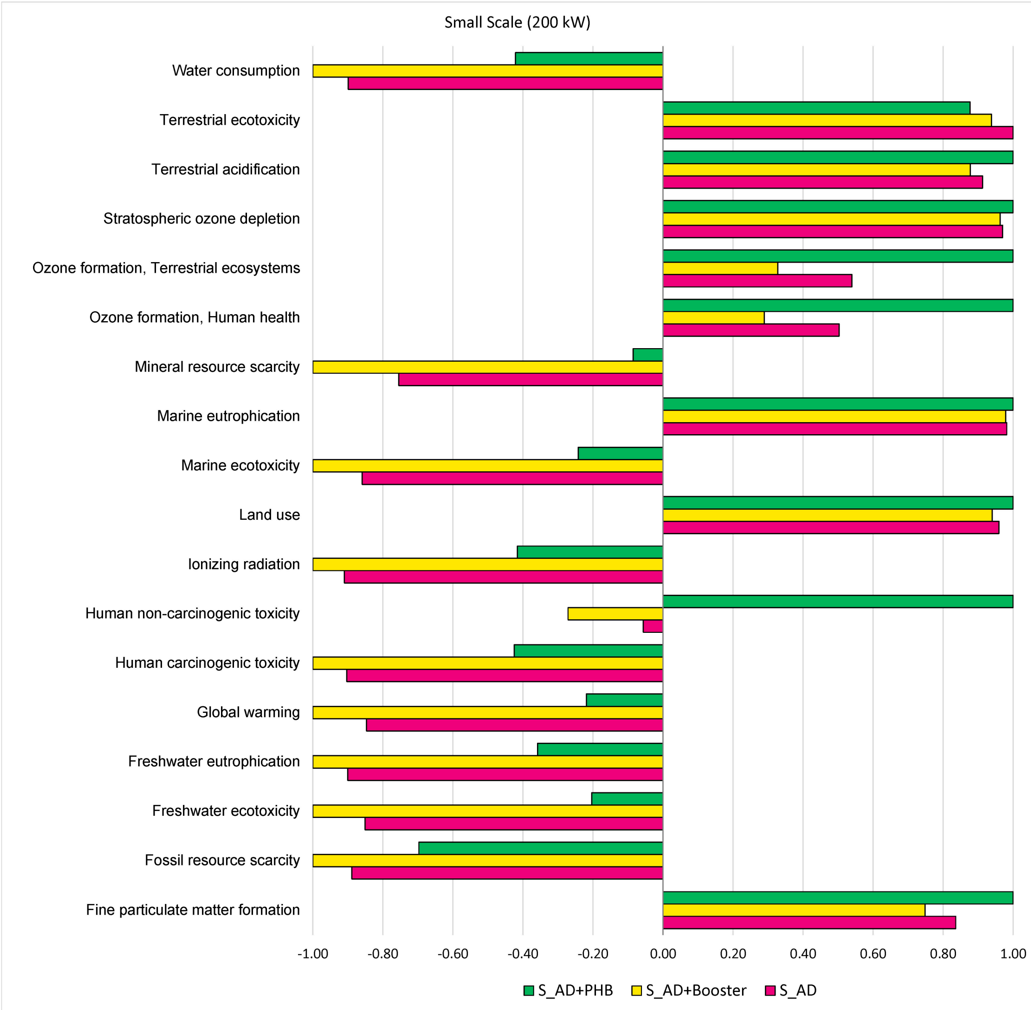

Figure A3. ReCiPE 2016 (H) midpoint results for the small-scale plant. Results are normalized per impact category to the worst or best performing scenario. Negative values show impact savings while positive values show burdens.

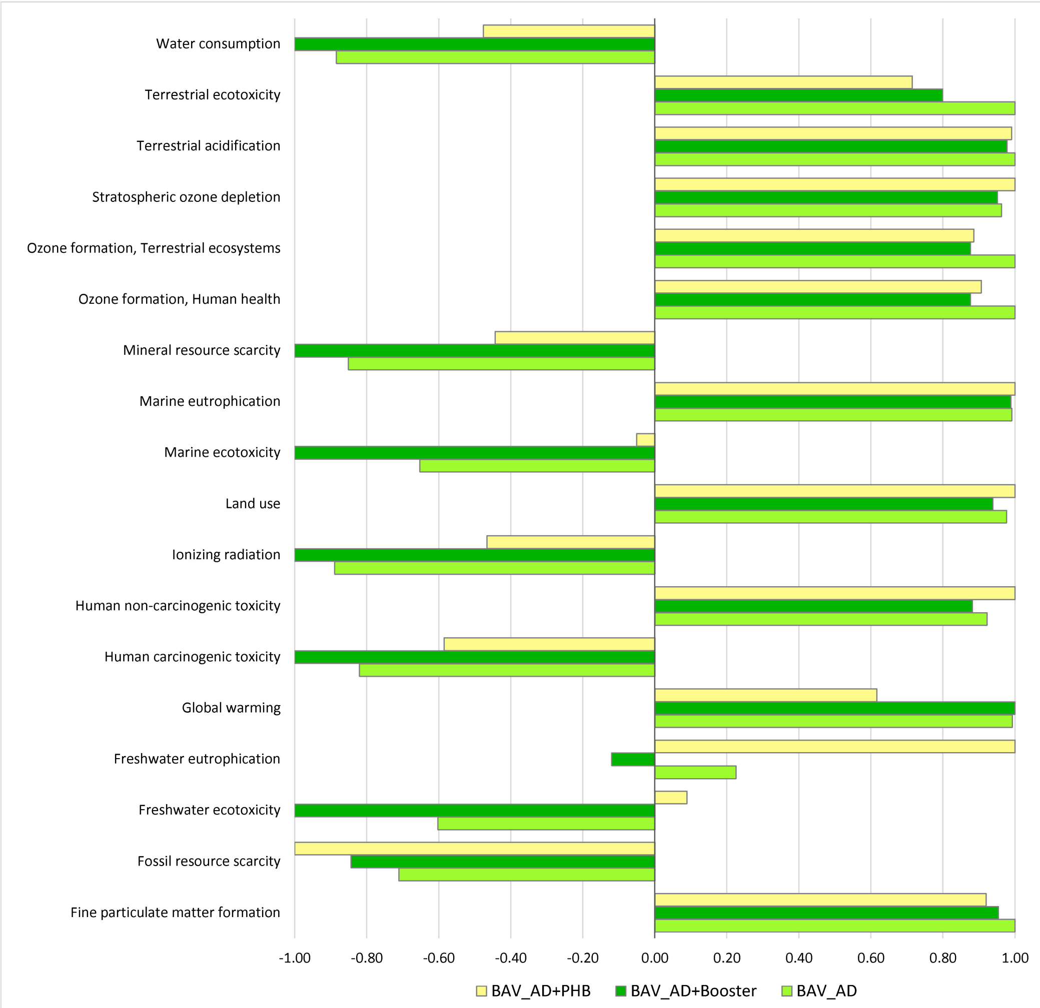

Figure A4. ReCiPE 2016 (H) midpoint results with theoretical green energy grid for the region of Bavaria. Results are normalized per impact category to the worst or best performing scenario. Negative values show impact savings while positive values show burdens.

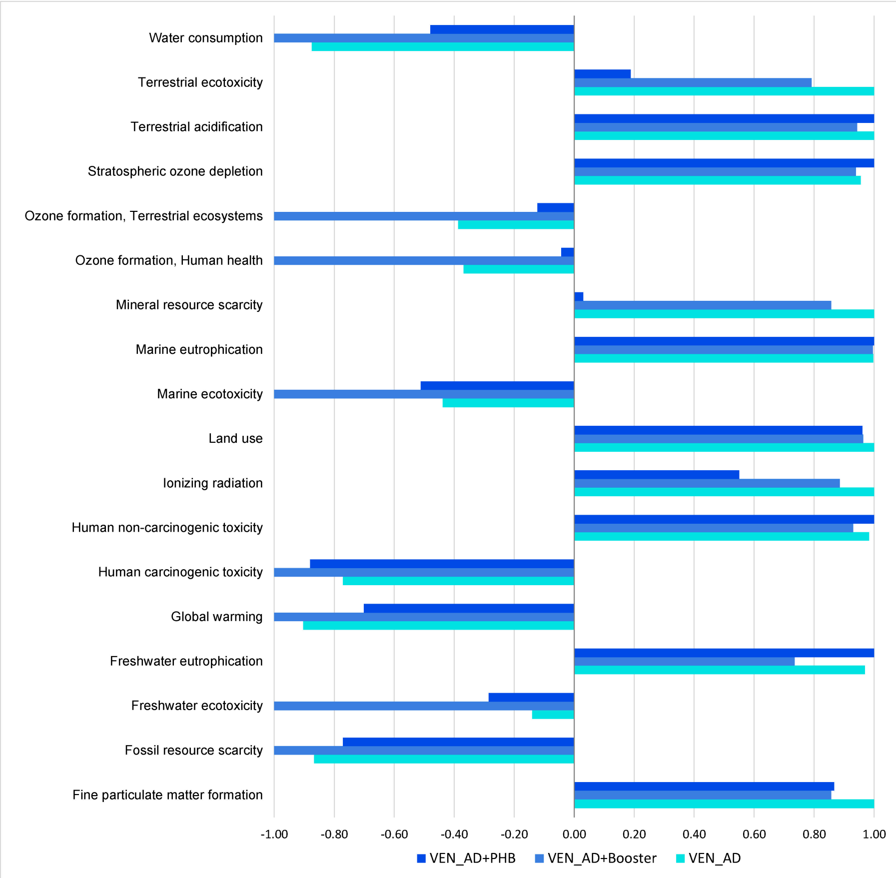

Figure A5. ReCiPE 2016 (H) midpoint results with theoretical green energy grid for the region of Veneto. Results are normalized per impact category to the worst or best performing scenario. Negative values show impact savings while positive values show burdens.

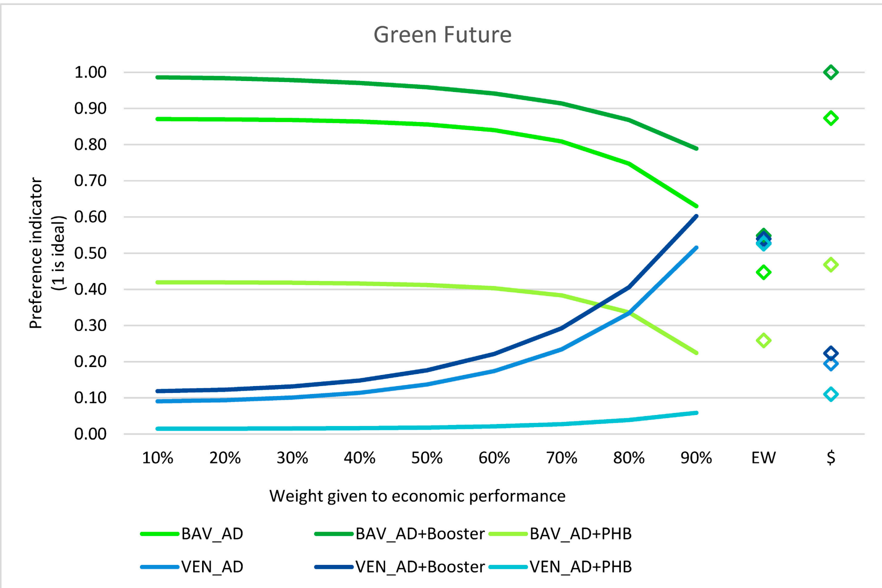

Figure A6. TOPSIS results for the regions with the theoretical green energy mix, with varying economic importance (10% to 90%), equal weights (EW), and internally normalized monetization ($) of endpoint damages.

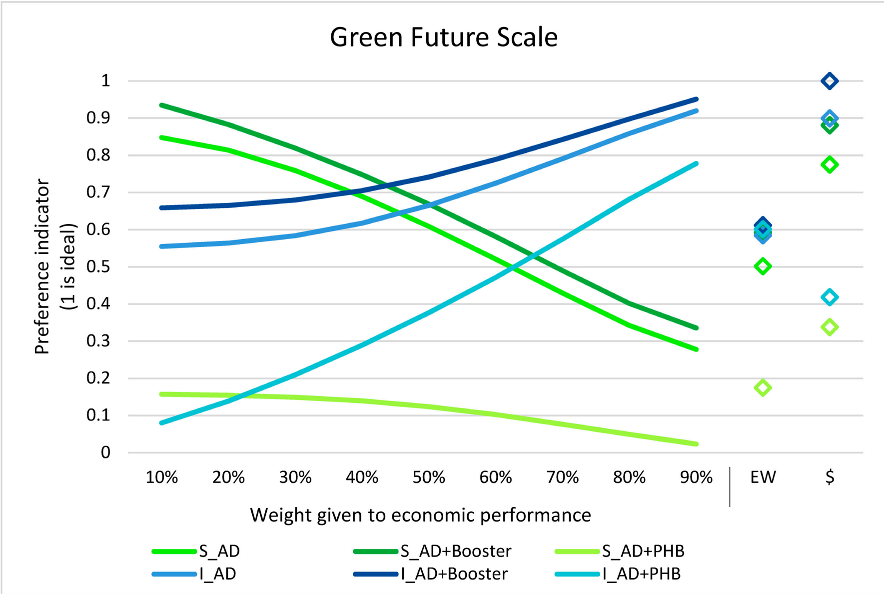

Figure A7. TOPSIS results for the two scales S = 200 kW and I 1000 kW, with the theoretical green energy mix, with varying economic importance (10% to 90%), equal weights (EW), and internally normalized monetization ($) of endpoint damages. Scenarios are named as S for small scale and I for industrial scale followed by each technology scenario (AD, AD + Booster, AD + PHB).

Table A7. Total amount of sustainable/technical feedstock potential in Mtonne/year, sorted from highest to lowest amount.

	Bavaria	Veneto
Cattle manure	35.08	7.58
Energy crop	17.65	1.04
Straw	7.09	0.42
Swine manure	4.73	0.92
Corn Stover	0.71	0.49
Sugar Beet	0.56	0.97
Rape	0.38	0.02
Forage	0.17	0.05
Sheep manure	0.14	0.02
Soybean straw	0.02	0.79
Pomace	0.02	0.31
Poultry manure	0.01	0.20
Leguminous residue	0.01	0.00
Vine shoots	0.01	0.12
Sunflower straw	0.01	0.03
Goat manure	0.01	0.01
Rice straw	0.00	0.02
Oil crop residue	0.00	1.12×10^{-5}
Industrial crop residue	0.00	2.26×10^{-3}

Table A8. ReCiPE 2016 (H) midpoint results for the regional assessment.

	Scenario Name						
Indicator	**BAV_AD**	**BAV_AD + Booster**	**BAV_AD + PHB**	**VEN_AD**	**VEN_AD + Booster**	**VEN_AD + PHB**	**Unit**
Fine particulate matter formation	3.27×10^6	2.69×10^6	4.04×10^6	-3.89×10^5	-5.44×10^5	-7.52×10^4	kg PM2.5 eq
Fossil resource scarcity	-2.06×10^9	-2.35×10^9	-1.70×10^9	-3.03×10^8	-3.49×10^8	-2.61×10^8	kg oil eq
Freshwater ecotoxicity	-2.54×10^8	-3.01×10^8	-1.13×10^8	-3.52×10^6	-5.72×10^6	-2.19×10^6	kg 1,4-DCB
Freshwater eutrophication	-1.12×10^7	-1.27×10^7	-5.39×10^6	-1.18×10^5	-1.52×10^5	-3.86×10^4	kg P eq
Global warming	-4.74×10^9	-5.49×10^9	-2.32×10^9	-6.40×10^8	-7.12×10^8	-4.42×10^8	kg CO2 eq
Human carcinogenic toxicity	-5.77×10^8	-6.54×10^8	-3.13×10^8	-1.48×10^7	-1.75×10^7	-1.07×10^7	kg 1,4-DCB
Human non-carcinogenic toxicity	-5.59×10^8	-1.75×10^9	3.52×10^9	4.02×10^8	3.56×10^8	4.72×10^8	kg 1,4-DCB
Ionizing radiation	-1.35×10^9	-1.53×10^9	-7.14×10^8	5.87×10^6	4.93×10^6	3.38×10^6	kBq Co-60 eq
Land use	1.21×10^7	1.14×10^7	1.31×10^7	1.95×10^6	1.93×10^6	1.76×10^6	m2a crop eq
Marine ecotoxicity	-3.62×10^8	-4.25×10^8	-1.67×10^8	-6.66×10^6	-9.65×10^6	-4.56×10^6	kg 1,4-DCB
Marine eutrophication	1.02×10^7	1.01×10^7	1.06×10^7	1.42×10^6	1.41×10^6	1.43×10^6	kg N eq
Mineral resource scarcity	-6.88×10^5	-8.63×10^5	-3.58×10^5	3.79×10^4	3.26×10^4	1.68×10^3	kg Cu eq
Ozone formation, Human health	-4.79×10^5	-1.75×10^6	1.65×10^6	-9.61×10^5	-1.23×10^6	-4.54×10^5	kg NOx eq
Ozone formation, Terrestrial ecosystems	-3.38×10^5	-1.62×10^6	1.68×10^6	-9.67×10^5	-1.24×10^6	-4.72×10^5	kg NOx eq
Stratospheric ozone depletion	4.86×10^4	4.79×10^4	5.07×10^4	5.93×10^3	5.83×10^3	6.21×10^3	kg CFC11 eq
Terrestrial acidification	3.26×10^7	3.09×10^7	3.57×10^7	7.66×10^5	3.04×10^5	1.82×10^6	kg SO2 eq
Terrestrial ecotoxicity	2.73×10^9	2.25×10^9	1.91×10^9	-7.79×10^7	-1.81×10^8	-1.27×10^8	kg 1,4-DCB
Water consumption	-2.27×10^{10}	-2.59×10^{10}	-1.26×10^{10}	-6.75×10^9	-7.73×10^9	-3.72×10^9	m^3

Table A9. ReCiPE 2016 (H) midpoint results for the scale assessment.

	Scenario Name						
Indicator	**S_AD**	**S_AD + Booster**	**S_AD + PHB**	**I_AD**	**I_AD + Booster**	**I_AD + PHB**	**Unit**
Fine particulate matter formation	0.08	0.07	0.09	0.00	0.00	0.03	kg PM2.5 eq
Fossil resource scarcity	−33.44	−37.61	−26.22	−16.66	−18.58	−13.79	kg oil eq
Freshwater ecotoxicity	−3.26	−3.83	−0.78	−0.11	−0.20	−0.02	kg 1,4-DCB
Freshwater eutrophication	−0.17	−0.19	−0.07	0.00	0.00	0.00	kg P eq
Global warming	−59.78	−70.58	−15.43	−37.69	−40.51	−25.06	kg CO2 eq
Human carcinogenic toxicity	−8.73	−9.67	−4.11	−0.80	−0.92	−0.52	kg 1,4-DCB
Human non-carcinogenic toxicity	−3.81	−18.33	67.59	52.53	50.63	57.37	kg 1,4-DCB
Ionizing radiation	−20.37	−22.38	−9.31	−0.10	−0.14	−0.28	kBq Co-60 eq
Land use	0.37	0.37	0.39	0.10	0.10	0.08	m2a crop eq
Marine ecotoxicity	−4.74	−5.52	−1.33	−0.24	−0.37	−0.10	kg 1,4-DCB
Marine eutrophication	0.37	0.37	0.38	0.07	0.07	0.07	kg N eq
Mineral resource scarcity	−0.01	−0.01	0.00	0.00	0.00	0.00	kg Cu eq
Ozone formation, Human health	0.04	0.02	0.08	−0.04	−0.05	−0.01	kg NOx eq
Ozone formation, Terrestrial ecosystems	0.05	0.03	0.08	−0.04	−0.05	−0.01	kg NOx eq
Stratospheric ozone depletion	0.00	0.00	0.00	0.00	0.00	0.00	kg CFC11 eq
Terrestrial acidification	0.61	0.59	0.67	0.24	0.22	0.31	kg SO2 eq
Terrestrial ecotoxicity	106.40	99.83	93.32	−5.38	−9.70	−9.08	kg 1,4-DCB
Water consumption	−331.10	−368.38	−155.31	−386.92	−428.04	−176.73	m^3

Table A10. ReCiPE 2016 (H) endpoint results for the regional assessment.

	Scenario Name						
Indicator	**BAV_AD**	**BAV_AD + Booster**	**BAV_AD + PHB**	**VEN_AD**	**VEN_AD + Booster**	**VEN_AD + PHB**	**Unit**
Fine particulate matter formation	2.06×10^{3}	1.70×10^{3}	2.54×10^{3}	-2.44×10^{2}	-3.41×10^{2}	-4.67×10^{1}	DALY
Fossil resource scarcity	-1.69×10^{8}	-2.06×10^{8}	-3.28×10^{8}	-8.80×10^{7}	-1.02×10^{8}	-8.53×10^{7}	USD2013
Freshwater ecotoxicity	-1.76×10^{-1}	-2.09×10^{-1}	-7.80×10^{-2}	-2.43×10^{-3}	-3.96×10^{-3}	-1.52×10^{-3}	species.yr
Freshwater eutrophication	-7.49×10^{10}	-8.53×10^{10}	-3.61×10^{10}	-7.90×10^{-2}	-1.02×10^{-1}	-2.57×10^{-2}	species.yr
Global warming, Freshwater ecosystems	-3.63×10^{-4}	-4.20×10^{-4}	-1.77×10^{-4}	-4.90×10^{-5}	-5.44×10^{-5}	-3.38×10^{-5}	species.yr
Global warming, Human health	-4.39×10^{3}	-5.09×10^{3}	-2.15×10^{3}	-5.94×10^{2}	-6.60×10^{2}	-4.10×10^{2}	DALY
Global warming, Terrestrial ecosystems	-1.33×10^{1}	-1.54×10^{1}	-6.49×10^{10}	-1.79×10^{10}	-1.99×10^{10}	-1.24×10^{10}	species.yr
Human carcinogenic toxicity	-1.92×10^{3}	-2.17×10^{3}	-1.04×10^{3}	-4.90×10^{1}	-5.82×10^{1}	-3.54×10^{1}	DALY
Human non-carcinogenic toxicity	-1.28×10^{2}	-4.00×10^{2}	8.03×10^{2}	9.16×10^{1}	8.13×10^{1}	1.08×10^{2}	DALY
Ionizing radiation	-1.15×10^{1}	-1.29×10^{1}	-6.05×10^{10}	4.98×10^{-2}	4.18×10^{-2}	2.87×10^{-2}	DALY
Land use	1.08×10^{-1}	1.01×10^{-1}	1.16×10^{-1}	1.73×10^{-2}	1.71×10^{-2}	1.56×10^{-2}	species.yr
Marine ecotoxicity	-3.80×10^{-2}	-4.47×10^{-2}	-1.76×10^{-2}	-7.00×10^{-4}	-1.01×10^{-3}	-4.79×10^{-4}	species.yr
Marine eutrophication	1.73×10^{-2}	1.71×10^{-2}	1.80×10^{-2}	2.41×10^{-3}	2.40×10^{-3}	2.42×10^{-3}	species.yr
Mineral resource scarcity	-1.59×10^{5}	-2.00×10^{5}	-8.29×10^{4}	8.78×10^{3}	7.54×10^{3}	3.88×10^{2}	USD2013
Ozone formation, Human health	-4.36×10^{-1}	-1.59×10^{10}	1.51×10^{10}	-8.74×10^{-1}	-1.12×10^{10}	-4.13×10^{-1}	DALY
Ozone formation, Terrestrial ecosystems	-4.36×10^{-2}	-2.09×10^{-1}	2.16×10^{-1}	-1.25×10^{-1}	-1.60×10^{-1}	-6.09×10^{-2}	species.yr
Stratospheric ozone depletion	2.58×10^{1}	2.54×10^{1}	2.69×10^{1}	3.15×10^{10}	3.10×10^{10}	3.29×10^{10}	DALY
Terrestrial acidification	6.92×10^{10}	6.55×10^{10}	7.57×10^{10}	1.63×10^{-1}	6.53×10^{-2}	3.86×10^{-1}	species.yr
Terrestrial ecotoxicity	3.12×10^{-2}	2.57×10^{-2}	2.18×10^{-2}	-8.80×10^{-4}	-2.06×10^{-3}	-1.45×10^{-3}	species.yr
Water consumption, Aquatic ecosystems	-1.37×10^{-2}	-1.56×10^{-2}	-7.61×10^{-3}	-4.08×10^{-3}	-4.67×10^{-3}	-2.25×10^{-3}	species.yr
Water consumption, Human health	-5.05×10^{4}	-5.75×10^{4}	-2.80×10^{4}	-1.50×10^{4}	-1.72×10^{4}	-8.26×10^{3}	DALY
Water consumption, Terrestrial ecosystem	-3.07×10^{2}	-3.50×10^{2}	-1.70×10^{2}	-9.11×10^{1}	-1.04×10^{2}	-5.02×10^{1}	species.yr

Table A11. ReCiPE 2016 (H) Endpoint results for the scale assessment.

	Scenario Name						
Indicator	**S_AD**	**S_AD + Booster**	**S_AD + PHB**	**I_AD**	**I_AD + Booster**	**I_AD + PHB**	**Unit**
Fine particulate matter formation	4.73×10^{-5}	4.24×10^{-5}	5.66×10^{-5}	2.24×10^{-6}	-1.85×10^{-6}	1.58×10^{-5}	DALY
Fossil resource scarcity	-2.77×10^{10}	-3.38×10^{10}	-5.28×10^{10}	-4.82×10^{10}	-5.39×10^{10}	-4.66×10^{10}	USD2013
Freshwater ecotoxicity	-2.26×10^{-9}	-2.65×10^{-9}	-5.40×10^{-10}	-7.75×10^{-11}	-1.41×10^{-10}	-1.68×10^{-11}	species.yr
Freshwater eutrophication	-1.12×10^{-7}	-1.25×10^{-7}	-4.47×10^{-8}	-2.19×10^{-9}	-3.14×10^{-9}	1.49×10^{-9}	species.yr
Global warming, Freshwater ecosystems	-4.57×10^{-12}	-5.40×10^{-12}	-1.18×10^{-12}	-2.88×10^{-12}	-3.10×10^{-12}	-1.92×10^{-12}	species.yr
Global warming, Human health	-5.54×10^{-5}	-6.54×10^{-5}	-1.42×10^{-5}	-3.50×10^{-5}	-3.76×10^{-5}	-2.33×10^{-5}	DALY
Global warming, Terrestrial ecosystems	-1.67×10^{-7}	-1.98×10^{-7}	-4.33×10^{-8}	-1.06×10^{-7}	-1.13×10^{-7}	-7.02×10^{-8}	species.yr
Human carcinogenic toxicity	-2.90×10^{-5}	-3.21×10^{-5}	-1.36×10^{-5}	-2.66×10^{-6}	-3.04×10^{-6}	-1.73×10^{-6}	DALY
Human non-carcinogenic toxicity	-8.74×10^{-7}	-4.18×10^{-6}	1.54×10^{-5}	1.20×10^{-5}	1.15×10^{-5}	1.31×10^{-5}	DALY
Ionizing radiation	-1.73×10^{-7}	-1.90×10^{-7}	-7.90×10^{-8}	-8.60×10^{-10}	-1.19×10^{-9}	-2.36×10^{-9}	DALY
Land use	3.31×10^{-9}	3.25×10^{-9}	3.45×10^{-9}	8.55×10^{-10}	8.46×10^{-10}	7.35×10^{-10}	species.yr
Marine ecotoxicity	-4.98×10^{-10}	-5.80×10^{-10}	-1.40×10^{-10}	-2.55×10^{-11}	-3.87×10^{-11}	-1.07×10^{-11}	species.yr
Marine eutrophication	6.31×10^{-10}	6.28×10^{-10}	6.42×10^{-10}	1.18×10^{-10}	1.18×10^{-10}	1.19×10^{-10}	species.yr
Mineral resource scarcity	-1.48×10^{-3}	-1.97×10^{-3}	-1.67×10^{-4}	5.79×10^{-4}	5.27×10^{-4}	-1.23×10^{-5}	USD2013
Ozone formation, Human health	3.77×10^{-8}	2.17×10^{-8}	7.49×10^{-8}	-3.71×10^{-8}	-4.72×10^{-8}	-5.28×10^{-9}	DALY
Ozone formation, Terrestrial ecosystems	5.87×10^{-9}	3.57×10^{-9}	1.09×10^{-8}	-5.31×10^{-9}	-6.77×10^{-9}	-9.10×10^{-10}	species.yr
Stratospheric ozone depletion	6.61×10^{-7}	6.57×10^{-7}	6.82×10^{-7}	1.84×10^{-7}	1.81×10^{-7}	1.94×10^{-7}	DALY
Terrestrial acidification	1.29×10^{-7}	1.24×10^{-7}	1.42×10^{-7}	5.03×10^{-8}	4.62×10^{-8}	6.57×10^{-8}	species.yr
Terrestrial ecotoxicity	1.21×10^{-9}	1.14×10^{-9}	1.07×10^{-9}	-6.10×10^{-11}	-1.10×10^{-10}	-1.04×10^{-10}	species.yr
Water consumption, Aquatic ecosystems	-2.00×10^{-10}	-2.23×10^{-10}	-9.38×10^{-11}	-2.34×10^{-10}	-2.59×10^{-10}	-1.07×10^{-10}	species.yr
Water consumption, Human health	-7.35×10^{-4}	-8.18×10^{-4}	-3.45×10^{-4}	-8.59×10^{-4}	-9.50×10^{-4}	-3.92×10^{-4}	DALY
Water consumption, Terrestrial ecosystem	-4.47×10^{-6}	-4.97×10^{-6}	-2.10×10^{-6}	-5.22×10^{-6}	-5.78×10^{-6}	-2.39×10^{-6}	species.yr

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Paper IV

PlastLCI for LCA: Accounting for ~~all~~ most impacts of plastic products.

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Abstract	A framework to inventory and account for plastic losses with special focus on secondary microplastic formation is introduced. The framework consist of a degradation module, which accounts for micro- and nano- plastic formation as particle distributions that continue to degrade for the time horizon of interest. An emission module accounts for the potential of the plastic particles to be emitted to air or ground compartments. Finally, an impact module characterizes microplastics impacts, as well as various decomposition gases from degradation of plastic in the environment, which can be integrated to existing life cycle impacts assessment (LCA) methods with ease. The framework allows for quantification of secondary microplastic in an LCA context and for further characterization of the impacts at endpoints in terms of human health and ecosystem damages. The framework is parametrized for easy application of local data and thus allows for a high level of regionalization, both in terms of input data and characterization of impact damages. The framework was tested on a case study of mulch film which shows that the per kg contribution to particulate matter and other impacts is low. However, when these impacts are scaled up to the European consumption of plastics and monetized it is evident that even small increases of particulate matter are costly for society and could potentially amount to millions of dollars per year in human health damages.
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Accounting for priority impacts of plastic products - PlastLCI a simulation study for advanced Life Cycle Inventories of plastics

Abstract

A framework to inventory and account for plastic losses with special focus on secondary microplastic formation is introduced. The framework consist of a degradation module, which accounts for micro- and nano- plastic formation as particle distributions that continue to degrade for the time horizon of interest. An emission module accounts for the potential of the plastic particles to be emitted to air or ground compartments. Finally, an impact module characterizes microplastics impacts, as well as various decomposition gases from degradation of plastic in the environment, which can be integrated to existing life cycle impacts assessment (LCA) methods with ease. The framework allows for quantification of secondary microplastic in an LCA context and for further characterization of the impacts at endpoints in terms of human health and ecosystem damages. The framework is parametrized for easy application of local data and thus allows for a high level of regionalization, both in terms of input data and characterization of impact damages. The framework was tested on a case study of mulch film which shows that the per kg contribution to particulate matter and other impacts is low. However, when these impacts are scaled up to the European consumption of plastics and monetized it is evident that even small increases of particulate matter are costly for society and could potentially amount to millions of dollars per year in human health damages.

1 Introduction

Plastic pollution has in the past decade come under the microscope of the public eye and the scientific community. Scientific studies examining various facets of plastic pollution have flooded the literature. Unlike other types of pollution, the problems caused by plastics are visible, which has perhaps motivated various countries to set targets for the elimination or reduction of some plastic products, such as plastic bags and disposable items [1]. In May 2018, a new proposal was passed by the European

Commission, banning several one-time use plastic items, and imposing stricter regulations on several other plastic products [2]. According to Giacovelli, (2018) there are now 60 countries that have either banned or imposed taxes on single use plastics. This puts into evidence how important the topic has become, both in terms of public perception and political willingness to act. Yet if plastics must be regulated due to increased evidence of negative impact [4], then their impacts must also be quantified. Plastics, which have been designed to optimize different material properties, such as flexibility, hydrophobicity, elasticity, and durability, among others, are well understood under laboratory conditions. However, when plastics are littered in the environment and exposed to the elements i.e. the combination of intermittent exposure to solar radiation, cooling, heating, drying, and rain, they begin to degrade, fragment and create microplastics. In turn, more and more studies of micro- and nano-plastic present evidence for the negative effects that these particles can cause on marine fauna [5–7]. By resent estimates, the amount of plastic entering the marine environment could be around 4.8 to 12.7 million MT in 2010 and is projected to increase by an order of magnitude by 2025 in a business as usual case [8]. Furthermore, an estimate for global mismanaged plastic predicts ca. 100 million MT per year in 2020 on average and reaching 220 million MT per year in 2060 if no preventative actions are taken to minimize the spill of plastic [9].

In order to accomplish truly sustainable and environmentally safe measures in regards to plastic products, it is necessary to evaluate them taking all possible issues into account. In this context, the less visible facet of plastic pollution, i.e. the issues caused by microplastics (MPs) and nanoplastics (NPs), is an expanding area of study. MPs and NPs, which may be present as particles or synthetic fibers starting in the nanometer range up to 5 mm, have been described as ubiquitous in the environment, by several authors [7,10,11]. These particles have been found everywhere from the poles to remote corners of the Pacific Ocean [12–14]. Their presence has been observed in a large variety of places e.g. on sea salt, honey, seafood, beer, breast milk and placenta [15–17]. Evidence of MPs' impact on human and ecosystem health is just beginning to emerge [17–21]. As more information comes to light, accounting for these impacts by means of life cycle assessment (LCA), a standardized methodology [22] designed to quantify the final damage of various substances, can be a step towards devising effective measures to regulate plastics. LCA is currently being used globally as a tool to support decision making in a policy context [23]. As it has moved from the academic realm, it has been adopted by for example, companies and governmental institutions, such as the European Commission's Joint Research Center, motivating various project like the LCA Initiative [24]. More recently, developments that incorporate life cycle thinking in the policy sector include the UN's Sustainable Development Goals, including goal 12 for sustainable consumption and production patterns [25]. Furthermore, life cycle thinking is being used to support green procurement and provide validity to greenhouse gas (GHG) claims, as it is the case for the Renewable Energy Directive recast (REDII)[26], and the Product Environmental Footprint (PEF) certification scheme. Thus, it seems a natural progression to employ LCA for quantification of potential problems due to plastics. Though LCA has come a long way, a need for further standardization is important and has been highlighted by various authors [27,28]. This is even more relevant when trying to quantify the multi-layered, interdisciplinary problems into which plastic develops when losses occur in nature. To date, a few attempts at devising numerical models to track losses of plastic in the environment and provide a plastic footprint have been published [29,30], and several methodologies are in development for providing an inventory of plastic losses [31]. However, to the best of our knowledge, most of these methodologies fail to make a connection between plastic loss and final impact, most of them do not consider secondary MP formation and none of them take into consideration the decomposition changes (size changes and gas products) that mismanaged plastic will experience throughout its lifetime. To date, only one attempt in which plastic losses have been given an environmental impact for ecosystem health was found [32]. Attempts to standardize inventory collection which, can be linked to a specific region of the world is also one of the missing components of most work found in the plastic loss inventories. Thus, many questions remain unanswered such as how various levels of infrastructure relate to plastic losses, or how various topographies influence redistribution of plastic in the environment, final fate and impact.

Presently, there are no guidelines on how to collect country specific inventory and include the impacts from MPs and NPs in LCAs of products. Moreover, the initial stages of plastic degradation leading to MP formation in the natural environment are poorly understood and rarely quantified. The issues related to permanence and thereby evolving impact potential of fragmented plastics is also lacking in the literature. Therefore, the aim of this work is to, create a framework for taking inventory of MPs and NPs that expresses regional differences, includes the time dependency of MP formation and emission and the subsequent changes of MPs in the environment, and quantifies the impacts to human health and ecosystem health with best available knowledge. The framework is demonstrated and tested on a small case study of low-density polyethylene (LDPE) mulch film, in two different countries.

2 Conceptual Framework

Figure 1 shows a visual representation of the conceptual framework developed to account for plastic impacts. The model consist of a degradation module, an emission module, and an impact module.

The degradation module includes UV damage to polymer surfaces by polymer type and plastic product usage. The model then accounts for further degradation of the polymer surface, as a function of exposure time, as well as consecutive degradation of the particles generated in the first instance i.e. month 1, year 1 or other time step. In this module, it is possible to link the degradation to a specific country, city or region by the use of site-specific irradiation data. Likewise, the emission module takes into account site-specific wind conditions and land cover, in order to predict a share of particles emitted to various compartments. Waste treatment capacity in the given location, is an important component of the inventory. Average waste treatment data is used in the model, which calculates a different potential for MP and NP formation depending on waste treatment methods, as well as influencing the possible share of plastic littered. Finally, the impact module takes the substances inventoried in the previous modules and produces impacts for the new emissions using conventional product system modelling software and the Life Cycle Impact Assessment (LCIA) methodology for assigning impacts to elemental flows.

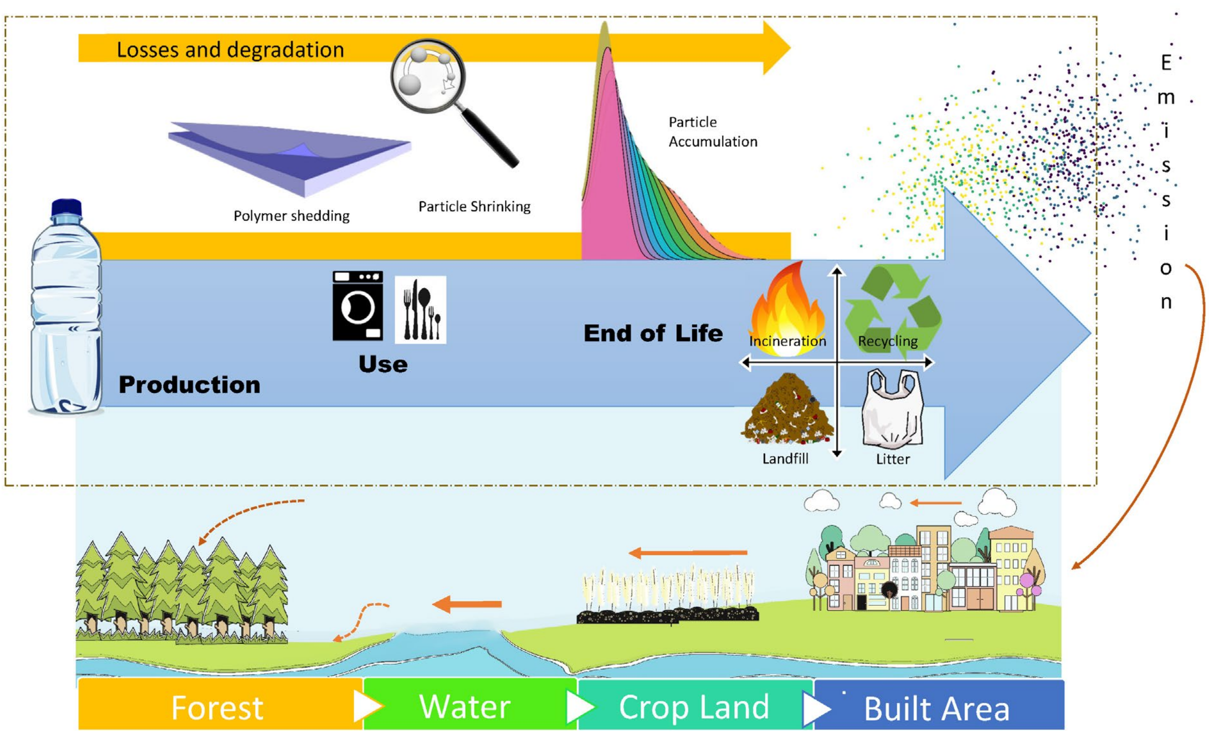

Figure 1 Conceptual framework for consistent inventory building for plastic loss and impacts in the environment. Dashed rectangle is the system boundary of the plastic's life cycle. The uppermost area shows the degradation aspects included in the model, such as progressive damage of the polymer surface, particle formation and degradation and particle accumulation through time. The different size solid arrows show the MPs potential for redistribution and emission to air, the dashed arrows show sinks.

3 Method

3.1 Degradation module

3.1.1 Particle formation and size distribution in UV conditions

Photo-degradation of different polymer matrices is extensively covered in the literature [33–36]. Modelling studies of for example, epoxy coatings have been used in the past to predict the lifetime of coatings when exposed to weathering, as well as particle distributions [37–39]. The approach taken by Lu et al (2018) is chosen here to approximate the initial degradation for polymers due to UV light. As defined in said work, the damage function to a polymer material is given by:

$$D_{total}(t) = E_o * \phi * t \tag{1}$$

Where D_{total} (dimensionless) is the effective damage dose to the polymer material with respect to E_o, the incident UV radiation dose (W m^{-2}), t, the time of exposure in seconds, and Φ , the quantum yield, which is the number of times an event occurs per photon absorbed, or in this case it can be interpreted as the ratio of bonds broken per photon at a given wavelength [37]. The quantum yield may be adjusted for each polymer by using the wavelength of excitement for the photosensitive groups known to each polymer [39,40]. For example, phenoxy groups in aromatic epoxy absorb the wavelength interval of 300-340nm, which corresponds to a quantum efficiency of 10^{-4} [37]. Whereas polystyrene's photo-oxidation is initiated by absorption by phenyl chromophore at 260 nm [41] and polypropylene (PP) absorbs in the ranges of 370 nm [42]. In general quantum efficiencies for various polymers range from 10^{-2} to 10^{-4} [43].

Equation 1 can be used to derive damage functions for the polymer material which may show a linear, power law or exponential response (more details available in Martin *et al.*, (2002) and Lu *et al.*, (2018)). Finally, equation 1 can be rewritten to derive a weight loss or thickness loss for the material in question [39]. The linear response is used in this work. Furthermore, the material loss occurs at the very surface of the polymer species, with ablation zones reportedly present from 2-100µm in thickness, such that polymer delamination happens gradually from the very surface of the polymer towards the inside [38].

Once a material loss has been calculated the model generates randomized particle diameter distributions equating the thickness loss (recalculated into a mass loss) predicted by equation 1. Assuming particles are spherical, a number of particles can be calculated using the density, ρ, of the polymer material and the particles volume. Where, the volume of a sphere is given by

$$V = \frac{4}{3}\pi r^3 \tag{2}$$

And V/ρ, gives the mass in µg of the average particle size (radius) in the particle size distribution. For simplification purposes, particles are assumed to be spherical, but it is clear that microplastic particles found in the environment come in different shapes, with some shapes possibly being more damaging than others [44].

Particle diameter distributions are adjusted to experimentally observed values whenever possible, since this has also been shown to be polymer specific e.g. polystyrene (PS) has been proven to produce nano-sized particles after just 14 days of 24h light exposure [45]. However, when it is not possible to obtain particle distributions from literature, then an initial distribution mimicking reported distributions in the marine environment is used as a starting point. Possible size distributions are summarized in Table 1.

Table 1 Sample of particle distributions found in the literature for a marine distribution and experimentally obtained values from accelerated weathering experiments of polyethylene (PE), polypropylene (PP) and expanded polystyrene (EPS) .

Radius Median[a] µm	**Marine[a] % by size**	**Radius Median[b] µm**	**PE[b] % by size**	**PP[b] % by size**	**EPS[b] % by size**
125	75%	150	97.170%	99.953%	99.984%
375	15%	400	2.830%	0.047%	0.012%
1500	10%	550	0.000%	0.000%	0.004%

[a] [46]

[b] Estimated from Song et. al (2017) [47]

The model then tracks each of the particles, at a constant degradation rate according to D_{total} for time t, where the time-step is dependent on the waste treatment pathway of the product. For litter particles may continue to degrade for 100 years or 500 years, depending on the time horizon of the LCA and the degradation rate. On the other hand, for sanitary landfill, an average UV exposure rate of 208 days per year was calculated based on management conditions for European landfills i.e. on average uncovered 4 days per week [48]. With each subsequent year after t_0 the polymer in question will proceed with polymer delamination i.e. a new set of particles will be generated each year for the time horizon of the LCA.

The sum of all micro- and nano- particles generated is then split into relevant damage size distributions and emission/fate of the particles is calculated.

3.1.2 Degradation into gaseous compounds

Gaseous products of decomposition such as CO_2, H_2O and CH_4, are part of the degradation module and should always be considered in relation to plastic degradation. Depending on degradation conditions being aerobic or anaerobic, the species of decomposition gases will differ [42,49]. Furthermore, morphology of the polymer i.e. whether it is a film, powder or fiber will have influence on the rate of degradation. Also, any biomass produced in the process due to microbial activity will also mineralize during the assessment's time horizon. For this work, gas production rates presented in Royer et al. (2018) are applied to the case study of mulch film, since this work presents gas production rates under various conditions. Average gas production rates under various conditions used for the case study are presented in Table 2.

Table 2 Average gas production rates for aged LDPE estimated from Royer et al 2018.

		Air		**Water**	
	Unit	**Dark**	**Light**	**Dark**	**Light**
Methane (CH_4)	C (nmol/g/d)	0.37	0.27	0.00	5.40
Ethylene (C_2H_4)	C (nmol/g/d)	0.14	48.16	0.00	5.10
Ethane (C_2H_6)	C (nmol/g/d)	0.21	0.16	0.00	1.40
Propene (C_3H_6)	C (nmol/g/d)	0.06	0.24	0.00	1.10

3.2 Emissions Module

Consider a MP source situated on the ground. The most important parameters, responsible for emission of the MPs from the surface into air are (1) the speed of the wind that the MPs is exposed to and (2) the size and shape of the MPs.

In this work the wind speed for one point in Denmark and one point in Italy over 11 years (time resolution of 1 hour) was obtained for 10 meter wind data, i.e. data for wind speeds at an altitude of 10 meter, from ERA4 from European Centre for Middle range Weather Forecasts (ECMWF). These data were transformed into two meter wind data, i.e. data for wind speeds at an altitude of 2 meter, for 7 different categories of land use for Denmark and Italy applying the wind shear formula [50]. The land use categories considered were bare soil, built area, cropland, forest, sand, shrub land, and water. For the second parameter (the shape) it was assumed that all the MPs were spherical.

Based on these assumptions a 2-dimmensional micro-physical model was developed [51]. The model is based on the theory of the oblique roll of the spherical object under the influence of air resistance. As input to the model, data from the MPs size distributions generated by the degradation module for 1 kg of plastic mulch film was used. The angle for the oblique roll in the model was 40%. This angle was chosen based on a study by Rice et al., 1995 [52].

The 2 dimensional micro-physical model determines whether the MPs will lift into the air or stay on the ground. The share emitted to air (in the range 0-10 µm) is considered a potential exposure to human health via inhalation. The remainder of the particle distribution is estimated to either be captured in soil or emitted to water ecosystems. There is a growing body of literature on the possible final fate of macro and MPs in the environment [10,13,30,53]. Due to the uncertainty related to determining the final fate of MPs in the environment, in this work, a crude value of 50% of the remaining particle distribution is estimated to be captured in soils, while the rest might end in waterways. Considering the long residence time of plastic in the environment and the light density of plastic materials, which increases the likelihood of their transport between compartments, it was deemed less likely that plastic sinks are permanent. Final fate for MPs and macro-plastics have been given various estimated values from only 10% reaching water bodies for tire abrasion particles [32], to between 10-40% of mismanaged plastic reaching the ocean [8]. More sophisticated modelling methods have been applied to predict sedimentation and buoyancy of plastic particles [54] and more granular estimates of plastic losses to watersheds [9]. It is clear from the wide range of values for plastic ending up in a particular compartment that this is a highly speculative area, but also that MPs have a high propensity for long distance travel as has been evidence by the work of [10] and [11].

3.3 Impacts Module

PM is a known air pollutant causing health effects in populations worldwide. PM consisting of "coarse particles" with aerodynamic diameters <10 µm, fine particles <2.5 µm and ultrafine particles of aerodynamic diameter <0.1 µm cause among other, respiratory damage due to, but not limited, to their small size, since their chemical composition and shape is also relevant. Fine particles and ultrafine particles are the most damaging since their small size allows them to penetrate deep in the lung. Ultrafine particles can penetrate the alveolar capillary membrane and reach any other organ in the body through the bloodstream. Thus, particles, though their point of entry is the respiratory track can lead to systemic effects causing damage in the rest of the body [55]. Furthermore, the final damage of PM is not only size related but also depends on their composition and structure. PM serves as a vector for heavy metals or other chemicals, depending on the source and may interact with allergens to induce asthmatic attacks [56]. In the context of MPs, additives such as bisphenol A and phthalates are just a few of the substances that may gain entry via inhalation. Furthermore, the biopersistence of MPs is another cause for concern, since they have been shown to accumulate in lung tissue, in for example lung biopsies of textile workers [57]. Within the LCIA area, mature characterization models are available that arrive at a final damage caused per unit of PM emitted. Thus, no further steps were taken to characterize the impact from MPs on human health, since we rely on the existing characterization models for this purpose. Though it is acknowledged that the combination of size/composition/shape of MPs and their final impact on human health is not thus far well characterized, which makes it likely that the impacts described in this work are underestimated. The emission module describes the share of particles of the whole particle distribution that is emitted to air or stays on the ground. The mass of the share emitted to air in the particle range of 0-10 µm is then used as input for the characterization models for further quantification of its induced impacts.

On the other hand, few studies have characterized the damage that losses of plastic might have on ecosystems [32,58–60]. For the most part, effect studies have been done in the laboratory with often higher concentrations than what is found in nature [61]. Toxicological effects of PE and PS particles have been observed in the laboratory, but are highly dependent on concentration. On the other hand, various studies have found negative effects on *Daphnia magna* communities and benthic organism at environmentally relevant concentrations [60,62,63]. In this work, the value for species loss derived by Ryberg et al. (2019) was applied to the MPs and macro-plastic loss calculated from the degradation and emissions module. As per the authors recommendations the value is adjusted by multiplying the lost mass of plastic by the fate factor and a species loss of 1.33×10^{-12} species.yr per year per kg of plastic (tire rubber in their work) [32].

3.4 The case study

Mulch film, which is typically used in agricultural applications for raising the temperature of soil and weed control was chosen as the product to test the PlastLCI model. As the model quantifies degradation due to UV damage, it was important to choose a product that is outside most of the year, or at times when not disposed of properly because it might be ploughed into fields or left on the field, all of the year [64]. For this purpose, an LCA was carried out that includes all life cycles of this product, that is to say the production of LDPE, use phase i.e. extending the film on agricultural land for 1 year, and end of life (EoL) with the following options: recycling, incineration, landfilling in sanitary landfill under European conditions, and litter. The baseline did not include litter as a possible end of life and did not include MPs formation during landfilling or during the use phase. Transport was excluded from all scenarios.

The LCA was performed from cradle-to-grave, with the system boundary extending from LDPE manufacturing gate to EoL gate. Co-products from EoL, that is to say, new LDPE granules from recycling and energy production from incineration, were not included in the study since this can be better categorized as an accounting LCA or situation A micro-level decision support [65]. The goal of the study is to explore the difference in impacts when including or not including impacts from MPs and littered macro-plastic, which also becomes MPs, concerning eco-design of products.

Two European regions were assessed, Denmark and Italy. The differences in waste treatment of these regions can be seen in Table 3. The recycling rates include a reject rate of 13% at sorting facilities and 17% of the incoming plastic waste at recycling facilities [66], which was estimated as the average European reject rate.

Table 3 Average regional waste treatment for Denmark and Italy. "-Slow" scenarios include MP formation via UV degradation while the "-base" scenarios do not include impacts from MPs.

	Recycling	**Incineration**	**Landfill**	**Litter**
DK-base	37%	60%	3%	0%
DK-slow	23%	65%	2%	10%
IT-base	31%	33%	36%	0%
IT-slow	28%	29%	32%	10%

A littering rate of 10% was applied to the non-baseline scenarios. It is difficult to estimate with accuracy how much of the plastic consumed in one region is littered or mismanaged. A littering rate of 2% was used by [8] for all European countries, while the share of mismanaged plastic varied from 15-40% of the waste produced by coastal populations. However, a recent study of mismanaged plastic waste with a spatial resolution of 1 km for the world, quantifies Europe's mismanaged plastic at ca. 11% of annual plastic waste generation or 3.3 million tons yearly (lower and upper range of 1.3-9.1 Mt) [9]. Thus, 10% was deemed a more appropriate value for this work, though this value was varied to test the sensitivity of the littering rate for overall results. The sensitivity of the model was also tested in regards to degradation rate. Values for degradation in the work of Chamas et al. (2020) [67], which include degradation rates for several polymers under various degradation conditions, were used as a point of comparison for the UV degradation rates calculated in the degradation module.

UV irradiance data was obtained from Solar Radiation Data website (SoDA) [68] for 3 points in each country of interest representing a wide coverage of latitudes in each country. UV data was averaged on a monthly basis for the three locations and used as input in the degradation module. Land use cover data, which is used in the emissions module to adjust wind conditions to the surface roughness of the area, was obtained from Eurostat [69].

The calculations were done using the data analysis tool R [70] where several scripts were developed for the assessment. Data generated from all three modules served as input to the LCA software OpenLCA [71] which was supplied with the Ecoinvent v3.6 database [72] and was used to construct the LCA model. Two characterization methods were used for the assessment: ReCiPe Hierarchist midpoint and endpoints [73] and ILCD Midpoint+ [74]. ILCD midpoints were then characterized to damage as endpoints with regionalized values for disability adjusted life years (DALY) and species.yr [75]. The damage at endpoints was then monetized using budget constrained ability to pay, which is the least uncertain way to assign a value to various damages [76]. Based on this method, a value of 65 000 USD_{2003} was used for species.yr and a value of 110 000 USD_{2003} per DALY [76,77].

4. Results

4.1. Model Results

The model provides various insights into the resulting fate and damage of MP particles formed through the degradation of plastic items. The case study focused on examining the fate and damage of particles arising from 1 kg of LDPE typically used as mulch film in agriculture to raise the temperature of the soil, protect against weeds, etc. [64]. It successfully links the waste treatment infrastructure and land cover of various countries to the final emission of MPs to the different compartments, making it possible to assess MP impacts in a regionalized manner.

4.1.1. Degradation rates and wind analysis

Summary statistics for wind data used in the emissions model per land cover is presented in Table 4. For simplification purposes, the 1st quantile was used as the minimum (Min) wind speed vector for the model and the maximum values were used as the high (Max). This is done to get as wide a representation as possible for wind speeds for each of the locations assessed, which are then implemented in the emissions module as minimum and maximum values for wind speed.

Table 4 Summary statistics of hourly wind speed in meters per second. Summary statistics are adjusted by the wind shear formula to 2 m wind speed, depending on land use type.

Denmark						
Land use type	Built Area	Cropland	Shrub land	Sand	Bare soil	Water
Min	0.00	0.00	0.00	0.00	0.00	0.00
1st Qu.	2.40	1.37	2.23	3.20	2.90	3.22
Median	3.44	1.96	3.18	4.57	4.15	4.61
Mean	3.58	2.04	3.32	4.76	4.32	4.80
3rd Qu.	4.64	2.65	4.30	6.17	5.60	6.21
Max	11.63	6.63	10.76	15.46	14.02	15.56
Italy						
Land use type	Built Area	Cropland	Shrub land	Sand	Bare soil	Water
Min.	0.01	0.01	0.01	0.02	0.02	0.02
1st Qu.	0.75	1.31	1.21	1.74	1.58	1.75
Median	1.09	1.91	1.77	2.54	2.30	2.55
Mean	1.23	2.16	1.99	2.87	2.60	2.89
3rd Qu.	1.58	2.77	2.57	3.69	3.34	3.71
Max.	5.71	10.01	9.26	13.31	12.07	13.40

Results from the degradation module predict a total yearly damage factor of 0.49 and 0.65 (dimensionless) for DK and IT respectively and a yearly damage factor of 0.28 and 0.37 (dimensionless) under landfill conditions, due to UV degradation. These degradation rates are used in the rest of the work as suggested by Lu et al. (2018) [39] and explained in 3.1.1 to derive a yearly mass loss as particle size distributions. The values agree with a higher overall irradiance falling on the Italian territory than in Denmark.

4.1.2. Particle repartition into various compartments

Surface roughness in combination with wind velocity determines the particle's ability to lift off the ground and potentially become a source of particle exposure for human health. Figure 2 shows how the various types of surfaces affect whether particles are emitted to air or stay on the ground, as a function of wind speed.

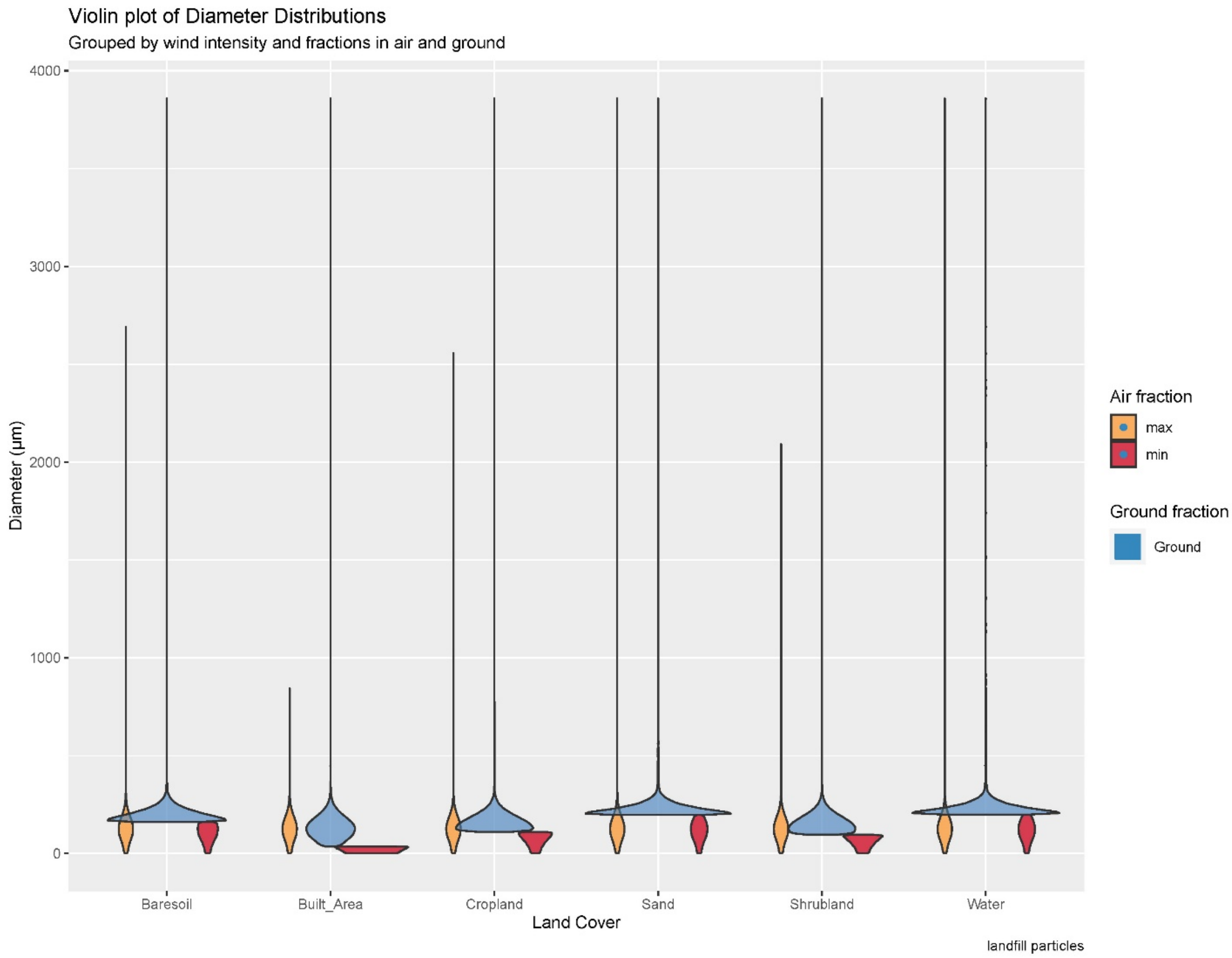

Figure 2 Violin plot of particle distribution count generated during sanitary landfilling of LDPE. Blue particles, with diameters higher than ~30 µm stay on the ground to various degrees depending on surface roughness. Red particles in the lower part of the particle distribution ca. <30 µm are lifted into the atmosphere even under low wind speed conditions (Min) while orange particles show the diameter sizes lifted into the air under high winds (Max) and include few particles of sizes > 500 µm in diameter.

It is clear from Figure 2 and Figure 3 that the smallest particles, those that are responsible for respiratory damage to human health (<10µm), are able to take off the ground under all wind conditions and land covers tested, as shown by the red distribution in conditions of low wind in Figure 2. By contrast, Figure 3 shows the diameter size of particles that are not able to lift off the ground. This includes the largest particles, mostly above 2500µm in diameter, for conditions of high wind (Max), with the exception of built up areas. Note, smaller particle ranges (purple) are also able to stay on the ground for low wind speed conditions (Min), though, particles of diameters lower than 10 µm are predicted to be emitted to air by the model.

Important to note is the fact that forest cover acts as a sink for MP and NP particles, since none of the particles are able to leave the ground (data not shown). This has important repercussion for geographical areas dense in forest cover.

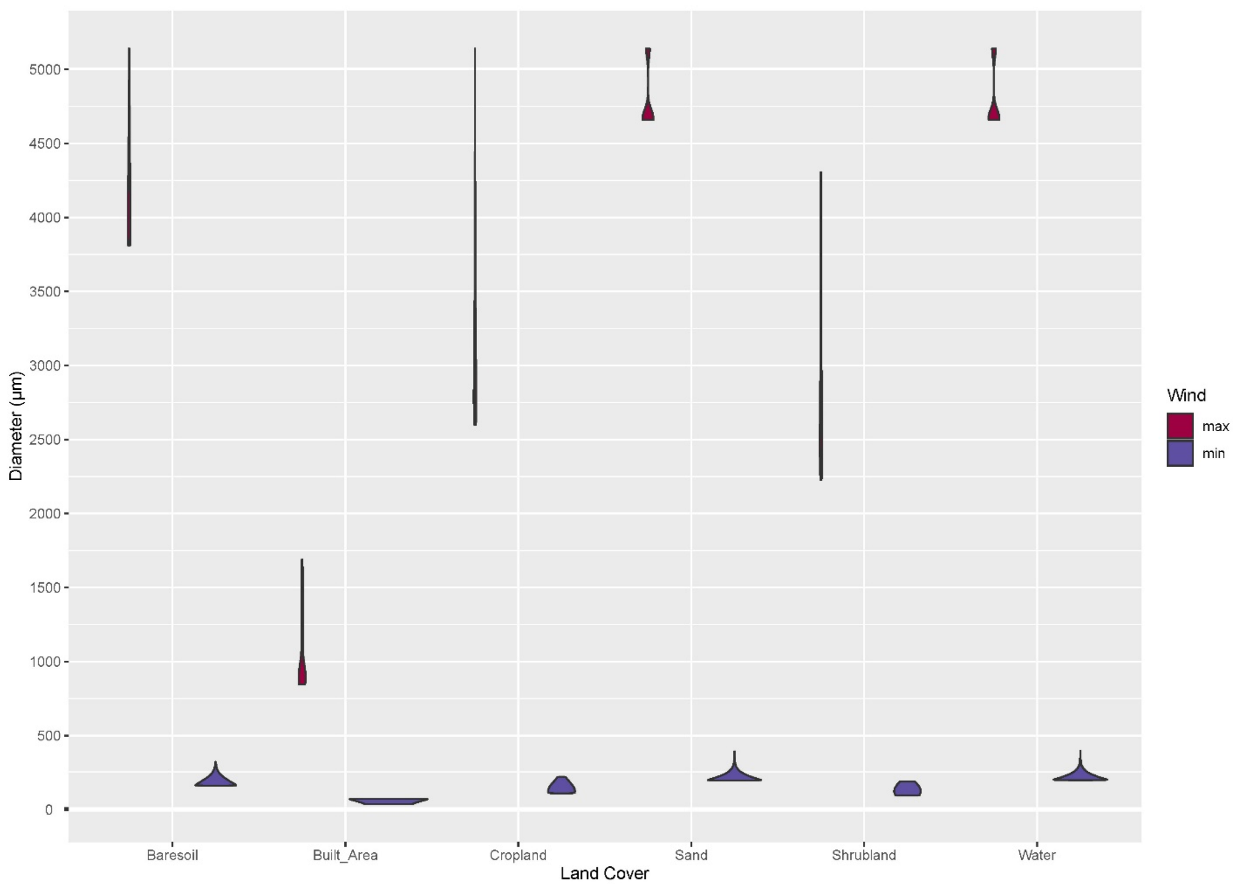

Figure 3 Violin plot of particle distribution count generated during littering of LDPE. Showing only particles that stay on the ground depending on high wind speed (max) and low wind speed (min).

The model groups particle by diameter sizes into different compartments, i.e. ground and air. The particles ending in the air compartment are further grouped into three groups according to size: PM_{10}, $PM_{2.5}$, and large particles in the air fraction (>11 µm in diameter).

The variation of the mass of particles ending in each of the compartments and standard deviation can be seen in Table 5.

Table 5: Share of mass of particle distribution emitted to air or remaining on the ground depending on wind intensity. Percentage share refers to the average mass in each compartment out of the total mass of the particle distribution in each life cycle (use phase, landfill and litter) and standard deviation in parenthesis.

	Wind Intensity	
Compartment	Min	Max
Ground	94% (± $1.43\text{x}10^{-03}$)	23% (± $2.37\text{x}10^{-02}$)
Air	6% (± $1.43\ \text{x}10^{-03}$)	77% (± $2.37\ \text{x}10^{-02}$)

In general, higher variation is observed under high Danish wind conditions (Max). The percentage of particles ending up in the air and ground compartments under Max conditions was 77% and 23%, while for Min conditions it was 6% and 94%, respectively. Furthermore, the percentage ending up in the PM_{10} fraction was higher for Min conditions than Max, since the mass fraction of the particle distribution ending up in the air compartment for low wind conditions is lower, thus PM_{10} is a larger share of the air compartment, $1.7\text{x}10^{-3}$ vs $1.4\text{x}10^{-4}$. The standard deviation for all PM shares is modest, thereby, the model is very stable and predicts a more or less uniform share ending up as PM for all life cycle stages assessed (Landfill, Litter, and the Use phase). About $5.03\text{x}10^{-6}$ % of the mass is in the range of $PM_{2.5}$ with Min conditions and $4.14\text{x}10^{-7}$ % for max conditions. The mass share of PM_{10} and $PM_{2.5}$ is very low, which is quickly understood when taking in consideration the mass of a single particle i.e. $4.83\text{x}10^{-13}$ and $7.55\text{x}10^{-15}$ kg, for a particle of 10 and 2.5 µm in diameter, respectively, and a density equivalent to PE. Thus, ~99% of the particle mass emitted to the air compartment belongs to particles of diameters >11 µm, which are heavier.

4.1.3. Particle evolution through the 100 year horizon

It is not enough to consider the effects of plastic particles for one year of degradation, since the formed particles in year 1 will continue to degrade over the following years, and in the case of littering, the release of a 1st set of particles in year 1 allows deeper layers of the polymer to be exposed to further degradation. Thus, in the subsequent years after the 1st set of particles is formed and is removed from the polymer surface, additional sets of particles are formed and continue to degrade the polymer. Additionally, all subsequent sets of particles continue to decrease in size during the 100 year horizon.

At a constant degradation rate, the 1st set of particle continues to degrade through 100 years, shown in Figure 4. Starting with an average diameter of ca. 130 µm at year 1, the 1st particle set reaches an average diameter of ca. 40µm in year 100. The particle distribution after 100 years has a considerably higher amount of particles in the damaging diameter range 0-10 µm, than after 1 year of degradation.

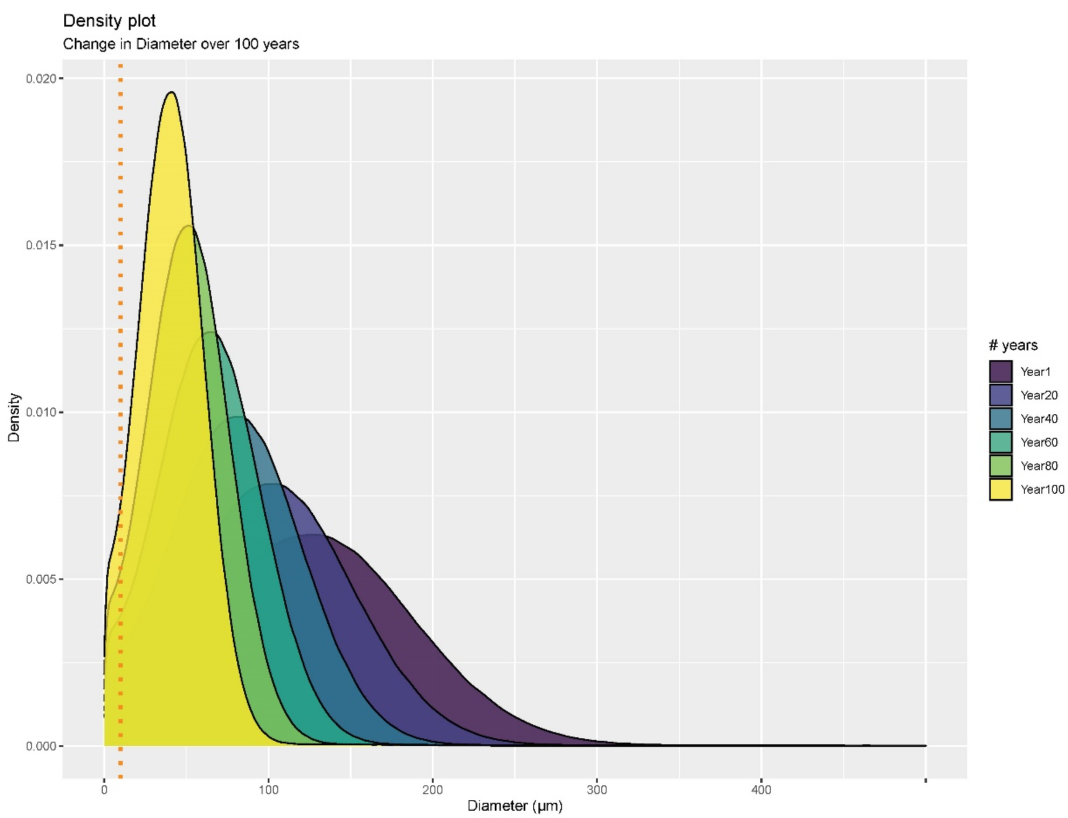

Figure 4 Density plot of the 1st set of particles formed due to UV degradation through a 100 year horizon.

To test the model, in order to see when the particle distribution would reach ca. 10 µm on average, the time horizon with the baseline degradation rate for Denmark was extended. Additionally, the degradation rate was tested with the values presented by Chamas et al. [67], since they use a very similar approach, but include additional types of degradation, other than UV, and accelerating factors of degradation in their model. Figure 5A shows the influence of a higher degradation rate on the size distribution of littered particles, hereafter referred to as the Fast degradation rate. The Fast degradation rate is ca. two orders of magnitude higher than the baseline degradation rate for Denmark. After about 18-20 years, the particle distribution with the faster degradation rate reaches an average size of 10 µm. This is in stark contrast to Figure 5B, where the particle distribution reaches ca. 10 µm on average after

200 years. This observation provides evidence for the importance of knowing how fast material degradation will occur under natural environment conditions.

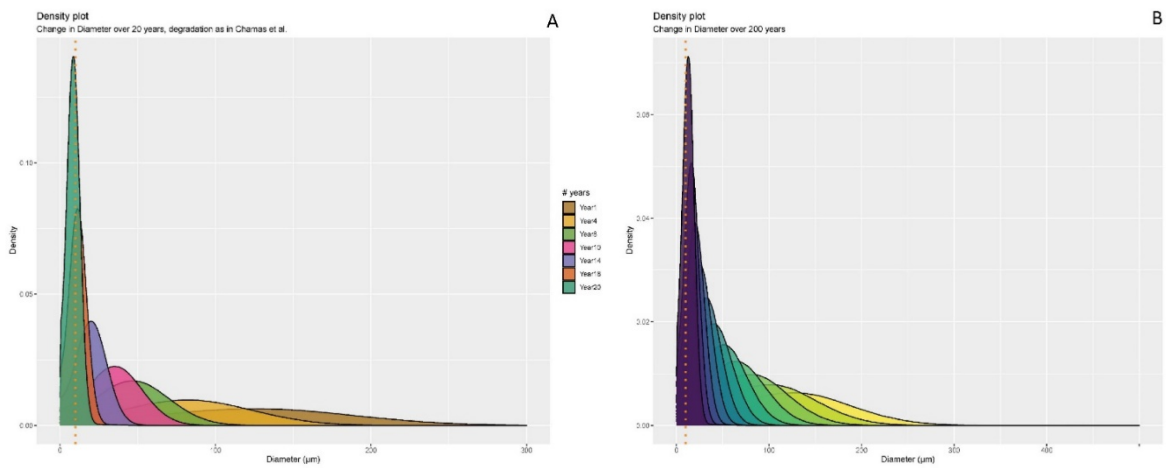

Figure 5 Density plots of particle distributions degrading through time, for A) a faster degradation rate as per Chamas et al. (2020) and B) the baseline degradation rate for Denmark with an extended time horizon of 200 years. The tails of the particle distributions have been cut at 300 µm and 500 µm for A and B, respectively.

The evolution of the total mass littered in comparison to the mass of particles in the PM 0-10 µm range can be seen in Figure 6. Figure 6A and Figure 6B show the logarithmic mass degradation. During this time step the mass of both PM ranges increases during the 100 year period and in Figure 6A it is a slow linear increase, while in Figure 6B the increase is sharp at the beginning and begins to level off toward the end of the time period. The slow increase in Figure 6A is due to bigger particles in the particle distribution degrading and becoming part of the PM range. The sharper increase in Figure 6B considers also the additional mass of additional particles sets being generated in the polymer and adding to the share of PM_{10} and $PM_{2.5}$. The same pattern is observed again in Figure 7, which shows the littered piece and PM particle mass evolution with the fast degradation rate described above. In the latter, it is possible to see the completion of the fate of the particle distribution. That is to say, a sharp increase in the PM levels followed by a drop as the total polymer mass continues to degrade and approaches zero. This pattern also explains the reason why a large part of the mass never becomes PM, since as PM increases there is also a share of the mass that essentially disappears from the particle distribution i.e. with constant degradation there is also a constant elimination of mass from the particle distribution or the mass of some of the particles becomes so small that it is practically undetectable. This is similar to experimental results observed by Song et al. [47] and colleagues who could only account for around 24%

of the initial mass of polymer after accelerated weathering experiments, and inferred that the unaccounted mass was in the submicron range and thereby undetectable.

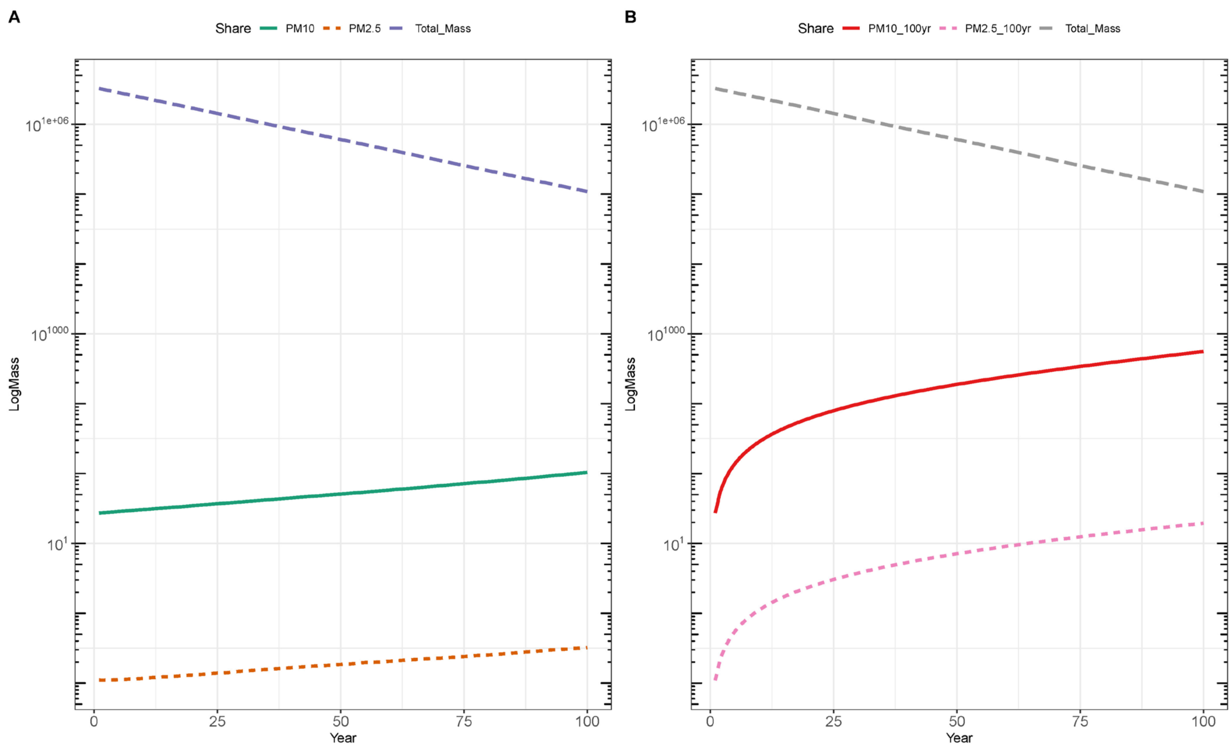

Figure 6 Degradation of total littered mass and formation of particulate matter in the 0-10 µm range for A) the 1st particle set as LogMass and total logmass of polymer and B) the 1st particle set and subsequent particle sets formed each year for 100 years as logMass.

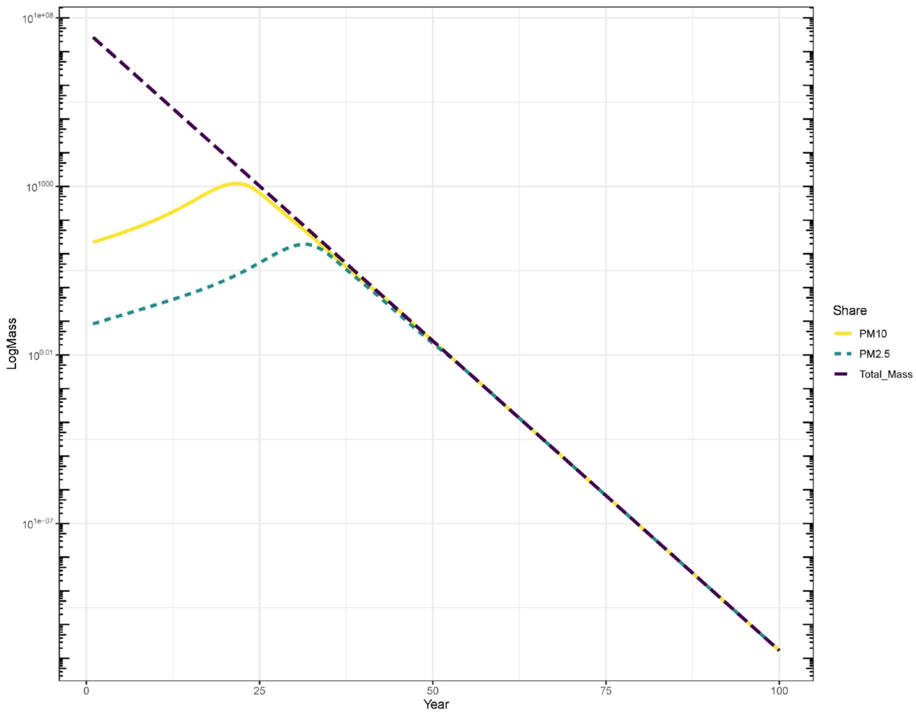

Figure 7 Degradation of the 1st set of particles with the Fast degradation in log scale.

After the 100 year period, and even when all particle sets are taken into account, PM_{10} makes up 0.024% and 1.24% of the initial littered mass for slow and fast degradation, respectively, while $PM_{2.5}$ makes up $8.66x10^{-07}$% and 0.012% of the initial mass for slow and fast degradation, respectively. That said, it is worth noting that the increase of PM_{10} and $PM_{2.5}$ within one year is of 592% and 376% respectively, for the Italian degradation rate during littering, and 280% and 194% for PM_{10} and $PM_{2.5}$, respectively, with the Danish degradation rate. However, the increase in PM_{10} and $PM_{2.5}$ mass is small, due to the very small masses of these particles and even when subsequent shedding of the polymer with a new particle set each year and the continued degradation of the sets thereof is accounted for, the mass of PM_{10} and $PM_{2.5}$ account for $2.2x10^{-5}$ kg and $8.07x10^{-8}$ kg, respectively, after 100 years of exposure. In comparison, the mass of PM_{10} is $5.6x10^{-7}$ kg and the mass of $PM_{2.5}$ is $1.9x10^{-9}$ kg after 1 year of exposure of the 1st particle set. Thus, the increase of PM is large, but does not amount to large masses due to the minuteness of particles.

4.2. LCA Results

PlastLCI relies mostly on recently published and thus existing characterization factors (CFs) for the developed impact assessment, i.e. substances tallied during the inventory are multiplied by a value (CF) which gives them a relative impact to a specific area of environmental interest a.k.a. an impact category.

Two different characterization methods were used to quantify the impacts induced by the elemental flows tallied here from MP formation. The two characterization methods included are ReCiPe 2016 and ILCD 2011, which are both standardized LCA methods, but differ in their classification of elemental flows (grouping) and CFs. For example, ReCiPe groups elemental emissions of ethane into two impact categories, Ozone Formation contributing to Human Health and Ozone Formation contributing to Terrestrial Ecosystems, as kg of NO_x-equivalents. While ILCD groups ethane into one Photochemical Ozone Formation impact category, as kg of non-methane volatile organic compounds (NMVOC)-equivalents. On the other hand, ReCiPe assigns a CF of 36 kg CO_2 equivalents to 1 kg of CH_4 emissions (the current consensus value according to the IPCC), while ILCD assigns it a value of 26 kg of CO_2 equivalents. Furthermore, ReCiPe lacks a CF for PM_{10} emissions, which are assigned a 0 contribution to particulate matter formation, while ILCD characterizes PM_{10} as 0.228 or 0.406 kg of $PM_{2.5}$-equivalents, depending on population typology (high or low population density). Thus, due to these know methodological differences it was deemed necessary to use both characterization methods, since ReCiPE is expected to underestimate impacts arising from particles, while ILCD will underestimate the global warming potential of the emissions of GHGs from plastic degradation, which are also included in PlastLCI.

The degradation of plastic in the environment leads to the creation of small particles, the release of NMVOCs such as ethane, ethylene, and propene and the release of the potent GHG, methane. These emissions were inventoried throughout the whole life cycle of 1 kg of LDPE used as mulch film and compared to a situation where the impacts from the degradation of plastic are ignored. The emissions contribute to three/four impact categories, listed first for ILCD and second for ReCiPe, i.e. particulate matter/fine particulate matter formation, Climate change/Global warming potential, and Photochemical Ozone Formation/Ozone formation for human or terrestrial ecosystems. The sensitivity of the model was tested for two variables, 1) an increase in the amount of littering, and 2) a fast or slow degradation rate.

4.2.1. Comparison to mulch film without MPs

Relative to a case where additional impacts of plastic are not taken into account, the addition of MP and NPs changes the overall impacts of the 1 kg of mulch film very little, as seen in Figure 8. As explained previously, the ReCiPe method does not assign an impact to most of the particles formed due to degradation, since these are in the PM_{10} range and it only accounts for $PM_{2.5}$. Photochemical ozone formation is also slightly underestimated. Thus, results from the ILCD method are given more focus from this point forward.

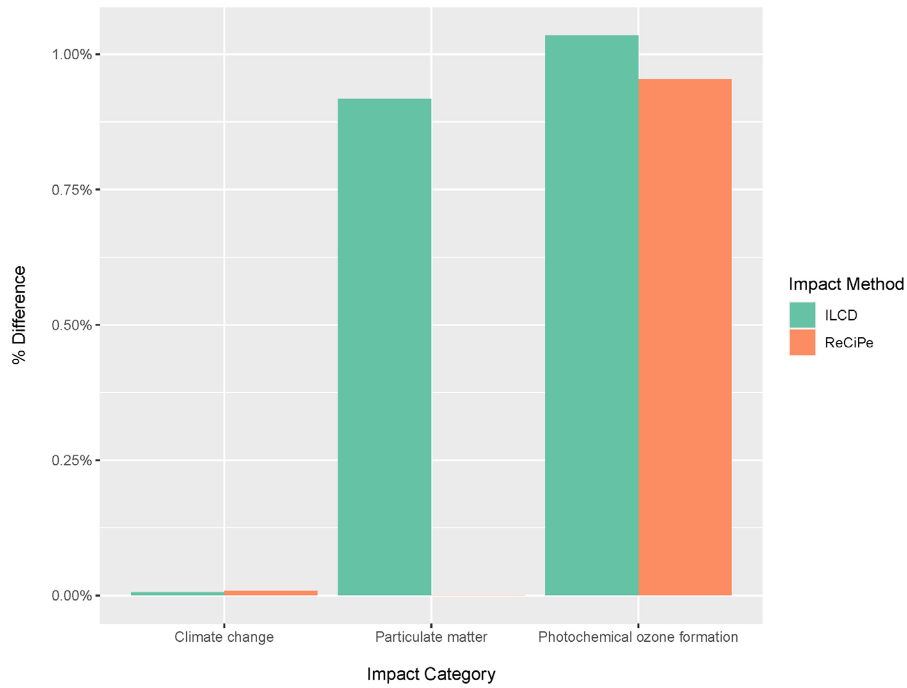

Figure 8 Relative change in Photochemical ozone formation, Particulate matter and Climate change potential relative to when degradation of plastics as MPs in the environment is not accounted. Difference between two life cycle impact assessment methods, ILCD and ReCiPe.

Looking at the contribution of each life cycle stage, here defined as the production of LDPE, the use of LDPE as mulch film and finally the disposal of mulch film via the waste treatment options of recycling, landfilling in sanitary landfill, controlled incineration, and finally the possibility of littering the mulch film. Figure 9 shows the contribution of each of the above named life cycle stages to three impact categories for LDPE without additional impacts (None panel) and for LDPE with the impacts of MPs under Danish degradation conditions (Slow panel).

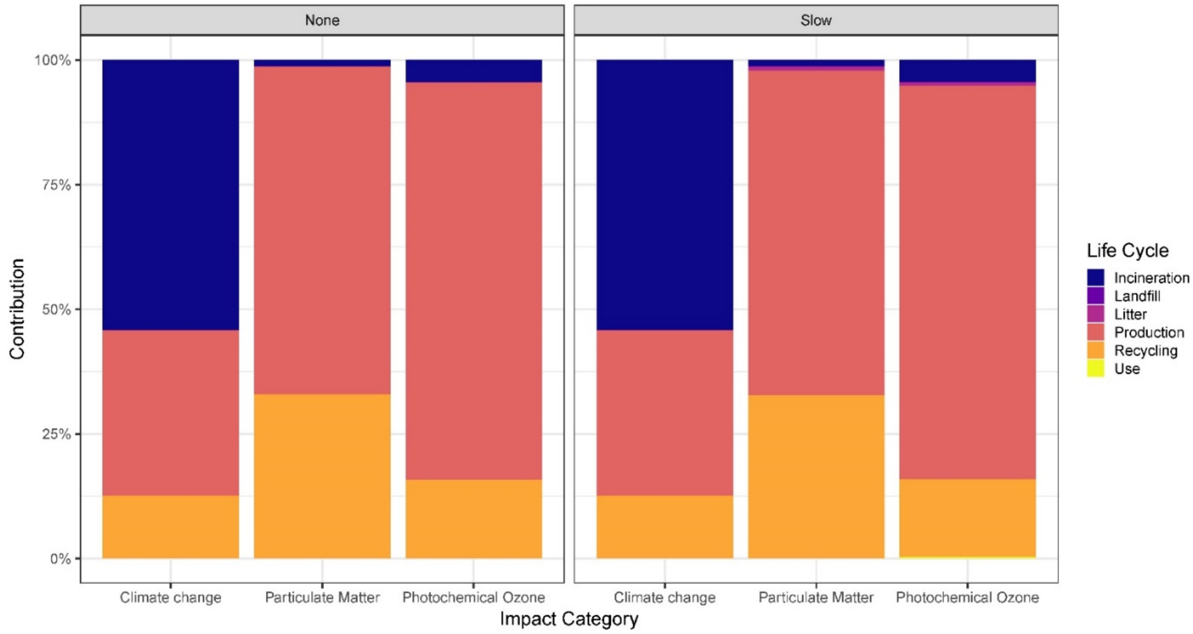

Figure 9 Contribution from each life cycle stage to the impacts of 1 kg of LDPE from cradle-to-grave for the plastic without additional impacts from the formation of MP, a.k.a. "None" for no additional degradation in the environment, and 1 kg of LDPE with MP impacts, here dubbed "Slow" according to the rather slow degradation for the baseline Danish case.

In comparison to production, recycling and incineration, the inventoried impacts coming from MPs and emission of NMVOCs and GHGs during the decomposition of LDPE in the environment, are small. Littering is the most burdensome of the MPs impacts, but accounts roughly for 1% of the impact for particulate matter formation and photochemical ozone formation. However, when the fast degradation rate is tested for sensitivity with the values for degradation reported by Chamas et al. 2020, noteworthy changes can be seen in particulate matter formation. With a faster degradation rate of MPs, the particulate matter burden increases by 40% compared to the MPs-less case, and 38% compared to the Slow degradation case. Moreover, the impacts of littering are now the second most burdensome impact contributing to particulate matter, second only after the production phase (Figure 10, Fast).

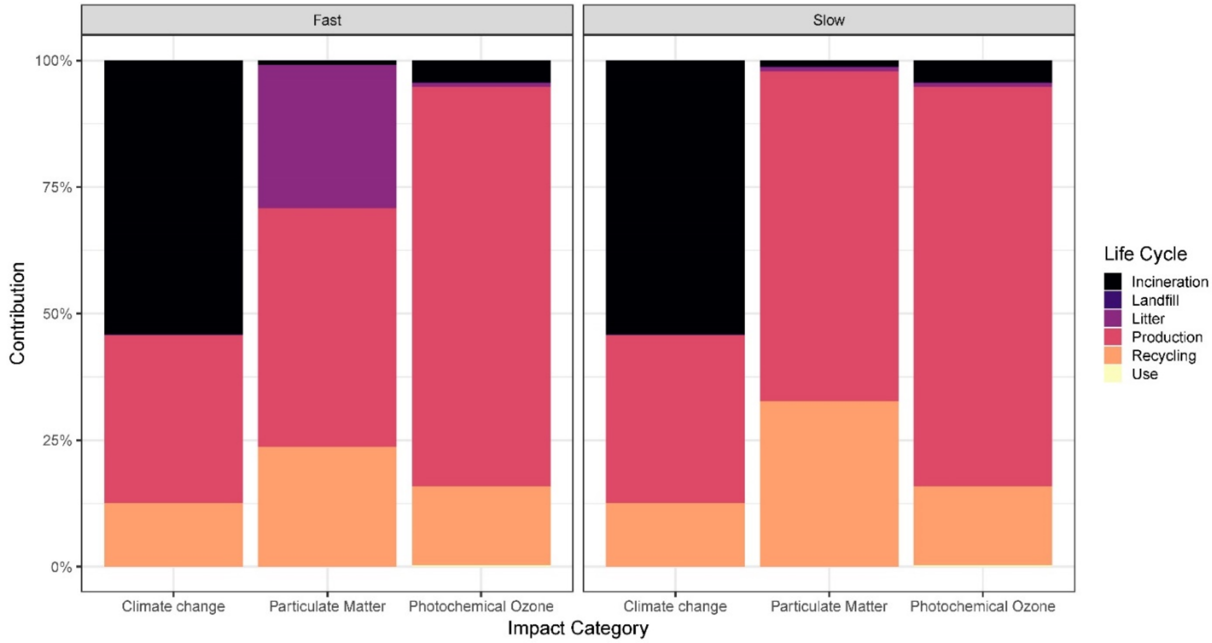

Figure 10 Contribution from each life cycle stage to the impacts of 1 kg of LDPE from cradle-to-grave for the plastic with a fast degradation rate (left) and slow degradation rate (right) with Danish waste treatment characteristics. Sensitivity was tested for only particulate matter formation.

4.2.2. Increased littering

Increasing the degree of littering, from the baseline value of 10% to 50%, 75% and 100% of the polymer, has the counterintuitive effect of lowering the overall impacts of the three impact categories studied. This is because when a larger share of LDPE is littered, a smaller share of the plastic is treated via recycling, incineration and landfilling. And, as can be observed in Figure 9, conventional EoL treatment options incur significant burdens. The LCA results show that results are rather impervious to a change in the littering rate, at least for the baseline “slow” degradation rate. A change of 40%, from 10% to 50% in the littering rate, incurs an increase of a mere 4% in the final impact of the three impact categories.

4.2.3. Regional differences

Particulate matter emissions from Italy are higher than for the Danish case, suggesting that the higher UV degradation rate in Italy is a more important factor than the forest cover of the locations assessed. Italy has a 33% forest cover compared to Denmark’s 11.4% of total land cover. The overall impact of waste treatment in Italy is lower, due to mostly a lower incineration rate. However, a higher rate of landfilling in Italy is visible, as there are higher impacts from the landfill contribution for Italy than Denmark.

4.2.4. Final damage assessment of MP impact

The increase in Disability Adjusted Life (DALY) years in Denmark, in comparison to the case when no MP impacts are included is of 0.06% per kg of LDPE mulch film, while species loss increases by about 0.01% for all scenarios. By contrast, for Italy the increase in DALY is of 0.5% per kg of LDPE in comparison to the Italian baseline. For the fast degradation tested under Danish conditions, the increase is of ca. 2.4% in comparison to the Danish baseline.

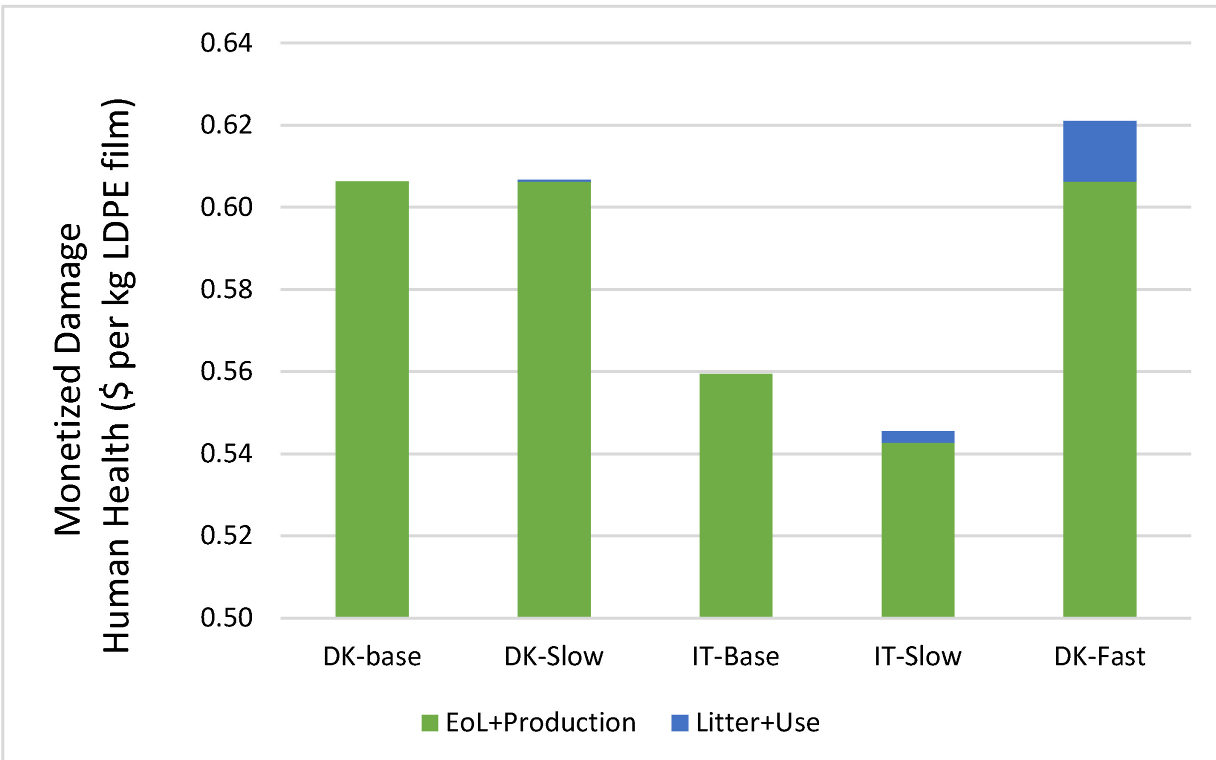

Figure 11 Monetized damage to human health per kg of mulch film. Derived from DALY valuated as 110,000 USD2003 per DALY. Scenarios with –base at the end do not include additional impacts from plastic litter and MPs. Slow refers to a slow degradation of plastic in the environment (only UV), and fast corresponds to a faster degradation rate, including various degradation factors.

When expressed in monetary terms (USD), the per kg contribution of monetized damage is low, only a few cents per kilo of LDPE, though more noticeable for IT and also for a faster degradation rate. Though these amounts may seem small when observed on a per kilo basis, it is informative to think about them in terms of, for example the EU's consumption of LDPE. Scaled up to European level, the additional impacts of plastic considered costs the EU between 3.5 million USD and 133 million USD per year in human health damages (slow and fast degradation, respectively) and around 800 USD in damages to species. If similar damages are considered for the whole of the EUs consumption of plastic (though this is better assessed on a per type of polymer basis), then the damage to human health could potentially cost from 20 million to 755 million USD, and ca. 4250 USD in damage to species. These values though highly uncertain should be considered carefully and refined as new information from risk assessment studies becomes available to LCA.

5 Discussion

The PlastLCI model may be a useful first step in generating complete inventories of plastic pollution that do not overlook the contribution of small particles. The conceptual framework presented can be applied to any plastic product, granted further data is needed for other polymers especially on potential gaseous decomposition products when degradation occurs in the natural environment. Furthermore, it was shown with the case study that it is fairly easy to perform the assessment in a regionalized manner, which is especially important for particulate matter contribution, since this is, for the most part, a localized phenomena (though it is also well known that PM can also travel far and wide [78]).

The case study showed that on a per kg basis, MPs formed during the use phase and landfilling are negligible in their potential for PM formation with the slow degradation rates predicted by the degradation module. Littering of the LDPE, however, has the potential to have a high contribution to particulate matter formation with a fast degradation rate and is not insignificant even with a slow degradation rate. The continued degradation of the particle distribution results in a large increase of particles in the PM_{10} and $PM_{2.5}$ ranges, but this increase does not show through in the LCA model, suggesting the PM formation impact category for both the ReCipe and ILCD methodologies may be highly unsusceptible to changes. The latter is a concern, since the LCIA methods routinely used for LCA should accurately represent the potential problems arising from PM. A CF factor of 0 for PM_{10} is incomprehensible given the evidence in literature for the importance of particles with aerodynamic diameters of <10 µm. Health problems arising from PM range from cardiovascular, to cerebrovascular and respiratory diseases, where small increases in PM have been shown to cause large increases in for example, cardiopulmonary mortality and respiratory admission [79]. However, due to this characterization approach, the Recipe methodology underestimates the damage caused by PM by an order of magnitude lower compared to the ILCD method.

In terms of NMVOCs, litter has the potential to be an important source of NMVOCs, which are precursors to tropospheric ozone formation and contribute to air pollution. This is true for LDPE which has been shown to degrade into compounds such as ethane and ethylene when exposed in air or water. If we contemplate the large quantities of plastic floating in the ocean and the recent predictions of future mismanage plastic worldwide, which are thought to reach 210 million tons on average per year by 2060 [9] it becomes clear that even small quantities on a per kg basis present a potentially large diffuse source of NMVOCs and the same can be said for PM formation. Furthermore, this work shows that particles of diameters <10 µm will be emitted to air where they will be a source of exposure. In this regard, the valuation of externalities presented in this work, which was estimated using a budget constrained ability to pay, may be useful in drawing attention to the high potential mismanage plastics may have in terms of monetary loss due to an increase in hospitalization and other costly health interventions.

Many questions remain unanswered in relation to plastic losses in the environment and their impacts. However, this work shows a viable way to include some of the impacts derived from the formation of secondary MPs from plastic products through their life cycles. A particularly pressing research need will be to determine with higher accuracy the degradation rate to which plastic products are subject when mismanaged, since this was seen to be a sensitive parameter of the contribution to PM. Furthermore, it is necessary to determine which gases are currently being emitted from other polymers types which undergo degradation in the natural environment and to what extent these contribute to different impacts e.g. global warming, NMVOCS. Lastly, this work analyzed the potential of particles to cause damage due to their small size, but the chemical composition of particles is an important, however, unassessed factor that may worsen the toxicological effects of PM. The addition of other chemicals and additives into plastic products which may then be inhaled should continue to be explored and integrated to LCA models, which may then be useful in improving product design or in providing guidance to decision makers.

6 Conclusion

A straightforward framework to account for secondary MP formation, emission and impacts throughout the life cycle of plastic products was presented. The framework allows for regionalized accounting and characterization of MP impacts, which show the impacts as endpoint damages by using already existing LCA methodology. The case study showed that the contribution of MP to particulate matter is low with a slow degradation rate, but may be an important contributor with a faster degradation rate that includes accelerating factors for polymer degradation in the natural environment. Furthermore, it was shown that small particles (<10 µm) are emitted to air under various land roughness and even at low wind speeds, and have the potential to become an source of exposure to human health. Decomposition of plastic in the environment may also be a potential source of NMVOCs, which might contribute to increase air pollution. Finally, valuation of damages at endpoints showed that when scaled up to European level, even small increases of PM are costly for society and could potentially amount to millions of dollars per year in human health damages.

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Supplementary Background Information to “Accounting for priority impacts of plastic products - PlastLCI a simulation study for advanced Life Cycle Inventories of plastics”

1 Sources of microplastic in the environment

Sources of microplastics in the environment, size ranging from 1µm up to 5mm, have been previously categorized as diffuse or point sources [1]. Point sources are, for example, waste water treatment (WWT) plants discharging purified water, which may still contain a large share of particles depending on the technology grade of the plant. Up to 99.9% can be removed in the more advanced WWT plants, but taking into consideration different levels of technology and the fact that only 15-20% of waste water is treated globally, waste water is an important source [2,3]. On the other hand, the sludge from these plants, which contains the brunt of particles, is often applied on agricultural land as biosolids, effectively making agricultural soils a sink for microplastics [4,5]. Nizzetto, Futter and Langaas, (2016) calculated that at the current rate of sludge application in Europe, where it is common practice to stabilize sludge into compost apt for agricultural use, about 63 000 to 430 000 tonnes of microplastics may be applied to land on a yearly basis.

Microplastics are further categorized as primary i.e. plastics that are already produced in the micro size range, such as exfoliating beads in personal care products made from polyethylene (PE), or extrusion pellets and acrylic and polyester beads used for various industrial uses. Both industrial and domestic products will be washed down drainage and end up in the sewage [7]. Secondary microplastics are likely to originate from everyday activities such doing the laundry, which will release synthetic microfibers in to the drainage in quantities of up to 1900 fibers per item [8]. As can be observed from these facts, it is likely that the most significant point source, both to water bodies and soils, is WWT works since these plants gather both primary and secondary microplastics in enormous quantities and it is unlikely that current sanitation techniques are able to remove all of it [6,9].

Some examples of diffuse sources include littering and accidental loses of plastic products during waste collection or losses from landfill sites exposed to the wind and rain events. Large macro-plastics become a microplastic source when they end up in nature. Fragmentation over time, results in smaller and smaller fragments since many plastic products become brittle when exposed to UV rays and changes in temperature [10]. Various human activities produce secondary microplastics. These include wear and tear of tires, dust from construction sites, and disintegration of plastic mulches and plastic tunnels used in agriculture. These travel through the air and eventually a fraction contaminates water bodies and soils. Recently, atmospheric fall out of MPs was proven [11–13].

MPs are measured in the environment by collecting, sieving or filtering. In the aquatic environment collection is often done with manta trawls and neuston nets, with a mesh size of 300 µm [14]. While for sediments, core analysis is a common collection method [15]. Analysis of collected MPs often includes flotation via density separation of the particles, which have densities lighter than sea water (for the most part), though biofouling causes them to sink. In the latter case, saturated solutions with higher densities can be used. The procedure to isolate MPs often combines flotation, and sequential filtration to achieve higher separation efficiencies [16]. For soil samples several techniques are used to first float the MPs and separate from organic matter (OM). Additionally, OM in the soil samples is dissolved using various type of treatments, including the use of acid or alkaline solutions, as well as enzymes [17]. Though techniques are improving, there is no standardized method for carrying out measurements and the lower limit of capture, due to for example mesh size, makes it difficult to measure very small µm and nm size particles [18]. Another hurdle is present when attempting to identify MPs. For this purpose, many methods have been employed including, visual identification, such as scanning electron microscopy, and spectral identification via Fourier Transform Infrared (FTIR) and Raman spectroscopies, which identify the polymers by their spectra. These methods are sometime combined with chemical identification methods such as pyrolysis and thermal desorption system gas chromatography, which increase the accuracy of identification. For more information on sampling and identification techniques the reader is referred to several reviews of these methods [16–18].

Previous study by [19] detected a parallel global increase of microplastics, as production of plastic products increased dramatically in the 50s and 60s. A large difference was seen from these decades, in comparison to the 80s and 90s where the increase was not as drastic. As plastic production continues to increase and its subsequent release to the environment, concentrations of microplastics in the environment will also continue to increase [10,20]. Moreover, a recent study on plastic waste production worldwide found a positive correlation between waste production rates and wealth while the correlation was negative for mismanaged waste, reflecting higher consumption levels in wealthier countries and at the same time better waste management systems (less mismanaged waste) [21]. Thus, plastic losses are intrinsically dependent on waste infrastructure, pointing out the importance of geographical granularity in assessments of plastic loss.

2 Degradability of conventional plastics and biodegradable plastics

2.1 Degradation pathways

Mechanisms for polymer degradation can be divided into biotic and abiotic pathways. When abiotically degraded, polymers may be subjected to hydrolytic, thermo-oxidative and photo- and photo-oxidative degradation, as well as mechanical and physical abrasion from natural elements. Biotic pathways, on the other hand, involve microorganisms that are able to use the polymer as carbon source for respiration and may produce extracellular enzymes capable of degrading polymer material. In part, the susceptibility of various polymer material to degradation is related to the polymer's "backbone", which may be composed purely of C – C bonds such as in the case of polystyrene (PS), polyethylene (PE), polyvinyl chloride (PVC) and polypropylene (PP), or is made up of heteroatoms, in polyethylene terephthalate (PET) and polyurethane (PU) [22]. Photo-degradation is, in most cases, the gateway that makes the polymer susceptible to other types of degradation. It starts when UV radiation breaks susceptible C – C bonds in the backbone, or at impurities in the backbone introduced during polymerization, causing a chemical reaction that produces free radicals. This in turn, leads in many instances to auto-oxidation (given that oxygen is present), chain scission, and finally formation of inert products such as aldehydes and ketones from olefins and many others. Photo-oxidation occurs at the polymer surface and leads to cracking, which exposes the inside of the polymer matrix to more degradation by any of the pathways described [23]. The monomers and oligomers produced by these reactions are many and different for each polymer type, some being more susceptible to biodegradation than others. Susceptibility to different kinds of degradation varies by polymer. Just to name a few, it has been observed that polymer structures that have repeating short and symmetrical units with hydrogen bonds, such as PE, PP and PET, are highly resistant to enzymatic attack, while PET undergoes both photo-oxidative and slow hydrolytic degradation and PVC is extremely sensitive towards UV light, which initiates dechlorination of its backbone [22]. The high variation between polymers reactions make it a difficult task to track degradation of polymers in the natural environment, which can be considered as unpredictable and highly heterogeneous. Moreover, blending of additives, UV-stabilizers and quenchers into plastic products is common practice, often carried out to increase the stability of polymers and lengthen shelf life of said products.

Biotic degradation, on the other hand, can only happen once the molecular weight of the polymer has been reduced sufficiently so that polymer fragments are small enough to pass through the cellular walls of bacteria or the molecular weight is first reduced by the production of extracellular enzymes that can accomplish the size reduction. Natural polymers such as cellulose and polyhydroxyalkanoates (PHA) depolymerize quickly, while polymers such as polylactic acid (PLA) require abiotic hydrolysis before bacteria are able to degrade it. Furthermore, PLA's monomers are known to recrystallize if temperatures are not high enough during degradation, making its degradation temperature dependent [24]. Whereas, natural polymers such as PHA degrade quickly in the natural environment due to the high number of bacteria species and fungi able to produce enzymes to degrade the polymer [25,26]. Biotic and abiotic degradation may take place in parallel or in chain.

2.2 Biodegradable plastics

Internationally agreed upon definitions for biodegradability where formulated at the International Workshop on Biodegradability of 1992, Annapolis, Maryland USA. This workshop gave birth to the ISO and ASTM standards which develop degradability tests for polymers [27]. Important agreed upon definitions from this meeting include, 1) The products of aerobic degradability are water, CO_2 and minerals 2) Degradability has to be consistent with the intended disposal pathway and 3) the material must degrade safely without causing negative impact [25]. Biodegradability can be further understood by looking at the different definitions developed under the many standardized tests that assess the degradability of material.

Robust tests to predict the ecotoxicity of substances are prescribed in the many sets of regulation governing compostable/biodegradable plastics. Regulation of the compostability of plastics is covered by different norms depending on the region of the world you find yourself in. In the USA it is defined by the International Organization for Standardization (ISO) 17088 [28] and by the American Society for Testing and Materials (ASTM) D6400 standard [29]. While in Europe it is regulated by EN 14995 [30]. When it comes to biodegradable polymers, the European norm is largely informed by the principles of the Registration, Evaluation, Authorization and Restriction of Chemicals (REACH), which aims to characterize individual substances in terms of human and environmental toxicity, as well as providing management guidance on disposal. In the case of ISO 17088 was modified so that components present in low concentrations, less than 10% of the overall material, are tested individually for biodegradation.

Some key findings from biodegradability norms governing compost include:

- 90% of mineralization must be achieved in 6 months where the product degrades into CO_2 and water
- 90% of the material must disintegrate into particles below 2mm in 3 months (12 weeks) in order to pass the level
- Toxicity of the material must be low on heavy metal content
    - Must contain limited amounts of substances with unknown biodegradability <1% concentration by weight and an overall sum of substances lower than 5%
    - Require that absence of polycyclic aromatic hydrocarbons and other persistent pollutants

A 90% mineralization rate within 6 months is accepted as safe because this level can only be attained by a fast and extensive degradation of intermediates into mineralization of inorganic end products, it is assumed that the remaining 10% will also follow this path [25]. If any recalcitrant substances are found then the eco-toxicological assessments must be applied. The standards allow substances with unknown biodegradation, as long as they are less than 1%. On the other hand, it is clear that due to the requirement that particles be smaller than 2 mm after 3 months, the existence of MPs in compost would potentially go unnoticed and be allowed under current standards.

2.3 Degradation kinetics

Several authors have quantified lifetime of polymers by using reaction kinetics, where k typically defines the rate at which the polymer species degrades in relation to parameters e.g. UV exposure, relative humidity, etc. [31–33]. These types of studies are highly relevant for environmental predictions of polymer degradation. As shown by Lambert *et al.*, (2013) and colleagues, multi-phase degradation kinetics were exhibited for latex exposed to winter and summer UV levels, as well as aquatic marine environment. Their experiment showed a breakpoint, where the rate of degradation described by the kinetic constant slowed down, at 44 days when the experiment was started in the summer and 105 days when started in the winter. Furthermore, hydrolysis of polymer bonds is a common step in the biodegradation of polymers such as PLA, PHB, and poly(ε-caprolactone)(PCL). For the bulk degradation of PLA, initial hydrolysis (bond cleavage) begins with uptake of water into the polymer, attaining steady-state hydrolysis, and subsequently an acceleration of the hydrolosis rate due to a higher level of acid chain ends, eventually leading to complete dissolution of the polymer [31]. Whereas, biodegradation of PHB starts at the surface of the polymer and occurs slowly until a threshold is reached for the polymer's mechanical integrity, after which point degradation occurs rapidly [35].

Aside from temperature, surface area has been demonstrated to be an important parameter for degradation kinetics. As a general trend, it has been shown that degradation of polymers occurs faster with decreasing size (larger surface area). Biodegradation of polybutylene sebacate (PBS) was shown to be dependent on surface area available to microorganisms, where pellets which had the least surface are degraded slowly [36]. Moreover, polymer thickness has been shown in various studies to have an influence on degradation rates, of nylon [34], PHA [37] PLA [24] and PHB(V) [38,39].

Empirical work describing degradation kinetics includes the work by [40] who calculate polymer degradation to be proportional to surface area. Thus the degradation rate, r_d is given by the differential equation:

$$r_d = \frac{d_m}{d_t} = k * SA$$

d_m: differential mass loss

d_t: unit of time

k: rate constant in ($kg\ s^{-1} m^{-2}$)

SA: surface area

The authors provide useful k constants for various types of polymers under various conditions of degradation, including PET, HDPE, LDPE, PP, PS, and some biodegradable plastics derived from a large literature review [40]. Degradation conditions include landfill/compost/soil, marine environment, biological degradation, and light induced degradation. Such work is an important contribution to determine environmentally relevant degradation rates for various polymer types.

3 Mechanism for MP dispersal and final fate: air, soil, water

3.1 Through the soil matrix

Important sources of MPs to land are losses from mismanaged waste, abrasion particles from tires and inputs to agricultural land through the application of sewage sludge, the latter being the most important point source [1]. Land receiving biosolids application was reported to have higher MP concentrations than those without application and the presence of MPs was detected even 15 years after the last biosolid application [4].

Once loaded onto the soil, microplastics have been shown to move throughout the soil matrix [4,41–43]. The way the particles will interact with soil will depend on various microplastic characteristics such as, hydrophobicity [42], size [44], charge, density and shape. Earthworms and micro-arthropods reportedly move microplastics along a horizontal axis [44,45], and smaller MPs moved more readily down the soil profile under lab conditions. Downward movement of MPs is also attributed to ploughing, cracks in the soil, and movement through macro-pores of decomposing roots and biopores left by biota. It is postulated that MPs would eventually reach ground water [46].

On the other hand, incorporation of MPs into soil macro-aggregates will serve as a sink and could potentially retain MPs for weeks to months [47]. Surface runoff will carry MPs to the aquatic compartment, but from there, it is not unlikely that the particles will make a full circle and return to land via flooding events, high tides, or wind erosion of dry sediments. It is also foreseeable that the amount of MPs remaining in the terrestrial compartment will exceed that entering the oceanic compartment for years to come [1].

More importantly, recent study showed that microplastics are transmitted through the terrestrial food web. Concentrations in soil, earthworm casts and chicken feces were studied and found to increase in that order [43]. MPs bioconcentrate in earthworms and potentially accumulate in earthworm tissue [48], while chickens mostly consumed macro-plastics from the soil surface, and the MPs found in chicken gizzards were thought to be generated during digestion. From this work, a consumption of around 840 particles/person was calculated from the consumption of chicken gizzards [43].

3.2 In the atmosphere

Little is known of microplastics in the atmosphere, but recent work by [11] and colleagues proved the atmospheric deposition of microplastic fibers to be an important source of MPs. The fibers where found to have a lognormal size distribution in the atmosphere, with fibers between 200-600 µm in length being more prevalent, though their lower limit of detection was 50 µm in length. The average diameter of the fibers varied from 7-15µm. Based on their results the authors calculated atmospheric deposition to be between 3 to 10 tons in the area of Paris and surroundings 2500 km^2, with 29% of fibers being of petrochemical origin. MPs have now been found in many remote areas of the world, where they have been carried by atmospheric currents. The North Pole has recently been described as a hot spot for sinking of MPs [49], while atmospherically transported MPs have also been found in remote mountain regions [12] and nature areas [13]. Furthermore, microplastics as an important component of urban dust have been identified as an important source of exposure for humans [50]. And, tire MPs have been estimated to make up from 0.1-10% of non-exhaust PM_{10}, and 3-7% of $PM_{2.5}$ [51]. Yet, other studies quantified indoor concentrations of MPs and found indoor concentration to be higher than outdoor concentrations [52]. Recently, a simulation of inhaled indoor air concluded that MPs make up a non-negligible share of inhaled particles, making up around 4% of all inhaled fibers [53].

The lognormal distribution of the particles in the atmosphere presented by Dris et al (2016) is similar to the distribution of aerosols [54]. Prevalence of aerosols is highest for ultrafine particles, including PM_{10} and $PM_{2.5}$, in the range of 1nm to 100µm, which will stay suspended for a longer than larger particles. Thus, intake of MPs via the airways is possible [55,56] and could be included into LCAs assessment as PM. The effects of PM are well established, and it is generally known that there is no level at which particulate matter is safe. In fact, an increase of PM_{10} of 10 µg/m3 showed an increase of 22% in lung cancer, while the same increase of $PM_{2.5}$ increases the chance by 36% [57].

3.3 Water

The aquatic compartment, including freshwater and marine water, has been extensively studied in relation to MPs. We refer readers to the work of Bergmann, Gutow and Klages, (2015), who presented extensive information of MPs in the marine compartment. We limit ourselves to a few important observations about the fate of MP in the aquatic domain.

Recent study modelled the fate of MPs starting from point sources in a river. The model was parametrized to initial polymer concentration, polymer density, particle size and presence of biofilm, but also collision frequencies and attachment efficiency to hetero aggregates [59]. This is important as the main mechanism of MP sink is believed to be through the formation of aggregates with other suspended solids and subsequent sedimentation [60]. Main model results showed particle size to be one of the most important parameters. Furthermore, retention was not monotonic in regards to particle diameter. Thus, retention was high for particle diameters <1µm and >50µm, with retentions of up to 60% and 100%, respectively. While particles in between this range (4µm) were highly mobile and exhibited up to 18% retention in sediments [59]. Furthermore, retention for particles with diameter 1-200µm depends on polymer density and increases with density. To sum up the authors concluded that nanometer and millimeter sized particles are very likely to be retained in river sediments, while micrometer size particles will be mobile and eventually reach coastal and marine ecosystems [59].

A study off the coast of California found that MP densities were highest at the bottom of the sea, while in the mid-depth water zone, densities were lowest [61]. Furthermore, plastic debris increased in the bottom of the sea after rain events [61]. In aquatic environments, floating debris is exposed to both photo degradation and hydrolytic forces, which makes material brittle. In areas where there is no sunlight, degradation is not biotic, but rather the polymers disintegrate into progressively smaller pieces [62]. While at the sea surf, mechanical abrasion also acts as a degradation force and may lead to an acceleration of MP formation [63]. An analysis of 5 km of coastline in Shandong Province, east China found a prevalent presences of MP in the sampled beaches. 50% of particles analyzed belonged to the 100-250 µm size range and polyurethane sponge MPs were detected for the first time. Surprisingly, unmanaged beaches contained the highest amounts of MPs, while highly managed beaches (tourist beaches) contained the least [64]. Distribution of MPs in waters of the East Asian sea coast of Japan were studied by trawling mesh and analyzing particle sizes. The size distribution is skewed left by the smallest particles being more abundant. Furthermore, after analyzing the size distribution the authors conclude that the origin of the MPs is from both the Yellow and East China seas, after repeatedly washing ashore and returning to the ocean [65].

4 Accounting for MPs through the life cycle of products

Here we provide guidance on how to account for potential MPs for plastic products. We take a point of departure in the individual product and product's disposal infrastructure. The latter is country specific as varying levels of development will influence how plastic products are disposed of around the world. The same is true for secondary microplastic arising from, for example, washing of garments, which might or might not be recovered at the local waste water treatment plant.

With the goal of quantifying the final microplastic emission from various plastic products, products are divided into two groups: P_1 is composed of all polymer types with the exception of polymers qualifying as P_2. Where P_2 are products composed of synthetic fibers and products which start out as microplastics such as microbeads in cosmetic products. Said subdivision is made because the dynamics of microplastic leaching during the use phase are differentiated for products of types P_1 and P_2. It is also important to note that P_1 or P_2 may also be composed of several polymers, so that P_1 may be 95% PE and 5% epoxy resin. The model does not account for additives and stabilizers.

Total microplastic leaching potential MP_{pot} is calculated as the sum of possible MP leaching occurances thorughout the products life cycle.

$$MP_{pot} = M_{pot,manufacture} + M_{pot,use} + M_{pot,EoL}$$

Where, $M_{pot,manufacturing,}$ $M_{pot,use,}$ $M_{pot,EoL}$ are potential microplastic leaching through the manufacturing, use and end of life stages of the life cycle of the plastic product in question.

4.1 Manufacture phase

Little is known of microplastics arising during the manufacturing stage of plastic production. To date, one study has been identified, where Lechner and Ramler, (2015) quantified up to 200 g of industrial microplastic per day being discharge into the river Danube. The authors further state that the amounts identified are within the legal limits for Austria, which is then estimated to allow up to 94.5 ton per year [7].

4.2 Use phase

Leaching of MPs during the use phase is product dependent. Factors can range from 0 when no leaching is expected from the products use, as in the case of disposable one time packaging, to thousands of fiber per wash cycle, as in the case of synthetic textiles [66,67]. For products in category P_1, where prolonged exposure to sunlight is expected, as in the case of mulch film and garden furniture, UV induced MP formation is calculated as described in section 3.1 of the accompanying paper. The total potential MP, $M_{pot,use}$ (kg), formation during the use phase is then given by equation 1, where m corresponds to the initial mass of the product and U_{leach} is the leaching factor for polymers of type P_2, while U_{wear} is the wear and tear factor for polymers of type P_1 (section 3.1), where applicable. Leaching factors for textiles, as well as other products may also be found in [68], where the authors provide MP inventory values for several types of products. These values can be used in conjunction with the methodology described here to obtain a complete inventory of MPs.

$$M_{pot,use} = m * U_{leach} + m * U_{wear}$$

4.3 End of life

An average approach is taken to handle quantification of MP during end of life (EoL). All products have an intended pathways for disposal, but unfortunately plastics are often incorrectly disposed of i.e. PET bottles are landfilled or incinerated instead of recycled, plastic bags or straws may be littered. Thus, an average approach where the EoL of the product mimics the national trend for waste management, including a fraction of mismanaged plastic (litter) can be representative for quantifying MP. The model is flexible enough to also check specific EoL disposal pathways for a specific value chain, if this was deemed necessary, e.g. 100% recycling.

Total rise of microplastics during EoL, $M_{pot,EoL}$, the sum of the mass of P_1 or P_2 post use phase, $mP_{1,post-U}$, multiplied by the fraction going to each specific treatment pathway, F_{treat}, derived from country specific waste management data, where F_{treat} is one of these four options: $F_{landfill}, F_{recycle}, F_{litter}$ and $F_{incineration}$ etc. The potential for secondary MP formation due to UV during each type of EoL, UV_{treat}, is only relevant for the fraction $F_{landfill}$ and F_{litter} , since prolonged periods of UV exposure are not expected for polymers being either recycled or incinerated. MP formation during treatment, is thus, denoted as $UV_{landfill}$ and UV_{litter}. Additionally, $UV_{landfill}$ and UV_{litter} are given by the damage equation (Eq 1) of the main paper.

$$M_{pot,EoL} = \sum_{0}^{t} mP_{1,post-U} * F_{i,treat} * UV_{j,treat}$$

As there are currently no reported values of littering from governmental institutions, a wide range of values is used in the literaturte for F_{litter}; the fraction of the plastic product that may be littered to the environment. F_{litter} is derived from Allen *et al.*, (2019), Jambeck *et al.*, (2015) and [21], where the authors use global production of plastic waste and amounts reported as treated, to derive a fraction of unaccounted waste. Using the values in Allen *et al.*, (2019) and assuming that 50% of the plastic produced stays in use for longer than one year, gives 27% of the plastic produced in Europe in 2016 as potential litter. Jambeck et al. on the other hand, applies a 2% of plastic in waste stream as litter fraction across all countries and has an additional fraction of mismanage plastic based on economic status, for which European countries contribute 0%. In this work, plastic littered is considered to be both accidental losses which is what is described by other authors as litter and mismanaged plastic which seems to be accidental losses connected to insufficient waste management infrastructure [69]. A more recent estimate of global plastic losses put's the European average loss at around 11% of plastic waste annually [21].

M_{pot} is reported both as a mass loss so that losses can be mass balanced, and as a number of particles with particle size distribution, which have implications for the final damage pathway (for more information refer to the main paper).

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PAPER V

A Methodology Concept for Territorial Metabolism-Life Cycle Assessment: Challenges and Opportunities in Scaling from Urban to Territorial Assessment.

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A Methodology Concept for Territorial Metabolism – Life Cycle Assessment: Challenges and Opportunities in Scaling from Urban to Territorial Assessment

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Abstract

To allow for the assessment of regional-scale geographically non-contiguous production system derived environmental impacts, a combined method of Territorial Metabolism – Life Cycle Assessment (TM-LCA) is proposed. By creating a two-pronged framework for the development of background system modelling, the TM-LCA method allows for process-based environmental impact modeling at a regional scale utilizing the concept of a production territory for the assessment of changes to durable production systems, such as energy infrastructure and agricultural systems. The TM-LCA framework creates the opportunity for direct assessment of environmental impacts, incorporation of system dynamics, and the use of multi-criteria decision analysis, which might be difficult or impossible to implement in other regional scale environmental impact assessment frameworks.

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Keywords: Urban Metabolism; Environmental Assessment; Regional Scale; Territorial Metabolism

1. Introduction

As the desire for more sustainable cities and regions increases, so too does the need for adequate and appropriate methods and frameworks for quantification of the environmental performance of such systems. The need for this type of sustainability quantification has led to the development of urban metabolism (UM) models (see Figure 1) and environmentally extended input-output models (EEIO), which can be of great utility, but are often somewhat lacking in terms of transparency and can often only quantify a limited number of environmental impact indicators [1]. Furthermore, studies based on EEIO models are not well suited for modeling prospective temporally dynamic systems. This is because any system dynamism would have to be incorporated into a characterization factor, in EEIO called the 'direct intensity vector' [2], that links an economic value with an environmental impact indicator. Normally, these factors are empirically derived from market data (i.e. trade statistics), which makes prospective assessment challenging, if not entirely precluded, as it would necessitate the mixing of both empirical and modelled data [2]. Due to the assumption of sector homogeneity, EEIO frameworks can also obfuscate the root cause of environmental impacts [2], making scenario generation difficult. While these limitations are present, UM or EEIO are often some of the only presently available methods for environmental assessment of large complex systems, such as systemic (regional scale multi producer or complex single producer) production, where a full process based LCA that encompasses the entire regional system would be impractical.

Nomenclature	
EEIO	Environmentally Extended Input Output
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
MCDA	Multi-Criteria Decision Analysis
TM	Territorial Metabolism
UM	Urban Metabolism

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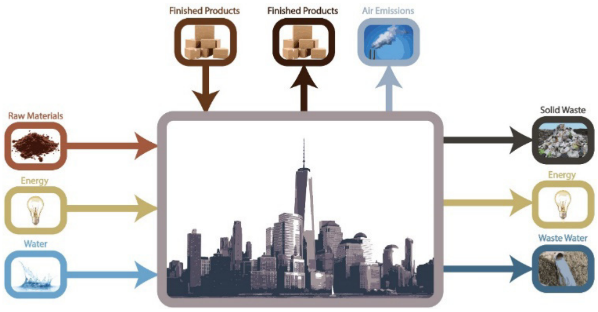

Figure 1: Conceptual visualization of a standard urban metabolism flow analysis

Table 1: Methods of large-scale environmental impact assessment. UM descriptions.

Method	Generalized characteristics
EEIO	Economically based system accounting utilizing a direct intensity vector to transform from economic flow to environmental impact. Can lack transparency and ability to define hotspots for improvement. Difficult to develop prospective temporal dynamics due to uncertainties introduced by mixing empirically measured and modeled estimates in the direct intensity vectors in dynamic EEIO. Due to the assumption of sector-homogeneity in EEIO, it is often not possible to assess a single producer.[2]
UM-Gen. 1	Based on material flow analysis. Can conflate material flow with environmental impact. Lacks incorporation of upstream and downstream processes.[1]
UM-Gen. 2	Based on UM with the inclusion of emergy assessment. Assumes all energy types are equal, missing variation in environmental impact from varying energy sources. Does not account for environmental impacts from other sources. [1]
UM-Gen. 3 (UM-LCA)**	Based on UM and incorporating LCA. Allows for greater transparency in determination of environmental impacts from given flows. UM Generation 3 is intended for assessment of systems at the scale of a city or urban area. Because of this, it lacks specific direction for expansion to larger scale assessment. [1]
TM-LCA***	Incorporates LCA in a UM-based method similar to a UM-G3 [1], but incorporates a framework allowing for the aggregation of multiple non-contiguous areas, which, when aggregated, are defined as a 'territory'.

In approaching a method for the quantitative assessment of the impacts of new system development (e.g. energy production or waste treatment) at a regional level, the methods developed in UM studies could offer utility in a more complex environmental impact assessment method. Recent study has shown that UM studies coupled with life cycle assessment (LCA), UM-LCA, can be effective as a tool for benchmarking the environmental performance of cities across a broader range of environmental impacts than previously possible with traditional UM studies [1], [3]. The increased range of potential impact quantification is particularly notable, as it is well documented that single indicator based environmental impact assessments, such as assessments based on greenhouse gas emissions, are often not representative of the entire environmental burden induced by a product/service or system [4], [5]. Additionally, by incorporating a process-based model, system dynamics could be introduced, allowing for prospective impact assessment for durable systems [6]. Furthermore, the incorporation of multi-criteria decision analysis (MCDA) in the impact assessment phase of the LCA would allow for better representation of environmental impacts [4].

In order to allow for the aforementioned incorporation of a process and life cycle based assessment system at a regional scale, a framework for implementation must be developed. By incorporating methodological elements from existing model types (as described in Table 1), such as UM or EEIO, an underlying methodological background can be developed. This forms a foundational model, to which a framework for implementation of territorial scale environmental assessment can be attached. Typically in an UM, material flows into and out of a geographically well-defined contiguous area are accounted for (Figure 1). The UM flow assessment approach can be effectively developed to an environmental impact indicator based assessment using the UM-G3 [1] method. However, when applied to a larger region, such an assessment can become too resource demanding (in terms of time, data, etc.). In order to manage this issue, we propose a new coupled-method of territorial metabolism-life cycle assessment (TM-LCA).

2. Methodology

In order to scale from urban metabolism to territorial metabolism, a framework for determining what elements should be included in the assessment should be made. In a traditional urban metabolism analysis, the flow of all materials into and out of a well-defined urban area are accounted for (Figure 1), but this might become either impractical or lack sufficient detail to be of use if applied at the larger scale of a region. In order to reduce the complexity of the system, while maintaining pertinent details, a scaling concept is applied to define the territory (Figure 2).

There have been a number of varying definitions of territory in relation to LCA [7], [8]. Typically, the territory refers to a "geographic space managed by local stakeholders [that] is characterized by a regional identity" [7]. For the purposes of this work, however a slight differentiation is made from this generalized definition to allow for more utility in the assessment. Rather than utilize the geo-political delineation of a territory (such as shown in the national and regional outlines in Figure 2), the territorial scale, or 'production territory', for the purposes of this work will be defined as the aggregated individual producers and the land within their geographic delineations contained in a defined region. Process data corresponding to the processes occurring among the aggregated producers (the territory) within the region and their interface with the surrounding background are modeled, while unchanging (non-related) background processes are ignored. This allows for the pinpointing of impact 'hotspots' while reducing the overall workload from what would be present if non-related processes were to also be included. The result of this division of modeled and un-modeled processes is an environmental impact assessment of a production territory. The approach only partially covering the territorial activities can hence not be used for an absolute assessment, such as a typical UM or EEIO would for the region, but is ideal for use in assessing the environmental implications associated with implementation of e.g. new production scenarios, new supply chain constellations or waste treatment technologies.

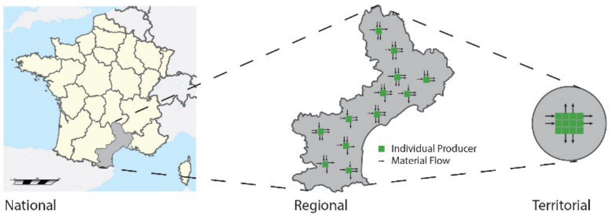

Figure 2: Scaling concept from national to regional allowing a process based aggregation of individual producers, shown in theoretical form for the Languedoc-Roussillon region in France.

2.1. Development from TM to TM-LCA

In order to integrate environmental impacts into the TM model, a conversion from material/service flow to impacts must be made. In EEIO and UM, there are differing methods allowing for quantifying the induced and avoided environmental impact (described in Table 1). To best-allow for the generation of scenarios for e.g. implementation of new technologies or production technologies, a process based LCA method is applied to the material and service flows across the boundary from the territory to the surrounding region. This can be accomplished by the incorporation of a standard database such as ecoinvent [9] for background processes in combination with user developed processes for scenario development. The development of these processes should as much as practically possible follow the ISO 14040 series standards [10]. It is likely, however, that the inclusion of e.g. system dynamics or MCDA will preclude some elements of the ISO standards. For example, in a dynamically developed product system where system expansion makes the product system excessively complex to assess, making allocation the only possible method. Or, if MCDA is used, then it is likely that explicit weighing would be included in published comparative results. In such a case, where ISO standards have been exceeded or compromised, it should be noted precisely.

2.2. Material Flow Data Development

In an ideal situation, primary life cycle inventory (LCI) data for all producers in a production territory would be available for use in the aggregated territorial process-based model. However, this is often not the case even when all producers in a production territory are owned by the entity wishing to complete a TM-LCA. In order to handle this shortcoming and increase the representativeness of the inventory used for the territorial model, a two-pronged approach is applied for modeling the territorial production processes where complete coverage by primary LCI data is not achievable. First, national or regional material flow data is applied and scaled to the territory. Processes are assigned to the material flows to create a material flow analysis based model. Then, if possible, a representative mix of individual producers are analyzed and modeled. The inventory data collected from individual producers is then scaled up to the level of the territory. This procedure provides indicator specific environmental impact ranges (Figure 3) that better reflect the actual environmental performance potential of the territory than if either (i.e. top up or bottom down) method were applied on its own.

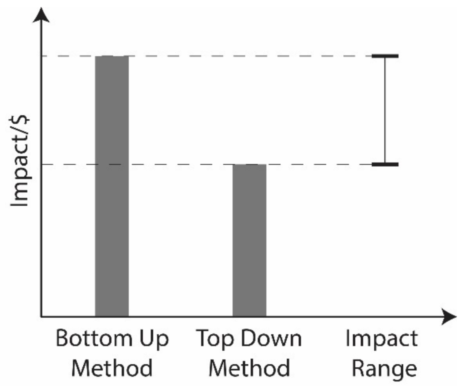

Figure 3: Bottom up and top down operational impact range visualization. Shown for economically normalized flow analysis.

To ensure completeness of the material flow analysis, ideally all material flows should be incorporated. This is, however, often not practically possible. Following the logic of ISO 14040 recommendations allowing for the omission of flows with less than 1% impact contribution [10], and in order to reduce data demands without compromising the resultant outcome, flows comprising 1% or less of the total mass-flow, or economic flow, depending on flow development method, could be omitted. This should not amount to more than 5% of the total flows. In addition, care should be taken to ensure that flows likely to produce a significantly stronger impact response per unit emitted are sufficiently assessed in a sensitivity analysis to ensure that their omission will not affect the results should they be included in the cutoff.

3. Discussion

For situations where a complex regional scale production system is to be assessed, such as in agricultural production, the TM-LCA method is well suited to provide detailed scenario analysis, in particular in the assessment of systems where multiple producers or a complex single producer cannot be adequately assessed using a standard LCA method. The environmental impact assessment inherent in the method can be extended to include non-environmental indicators, making it an ideal method for both regulatory bodies and commercial enterprise. This is made possible because the TM-LCA method is process-based, rather than e.g. homogeneous-sector economic flow characterization based as in EEIO, which creates the opportunity for further development of the model. This could include the incorporation of temporal system dynamics in the LCI, which might increase the validity of such a model [6]. Expansions could also include the incorporation of alternative methods of damage assessment such as MCDA, which could further increase the validity of the results if e.g. carbon-tied and non-carbon-tied alternative scenarios are being tested [4]. By using MCDA, there is also the opportunity to incorporate non-environmental factors into such a model, such as life cycle costing or ease of implementation metrics.

3.1. Potential challenges,

While the TM-LCA method offers great utility, there are also a number of challenges. One of the primary challenges in implementing a TM-LCA is data availability for the foreground processes and the representativeness of background processes used from existing databases. Care should be taken to check if a background processes are a dominant contributors to the results. If such a case occurs, the significant background process should be properly vetted to ensure that it is adequately representative; else, primary data should be procured. Furthermore, the completeness of the model must be ensured. In particular, if a flow cutoff is used to simplify the model, it should be demonstrated that this would not have a material effect on the results.

3.2. Potential Limitations, and Drawbacks

Apart from the challenges that face the TM-LCA method, a number of limitations and drawbacks should also be taken into account. One of the most apparent limitations is that this method, in most cases, will not provide an assessment of the entire region. As such, it cannot be used in lieu of UM assessment or EEIO. So, if such information is desired of an assessment a method such as UM-G3 [1] would be better suited. Also, the method is data driven and hence as LCI data becomes more and more diverse and hence case representative, the higher the quality of the conclusions that might be drawn from a regional assessment. As such, the results should be seen in an appropriate light with regard to data quality. Furthermore, the TM-LCA method requires more data collection than a standard process LCA for a typical single-producer's product, as such it will likely take more time and potentially incur more costs. Because of this, the potentially added value in relation to a product LCA should be considered in the decision to implement a TM-LCA.

3.3. Implications for production management and regional governance

The inclusion of process based life cycle assessment methods in the TM-LCA method allows for a number of assessment tools and perspectives that would otherwise be difficult to implement/represent with traditional UM or EEIO methods. For instance, because environmental impacts are directly measured in a process-based analysis, environmental hotspots in production can be identified, giving both governing agencies and producers valuable information. The process based impact assessment also allows for system dynamics to be incorporated, which is an important element in accurately assessing durable system implementations [6]. Furthermore, the inclusion of MCDA could also improve the ability to compare environmental impacts of certain types of scenarios [4] and allow for non-environmental impact assessment such as life cycle cost or ease of implementation. The potential for inclusion of these elements, as well as the target geographic scale, make the TM-LCA method ideal for informing regional governance bodies, trade associations, and large scale commercial producers.

4. Conclusions

The TM-LCA method is of great utility when applied for the comparison of regional scale sector or large-scale single producer implementation of alternative production methods and technologies as well as other system-scale change alternatives such as the implementation of alternative waste management technologies. By setting a consistent method for such assessments, the variability inherent in the implementation of ad-hoc solutions is avoided, which will become more imperative as the use of LCA in increasingly complex and varied types and scales of systems continues to broaden.

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PAPER VI

Techno-environmental assessment of the green biorefinery concept: Combining process simulation and life cycle assessment at an early design stage.

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Techno-environmental assessment of the green biorefinery concept: Combining process simulation and life cycle assessment at an early design stage

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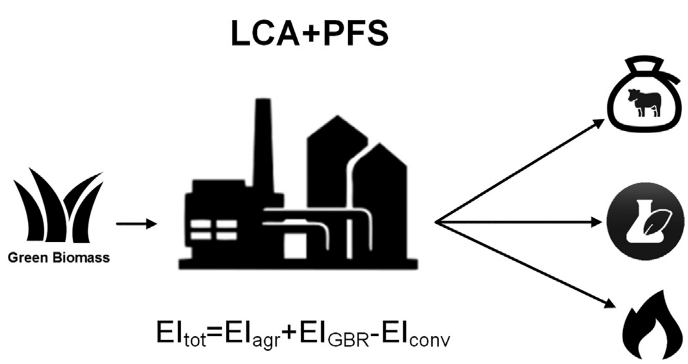
HIGHLIGHTS

- PFS + LCA combined to screen the environmental performance of different GBR setups.
- The GBR environmental profile is highly affected by the press-pulp utilization.
- Environmental savings to conventional products depends on the GBR configuration.
- Configurations prioritizing protein extraction efficiency lead to highest savings.
- Local protein-rich feed production can lead to reductions of climate change impacts.

GRAPHICAL ABSTRACT



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ABSTRACT

The Green biorefinery (GBR) is a biorefinery concept that converts fresh biomass into value-added products. The present study combines a Process Flowsheet Simulation (PFS) and Life Cycle Assessment (LCA) to evaluate the technical and environmental performance of different GBR configurations and the cascading utilization of the GBR output. The GBR configurations considered in this study, test alternatives in the three main steps of green-biorefining: fractionation, precipitation, and protein separation. The different cascade utilization alternatives analyse different options for press-pulp utilization, and the LCA results show that the environmental profile of the GBR is highly affected by the utilization of the press-pulp and thus by the choice of conventional product replaced by the press-pulp. Furthermore, scenario analysis of different GBR configurations shows that higher benefits can be achieved by increasing product yields rather than lowering energy consumption. Green biorefining is shown to be an interesting biorefining concept, especially in a Danish context. Biorefining of green biomass is technically feasible and can bring environmental savings, when compared to conventional production methods. However, the savings will be determined by the processing involved in each conversion stage and on the cascade utilization of the different platform products.

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1. Introduction

In recent years, the utilization of biomass for the production of feed, fuel and fibers has been suggested as one of the most promising solutions to fight climate change and reduce our dependence on petroleum derivatives. Several strategies have been proposed for the upgrade of biomass feedstock into a large array of products. Initial research, focusing on the utilization of food crops for the production of biofuels showed the technology to be technically and economically feasible (Worldwatch Institute, 2006). However, concerns were quickly raised regarding competition for land (i.e. the "food vs fuel dilemma") (OECD, 2008) and environmental savings, in comparison to traditional fossil fuels, were shown to be small (if any) when land use changes were included in the environmental sustainability assessment (Fargione et al., 2008; Searchinger et al., 2008). Since then, interests have shifted to the utilization of non-edible crops and crop residues in biorefineries: the so-called second-generation feedstocks. The goal of a biorefinery is to utilize all biomass fractions in order to maximize the product yield per biomass input, in the same way conventional refineries have been optimized to produce a multitude of products by exploiting all the crude oil components.

Biorefineries can be classified depending on the type of biomass feedstock they use (Bell et al., 2014; Cherubini et al., 2009; Kamm, 2013). The most prominent biorefinery concepts are: "yellow" biorefineries that utilize "dry" lignocellulosic materials; "green" biorefineries that utilize nature's "wet" grasses and immature crops; "blue" biorefineries that use algae; and "grey" biorefineries that utilize food waste.

Of particular interest, especially for the Danish context, is the green biorefinery. The green biorefinery aims at exploiting certain biomass components, which are generally lost during the maturation or drying of the biomass. Those components are generally water-soluble compounds, which become hard to fractionate when water is removed from the plant cell. The green biorefinery generally fractionates a "wet" biomass into a liquid stream and a solid stream (Xiu and Shahbazi, 2015). From these two streams, different cascading products can be generated depending on the processes involved (Kromus et al., 2006).

Three aspects make the green biorefinery concept interesting for the Danish scenario, as it can:

1) Decrease import dependency on protein-rich feed for the extensive Danish livestock sector.
2) Stimulate the local agricultural sector.
3) Increase synergies between different agricultural sectors (i.e. pig and poultry husbandry, dairy production and crop farming).

Due to intensive livestock production in Denmark, approximately 36 million tons of feed-products were consumed in 2015. While roughage and cereals are almost entirely produced in Denmark, approximately 80% of the protein-rich feed is imported (Bosselmann et al., 2015), and local production consists mainly of rape cakes and other by-products from the food industry. Soya by-product imports (cakes) account for approximately 50% of the total protein-rich feed consumption and 62% of the total import of protein-rich feed. Soya is imported mainly from South America directly, or re-exported from other EU countries, and a minor import comes from USA (Termansen et al., 2016). Fig. SI-5 in the Supporting Information (SI) shows the soy-based feed import to Demark in 2015. Due to socio-political concerns and environmental problems connected to soy production, as well as the added benefit of not paying for soya imports (Cong and Termansen, 2016), there is an active interest to reduce import dependency and look for local alternative protein sources (Hörtenhuber et al., 2011; Lehuger et al., 2009). In the SI, Table SI-17 presents the consumption of the most important protein-rich feed in Demark between 2010 and 2015.

The second aspect is the intensification of Danish agriculture. Cereals occupy a majority of Danish farmland and knowhow on cereal cultivation is estimated to be already at its maximum. Thus, limited improvement can be achieved in countries where intensive farming is already practiced. Annual crops such as cereals cannot use a significant part of the solar radiation during the growing season for photosynthesis and biomass production. Cereal crops mature from mid-July, are harvested in August, are re-sown in September and green fields are not seen until the very end of the year (Termansen et al., 2016). Grasses on the contrary, are perennial crops that can utilize solar radiation available year-round, achieving higher DM yields on a yearly basis (Gylling et al., 2016). Having a permanent soil cover, not only allows for higher yields, but can also bring benefits by minimizing nutrient losses. Eriksen et al. (2014) compiled a catalogue of measures, which may be used to mitigate nitrogen leaching in Denmark. In this context, the conversion of land from cereals to permanent intensive grass has been suggested as a relevant mitigating measure. In addition to reductions of nitrogen leaching, the transition from annual crops to perennial crops is also expected to lead to an increase in the carbon stock in the soil, due to the larger root system, and it will also lead to a decrease in the pesticide use compared to cereals (Jørgensen and Lærke, 2016).

Finally, the green biorefinery concept could improve synergies between pig and poultry farmers, dairy farmers and crop producers, on the one hand developing alternative and local protein sources and on the other intensifying the use of arable land (Cong et al., 2017; Gylling et al., 2016).

Given the potential of GBR to bring benefits across sectors, several projects have focused on developing the technology. Despite the common basic technology, i.e. fractionation of a wet feedstock into a liquid and a solid fraction, different process configurations and different target products have been developed, see Table 1. Several techno-economical assessments have been published, suggesting that the GBR could be feasible and economically competitive (Kamm et al., 2016; O'Keeffe et al., 2012; Sinclair, 2009). However, until now, knowledge on the best configuration remains limited, due to limited penetration and implementation in the biorefinery market. Furthermore, few studies have looked at the environmental sustainability of the GBR system (Cong and Termansen, 2016; Corona et al., 2018; Parajuli et al., 2017a) and none have focused on finding the most sustainable GBR value-chain.

Therefore, the present study performs a techno-environmental assessment of different GBR configurations. In order to estimate the technical performance of different GBRs, at an early design stage, a Process Flowsheet Simulation (PFS) of different GBR configurations was developed. The PFS, based on experiments and production trials performed at a pilot plant in (Foulum, DK), estimates material and energy input, as well as quantity and quality of the products for each configuration. The PFS's results were used to populate the inventory of the LCA model, in order to screen the best configuration in terms of environmental performance, to identify hotspot and focus points for the technology developers within the conversion pathway. Finally, a sensitivity analysis was used to look at the effect of process optimization on the environmental performance of the GBR.

2. Materials and methods

2.1. Description of the green biorefinery

A GBR pathway can be described in five main steps:

1) Biomass cultivation
2) Fractionation
3) Precipitation
4) Protein separation
5) Downstream processing of the GBR output

Table 1
Overview of the different GBRs concepts developed in Europe.

Country	Product from cake	Product from juice	Product from residuals	Reference
Netherlands	Ruminant feed	Protein feed	Biogas	www.grassa.nl
Germany	Feed pellets	Protein feed/food	Biogas	(Kamm et al., 2010)
Germany	Composite material	Protein feed	Fertilizer	www.biowert.de
Switzerland	Composite material	Biogas	Biogas	(Sharma and Mandl, 2014)
Austria	Biogas	Lactic acid Amino acid	Biogas	(Ecker et al., 2012)
Ireland	Composite material	Protein feed Lactic acid	Biogas	(O'Keeffe et al., 2012)
Denmark	Silage Feed	Protein feed	Biogas	(Ambye-Jensen and Adamsen, 2015)
Denmark	Grass pellets	Lysine	Lysine	(Andersen and Kiel, 2000)

A general description of each step is presented in the following paragraphs. Additional information is available in the SI.1. The studied GBR has a capacity of 20,000 ton_{DM}/yr, and is assumed to operate between May and October. Data for the different processes in the GBR, as well as estimates of a realistic process optimization, were obtained from experiments and production trials performed at a pilot plant facility at Aarhus University, located in Foulum, Denmark (Hermansen et al., 2017). Energy consumption for each process in the GBR value-chain is presented in Table 2. Data for alternative cascade utilization of the press-pulp and for the production of human-grade protein were based on values taken from literature (Kamm et al., 2009; Kromus et al., 2004).

2.1.1. Feedstock

Alfalfa (*Medicago sativa*) was assumed to be a representative feedstock for the GBR process, as it is one of the most important foraging crops in Europe, covering 7.12 million ha in 2010 (Kamm et al., 2016). The inventory for the cultivation of alfalfa in Denmark and transportation to the biorefinery gate was based on Parajuli et al. (2017b). Alfalfa is assumed to be a rotational crop, with a rotation cycle of three years and three harvests per year (Jørgensen et al., 2011). Yearly yield of alfalfa was assumed to be 12 ton_{DM}/year. The complete inventory for the agricultural stage of alfalfa is available in the supporting information, SI-2.1.

2.1.2. Fractionation

The fractionation step aims at separating the liquid stream i.e. the press-juice, from the solid fiber-rich fraction, i.e. the press-pulp. The biomass is initially shredded and fed into a screw-press were it is mechanically separated into the liquid and solid stream. The fractionation step can be performed once or twice, with a washing step in-between. In this study, the baseline consists of a single-step pressing. As an alternative, after the first pressing, water is added to the press-pulp to achieve a $25\%_{DM}$ concentration, which is then re-pressed, resulting in a two-step pressing (O'Keeffe et al., 2011b). The latter allows for a higher extraction of soluble components, proteins and carbohydrates. One-step pressing has a protein extraction efficiency of 45%, while with a two-step pressing the overall extraction efficiency can be increased to 65% (DCA, 2016). After the fractionation stage, the two streams are separated and sent to the following downstream processes.

Table 2
Energy consumption of the different biorefinery processes.

Process	Unit	Energy	Source
Shredder	kWh/ton_{DM}	20	BioValue
Fractionation	kWh/ton_{DM}	11.1	BioValue
Anti-foaming agent	$Kg/m^3_{press\text{-}juice}$	1	BioValue
Thermal precipitation	$MJ/m^3_{press\text{-}juice}$	294	BioValue
Pumping	$kWh/m^3_{press\text{-}juice}$	1	BioValue
Biological precipitation	$MJ/m^3_{press\text{-}juice}$	84	BioValue
Centrifugation	$kWh/m^3_{press\text{-}juice}$	11	BioValue
Drying heat (35%–60% DM)	MJ/ton_{water}	2000	(Grabowski and Boye, 2012; Mujumdar, 2014)
Drying heat (60%–95% DM)	MJ/ton_{water}	5000	(Grabowski and Boye, 2012; Mujumdar, 2014)
Drying electricity	$kWh/ton_{product}$	2.64	(Grabowski and Boye, 2012; Mujumdar, 2014)
Human-grade protein separation and drying	$kWh/ton_{product}$	428	(Kamm et al., 2009)

2.1.3. Press-pulp utilization

The press-pulp- or fiber-rich fraction- mainly contains the insoluble components of the biomass. This fraction is rich in structural carbohydrates, predominantly cellulose, and contains the fiber-bound proteins, as well as residual non-separated soluble proteins. This study analysed three different cascade utilizations for the press-pulp. Studied utilizations include:

1) Use as ruminant feed
2) Use as composite material for insulation
3) Use as feedstock for lysine production

2.1.3.1. Utilization as ruminant feed. In this scenario, the press-pulp is ensiled and used as animal feed. The scenario aims at utilizing the carbohydrates and fiber-bound protein content as feed for ruminants, which requires that the press-pulp is ensiled and stored for further use. When utilized as ruminant feed, the press-pulp must meet a minimum protein content requirement of 140 g/kg DM of the dried press cake (Kamm et al., 2010). The ensiled press-pulp is assumed to replace alfalfa silage.

2.1.3.2. Utilization as composite material for insulation. This scenario aims at utilizing press-pulp fibers in thermal insulation panels. The press-pulp is initially dried to reach a DM content of 92%. Subsequently, the dried fibers are mixed with Borax to increase fire resistance, and be compliant with current fire and safety standards for buildings. The product is assumed to enter the insulation material market and replaces other conventional insulation panels e.g. panels made from mineral wool. Inventory for this scenario is based on (Biowert, 2014; Kamm et al., 2009; Kromus et al., 2006, 2004).

2.1.3.3. Utilization as fermentation feedstock. This scenario aims at exploiting the carbohydrate content of the press-pulp by producing sugars for downstream fermentation processes. The targeted fermentation product is lysine. The press-pulp first undergoes a hydrothermal pre-treatment to break down the biomass structure. The biomass is subsequently treated with enzymes, to induce hydrolysis of the carbohydrates into shorter-chain sugars that can be metabolized by yeast or bacteria during the fermentation process. The sugars from the hydrolyzed press-pulp are used as carbon source for the fermentation organisms, replacing the conventional carbon source used in lysine production, which is glucose syrup (Anaya-Reza and Lopez-Arenas, 2017; Blonk Consultant, 2010; Leiß et al., 2010). Inventory for this scenario is based on (Bentsen et al., 2006; Blonk Consultant, 2010; Larsen et al., 2012; Wang et al., 2018).

Table 3
Overview of the tested scenarios in the PFS and LCA model. Configuration scenarios are indicated with a number, while utilization scenarios are indicated with a letter.

Scenario name	Pressing	Precipitation	Protein separation	Solid fraction utilization
Baseline (1.T.1.S)	1step	Thermal	1step	Feed
1.T.1.F[a]	1step	Thermal	1step	Fermentation
1.T.1C	1step	Thermal	1step	Composite
1.T.2.S	1step	Thermal	2Step	Feed
1.T.2.F	1step	Thermal	2Step	Fermentation
1.T.2.C	1step	Thermal	2Step	Composite
1.B.1.S	1step	Biological	1Step	Feed
1.B.1.F	1step	Biological	1step	Fermentation
1.B.1.C	1step	Biological	1step	Composite
2.T.1.S	2Step	Thermal	1Step	Feed
2.T.1.F	2Step	Thermal	1Step	Fermentation
2.T.1.C	2Step	Thermal	1Step	Composite
2.T.2.S	2Step	Thermal	2Step	Feed
2.T.2.F	2Step	Thermal	2Step	Fermentation
2.T.2.C	2Step	Thermal	2Step	Composite
2.B.1.S	2Step	Biological	1Step	Feed
2.B.1.F	2Step	Biological	1Step	Fermentation
2.B.1.C	2Step	Biological	1Step	Composite

[a] The abbreviations used as scenario names signify (x,T,y,F): were x indicates the number of pressing step (one or two), the middle letter indicates the type of precipitation process used (T thermal, B biological), y indicates the separation steps (one or two), and the last letter indicates the solid fraction utilization (S silage, C composites, F fermentation).

2.1.4. Press-juice utilization

The press-juice contains most of the soluble content of the original biomass, such as soluble carbohydrates and protein. Depending on the targeted compound i.e. generally the soluble proteins, different downstream processes can be used for separation from the press-juice. The proteins are precipitated from the press-juice to facilitate the separation and are subsequently dried, while the residual press-juice is sent to the biogas plant for anaerobic digestion.

2.1.4.1. Protein precipitation from press-juice. Two different precipitation methods have been included in the model, thermal and biological precipitation. During thermal precipitation, the press-juice is heated to 80 °C by steam and heat exchangers. At this temperature, proteins denature and coagulate into larger agglomerates that will settle or float in the supernatant surface. Alternatively, precipitation can be induced by lowering the pH of the solution. In this scenario, lactic acid bacteria are inoculated in the press-juice, which acidifies the fermentation solution and brings the pH down to 4 to make the proteins coagulate (Santamaría-Fernandez et al., 2017). This second scenario has lower energy needs, but also a lower precipitation efficiency.

2.1.4.2. Protein separation. For the protein separation process two main process set-ups have been included in the model. In the baseline scenario, a simple decanter centrifuge separates the protein from the press-juice (Termansen et al., 2016). The resulting product has a protein content of approximately 46–50% DM, which can be used as protein-rich feed for monogastric animals (e.g. pigs) in substitution of other protein-rich feed and has a similar composition and protein content to soybean meal (Kragbæk, 2014). The combined protein extraction efficiency from both the decanter centrifuge and thermal or biological precipitation processes ranges from 90% if thermal precipitation is used to 70% for the biological precipitation.

An alternative scenario has been proposed by Kamm et al. (2016), where two different protein products are produced; an "animal-grade" protein-rich feed that can be used for monogastric animals; and a "human-grade" protein-rich food that has higher quality and purity, and can be used by humans. This two-step process requires milder heating, at 60 °C, in the precipitation stage and cannot be performed with biological precipitation. In the first stage, the animal-grade protein is separated by centrifugation, with a protein separation efficiency of 70%. The quality of this feed is similar to the protein-rich feed of the baseline protein separation process. In the second stage, the human-grade protein is separated from the remaining liquid stream by acid

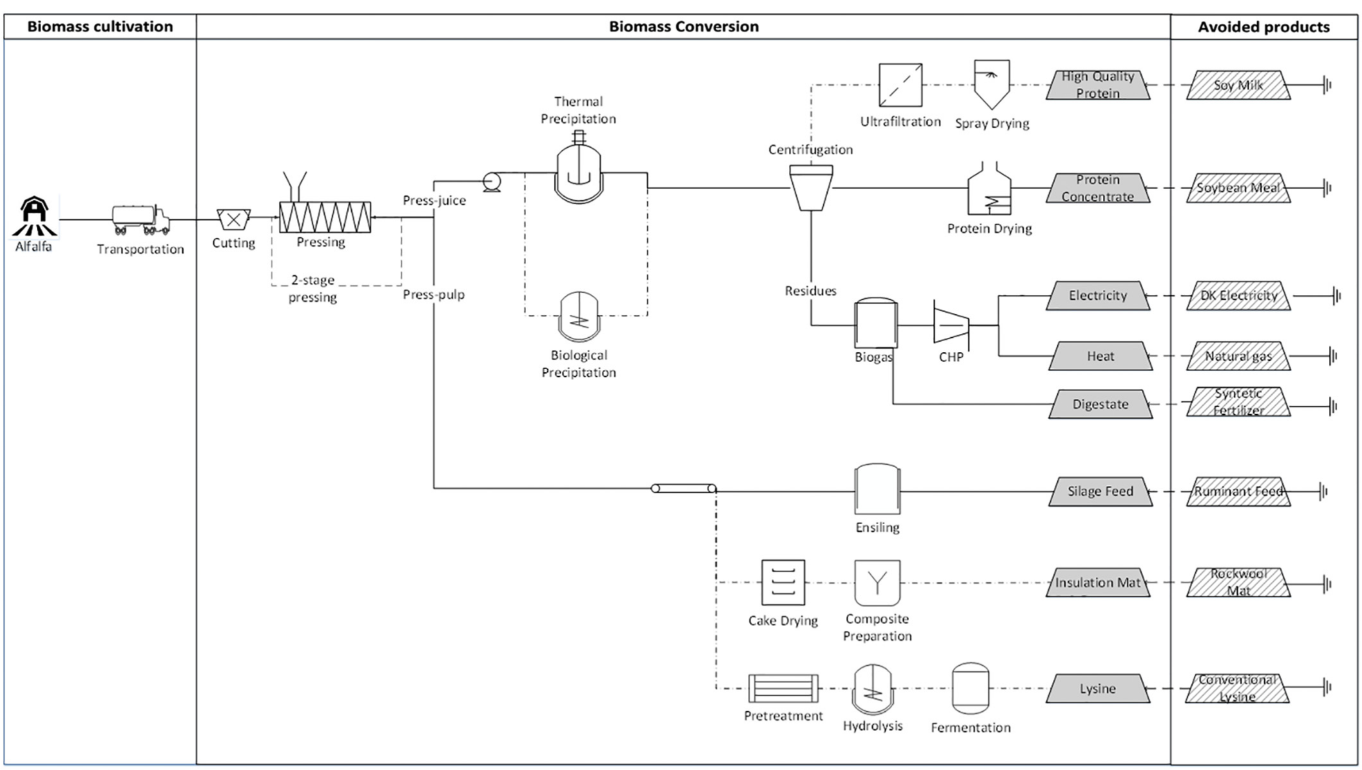

Fig. 1. System boundaries of the LCA study. The system boundaries include biomass cultivation, biomass conversion in the GBR and the conventional products avoided by GBR products. The biomass conversion section shows the most important conversion processes. The baseline scenario follows the solid arrows between processes, while the alternative scenarios are identified with dotted lines. Dark grey boxes show the GBR products while white dashed boxes show the avoided products displaced by the GBR products.

Table 4
Overview of the conventional products replaced by the GBR output. The last column lists the property for which the substitution factor is calculated. Details of the calculation of the substitution factor are presented in SI-2.3.

GBR Product	Replaced product	Substitution factor
Animal-grade protein	Soybean meal	Nutritional Value (digestible protein content)
Human-grade protein	Soymilk	Nutritional Value (digestible protein content)
Feed from press-pulp	Alfalfa silage	Nutritional Value (digestible energy content)
Composite from press-pulp	Rockwool insulation panel	Insulation properties (thermal insulation)
Lysine from press-pulp	Lysine from glucose syrup	Sugars-to-lysine productivity

precipitation and membrane filtration. Protein separation efficiency of this second step is 90% of the available protein in the residual press-juice.

2.1.4.3. Drying. After the separation process, the animal grade protein product has a DM content of approximately 34% and the human grade protein product a DM content of approximately 24%. All products are dried until a 95% DM content is achieved. A drying step has yet to be implemented at the pilot plant at AU Foulum. Estimates of energy consumption for this step are therefore based on general energy usage in industrial water removal operations, as reported in the literature (Grabowski and Boye, 2012; Mujumdar, 2014), as well as on communications with the green pellet industry in Denmark (Dangrønt products A/S). The estimated energy consumption is divided into two steps, (i) from 35 to 60% DM and (ii) from 60 to 95% DM, due to differences in efficiency of moisture removal at lower dry matter concentrations. It is thereby estimated that 2000 kJ/kg of evaporated water are used for the first stage and 5000 kJ/kg for the second stage. Furthermore, it is estimated that the consumption from external energy sources in the future may be reduced by a factor of five to ten. This estimated energy optimization would be a result of e.g. process development, heat integration with energy production such as biogas, and mixing the wet product with other dried feed ingredients, which would aid the subsequent drying process. (Grabowski and Boye, 2012). In the model's baseline, a semi-industrial scale of the process is represented by assuming a fivefold reduction of heat consumption, and 50% of the heat used in the drying process is recycled in the system from drying to precipitation (Grabowski and Boye, 2012; Mujumdar, 2014). The influence of heat optimization is further analysed in the sensitivity analysis, Section 2.2.4.

2.1.4.4. Utilization of the residual juice. The deproteinated press-juice still contains valuable organic compounds that can be fermented by anaerobic digestion in a biogas plant. The amount of biogas produced by the residual fraction was estimated using Buswell's formula (Buswell and Mueller, 1952), assuming a biogas conversion efficiency of 70%, in a hypothetical 2-stage wet mesophilic anaerobic digestion plant (Hamelin et al., 2014). The biogas produced is supplied to a combined heat and power engine (CHP) with an electrical efficiency of $\eta = 40\%$ and thermal efficiency of $\eta = 45\%$ (O'Keeffe et al., 2011b). The digestate was assumed to be subsequently spread on the field within the catchment area supplying the GBR, hence avoiding the production of conventional fertilizers (O'Keeffe et al., 2011a). Calculation of the fertilizer potential of the digestate was taken from (O'Keeffe et al., 2011b) and adapted to the selected biomass, based on its biochemical composition of the deproteinated press-juice.

2.2. LCA model description

2.2.1. Goal and scope definition

This study aims to compare different GBR configurations at the early stages of biorefinery design. Furthermore, the study attempts to answer the research questions: "*What are the environmental impacts connected to different GBR configurations*?" and "*What is the best utilization for the press-pulp and press-juice*?" To answer these specific questions, the functional unit selected for the analysis is the "*Production and conversion of 1ton$_{DM}$ of alfalfa biomass in the GBR*". Thus, the LCA can be used to estimate the environmental impacts associated with the production and conversion of the feedstock, while using different GBR configurations and/or targeting different final products. In Table 3, an overview of the different scenarios tested in the PFS and LCA model is presented.

2.2.2. System boundaries and life cycle inventory data

The system was analysed from cradle to biorefinery exit gate. Fig. 1 shows the system boundary and the unit processes included in the analysis. The system was expanded to include the substitution of conventional products by the GBR products. The PFS model calculated the inventory for biomass conversion in the biorefinery in terms of product

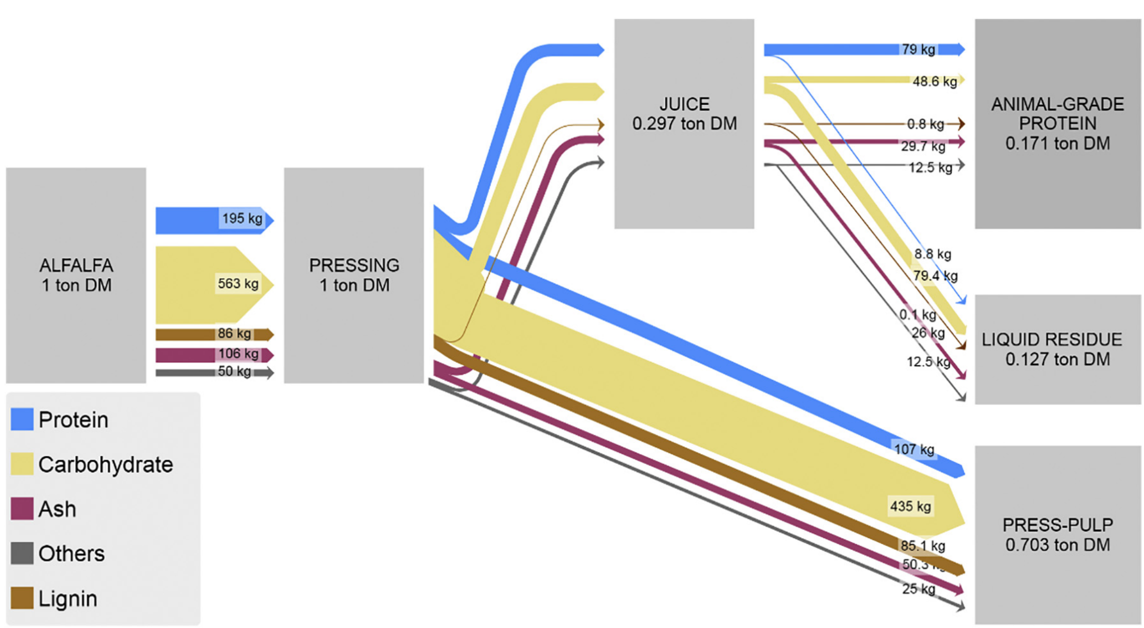

Fig. 2. The Sankey diagram shows, for the baseline scenario, how the biomass components are separated during the GBR process and the composition of the final GBR products.

Table 5
Breakdown of GBR product quantities in the different configuration scenarios from the PFS analysis. Amounts shown per 1tonDM of converted feedstock biomass.

Scenario name	Animal-grade protein [kg]	Human-grade protein [kg]	Press-pulp [kg]	Residues [kg]
Baseline (1.T.1)	$1.71 * 10^2$	[−]	$7.03 * 10^2$	$1.27 * 10^2$
1.B.1	$1.53 * 10^2$	[−]	$7.03 * 10^2$	$1.44 * 10^2$
1.T.2	$1.53 * 10^2$	$3.55 * 10^1$	$7.03 * 10^2$	$1.09 * 10^2$
2.T.1	$2.06 * 10^2$	[−]	$6.64 * 10^2$	$1.31 * 10^2$
2.B.1	$1.80 * 10^2$	[−]	$6.64 * 10^2$	$1.56 * 10^2$
2.T.2	$1.80 * 10^2$	$4.60 * 10^1$	$6.64 * 10^2$	$1.10 * 10^2$

yields and quality, and consumption of energy and auxiliary materials, while the EcoInvent 3 life cycle unit process database (Wernet et al., 2016) was used to supply the background data.

2.2.3. Environmental impact categories and the LCA methods

The environmental impact categories used in this study are: Global Warming Potential (GWP), Eutrophication Potential (EP), Non-Renewable Energy (NRE) use, Agricultural Land Occupation (ALO), and Potential Freshwater Ecotoxicity (PFWTox). The selection of the environmental Impact Categories considered in this study was based on Parajuli et al. (2015) and to be in line with the LCA data used for the cultivation stage provided by (Parajuli et al., 2017b), which is used in this assessment. For the first three impact categories (ICs) the “EPD” method (Environdec, 2015) was used. For the calculation of climate change impacts, the contributions from indirect land use changes (ILUC) induced by the occupation of arable land for the production of the biomass and those avoided by the displaced conventional products, were also included. ILUC factor was taken from Schmidt et al. (2015). The ReCiPe method (Huijbregts et al., 2015) was used to estimate agricultural land occupation impacts, while the ILCD method (JRC, 2011) was used for the PFWTox impacts.

2.2.4. Sensitivity analysis

In the sensitivity analysis, the parameters connected to the heat optimization, which has not been implemented at the pilot plant yet, were varied in the GBR model to observe the effects of different plant maturity levels on the NRE impact score. The effect of optimization values from 1 to 10 and heat recycling from 0% to 100% were evaluated in the baseline scenario. This allows simulating the biorefinery at different optimization/maturity levels: from a non-optimized/lab-scale level (optimization = 1, recycling 0%) to semi-industrial/pilot plant (optimization = 5; heat recycling = 50%) and finally to optimized/industrial level (optimization = 10, recycling = 100%) calculated after Kamm et al. (2016).

2.2.5. Substitution factor

Each of the GBR products is assumed to replace a conventional product. To estimate the impact due to the avoided production of conventional products, substitution factors were calculated based on the individual product's quality and the market that the GBR products enter. Table 4 shows the conventional products replaced by the GBR products and the basis for calculating the substitution factor. SI-2.3 describes how the substitution factors were calculated.

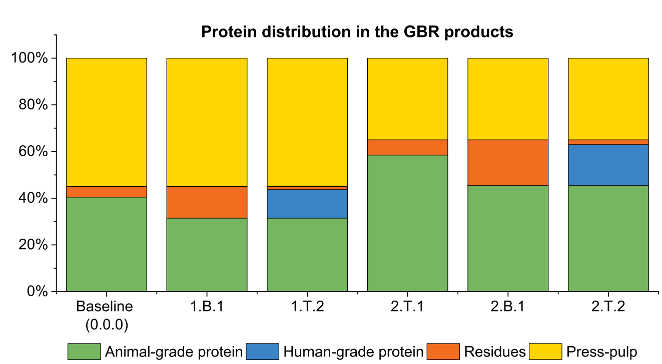

Fig. 3. PFS results: Protein distribution between the GBR products in the studied scenarios.

3. Results and discussion

3.1. Process flowsheet simulation results

3.1.1. Product yield and composition

The PFS model was used to estimate the quantity and composition of the GBR's outputs, as well as energy and auxiliary materials consumption in the GBR. Results are calculated for the conversion of 1ton_{DM} of biomass. Fig. 2 shows a Sankey diagram of the baseline scenario. Results for the other configuration scenarios are presented in the supporting information, SI-3.

From Fig. 2, it is possible to observe that the fractionation process mainly separates the soluble biomass components from the insoluble. Soluble components end up in the press-juice, while insoluble ones, such as lignin, cellulose and hemicellulose, mostly end up in the press-pulp. In the baseline scenario, approximately 70% of the DM ends up in the cake (press-pulp), 17% in the protein-feed and 13% in the residuals. Approximately 40% of the protein available in the whole biomass is separated into the animal-grade protein. The animal-grade protein has a protein content of 46$\%_{DM}$ in the animal feed, which is similar to the protein content in soybean meal (Nemecek and Kägi, 2007) and in line with previous works ((Kamm et al., 2009). Of the remaining protein, approximately 55% is separated into the cake and 5% ends up in the residual liquid. It is important to note that a substantial part of the protein found in the cake is fiber-bound, and thereby can only be digested by ruminants, as it is impossible to separate this type of protein by mechanical means only (Dotsenko and Lange, 2016). Insoluble carbohydrates, fiber bound proteins, and lignin are the main components of the press-pulp, with carbohydrates making up the largest share of 62% press-pulp dry matter. Lignin accounts for 12$\%_{DM}$ of the press-pulp, which accounts for 99% of the lignin content in the original biomass.

Table 5 shows the output of the GBR, from the conversion of 1ton_{DM} in all configuration scenarios. The yield of animal-grade protein varies from 153 kg/ton_{DM} for scenarios 1.B.1 and 1.T.2, to 206 kg/ton_{DM} for the 2.T.1 scenario. In comparison to the baseline, the yield varies within a range of −11% for the lowest yielding scenario to +20% for the highest

Table 6
PFS results: quality parameters for the LCA model.

Parameter	Unit	1.T.1	1.B.1	1.T.2	2.T.1	2.B.1	2.T.2
SF animal protein	kg/$kg_{soymeal}$	0.95	0.81	0.81	1.15	1.01	1.01
SF human protein	kg/$kg_{soymilk}$	[−]	[−]	1.40	[−]	[−]	1.56
SF cake-silage	kg/kg_{silage}	0.96	0.96	0.96	[−][1]	[−][1]	[−][1]
SF cake-composite	kg/$kg_{rockwool}$	0.90	0.90	0.90	0.90	0.90	0.90
SF cake-lysine	kg/kg_{syrup}	0.79	0.79	0.79	0.84	0.84	0.84
Specific biogas yield[2]	m^3/kg_{DM}	0.25	0.26	0.25	0.26	0.26	0.25

[1] Excluded because the protein content of the press-pulp below the minimum nutritional threshold.
[2] Based on Volatile Solid (VS) content of the residual fraction.

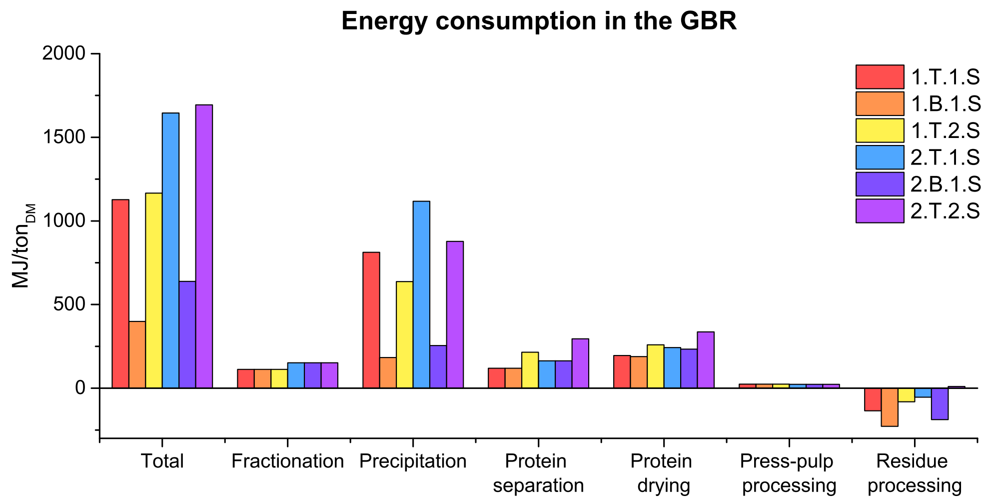

Fig. 4. Total heat and electricity consumption in the GBR calculated from the PFS analysis. Positive values indicate energy consumed; while negative values indicate energy produced.

yielding one. The yield of animal-grade protein is similar for scenarios 1.B.1 and 1.T.2, however, a more advanced processing method allows for human-grade protein to be produced in 1.T.2, resulting in an overall protein output of 189 kg. Press-pulp yield is only affected by the number of times fractionation is carried out. Pressing twice decreases the press-pulp yield by approximately −6%, while at the same time increasing the protein-feed yield by approximately +18% compared to the baseline. Moreover, when looking at the GBR residues, lowest output of residues is found when the two-step protein separation process is in place; i.e. in the 1.T.2 and 2.T.2 scenarios. On the other hand, the output of residues is highest when biological precipitation is used because of the lower efficiency of this process i.e. scenarios 1.B.1 and 2.B.1. The yield of GBR output is in line with previous publications (Kamm et al., 2009; O'Keeffe et al., 2011b).

Fig. 3 shows a detailed analysis of the protein distribution in the GBR products. In the baseline scenario, the overall protein extraction efficiency, i.e. the protein that ends up in the protein-rich feed, is approximately 41%. By using biological coagulation, the overall protein extraction efficiency decreases to 32%, while increasing the protein in the residues from 5% to 14%. Contrastingly, using a two-step protein separation increases the protein extraction efficiency to 44%; 32% in the animal-grade and 12% in the human-grade protein feed.

Two-step fractionation allows for an increase in the protein extraction efficiency of approximately +18% for scenario 2.T.1, +5% for scenario 2.B.1 and + 23% in scenario 2.T.2, compared to the baseline. Since more protein is extracted in the press-juice, the protein content in the press-pulp decreases. Thus, the protein content of the press-pulp drops from 15%$_{DM}$ with one-step fractionation to 10%$_{DM}$ with two-step (see Table SI-10). This results in a protein content lower than the minimum requirement for animal feed as stated by Kamm et al. (2010) of 140 g/kg. Hence, the utilization of the press-pulp as ruminant feed in the 2.X.X scenarios is excluded in the LCA results.

Table 6 shows substitution factors (SF) and biogas yields calculated for the LCA model from the PFS analysis. SFs for the animal-grade protein to soybean meal vary from 1.15 to 0.81 kg/kg$_{soymeal}$ depending on the GBR configuration scenario. The cake-silage SF is slightly below one when compared to alfalfa silage, because part of the protein originally available in the biomass has been removed into the press-juice, decreasing its nutritional value. When utilized as composite, all scenarios have a similar SF, since it is assumed that a reduction of protein and other soluble products in the press-pulp does not affect the thermal properties of the composite.

For the fermentation utilization, the SF is correlated to the carbohydrate content in the press-pulp. Two-step fractionation increases the carbohydrate content per DM of press-pulp and thereby the SF for lysine increases.

The biogas yield has a very limited variation between the studied scenario and ranges from +4% in scenario 2.B.1 to −1% in scenario 1.T.2. Specific biogas yields are in line with previous results found in the literature (Santamaría-Fernández et al., 2017).

3.1.2. Energy consumption in the GBR

Fig. 4 presents the GBR energy footprint in the different configuration scenario. The figure shows total heat and electricity consumption

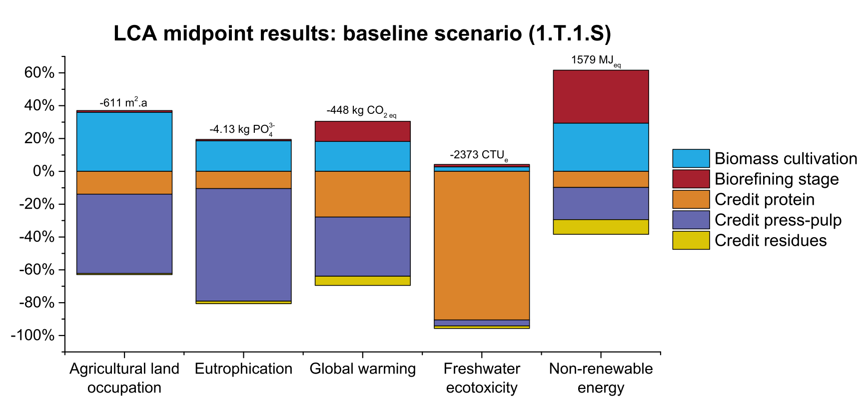

Fig. 5. Midpoint LCA results for the conversion of 1 ton$_{DM}$ alfalfa for the baseline scenario (1.T.1.S). The figure shows the percentage contribution of each life cycle stage and the total score for each impact category.

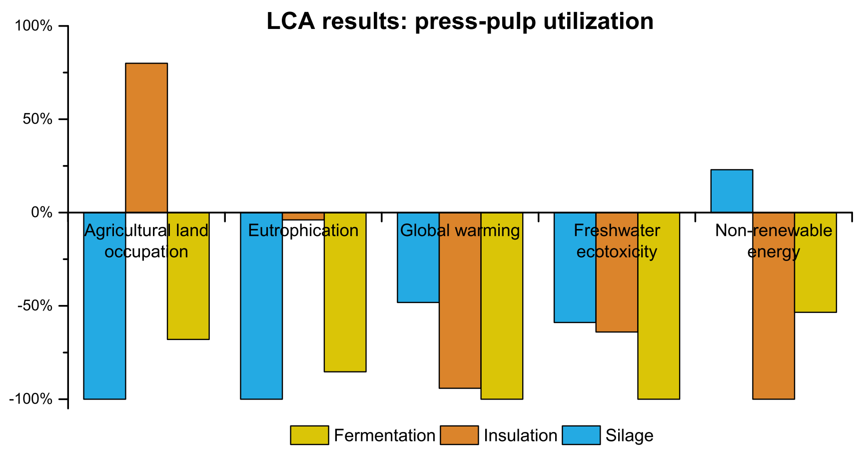

Fig. 6. Midpoint LCA results for the different downstream utilization scenarios of the press-pulp. Results are shown for the conversion of 1ton$_{DM}$ of alfalfa, for the baseline GBR configuration and are internally normalized to show contributions relative to maximum savings.

for each configuration scenario. Results are shown for the utilization scenario where the press-pulp substitute animal feed. Figs. SI-2 and SI-3 in the SI show the analysis for electricity and heat separately.

The total energy consumption in the GBR ranges from 399 MJ/ton$_{DM}$ in scenario 1.B.1 to 1694 MJ/ton$_{DM}$ for scenario 2.T.2. The lower range of energy consumption is in line with results presented by Kamm et al. (2009), while the upper range is higher in this study due to differences in precipitation temperatures. While Kamm et al. (2009) takes into consideration a pre-heating step with recycled heat and lower precipitation temperature, which result in an overall lower energy consumption, the present study had one heating step with a precipitation temperature of 80 °C, which leads to the high energy consumption at the upper range. The most energy consuming process is the coagulation followed by drying and protein separation. Pressing the biomass twice increases the energy consumption by approximately +45%. However, the increase occurs in the precipitation step, as an extra washing step leads to a larger water content that has to be heated during thermal precipitation. Therefore, 38% of the added energy consumption is present during this step, while only 7% is due to the energy consumed in the additional fractionation process. Furthermore, the energy consumption of protein separation and drying shows a slight increase, due to the higher protein content of the press-juice. The use of biological precipitation, which requires a lower operating temperature, allows a reduction in energy consumption of −65% for the 1.B.1 and − 43% for the 2.B.1 scenarios compared to the baseline. The energy produced by anaerobic digestion of the residues has a limited contribution since the Volatile Solid (VS) content available in this stream is very low. Part of the heat produced is used internally in the biogas plant to warm up the residues. For the most advanced scenario, i.e. 2.T.2, the energy produced from biogas does not offset the heat requirement in the biogas plant, resulting in an overall net heat consumption for processing the residues.

3.2. LCA results

3.2.1. LCA results baseline

Fig. 5 shows midpoint LCA results for the baseline scenario. Negative values represent environmental savings, while positive values show burdens to the environment compared to the production of the conventional products. The net results are negative throughout the ICs, with the exception of Non-renewable energy (NRE), since the avoided production of conventional protein-feed, silage and energy replaced by GBR products, is associated with larger impacts than those induced by

Table 7
LCA results of the different GBR configurations. The 15 different scenarios have been grouped into the three utilization options analysed. Color-coding identifies best and worst performing configuration scenario within each utilization scenario moving in gradients from green to red, respectively for each impact category and press-pulp utilization option.

Press-pulp for silage production							
Impact category	Unit	0.0.0.S	0.1.0.S	0.0.1.S	1.1.0.S	0.0.1.S	1.0.1.S
Agricultural land occ.	m^2a	$-6.11*10^2$	$-5.38*10^2$	$-7.36*10^2$	[–]	[–]	[–]
Eutrophication	kg $PO_4^{3-}{}_{eq}$	$-4.12*10^0$	$-3.98*10^0$	$-4.48*10^0$	[–]	[–]	[–]
Global warming	kg CO_{2eq}	$-4.44*10^2$	$-4.27*10^2$	$-6.70*10^2$	[–]	[–]	[–]
Freshwater ecotoxicity	CTU_e	$-2.37*10^3$	$-1.84*10^3$	$-3.45*10^3$	[–]	[–]	[–]
Non-renewable energy	$MJ_{primary}$	$1.63*10^3$	$9.66*10^2$	$-6.07*10^2$	[–]	[–]	[–]
Press-pulp for composite material production							
Impact category	Unit	0.0.0.C	0.1.0.C	0.0.1.C	1.0.0.C	1.1.0.C	1.0.1.C
Agricultural land occ.	m^2a	$4.89*10^2$	$5.62*10^2$	$3.67*10^2$	$3.45*10^2$	$4.50*10^2$	$1.67*10^2$
Eutrophication	kg $PO_4^{3-}{}_{eq}$	$-1.54*10^{-2}$	$1.28*10^{-1}$	$-3.66*10^{-1}$	$-3.26*10^{-1}$	$-1.20*10^{-1}$	$-8.36*10^{-1}$
Global warming	kg CO_{2eq}	$-8.71*10^2$	$-8.50*10^2$	$-1.09*10^3$	$-9.42*10^2$	$-9.13*10^2$	$-1.26*10^3$
Freshwater ecotoxicity	CTU_e	$-2.58*10^3$	$-2.05*10^3$	$-3.66*10^3$	$-3.65*10^3$	$-2.88*10^3$	$-5.21*10^3$
Non-renewable energy	$MJ_{primary}$	$-6.83*10^3$	$-7.44*10^3$	$-8.99*10^3$	$-6.05*10^3$	$-6.96*10^3$	$-9.21*10^3$
Press-pulp as fermentation feedstock							
Impact category	Unit	0.0.0.F	0.1.0.F	0.0.1.F	1.0.0.F	1.1.0.F	1.0.1.F
Agricultural land occ.	m^2a	$-3.92*10^2$	$-3.20*10^2$	$-5.40*10^2$	$-5.39*10^2$	$-4.34*10^2$	$-7.43*10^2$
Eutrophication	kg $PO_4^{3-}{}_{eq}$	$-3.46*10^0$	$-3.32*10^0$	$-3.87*10^0$	$-3.80*10^0$	$-3.60*10^0$	$-4.37*10^0$
Global warming	kg CO_{2eq}	$-9.18*10^2$	$-9.01*10^2$	$-1.15*10^3$	$-1.04*10^3$	$-1.01*10^3$	$-1.37*10^3$
Freshwater ecotoxicity	CTU_e	$-3.85*10^3$	$-3.32*10^3$	$-5.11*10^3$	$-4.94*10^3$	$-4.17*10^3$	$-6.68*10^3$
Non-renewable energy	$MJ_{primary}$	$-3.54*10^3$	$-4.21*10^3$	$-5.85*10^3$	$-3.31*10^3$	$-4.22*10^3$	$-6.62*10^3$

the GBR value chain. For NRE, the system has a positive score, which is connected to the agricultural inputs and the energy consumption in the biorefining process.

Savings related to substituted protein dominate the overall score for the freshwater ecotoxicity impact category (PFWTox IC) and are two orders of magnitude higher than the induced impacts from the agricultural and GBR steps. There are two reasons for this: (i) the feedstock biomass used in the biorefinery has lower pesticide application rates compared to highly industrialized crops like soy and corn/maize that are substituted (ii) implications from the inventory modelling approach used for the pesticide emissions. For the biomass converted in the GBR (i.e. the induced impacts), PFWTox is modelled using PestLCI (Dijkman et al., 2012) to quantify the pesticide emissions at the field level (Parajuli et al., 2017b). In contrast, for the avoided crops (e.g. soy) the modelling approach is based on the EcoInvent guidelines, which assume that 100% of all pesticides applied to the field are emitted to the soil. It is not possible to apply the PestLCI model to the credited EcoInvent processes, since the avoided crops are grown outside Europe and the pesticide model cannot yet assess other geographical regions than Europe.

Looking at the induced impacts, the agricultural stage plays a major role, contributing with a range of 97% for Agricultural land occupation (ALO) to 48% for NRE. The biorefining stage shows high contribution in the energy related ICs, such as GWP and NRE, where it induces a total impact of 40% and 52%, respectively for each IC.

Regarding the avoided impacts, i.e. the credits connected to the replacement of conventional products, the largest credits arise from the production of silage feed. This is due to the large yield (biomass-to-product) of this product compared to the others products, as approximately 70% of the feedstock ends up as silage feed. Thus, the avoided impacts induce savings in the range of −85% for EP and − 4% for PFWTox. For climate change, the protein product contributes 40% of the avoided impacts, demonstrating that the production of local, protein-rich feed alternatives to soybean meal can lead to a reduction of climate change related impacts.

3.2.2. Utilization of the press-pulp

Fig. 6 shows impacts of the baseline GBR configuration scenario (1.T.1) for different downstream utilizations of the press-pulp. A similar trend is observed for the other configuration scenarios (see SI-4).

The environmental performance of the system changes depending on the type of downstream application and consequently according to the type of avoided product. If the press-pulp is used for silage production, and thereby the avoided product is an agricultural product, the largest impact reduction potential can be achieved in the agricultural related impact categories, such as ALO and EP. If the press-pulp is used for the production of insulation panels, there are large reductions in the energy related impact categories, i.e. GWP and NRE, since the pulp based product replaces an energy intensive product. Thus, burden shifting is observed between the silage and insulation applications, since benefits for agriculturally related ICs are exchanged for burdens in the energy related ICs. Lastly, the best overall performance between the studied utilization scenarios occurs when the press-pulp is used for lysine production, where the net scores are below zero across all impact categories.

3.2.3. GBR configuration

LCA results of 15 different GBR scenarios are presented in Table 7. The 2.T.2.F scenario exhibit the best environmental performance in the ALO, EP (together with 1.B.1.S), GWP and PFWTox impact categories. For NRE, scenarios 2.T.2.C and 1.T.2.C have the lowest score. The worst performing scenarios are 1.B.1.C in ALO and EP, 1.T.1.S in GWP and NRE and 1.B.1.S in PFWTox.

Table 7 reveals a visible pattern and highlights the importance of protein recovery for the overall LCA results. It is evident, by observing the color pattern, that biorefinery configurations that prioritize protein extraction efficiency reach the largest savings across the ICs, as can be observed by looking at the scenario group X.X.2, which has two-step protein separation for all scenarios. Moreover, scenario 2.T.2 which has two-step fractionation and two-step protein separation and thereby has the highest protein recovery is the best performing scenario.

The following paragraphs describe the variation in the LCA results between baseline and alternative processes in each biorefining stage. Results are presented for the utilization scenario where the press-pulp is used for lysine production. A similar trend can be seen for the other utilization scenarios (see SI-4).

By using two-step fractionation and thereby increasing the protein content in the press-juice, savings are achieved throughout all ICs except for NRE, as shown in Fig. 7. The choice of fractionation frequency affects the system in three different ways: in the biorefining stage, in the savings obtained by replacing conventional protein feed and in the amount of biogas produced.

Firstly, two-step fractionation leads to higher impacts, since it increases energy consumption at the biorefining stage. However, this is counter-balanced by a higher yield of protein in the feed products and biogas, which leads to an overall lower impact score for the alternative scenario for all ICs except for NRE. Having one or two fractionation steps does not, however, change the fermentation potential of the press-pulp, since the cellulose and hemicellulose content in the press-pulp is not

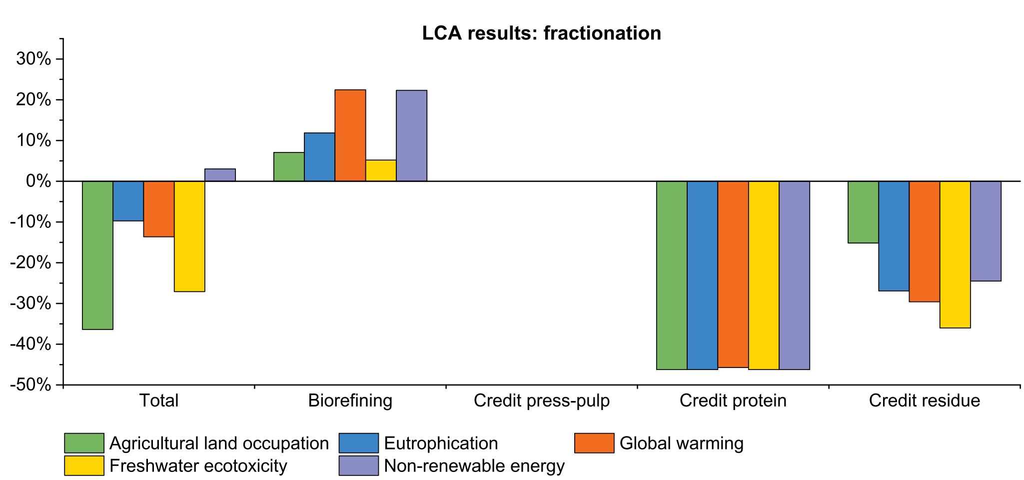

Fig. 7. LCA results assessing the effects of one-step vs. two-step fractionation. Results are presented as a percentage difference between one-step and two-step fractionation. Results shows the difference on the total LCA score (first column) and on the affected processes (biorefining, credit press-pulp, credit protein and credit residues).

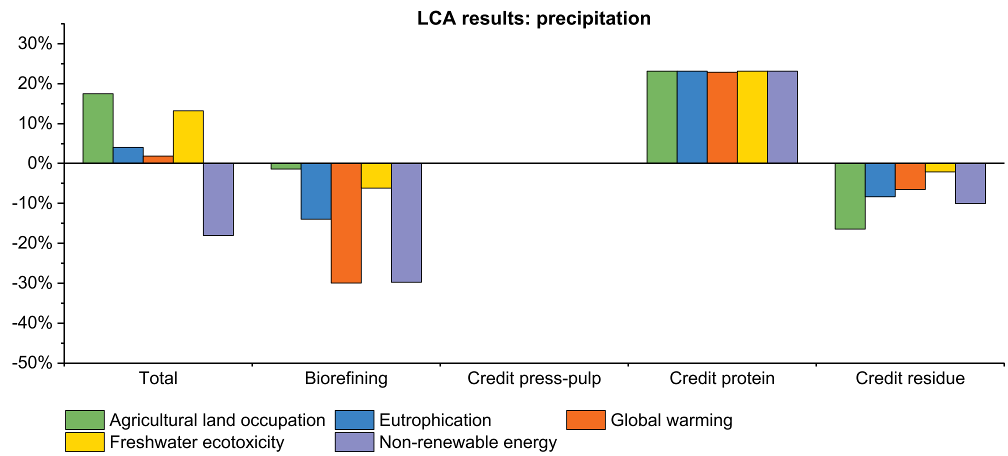

Fig. 8. LCA results of thermal vs. biological precipitation. Results are presented as a percentage difference between biological and thermal precipitation. Results shows the difference on the total LCA score (first column) and on the affected processes (biorefining, credit press-pulp, credit protein and credit residues).

affected by this procedure. Hence, the savings connected to press-pulp utilization remain unaltered.

As with fractionation, the same life cycle stages are affected by the choice of precipitation method, however the trend is inverted when biological precipitation is employed, see Fig. 8. The use of biological precipitation induces higher environmental impacts across all ICs, except for NRE. Despite having lower energy consumption, biological precipitation has a lower overall protein precipitation efficiency. Hence, the reduction in energy consumption is not enough to counteract the reduction in product yield, resulting in a net overall impact increase in all IC, except for NRE, which is strongly reciprocal to energy consuming processes.

By using a two-step protein separation process, the production of a higher value product i.e. human-grade protein, is possible, which leads to a reduction in the environmental impacts of the GBR across all ICs, see Fig. 9. However, the GBR stage becomes more burdensome, due to higher energy consumption needed in the extra protein separation stage, and because a part of biogas credits are lost, since the residual juice has a lower VS content. On the other hand, the production of human protein fully counteracts the higher impacts induced in the other life cycle stages and results in a net overall impact reduction.

3.2.4. Sensitivity analysis

The results of the sensitivity analysis connected to the heat consumption in the GBR are presented in this paragraph and in Fig. 10. The NRE score ranges between 2480 MJ/ton$_{DM}$, +57% compared to the baseline, and 1479 MJ/ton$_{DM}$, −6% from the baseline. It can be observe that a variation of the parameter connected to heat optimization has larger effect when there is limited heat recycling. Furthermore the NRE score is positive even in the most optimized scenario (i.e. optimization parameter = 10 and 100% of heat recycling) suggesting that the GBR system still has a lower performance in this IC compared to the conventional products. As shown in (Corona et al., 2018) this could be avoided by a different energy source for heat production, e.g. substituting natural gas with biomass or biogas.

4. Conclusion

Maximizing product yield proved to be the most important environmental optimization parameter for the GBR, even more important than reducing the biorefinery's energy consumption. This can be observed when the protein content of the products is increased by applying either two-step fractionation, advanced protein separation, or both. Decreasing the biomass-to-product ratio influences the credits gained by substituting conventional products, which in the case of scenario 2.T.2 counteracts the higher energy consumption. On the other hand, biological precipitation results in low protein yields, which cannot be counterbalanced by the reduction in the energy consumption, showing once more that focus in the designing phase should lie on increasing the

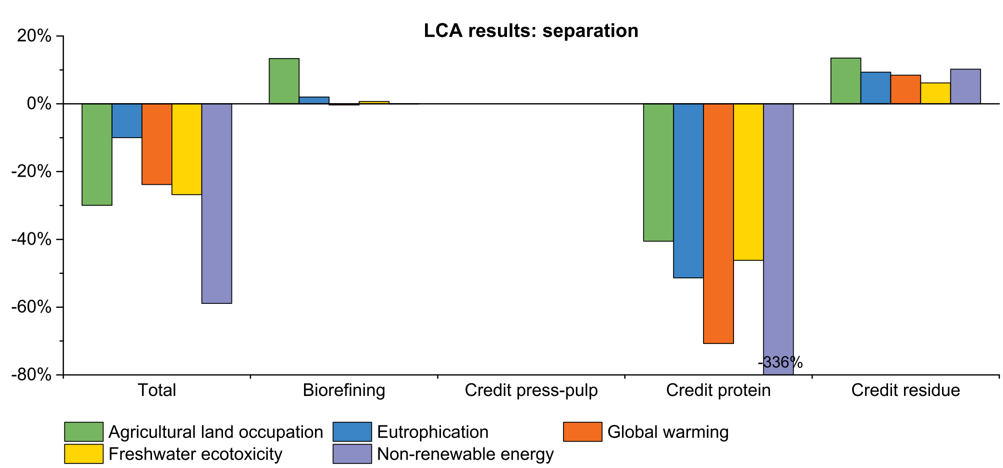

Fig. 9. LCA results assessing the effect of one-step vs. two-step protein separation. Results shows the difference on the total LCA score (first column) and on the affected processes (biorefining, credit press-pulp, credit protein and credit residues).

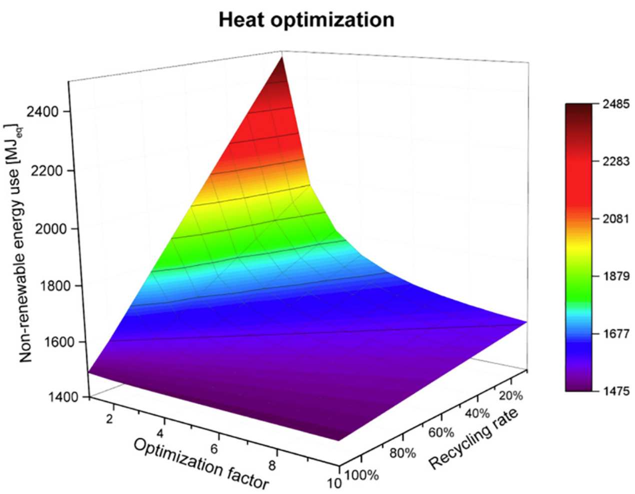

Fig. 10. Sensitivity analysis results for non-renewable energy. The figure shows the NRE score varying the parameters connected to the heat consumption in the GBR. The heat optimization parameter (x-axis) has been varied between 1 and 10 while the heat recycling (y-axis) has been varied between 0% and 100%.

efficiency of product recovery in order to attain a more sustainable biorefinery. However, this does not rule out biological precipitation as such, but instead suggests an optimization of the separation step after the biological precipitation e.g. using membrane technology.

The utilization of the press-pulp was shown to be another important optimization parameter with extensive influence on the environmental profile of the GBR system, since approximately 70% of the biomass ends up in the pulp. If the press-pulp is used to replace conventional energy-intensive materials e.g. mineral wool, large savings are achieved on energy related ICs (GWP and NRE), while if the replaced product is agricultural, large savings are observed on agriculture related ICs (ALO, EP).

Green biorefining is shown to be an interesting biorefining concept. Biorefining of green biomass is technically possible and can bring environmental savings, when compared to conventional production methods. However, those savings are very much determined by the processing involved in the conversion stages and on the cascade utilization of the different platform products. An insight in the environmental implications of the different products and GBR configurations is the first step that complemented with an economic assessment of profitability can bring about sustainable choices for future bio-production.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.03.357.

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PAPER VII

Using Life Cycle Assessment to quantify the environmental benefit of upcycling vine shoots as fillers in biocomposite packaging materials

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Using Life Cycle Assessment to quantify the environmental benefit of up-cycling vine shoots as fillers in biocomposite packaging materials

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Abstract

Purpose The objective of the present study was to better understand the potential environmental benefit of using vine shoots (ViSh), an agricultural residue, as fillers in composite materials. For that purpose, a comparative life cycle assessment (LCA) of rigid tray made of virgin poly(3-hydroxybutyrate-co-3-hydroxyvalerate) PHBV, polylactic acid (PLA) or polypropylene (PP) and increasing content of ViSh particles was performed. The contribution of each processing step in the life cycle on the different environmental impacts was identified and discussed. Besides, the balance between the environmental and the economic benefits of composite trays was discussed.

Methods This work presents a cradle-to-grave LCA of composite rigid trays. Once collected in vineyards, ViSh were dried and ground using dry fractionation processes, then mixed with a polymer matrix by melt extrusion to produce compounds that were finally injected to obtain rigid trays for food packaging. The density of each component was taken into account in order to compare trays with the same volume. The maximum filler content was set to 30 vol% according to the literature and industrial data. The ReCiPe 2016 Midpoint Hierarchist (H) methodology was used for the assessment using the Cut-off system model.

Results and discussion This study showed that bioplastics are currently less eco-friendly than PP due to the fact that LCA does not take into account, in existing tools, effects of microplastics accumulation and that bioplastics are still under development with low tonnage. This study also demonstrated the environmental interest of the development of biocomposites by the incorporation of ViSh particles. The minimal filler content of interest depended on the matrices and the impact categories. Concerning global warming, composite trays had less impact than virgin plastic trays from 5 vol% for PHBV or PLA and from 20 vol% for PP. Concerning PHBV, the only biodegradable polymer in natural conditions in this study, the price and the impact on global warming are reduced by 25% and 20% respectively when 30 vol% of ViSh are added.

Conclusion The benefit of using vine shoots in composite materials from an environmental and economical point of view was demonstrated. As a recommendation, the polymer production step, which constitutes the most important impact, should be optimized and the maximum filler content in composite materials should be increased.

Keywords: Biocomposite · Life cycle assessment · Packaging · Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) · Vine shoots · Extrusion

1. Introduction

In viticulture, every winter after pruning, large quantities of vine wood are produced that are currently underutilized. Pruning of vine shoots (ViSh) is necessary in order to improve growing conditions for the plant, as well as to increase the yield and quality of grapes. Vine shoots can be from 1 to 2 meters long, and production amounts to between 1 and 2.5 tons of dry matter per hectare per year [1]. The productivity of the vine plant depends on the region where it grows, the pruning method and the vine species. In Languedoc-Roussillon (LR), a wine region in the south of France, ViSh production amounts to 500 000 tons every year [2]. Currently, management of vine shoots in France is done by either collecting and burning the ViSh or leaving them on the vineyards where they are cut roughly and used as organic fertilizer [3]. When used as biofertilizers, ViSh should be considered as by-products and not waste. However, their use as soil amendment can be problematic, as decomposing ViSh may serve as vector for diseases for the following vine crop [4]. Furthermore, it is worth noting that ViSh is not the most judicious biofertilizer since its biodegradation, i.e. mineralization in soil, comes in competition with the vine's growth as regards the nitrogen consumption [5]. Less commonly, ViSh are used as fuel wood or compost, which are considered low value uses for this potential resource. Regarding the ambitious goals set by the European community for a bioeconomy, which include the decarbonization of the economy by an 80-95% decrease of CO_2 emissions by 2050 [6], ViSh present a valuable resource for implementing decarbonizing recovery strategies. These strategies can be achieved in a biorefinery context, where cascading treatments of ViSh are investigated to produce added-value products, including the production of lignocellulosic fillers for biocomposite applications [7]–[9]. Lignocellulosic fillers from agricultural residues present the advantages, in addition to their fully biodegradability in natural conditions, to have a lower density than conventional inorganic fillers and to be highly available at a low price, with no competition with the food sector [10]. ViSh present a great opportunity in the field of biocomposites, a potential application being rigid food packaging biodegradable in natural conditions [11], [12].

On the other hand, the global plastic market is continuously growing reaching 350 million tons in 2018, with 40% of the production used in the packaging sector [13]. The massive amount of plastics used each year results in a constant accumulation of plastic wastes in our environment [14]. The associated effect of this on ecosystems, wildlife, and humans is worrying even if not yet fully understood. For this reason and the concern about global warming, fully biosourced and biodegradable materials such as biocomposites are emerging as a possible solution to tackle the problem of accumulation of plastic in our environment and to reduce greenhouse gas emissions. Poly(3-hydroxybutyrate-co-3-hydroxyvalerate), called PHBV, is a promising bacterial biopolymer that is biodegradable in soil and ocean, and that can be synthetized from all kinds of carbon residues. PHBV can be combined with natural fillers to give fully biodegradable biocomposites, for example for rigid trays applications [15], [16]. Moreover, PHBV displays similar mechanical and barrier properties as polyprolylene (PP) and can therefore substitute this fossil and non-biodegradable conventional polymer [17]. A competitor to PHBV is poly(lactic acid) (PLA), which is the most widely commercialized bio-sourced plastic currently in the market. However, it is worth noting that PLA is not fully biodegradable in natural conditions, but only compostable in industrial conditions [18], which requires collection and sorting in order to achieve a valuable end-of-life management and does not avoid concerns related to plastic accumulation from littering or leakage.

The development of biocomposites is largely motivated by either an improvement of the overall technical performance, insisting on mechanical properties, a decrease of the overall cost of materials, and the improvement of the carbon footprint, by replacing a part of non-renewable fossil resources [19]. Biocomposites are thus generally presented as eco-friendly materials. However, most of the time, the environmental benefit is not quantitatively proven [20]. It is thus necessary to ensure they are actually capable of mitigating the abovementioned environmental problems, as the use of bioplastics and natural fillers to produce biocomposites does not automatically make them sustainable. In order to quantitatively verify environmental claims made about biocomposites and other innovative materials, it is possible to carry out environmental assessments. Life cycle assessment (LCA), which is a holistic tool capable of measuring environmental impacts of products and services, can be applied to emerging biomaterials [21]. It investigates the inputs (i.e. resources and energy) and outputs (i.e. waste gases, wastewater and solid waste) across the entire life-cycle stages (cradle-to-grave). LCA allows to locate “hot spots” in the life cycle and avoids the problem shifting from one life cycle stage to another while accounting for all types of emissions and resource consumption [22]. Its main limits are the collection of data that can be difficult and the initial assumptions that need to be justified. Most of the LCA carried out for biocomposites focused on the comparison of natural fillers with synthetic fibers [20], [23], [24], especially for applications in the automotive industry [25]–[27]. Generally, natural fillers tend to have a better environmental performance than glass fibers, notably thanks to the weight reduction of the composites and their low energy demand for production [27].

There are fewer papers in the literature regarding the environmental advantage of incorporating natural fillers in polymer matrices. In a previous study considering 1 kg of material as functional unit, the environmental impacts of materials made of virgin polyolefins (PP and HDPE) and biocomposites with natural fillers (derived from rice husks and cotton linters) were compared [28]. LCA showed that composites displayed lower environmental impacts in all impact categories, except eutrophication, due to the use of fertilizers for rice cultivation. Similarly, it was shown that the incorporation of either wood flour or wood fiber allowed for reducing the environmental impacts of HDPE [29] and PP [29], respectively, in proportion to the filler content.

LCAs of vine shoots and their incorporation in composites were not found in the literature. The combustion of ViSh and induced emissions have previously been studied [30], [31] without LCA tools. More recently, Gullón et al. performed a LCA of the valorization of vine shoots into antioxidant extracts, and other bioproducts from a biorefinery perspective [32]. They determined that ViSh production related processes should be burden free in the biorefinery system since the environmental impacts were entirely allocated to the grape harvesting, as ViSh were considered agricultural waste [33], [34].

Concerning PHBV, no process data is currently available in the Ecoinvent database. However, as shown by Yates et al. [35], several LCAs about bioplastics including PHBV are available in the literature. Inventory data from these papers can be used [35], [36].

In this context, the objective of the present study was to better understand the potential environmental benefit of using vine shoots as raw resources for the production of lignocellulosic fillers for biocomposite applications. For this purpose, a comparative life cycle assessment was carried out, first on rigid trays made out of virgin PHBV, polylactic acid (PLA) or polypropylene (PP). Then, the effect of ViSh incorporation in these 3 polymer matrices was studied by considering a cradle-to-grave approach. The contribution of each life cycle step was identified and discussed. Besides, the balance between the environmental and the economic benefits of composite trays was discussed.

2. Methodology

2.1. Goal and scope

The aim of this article was to determine to what extent addition of ViSh fillers in packaging trays was environmentally beneficial compared to trays produced entirely from virgin plastics. For that purpose, the environmental performance of packaging trays produced in France from either 100% virgin plastics or related ViSh-based biocomposites was assessed. Three polymer matrices, i.e. PHBV, PLA and PP, and different filler contents were considered in the predictions.

2.2. Functional unit and system boundary

The functional unit was a tray of standard model (176 x 162 x 40 mm, GN 1/6 type), 25 cm^3 in volume, for single use packaging, produced by injection molding. It was assumed that all the considered trays had the sufficient properties to provide the same service. The volume of the trays was thereby kept equal throughout the assessment. However, due to the intrinsic densities of the considered materials, the final weight of the trays varied according to the nature and the proportion of each constituent (Table 1).

The scenarios included in this study were trays of virgin PHBV, PLA and PP, and trays of PHBV, PLA and PP filled with milled vine shoots.

The main properties of the raw materials are presented in Table 1. They correspond to commercial grades PHBV (PHI002 from Natureplast), PLA (PLI 003 from Natureplast) and PP (PPH9020 from Total Petrochemical). The density of ViSh was experimentally determined, as explained in Supplementary Data.

Table 1. Different properties for the components of the studied biocomposites.

	Density ($g.cm^{-3}$)	Weight (g) (25 cm^3 tray)	Melting temperature (°C)	Degradation (°C)	Young's modulus* (GPa)	Stress at break* (%)	Strain at break* (%)
PHBV	1.23	30.75	170	200	4.2	40	3.2
PLA	1.24	31	150	250	3.5	45	3
PP	0.91	22.75	165	320	1.7	37	8
ViSh	1.36	-	-	230	na	na	na

* according to the standard ISO 527

It was previously shown that increasing the content of ViSh in PP [37], PE [37] or PHBV [12], [38] resulted in a slight decrease of the mechanical properties of the materials. Ahankari *et al.* [39] studied the reinforcement of PHBV and PP with agro-residues and recommended to incorporate filler contents lower than 40 wt% to avoid a decrease in mechanical properties, due to an increased filler agglomeration in the polymer matrix. Confirming this, Berthet *et al.* [40] observed that the processability of PHBV/wheat straw biocomposites became difficult when the filler content was above 40 wt%. Authors usually considered weight filler contents. However, considering that the volume of the injected molding tray remains constant whatever the matter, it was considered that the use of volume filler contents was more pertinent to compare the different formulations. Given that, it was assumed that the maximum ViSh filler content to get the enough properties for the tray application was 30 vol% for all the composites. This was also in accordance with the filler content currently used in commercialized composites (*Vitis valorem*, Meursault, France, PLA or PP- Sarmine® products). This set limit of 30 vol% corresponded to weight contents of 32 wt% for PHBV and PLA, and 39 wt% for PP (for a given filler volume content and a tray volume, the filler weight content depends on the density of each constituent).

Figure 1 displays the system boundary considered in the present study, with the different life cycle steps that were included. It was assumed that the collection of vine shoots and the production of the trays were done in the Languedoc-Roussillon region of France. In case of 100% virgin plastic trays, the steps encased by dashed lines in Figure 1 were irrelevant because they concerned the ViSh treatment and compounding steps.

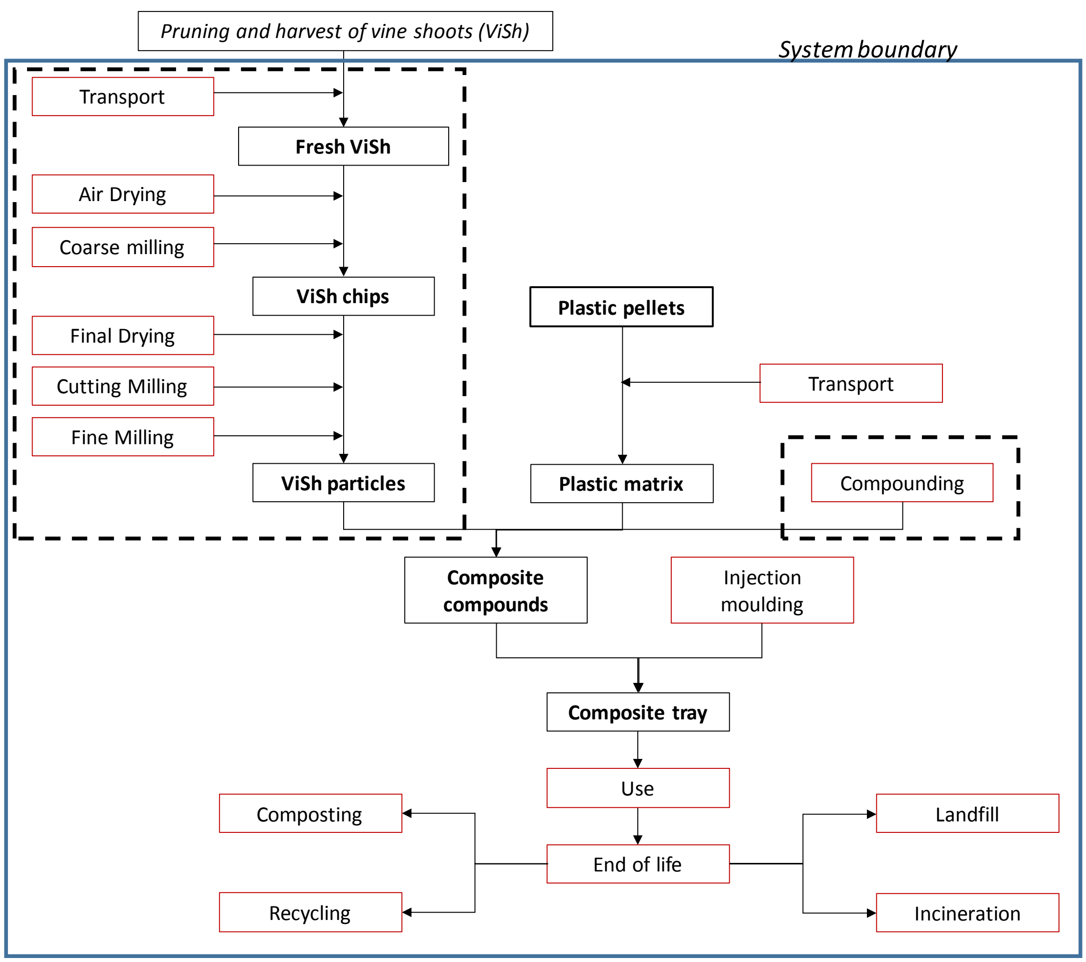

Figure 1. Boundary of the studied system

2.3. System description and inventory

All background data used in the assessment were obtained from the Ecoinvent v.3.4 database with the Cut-off system model and processed using the LCA software Simapro v.8.5. The ReCiPe 2016 Midpoint Hierarchist (H) methodology was used in the interpretation phase of the assessment. In accordance with the geographical boundary, all the electricity used in the system was assumed to conform to the French energy mix.

2.4. Raw materials

Polymer matrices were PHBV, PLA and PP. Ecoinvent processes data recorded for fossil-based PP and PLA from maize grain were used in the LCA. Inventory for PHBV made from sugar cane was obtained from the work of Harding et al [36]. Transport of plastic matter to the production facility was taken into account using the "Background data for transport" sheet from Ecoinvent as the specific transport mode was unknown [41].

For all tested scenarios, lignocellulosic fillers were obtained from the dry milling of ViSh collected in the Languedoc-Roussillon region. It was assumed that ViSh came from the same varieties. In keeping with status quo practices, ViSh were collected during the winter after pruning and initially had a moisture content of 40 wt% (w.b.).

Vine shoots are viticultural residues that can be seen as by-products if they are used as soil amendment, wood fuel or compost, or as a waste if they are simply burnt without energy recovery. The pruning is a necessary process that is independent from the fate of the ViSh. It is difficult to estimate the exact proportion of burnt ViSh because this practice, which is a common fate for ViSh, is in theory forbidden, but derogations and tolerances still exist [42]. According to FranceAgriMer, burning of ViSh accounts for between 25 and 50% in France [3], [43]. In the present study, ViSh burnt on site or without valorization were considered. In that case, the collection of the ViSh happens anyway in order to remove ViSh from the vineyards and it was therefore considered a part of the grape cultivation production system. Besides, ViSh have no market value and thereby, zero environmental impact would be allocated to them. ViSh were thus, considered burden free in the present system. Additionally, ViSh being produced in a wine-grape production system, all the environmental impacts of production were ascribed to the production of wine-grapes. Finally, transport of ViSh from the field to the filler producing site was assumed to be done by a 3.5-7 t lorry with an average distance of 10 km according to *Vitis Valorem* (France) information.

2.5. Production of biocomposite trays

Practical information about the handling of ViSh as raw material for the production of biocomposites was provided by *Vitis Valorem* (France). Commonly, ViSh are first air-dried outdoors for seven months, between January and August. The corresponding land use was determined considering that the ViSh are arranged on the ground reaching an average height of 2 meters, with an apparent density of 30 $kg \cdot m^{-3}$. Only manual labor was used during this step. At the end of this period, the moisture content of ViSh was 20 wt% (w.b.).

Coarse milling with a common wood chipper (Greentec 952, Ufkes Greentec BV, Netherlands) was utilized to mill the ViSh. The throughput was set at 2000 kg·h^{-1}, and 10% of the initial ViSh mass were lost during the milling process. Output chips sizes ranged between 3 and 6 cm in their largest dimension. The output is called "ViSh chips".

An additional drying step was required to reduce the moisture content of the ViSh to 5 wt% (w.b.) after air drying. An existing drying process from the Ecoinvent database was used (see Supplementary data), modified to utilize the French electricity grid.

After coarse milling, a finer milling process in two steps is needed in order to obtain particles of between 0.3 and 0.05 mm in size. First, ViSh were milled using a cutting mill type SM 300 (Retsch, Germany) with a 2.0 mm sieve and secondly they were milled with a fine impact mill (CUM 150, Netzsch Condux, Germany). The final output is hereafter called "ViSh particles". Data for milling were provided by *SD-Tech Group* (Alès, France).

Flexible Intermediate Bulk Containers (FIBC, commonly known as "Big Bags") were used to store the ViSh chips after coarse milling, ViSh particles after fine milling and composite granules after compounding. It was assumed that each FIBC was used 3 times per year during a period of 5 years before being discarded. Each FIBC had a mass of 2.5 kg with a capacity of 1 m^3 and it is made from PP. ViSh chips after coarse milling, fine milled ViSh particles and composite granules had a bulk apparent density of 200 kg·m^{-3}, 420 g·m^{-3} and 700 g·m^{-3} respectively.

During the compounding step, the plastic was mixed with ViSh fillers in an extruder. The extrusion process in Ecoinvent was adapted with data from *Vitis Valorem,* which uses a compounder, model ZSE 160 HP (Leistritz, Nuremberg, Germany). Electricity consumption of the compounding step was 300 kWh.t^{-1} and the yield is 97.6%. In the assessment, the same yield and energy data is used for all compounding regardless of composite granule type. No plasticizer nor additive was used.

Trays were assumed to be produced by injection molding of compounds. The injection molding process in Ecoinvent was modified to provision electricity from the French electricity mix. The yield was assumed to be 99.4% because scrap and waste could be recycled in a nearly closed loop.

All the previously described steps (from air-drying to injection molding) were assumed to occur at the same location.

2.6. Use phase

It was assumed that the use phase of the biocomposite trays was the transport from the factory gate to the place at which they are used as food packaging and then to the distribution site. These transports were assumed to be done by a 32 ton lorry with an average distance of 100 km for each transport stage [44]. The use by the consumer was assumed to be the same for all assessed materials and thus was left out of the assessment.

2.7. End of life

The end of life (EoL) of each tray was defined according to French practices for municipal waste [45] and considering the characteristics of the materials and existing facilities (Table 2). With regard to transport in the end of life, it was estimated that the trays travelled on average 100 km from household to a waste treatment center [46]. Transport was assumed to happen by a 16-32 t lorry, EURO5 from Ecoinvent.

Table 2. Current possible end of life of the different trays (in weight %) from [45]

Tray material	Landfill	Incineration	Recycling	Composting
PP	34.6%	36.5%	28.9%	0.0%
PP-ViSh composite	48.7%	51.3%	0.0%	0.0%
PHBV	38.0%	40.0%	0.0%	22.0%
PHBV-ViSh composite	38.0%	40.0%	0.0%	22.0%
PLA	38.0%	40.0%	0.0%	22.0%
PLA-ViSh composite	38.0%	40.0%	0.0%	22.0%

Concerning composting, only industrial compositing was included due to the lack of data for home composting. The incineration process of Ecoinvent was adapted to account for CO_2 emissions and the nature of carbon (biogenic or fossil). Anaerobic digestion could be an end-of-life option for bioplastics and biocomposite trays, but was not included in the possibilities because it is not widely used in France, and it is more dedicated to agricultural wastes than composite materials.

A more detailed inventory for the production of biocomposites is given in the supplementary inventory (SI) of this paper.

3. Results and discussion

3.1. Environmental impact of 100% virgin plastic trays: Comparison of PHBV, PLA and PP

First, the environmental performances of 100% plastic trays without ViSh fillers were compared (Figure 2). Trays made of PP displayed lower impacts than PLA or PHBV trays in all the categories except for *fossil resource scarcity*. This could be explained by the fact that the density of PP (0.91 $g.cm^{-3}$) was lower than those of PHBV or PLA (1.23 and 1.24 $g.cm^{-3}$, respectively). Thus, in order to get the same tray, i.e. with the same volume, a smaller amount of PP (in mass terms) was needed, i.e. 22.75 g instead of 30.75 g for PHBV (Table 1). Similar results were found showing that when compared by volume rather than weight, PHBV had higher environmental impacts than PP or PE [47]. Moreover, the production of 1 kg of either PHBV or PLA had higher impacts than PP. Impacts for *stratospheric ozone depletion, freshwater and marine eutrophication, land use, and water consumption* were very low for PP, in comparison to the bioplastics because the life cycle of PP did not have agriculture activities which heavily impacted the above named impact categories. On the other hand, the *fossil resource scarcity* impact for PP was the highest because it PP is entirely made from fossil resources. In regards to PHBV, results showed that PLA had the highest impact for 13 out of the 18 categories.

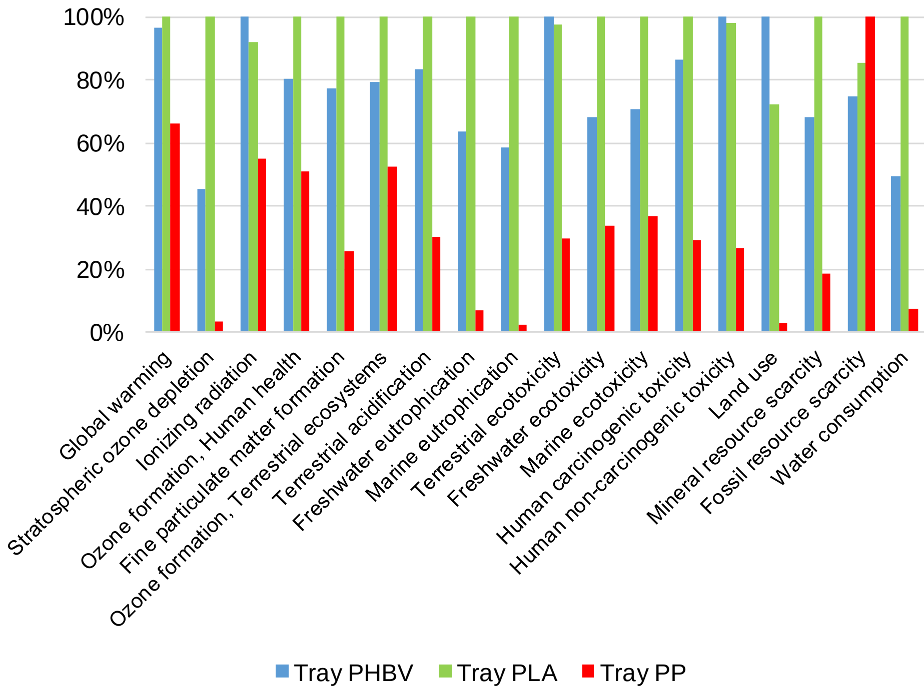

Figure 2. Environmental impact for all impact categories of the ReCiPe 2016 (H) method, for 100% virgin plastic trays.

The impact of each production step on *global warming* is presented in Figure 3. The production of polymer pellets appeared as the most impacting step of the process accounting for more than half of the burden for PP and more than 80% for PLA and PHBV. The PP tray impacts were 30% lower compared to bioplastic trays. This suggests that the substitution of traditional plastic trays with bio-based materials does not always result in a lower environmental impact. Nevertheless, conventional plastic industries have a high degree of optimization which is not the case of bioplastics that are produced in low tonnage. This is exemplified by PP, a petrochemical matrix polymer, which the production has been highly improved, whereas the development of biopolymers is recent and they have not yet reached the same level of technological maturity. Further research on the optimization of the bioplastics processing toward their environmental improvement should be conducted [28]. In the future, their environmental impact is expected to reach lower levels than those reflected in the present study.

The use phase had low impact in the overall life cycle, representing less than 0.5% of the *global warming* for each formulation of tray. It is interesting to note that the end of life was more important for PP, accounting for 26% of the total burden, than for bioplastics (2%). This was mainly attributed to the incineration process. Incineration was more favorable to bioplastics and biocomposites because the carbon released was biogenic, unlike that from to fossil-based plastics. The landfilling contribution to *global warming* was low, representing less than 5% of the PP end of life impacts, because PP was not assumed to be decomposed in the landfill. It must be noted that recycling of PP is an empty process because of the cut-off at recycling. The recycling benefit and costs are allocated to the production of new PP material.

In the present study, it was considered that all the plastic wastes were managed without littering, but in reality a non-negligible proportion of plastic waste ends in nature. In the world since 1950, 79% of plastic waste was accumulated in landfills or natural environment [14]. Long-term impacts such as the accumulation of micro-plastics in the environment are currently not taken into account in LCA. The advantage and benefits of using bioplastics that fully biodegrade in natural conditions are thus not quantified nor included in the analysis. This is particularly relevant for PHBV, which is fully biodegradable in soil and does not require industrial composting, contrary to PLA [48]. Furthermore, gas emissions from petrochemical polymer degradation, which have recently been demonstrated to produce methane and ethylene emissions under sunlight conditions in both water and air, are also not accounted for in LCA [49].

The nutrient contents of bioplastics (e.g., nitrogen, phosphorus, etc.) are so small that the benefit for reducing fertilizer use can be ignored. However, the sequestration of carbon in soil and the soil improvement properties are potential benefits of organic compost [23]. Nevertheless, these are difficult to quantify and are considered outside of the scope of the present work.

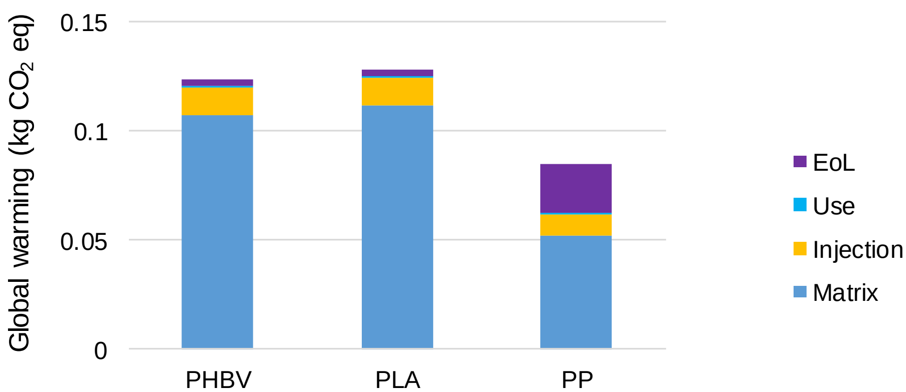

Figure 3. Global warming impact of one 100% plastic tray (without fillers)

3.2. Effect of the incorporation of ViSh fillers on the environmental performance of trays

A composite is the combination of two components: a matrix that constitutes the continuous phase, i.e. either PHBV, PLA or PP in the present study, and fillers that corresponds to the dispersed phase, i.e. ViSh particles in the present study. The *global warming* impact for 1 kg of material is displayed in Figure 4 for the 4 possible constituents of composite materials. It clearly appeared that ViSh fillers had a lower impact (0.26 kg CO_2eq/kg) than the polymer matrices (respectively 3.47, 3.58 and 2.29 kg CO_2eq/kg for PHBV, PLA and PP). The ViSh impact was almost 9 times lower than those of PP matrix. This was due to the advantage of using agricultural residues that only required transport, drying and milling.

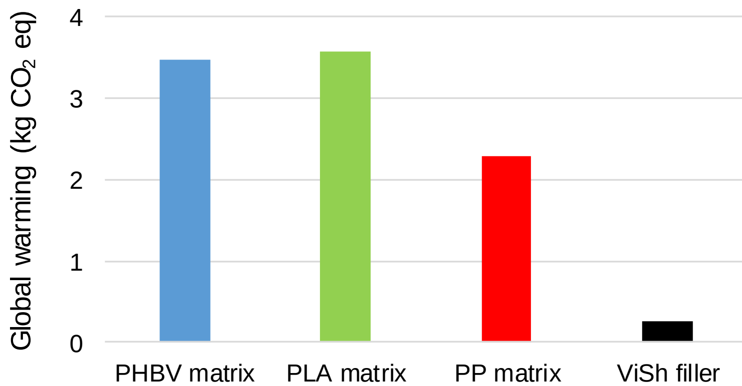

Figure 4. Global warming impact (kg CO_2eq/kg) of 1 kg of each composite component

Figure 5 shows how the *global warming* impact was affected by an increasing filler content in biocomposites. Similar figures for the other impact categories are available in SI. A decreasing burden of the composite with increasing filler content was observed. The incorporation of ViSh appeared to be beneficial concerning *global warming*. It is worth noting that the production of composites required an additional compounding step and that the density of ViSh was 50% greater than that of PP, i.e. 1.36 $g \cdot cm^{-3}$ for ViSh against 0.91 $g \cdot cm^{-3}$ for PP. The burden incurred by the compounding step was visible for composites with very low filler contents. As the production of biocomposites induced an additional use of energy, in all cases, composite with 1 vol% of ViSh had a higher *global warming* impact than respective virgin polymer matrices. The negative impact of both the additional compounding step should be thus compensated by the incorporation of increasing contents of ViSh particles in the polymer matrix. The magnitude of the decrease in impacts varied depending on the matrix type. For PHBV, PLA and PP, the slope was respectively 1.00, 1.05 and 0.68 mg CO_2eq/%ViSh. Then, the use of ViSh was beneficial from 5.5 vol% for PHBV and PLA, whereas the ViSh benefit in PP was first observed for a volume filler content of 20.0 vol%. PHBV-based composites had a lower contribution to *global warming* than 100% virgin PP tray, starting from a PHBV matrix with ViSh content of 44 vol%. However, this filler content is too high to be considered realistic, when taking into account the processability of the materials and their resulting mechanical properties. *Global warming* of PP-based composites was higher than PHBV-ViSh composites, only when reaching a ViSh content of 98.5 vol% and higher, which was of course a non-realistic formulation.

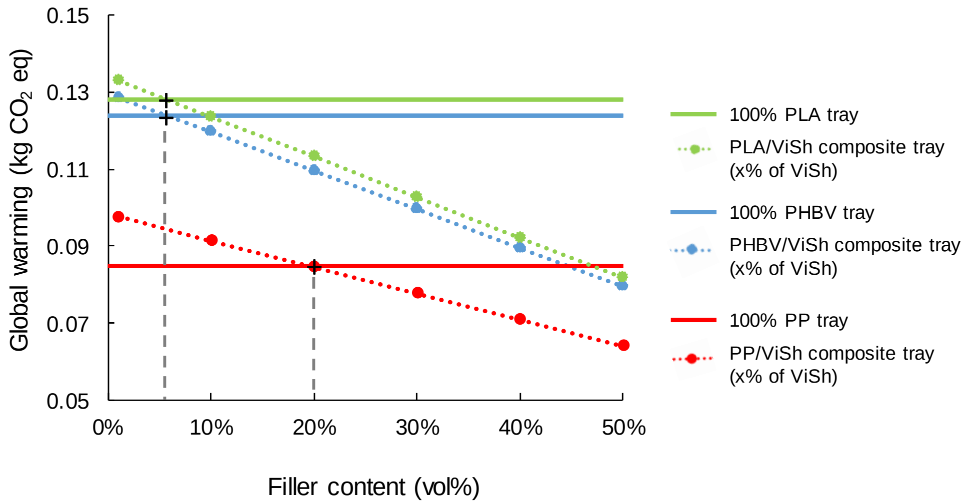

Figure 5. Global warming impact (kgCO_2 eq) as influenced by the filler content (vol%) for composite trays.

The filler content from which the addition of ViSh in the composite resulted in a benefit for all impact categories is displayed in Figure 6. PHBV and PLA displayed similar results; the incorporation of ViSh improved the environmental impacts for all the categories except for *ionizing radiation*. If *ionizing radiation* was to be used as a single score indicator, then biocomposites would never exhibit lower impact than 100% virgin plastic trays because of the electricity needed for the milling, drying and compounding steps of ViSh. The high *ionizing radiation* impact is mainly due to the French electricity mix, which is largely produced from nuclear power. In case of PP, PP-based composites trays can be better than 100% PP trays in 10 categories over 18. The ViSh burden was higher than PP matrix in 4 categories which explains the higher impact of composite in *strastospheric ozone depletion, ionizing radiation land use* and *mineral resource scarcity*. Similarly, the compounding step was responsible for the higher impact in *water consumption* and *terrestrial ecotoxicity*. Finally, *freshwater and marine eutrophication* burden was due to the end of life of the composite. The black dash line in Figure 6 represents the limit of acceptable filler content of 30 vol% in the composite to ensure the functional unit. Thus, *freshwater* and *marine ecotoxicity* and *human non-carcinogenic toxicity* were other impacts that PP-based composite could not improve.

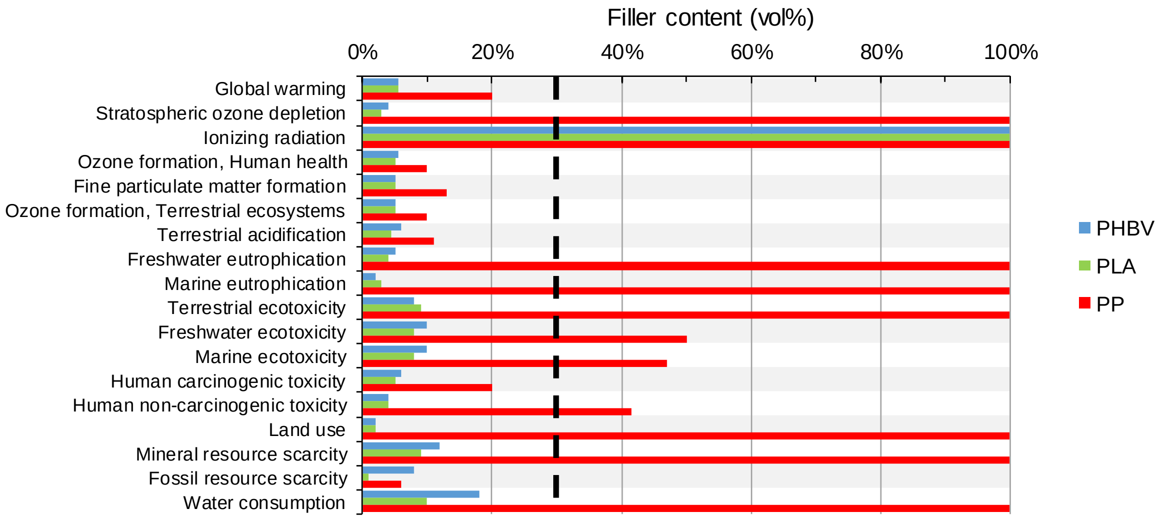

Figure 6. Filler content (vol%) from which a composite tray results in lower environmental impacts than a 100% virgin plastic tray for each assessed impact category. The black dashed line represents the physical limitation of filler content (30 vol%) in the composite to ensure the functional unit. When bars reached a filler content of 100%, no benefit can be realized by the addition of filler.

According to results presented in Figure 5 and Figure 6, it could be concluded that increasing as much as possible the ViSh filler content in the composites, while respecting the restrictions set by material properties, was globally the best for the environment.

The environmental performance of composite trays filled with 30 vol% of ViSh particles was assessed in detail (Figure 7). The 100% virgin PP tray was also added as reference. As previously described in the 3.1. part, results were largely influenced by the nature of the matrix, mainly due to differences in density. PLA composites exhibited the highest environmental impact except for *ionizing radiation*, *terrestrial ecotoxicity*, *human non-carcinogenic toxicity* and *land use* where PHBV exhibited the worst impacts. As expected, PP-based materials exhibited the highest impacts concerning *fossil resource scarcity*.

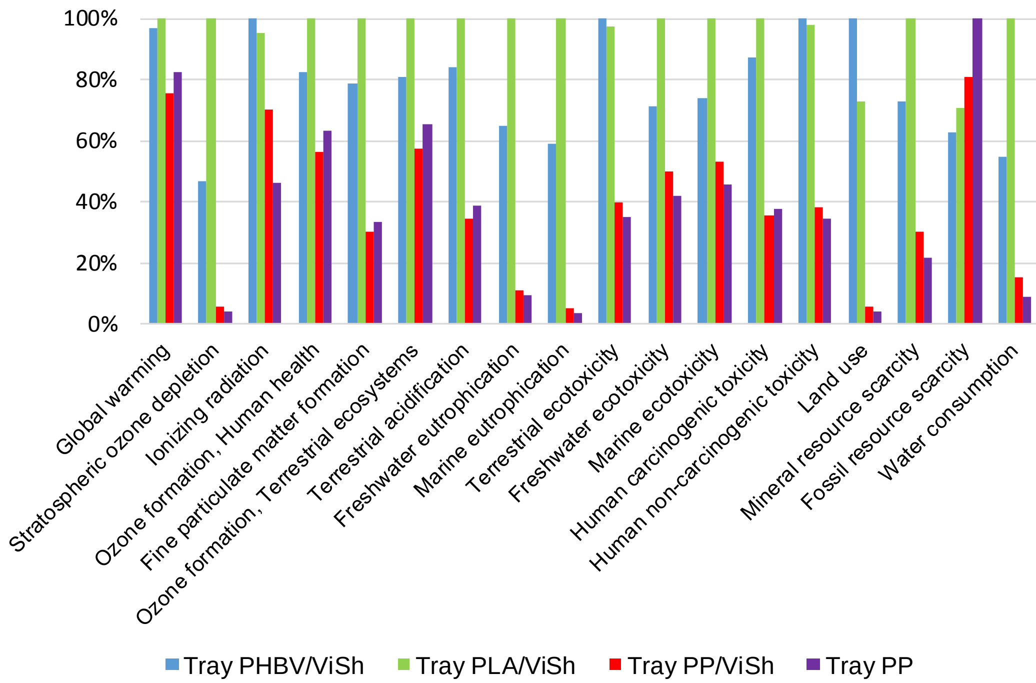

Figure 7. Environmental impact of composite trays filled with 30 vol% of ViSh fillers (all impact categories)

As shown on Figure 8, *global warming* impacts of trays with 30 vol% ViSh fillers were significantly lower than those of trays made from 100% virgin plastics. This was in line with a previous study on the production of biocomposites with wheat straw [50]. The contributions were divided in three categories: (i) raw materials for matrix and ViSh fillers, (ii) processing for compounding and injection steps, and (iii) use and the end of life. The incorporation of 30 vol% of fillers reduced the *global warming* burden of the raw materials by 25% compared to 100% plastic tray. Moreover, the end of life impacts was also reduced for bioplastics. In case of PP-based composite, PP could not be considered recyclable anymore, due to the presence of ViSh filler, inducing a slight increase of the EoL impact. On the other hand, the higher density of the composite materials relative to the pure plastics resulted in higher impacts from the injection molding step. The addition of ViSh came with an additional step of compounding, which had a relatively low impact compared to the injection molding process, as it represented 20% of the burden of the processing. The incorporation of 30 vol% of ViSh in trays reduced their *global warming* effects by 19.6%, 19.9% and 8.5% for PHBV, PLA and PP based trays, respectively.

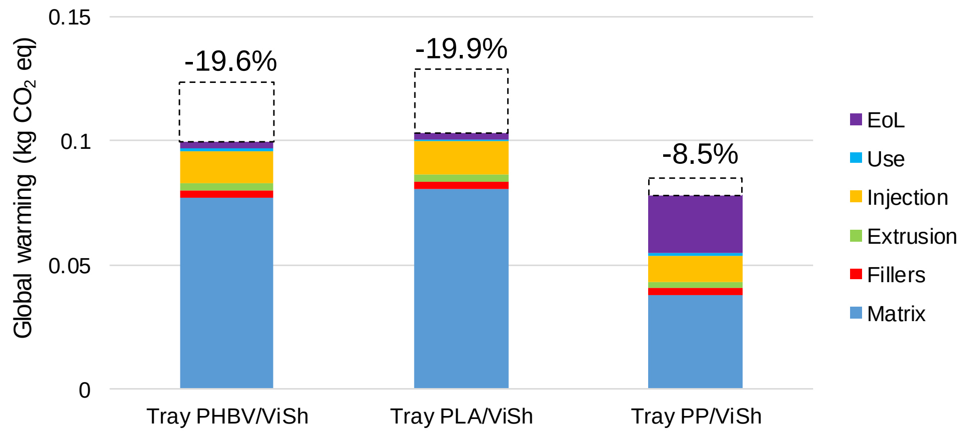

Figure 8. Global warming impact of trays with 30 vol% filler. The percentages above the bars indicate the reduction of the impact compared to trays without ViSh filler.

3.3. Identification of the hot spots

3.3.1 ViSh filler production: contribution of each step on the environmental impact

The main contributor to the environmental impacts of ViSh particles was the milling steps (Figure 9). Milling represented 72% of the *global warming* impact, followed by the drying steps, with a contribution of 22%. The most burdensome type of milling was coarse milling, though there was no impact for *ionizing radiation* because the energy came from diesel fuel. This was contrary to electricity powered cutting and fine millings. The final drying step also consumed energy, but in the form of heat from steam in chemical industry, which explained the low impact value in the *ionizing radiation* category.

The impact of ViSh transport was low in all the categories because it was considered that the production of trays took place in the same region (Languedoc-Roussillon) as the generation of ViSh, allowing for short transportation distances.

Air drying showed burdens in only one impact category, since it only required space to spread the vine shoots on the floor without the help of machinery. Thus, this step only appeared in the category land use and represented 56% of it.

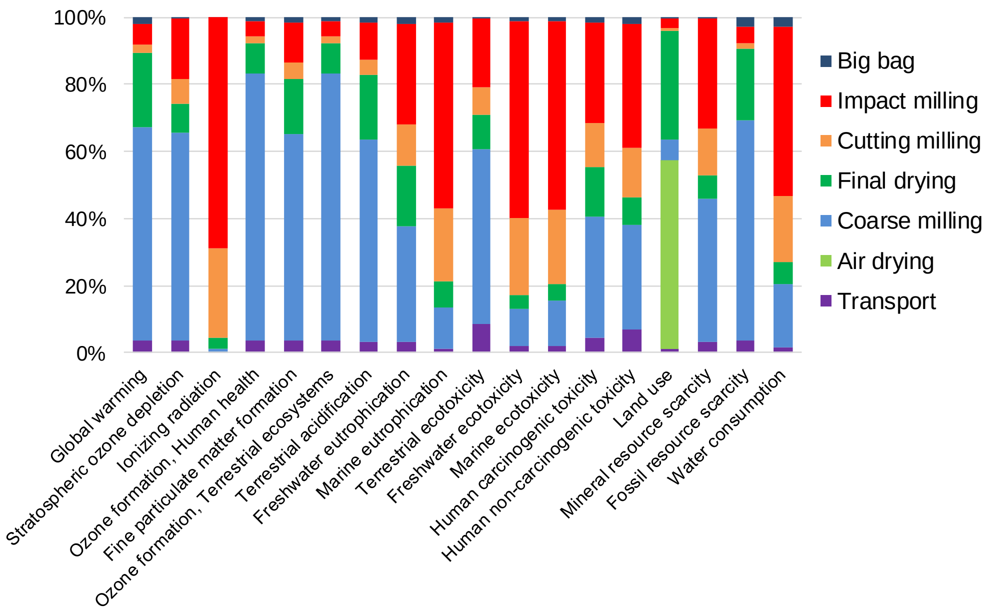

Figure 9. Contribution for ViSh filler production

3.3.2. Polymer/ViSh (30 vol%) composite trays production: contribution of each step on the environmental impact

The analysis of the biocomposites burden clearly showed the strong contribution of the components of the composite and especially the matrix (Figure 10). The contributions of PLA are not shown in Figure 10 to increase clarity and because the results were very close to those of PHBV composites.

For PHBV-based composites, the production of the polymer matrix was the most impacting element for 15 categories, ahead of the end of life (*freshwater and marine ecotoxicity*) and the injection molding (*ionizing radiation*). In case of PP-based composites, results were more balanced with 9 categories dominated by the matrix, 4 by the injection molding or end of life and 1 by the compounding (*water consumption*). When comparing *global warming* potential, the polymer matrix caused the largest contribution to environmental impact for the composite trays. The *global warming* impacts associated with polymer production outweighed those from the filler, manufacturing or end of life.

As expected, *ionizing radiation* impacts were mainly due the manufacturing steps: injection molding and compounding. These processes required electricity. In case of PLA and PHBV composite, land use impact was principally on account of their production requiring respectively corn and sugar cane as carbon source.

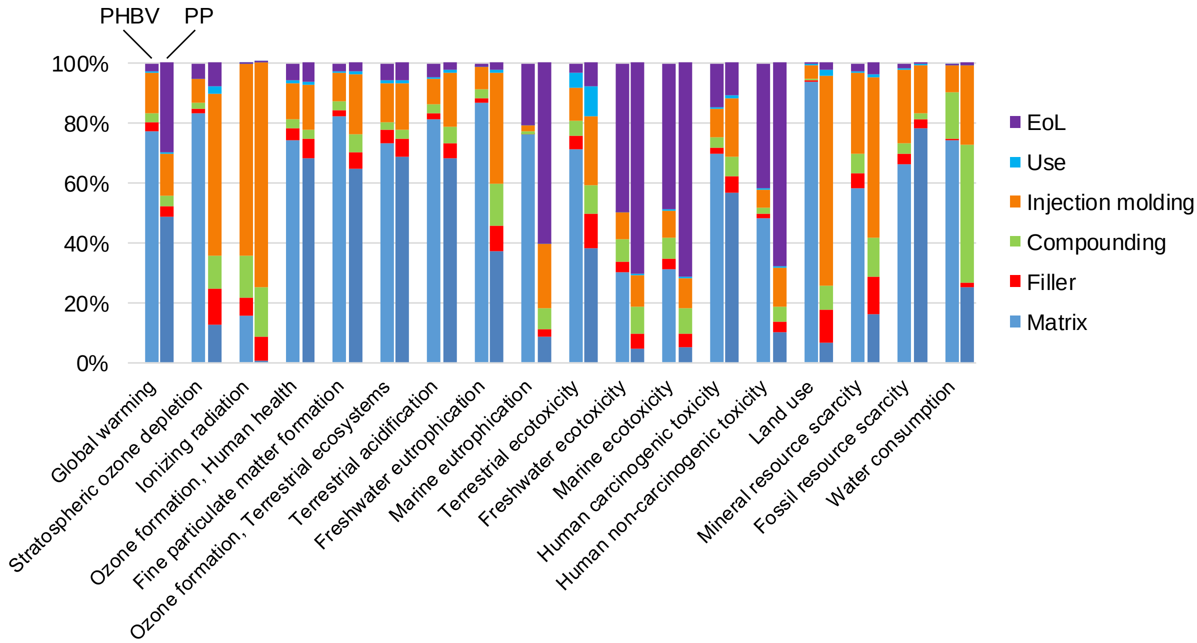

Figure 10. Contribution for PHBV and PP-ViSh (30 vol%) composites

3.4. Economic vs environmental balance analysis

The price of the different trays was estimated from data given by industrials (Table 3). From an economical point of view, the incorporation of 30 vol% of fillers reduced the price of a PHBV tray by 25.4%, and to a lesser extent in case of PP (12.0%) because the price of raw PP is much lower than PHBV (Table 3). It is interesting to note that the injection molding accounted for a large share of the price, ranging from 12% for 100% PHBV trays to 50% for PP-based composite trays. On the contrary, in case of composite materials, the additional price of compounding was almost negligible. The factory price of final trays was not only a rule of mixtures with the price of raw materials. Thus, the addition of ViSh in trays reduced the final price but not as much as expected according to the price of raw materials. There are two reasons for this: the price of injection molding, which was constant, and the density of ViSh was higher from the one of plastics.

Table 3. Price of the studied composite trays. ViSh is 0.30 €/kg (*Vitis valorem, ADEME*), the compounding is 0.04 €/kg (IPC) and the injection molding is 0.03 €/p (Fürstplast).

	Price (€/ton)	Price 100% plastic tray (€/100p)	Price 30 vol% ViSh filler tray (€/100p)	Reduction of the price due to 30 vol% of ViSh filler
PHBV	7 750[a]	26.95	20.11	-25.4 %
PLA	2 800[b]	11.73	9.46	-19.4 %
PP	1 240[c]	6.94	6.10	-12.0 %

[a] NaturePlast, grade PHI 002, 2019
[b] NaturePlast, grade PLI 003, 2019
[c] French customs department, 2017

4. Conclusion

This study assessed the environmental impacts of composite trays made of PP, PLA or PHBV, and increasing content of ViSh particles, based on a comparative life cycle assessment (LCA). It was shown that bioplastics matrices, i.e. PLA and PHBV, which are considered to be eco-friendly, displayed higher environmental impacts than fossil-based polypropylene. This result should be tempered by the fact that long-term impacts such as plastic accumulation are not considered and that the production of bioplastics is still at a much lower level of technological development. In the case of PHBV, the only truly biodegradable bioplastic among the three studied, it is expected that production processes will be optimized, in such a way to decrease their environmental impacts. It is therefore difficult to draw a general conclusion about the environmental efficiency of bioplastics compared to conventional plastics due to the expected evolution of the database. As described by Yates et Barlow in a critical review on biopolymers [35], it is complex to compare their environmental impacts with other studies for different reasons: updated eco-profiles, feedstocks used, sources of energy, etc… There is currently no factor that quantifies the effect of plastic debris on biodiversity [51]. The biodegradability of PHBV can thus not be assessed in the LCA framework. However, there is ongoing research on this issue (for example, the Marilca initiative supported by the Life Cycle Initiative of the UN Environment [52]). One has to wonder how the conclusions of this work will change when such data will become available. The interest of a biodegradable material, compared to a non-biodegradable material but recycled, may seem low from a short-term life cycle analysis point of view. It is neglected the fate of the recycled material which, after a few cycles, will eventually be released into the environment (the recycling of plastic, whether closed short loop or long loop, is limited in time).

The incorporation of increasing contents of ViSh particles in plastic trays resulted in a reduction of environmental impacts despite the additional processing steps required to produce ViSh fillers and the higher density of ViSh compared to the three polymer matrices under consideration. Trays with a higher filler content are therefore heavier inducing more matter to be processed. Despite that fact, this study proved the interest of using agro-residues in composites. Concerning global warming, composite trays had less impact than virgin plastic trays from 5 vol% for PHBV or PLA and from 20 vol% for PP. Regarding PHBV, the only biodegradable polymer in natural conditions in this study, the price and the impact on global warming are reduced by 25% and 20% respectively when 30 vol% of ViSh are added. The maximum filler content of 30 vol% should be increased to reduce the environmental impacts even further.

It can be concluded that most of the research efforts should be devoted to the optimization and scale up of the bioplastics production, PP production being already optimized. The use of cleaner energy should help to achieve this goal but also reduce the impact of injection molding step. Finally, the end of life should be also improved by increasing recycling for PP, ensuring separate collection for composting of PLA, and home composting for PHBV.

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PAPER VIII

Testing the no agricultural waste concept – an environmental comparison of biorefinery value chains in various regions.

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Advanced draft.

Testing the no agricultural waste concept – an environmental comparison of biorefinery value chains in various regions.

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Abstract

Although there is great opportunity, the bioeconomy is not a silver bullet in the quest to solve various environmental problems. This assessment tests the no agricultural waste concept, an agricultural system where all residues are utilized within a value chain, to elucidate whether the concept does indeed improve environmental performance across various regions, and if so, explores how various biorefinery concepts might be organized into various value chains to attain environmental benefits. In order to valorize this, the study illustrates how to do a step-wise assessment in order to design biorefinery set-ups based on their feedstock compatibility and region of implementation. The results show that no agricultural waste systems do not always result in environmental benefits, especially when environmental impacts are measured via a holistic interpretation of environmental damages, namely monetizing environmental damages. Furthermore, disagreement is shown when comparing environmental impacts interpreted via global warming potential impacts and monetized damages. The performance of the various biorefineries was highly affected by the degree of decarbonization present in the energy grid of each region. While energy intensive biorefineries are able to provide benefit in terms of global warming savings, tradeoffs are observed where impacts are shifted to other areas of environmental impact. Despite these tradeoffs, across multiple regions, there is great potential for large-scale implementation of biorefineries as a tool for ameliorating environmental damages.

1. Introduction

The bioeconomy is perceived as one way to reduce environmental impacts, in particular emissions of CO_2 caused by the use of fossil fuels. The first European Bioeconomy strategy was launched in 2012 and updated in 2018 (European Comission, 2018). The overall scope of the strategy remains the same in the updated version, which is to strengthen and scale up bio-based sectors while respecting the planets ecological boundaries. However, the updated version has more focus on the deployment of local bioeconomies in the EU as well as increased focus on the sustainability issues related to the bioeconomy. In addition to the strategy of the European Union, several national and regional bioeconomy strategies have been developed to support the bioeconomy in a particular country or region (de Besi and McCormick, 2015; Motola et al., 2018). Even though strategies are developed for the entire EU or individual nations, they are most likely to be implemented regionally (de Besi and McCormick, 2015; Motola et al., 2018). Thus, the regional perspective is important when, for example building biorefineries or other facilities for biomass processing, which may need to consider local conditions for optimal operation. However, as several studies have shown previously (Jørgensen et al., 2012; Ögmundarson et al., 2020) being bio-based is not necessarily equal to being sustainable. Also, the degree of environmental performance attained while implementing bioeconomy related activities will vary depending on the region where it is implemented (G. Croxatto Vega et al., 2020; Croxatto Vega et al., 2019).

Despite the bioeconomy not always supporting it, sustainability is an important aspect of the bioeconomy. Bio-based products and bioenergy have the potential to induce better environmental performance, while at the same time supporting regional economies. On the other hand, an excessive extraction of biomass is environmentally unsustainable and bio-based processes sometimes consume more energy and input materials than conventional fossil-based processes (Yates and Barlow, 2013). Thus, it is important to assess the sustainability of single biorefinery concepts in a regional context to find the most suitable options.

A paradigm shift in attitudes towards the bioeconomy occurred in 2008, after the work of Searchinger et al., (2008) showed that ethanol from crops performs worse than fossil fuels, from an environmental perspective, if land-use changes are included. This propelled a shift of focus from 1st generation crops, which have competing uses, to 2nd generation biomass such as crop residues that do not cause land-use changes. Agricultural residues are abundant and there is great potential to increase the efficiency of their use. However, there are regional differences in how agricultural residues are managed, which determine how sustainable their use becomes. Among the many considerations, these depend on technical/environmental factors such as yields, and farming intensity (O'Keeffe and Thrän, 2020), and legislative differences i.e. regulation of mineral fertilizers, caps on certain crop uses, etc. (Thrän et al., 2020). Furthermore, even though climate change is still a high priority, there are other types of environmental impacts that are important and can also be targeted through a bioeconomy. For example, it has been shown that though climate burdens may be reduced through the use of biomass resources, other impacts such as acidification and eutrophication from the use of fertilizers can be increased when alternative bio-based options are utilized (Corona et al., 2018; Dressler et al., 2012). It is important to consider the full array of environmental issues in addition to climate change to avoid environmental burden shifting.

The aim of this work, is to test the possibility of utilizing the majority of agricultural residues of a given region to maximum environmental potential, thereby testing the thesis of no agricultural waste. This asks the question: is it possible and always beneficial to utilize the majority of the agricultural residues of a region? If so, we try to answer the how. That is to say, which biorefinery combinations result in the highest environmental benefits for the region. This is done by applying the life cycle assessment (LCA) methodology in two separate steps. First, we assess various processing options for agricultural residues and find region-specific compatible biorefinery setups. This is then used with a decision metric to make a logical conclusion on how best to utilize the available feedstock in each region. Second, we assess the combination of biorefinery setups scaled up to the specific regional context i.e. using all the regionally available feedstock. Lastly, to ensure there is no disproportionate burden shifting, e.g. from climate change potential to eutrophication, we monetized environmental damages and provide single score indicators to aid decision support. Through this process, we design scenarios that fit the specific regions based on agricultural residues present there and other regional considerations that influence the final impact of biotechnologies.

2. Methodology

In order to design a system with no agricultural waste, which is to say that all agricultural by-products are utilized, it is necessary to optimize the paring of feedstock to technology with a clear background or regional context in mind. That is to say, it is necessary to understand feedstock availability, existing infrastructure, and the system that supplies the necessary inputs for potentially applied technologies. The feedstocks considered for this study were cow manure, pig manure, poultry manure, wine pomace, vine shoots and straw. These were chosen because of their status as second generation feedstocks which are, for the most part, not in competition with animal feed and other uses. In brief, the approach followed here is to first test the technologies with each feedstock in order to get an idea of feedstock to technology compatibility. This step is essentially a mini-LCA for each technology with each feedstock in each region of interest. The mini-LCAs are then used with a decision metric to make a logical conclusion on how best to utilize the available feedstock in each region to arrive at the highest possible environmental benefit. These steps are described more in detail in the following subsections. Furthermore, the technologies are not equally compatible with each feedstock type assessed, eliciting different combinations for the biorefinery setups. These are described in detail in section 2.2 (Compatibility Matrix).

2.1. Technology Description

Four main technologies form the pillars of the biorefinery combinations assessed in this work. The technologies are: Anaerobic digestion (AD), AD with additional polyhydroxyalkanoates (PHA) production (AD+PHB), solvent extraction of polyphenols (Poly), and Filler production. These are each further described below.

2.1.1. Anaerobic Digestion

The AD used in this study is based on a model developed in Superpro Designer v.10 (Intelligen Inc, 2018) of conventional AD, where organic materials are converted into CO_2 , CH_4 and trace gases via biological conversion routes. The parametrization of the model is described more in detail in Croxatto Vega et al (G. Croxatto Vega et al., 2020). The model includes a two-step AD set up, with a fermentation step, followed by completion of AD. The products from this process are biogas and digestate.

2.1.2. Anaerobic Digestion with polymer production

Conventional AD can be split after the fermentation step into a liquid and solid fraction, via the use of a screw press. The liquid fraction where most of the volatile fatty acids (VFAs) produced are found are then re-routed. In this case the VFAs are fed to two bio-oxidation tanks to perform a selection of specific bacteria capable of producing polyhydroxybutyrate (PHB). The bacteria are fed via the feast and famine (Majone et al., 2017) method so that PHB can accumulate within bacterial cells. Finally, an extraction step kills the bacteria and the polymer is recovered. The model was built using Superpro Designer, further information can be found in (G. Croxatto Vega et al., 2020).

2.1.3. Solvent extraction of polyphenols

Extraction of polyphenols is accomplished by the use of solvents, which are mixed with wine pomace that has undergone the wine production process. The amounts of solvent used was optimized in Ferri et al., (2020) and previously analyzed with LCA by the authors (G. C. Croxatto Vega et al., 2020). From this previous work the most promising solvent extraction set-up was chosen for scaling up to the regions in this work, i.e. a solvent extraction with a mix of water and acetone as solvent, in a ratio of 5:1 of solvent to dry matter (DM) content. The polyphenol extraction process consists of a grinding step for pomace grinding, an extraction step in an airtight vessel at 50°C, and a polyphenol separation step where the polyphenol is removed under vacuum and the solvent is recovered.

2.1.4. Filler production

Filler production consists of a series of grinding and drying steps to go from the raw material to dry particles that can be used for the production of composites together with a polymer matrix. Two grinding steps are necessary, the first achieves size ranges of 3-6 cm in size, while the second achieves particle sizes of 0.05-0.3 mm, which is the market ready size. Starting from the raw material, drying is done until a 95% DM content is achieved. This is done because moisture is not a desirable property for composite production.

2.2. Compatibility Matrix

Figure 1 shows the technology to feedstock compatibility, as well as a simplified process flow diagram of each technology, product and substituted product. As mentioned previously, the feedstock is first tested for compatibility with each technology in each region. The particularities of each region also have some influence in the substituted products as well as on the background electricity and heat provisioning for the assessed systems. The regions specific particularities are described further in section 2.4.

The most versatile of feedstocks, as seen in Figure 1, is wine pomace, since it can effectually be used in filler production, polyphenol production, AD or AD+PHB, while the least versatile is vine shoots, which can only be used for filler production. The different combinations depicted in Figure 1 yield a total of 6 biorefinery set-ups to be explored with 6 different feedstocks. This means that in phase 1 of this assessment, 16 mini-LCAs are done, per region, since for some feedstock, e.g. manures, only 2 biorefinery setups work (AD and AD+PHB), while for wine pomace all biorefinery setups come into play.

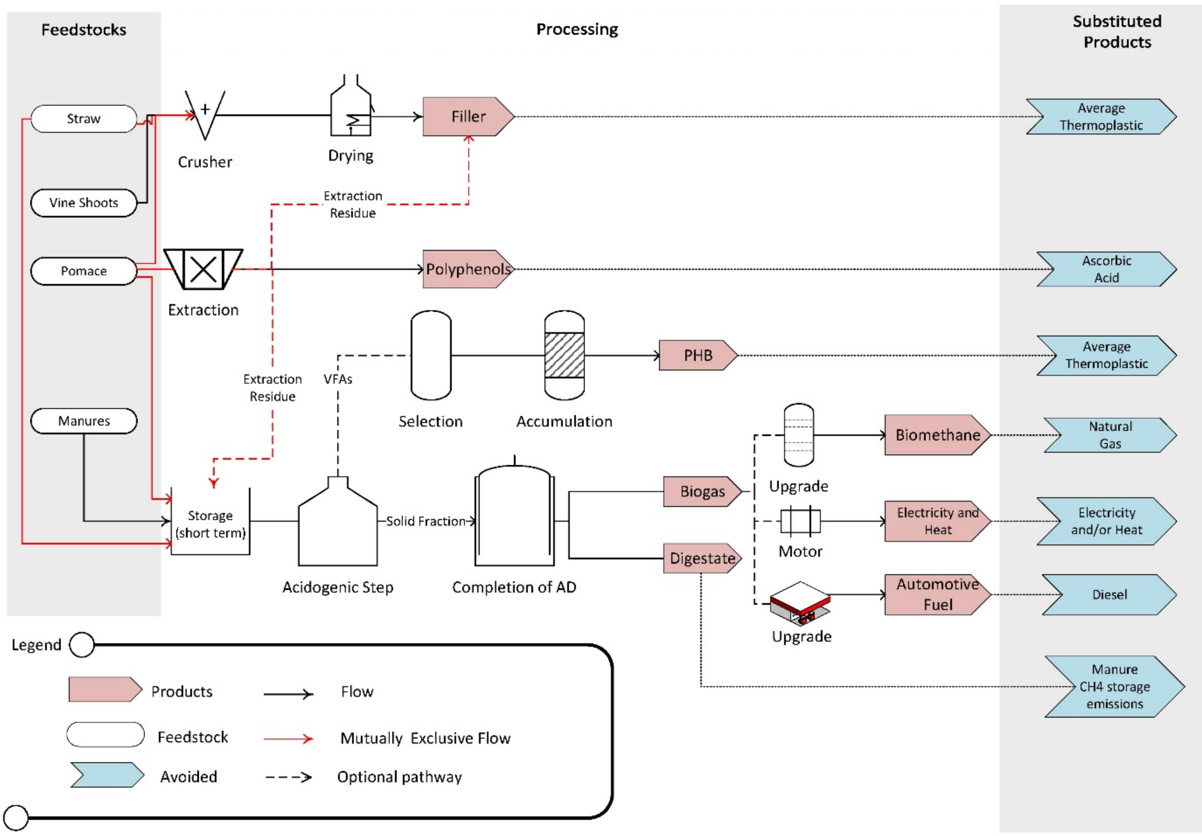

Figure 1 Technology to feedstock compatibility, showing possible combinations for the biotechnologies assessed.

2.3. Scope and System Boundaries

The functional unit for the mini-LCAs is the treatment of one ton of each feedstock in the region of interest, while at the regional level the functional unit becomes the treatment of the regional feedstock composition as determined by each region. Average inventory data were used for the assessment, since structural changes are not expected to occur as consequence of the biorefinery combinations.

The system boundary for the study is from cradle to grave, thus, residue production, treatment and end of life (EoL) of the products produced and of the substituted products are all assessed. Co-production and product substitution were handled via system expansion. The residues come into the system mostly burden free with the exception of straw, which is a commodity, thus it carries part of the burden from straw production. The Ecoinvent process for straw production in Europe was used for this purpose for all regions, since it is the most representative available data. The main products produced by the biorefinery setups are filler, PHB, biogas, digestate and polyphenols. The biogas produced is utilized in different ways depending on the region where it is produced (see Table 2). On the other hand, PHB and polyphenols substitute their conventional counterparts at a 1:1 ratio. In the case of PHB this is because it is assumed to substitute granules at factory gate, rather than assigning a specific application which might not come to be. Polyphenols are assumed to substitute ascorbic acid (Ferri et al., 2013; Rahim et al., 2008). Filler, on the other hand, substitutes global thermoplastic production, since the types of filler modelled in this work do not improve the material properties of the composite (Girones et al., 2017). The substitution is modelled as a 0.3:1 ratio of filler to thermoplastic by mass, based on the technical maximum amount that can be mixed into the polymer matrix (Berthet et al., 2015). Digestate is thought to substitute manure, thus offset from avoided storage of manure is considered, while emissions arising from application are excluded from the assessment, since they most likely fall within the same emission range (Möller and Müller, 2012; Nkoa, 2014).

2.4. LCIA Methods

Once the inventory is complete for each region, feedstock and technology, impacts are calculated using the ReCiPe 2016 Hierarchist (Huijbregts et al., 2016) life cycle impact assessment (LCIA) method at midpoints and endpoints using the OpenLCA Software (GreenDelta, 2019). Two approaches are taken to evaluate the results from the mini-LCAs of the feedstock/technology compatibility per region. First, impacts are summarized for global warming potential (GWP), using the midpoint ReCipe 2016 LCIA method. Second, ReCiPe endpoint damages are monetized to produce a monetized environmental damage (MED) single score that includes the influence from all impact categories. Monetization of endpoint results is carried out by using the value of 65,000 USD_{2003} per species year, which is a willingness to pay valuation of species.yr lost or gained derived by Weidema, (2009) and applied to ecosystem damages. Similarly, a value of 110,000 USD_{2003} is applied for disability adjusted life years (DALY), to express human health damage (Dong et al., 2019; Weidema, 2009). Resource scarcity is already expressed in monetary terms and it is thereby left as is for inclusion in the MED.

2.5. Regional Scale up

After impacts are obtained on a per ton of specific feedstock per technology, it is possible to compare the environmental impact reduction potential of each feedstock and technology combination in terms of GWP and MED taking all impact categories into account, as described in section 2.3. With these results, an objective choice is made regarding the application of technologies that brings about the highest potential environmental savings for the region in question. The mini-LCAs serve as a tool to prioritize the routing of feedstock to those technologies that are most compatible with said feedstock, thereby, resulting in higher impact savings. The inventory collection for each region assessed as well as regional differences are presented in the next section.

2.5.1. Regional Inventories

Four European regions were assessed i.e. Bavaria (DE), Languedoc-Roussilon (FR), Veneto (IT) and Skåne (SE), as well as Oregon State in the United States. Available amounts of agricultural residues in Bavaria, Languedoc-Roussillon and Veneto were estimated based on data from Eurostat (Eurostat, 2018). Skåne is not a defined region in the Eurostat database, thereby, national statistics were used (Jordbruksverket, 2019). Straw was calculated based on a ten year average for cereal production for all regions. The amounts of straw production were estimated using residue to crop ratios from the literature (Scarlat et al., 2010; Thorenz et al., 2018). Residue amounts were then turned into sustainable and technically feasible collection amounts. This was done by subtracting amounts of residues that must stay in the field to uphold soil quality, and residue amounts used for competing uses. This includes for example, the use of straw for bedding and feed for cattle and swine, which is calculated on a per head of livestock basis. A sustainable removal factor of 30% of produced residues (in fresh weight), was used for the European regions.

Manure production amounts were calculated using the factor for manure production on a per head of livestock type basis (Scarlat et al., 2018), multiplied by the number of days said livestock spends inside stables, since the manure produced on green pastures is considered uncollectable. For Skåne a grazing period of 120 days was assumed which is in accordance with national regulation (Jordbruksverket, 2020). The same grazing period was assumed for all non-organic cattle in all European regions. For all organic cattle in European countries, it was assumed that all cattle grazed year round. While this may not be accurate, as weather patterns might not permit, the percentage of organic cattle was very low, and a change in this factor would not be expected to cause more than 1% change in the total values for manure capture. For poultry and swine, it was assumed that the animals were housed year round.

For the state of Oregon, data on produced manure and residues was mostly collected from an inventory produced for the Oregon Legislature on the potential for Biogas production from different residues in the State (Oregon Department of Energy, 2018). For the manures availability, only confined animal feed operations were included, with grazed animals manures considered unrecoverable, and each specific operation was analyzed to produce a high quality estimate of manure capture potential. Since the values in this report are aggregated for crop residues, it was necessary to remove corn stover residues, which were not included in this assessment. In order to do so, data on corn production for 2017 was collected (United States Department of Agriculture and National Agricultural Statistics Service, 2017), which is the same data source as was used in the Oregon Department of Energy, report (2018). To calculate the mass of stover residues to remove from the inventory, a crop:residue ratio of 1 was used to convert primary production of corn to corn stover residues. The assumed sustainable removal rate of residues for Oregon was already included in the renewable natural gas inventory report (Oregon Department of Energy, 2018), and is 50%.

Additionally, data on regional differences in waste treatment was collected. This is needed to perform a cradle-to-grave assessment, since, for example, the production of PHB is modelled through EoL with the regionally specific waste treatment of plastics in each region. On the other hand, PHB in the AD+PHB scenarios substitutes the production and EoL of conventional thermoplastics. The same is true for the filler material, though in this case the filler is treated by mostly biological treatment routes. A description of the EoL treatment options applied to the products manufactured in the assessed systems is provided in Table 1, for avoided thermoplastic production, filler, and PHB. The average plastic treatment values (PlasticsEurope, 2018) have been adjusted for filler and PHB. For filler, the values from Plastics Europe were adjusted proportionally after removing recycling as an option. Taking today's infrastructure into consideration, only incineration or landfilling are viable treatment options for composite materials, since mixed composite materials are not easily separated into pure components for recycling and are likely not fully biodegradable. For PHB, a composting EoL is possible, and thereby, the values from Plastics Europe were adjusted with Eurostat composting rates for municipal solid waste treatment (Eurostat, 2020).

Table 1. EoL treatment options applied in the LCA model. Global thermoplastic for avoided plastic production, due to PHB production. PHB for EoL of the produced PHB and Filler for EoL of produced filler (PlasticsEurope, 2018).

	Global thermoplastic				
	Recycling	Incineration	Landfill	Litter[2]	
DE	26.50%	70.70%	0.80%	2.00%	
FR	16.40%	49.30%	32.30%	2.00%	
IT	21.30%	41.60%	35.10%	2.00%	
SE	26.80%	70.50%	0.80%	2.00%	
OR[1]	20.60%	14.80%	62.70%	2.00%	
	Filler				
	Recycling	Incineration	Landfill	Litter[2]	
DE	0.00%	96.80%	1.20%	2.00%	
FR	0.00%	59.40%	38.60%	2.00%	
IT	0.00%	53.40%	44.60%	2.00%	
SE	0.00%	96.90%	1.10%	2.00%	
OR[1]	0.00%	21.10%	78.90%	2.00%	
	PHB				
	Recycling	Incineration	Landfill	Litter[2]	Composting
DE	0.00%	60.20%	0.70%	2.00%	37.40%
FR	0.00%	41.60%	27.20%	2.00%	29.50%
IT	0.00%	34.50%	29.10%	2.00%	34.80%
SE	0.00%	61.00%	0.70%	2.00%	36.70%
OR[1]	0.00%	8.40%	88.80%	2.00%	0.00%

[1] (Shepperd et al., 2018)
[2] (Jambeck et al., 2015)

Another area in which the regions differ is in the methods for utilization of the biogas produced in the systems assessed. For example, in Bavaria where the biogas sector is at a mature stage, biogas burned in combined heat and power motors produces electricity and heat and a large percentage of the heat is utilized for heating nearby villages and industry (FNR - Fachagentur Nachwachsende Rohstoffe e.V., 2019). Conversely, in Italy biogas is mostly used for the production of electricity, while heat is a waste by-product, though the same type of motor is used (Benato and Macor, 2019). Table 2 describes the utilization of biogas when this is a product of the system assessed. The only two regions where the co-produced heat is utilized are Bavaria and Skåne. For these two the heat is utilized at a ratio of 0.52 kWt/kWe (FNR - Fachagentur Nachwachsende Rohstoffe e.V., 2019) and 0.8 kWt/kWe, for Bavaria and Skåne, respectively.

Table 2 Biogas utilization in the various regions.

	DE[1]	FR[2]	IT[3]	SE[4]	OR[5]
Cogeneration	90%	100%	98%	1%	33%
Fuel	4%	0%	0%	76%	0%
Heat	4%	0%	0%	5%	0%
Export	2%	0%	2%	0%	0%
Natural gas	0%	0%	0%	14%	67%
Flaring	0%	0%	0%	4%	0%

[1] (FNR - Fachagentur Nachwachsende Rohstoffe e.V., 2019)
[2] Own estimate
[3] (Bozzetto et al., 2017)

[4] (Swedish Energy Agency, 2018)
[5](United States Environmental Protection Agegency, 2020)

2.5.2. Regional Technology Selection

In order to select the most environmentally preferable technology implementation for a given region, an order of preference metric was used. This metric was carried out through a sequence of logic consisting of a series of binary questions. The first of these questions is 'is this technology shown to be the most environmentally preferable (based on the given environmental impact measurement, technology, and feedstock pairing)'. If the answer is yes, then it is assumed that as much of the feedstock as technically possible should be used in said technology, barring an overriding factor. If the answer is no, then the next available technology for the feedstock is queried in the manner previously described. Once the most preferable technology for a given feedstock-region pairing is selected, it is determined if the maximal use of said feedstock would preclude the most preferable use of any other assessed feedstock available in said region. If no preclusion is found, then it is assumed that a maximum technically feasible amount of the given feedstock should be allocated to said feedstock's most environmentally preferable technology. If preclusion of another technology that is most preferable for another feedstock is found, then the two competing technologies are compared. This is carried out as follows: if, based on a given region and its feedstock availability, there are two competing feedstocks, F_a and F_b, with two technologies, T_a and T_b, the potential environmental impact of utilization of a technically maximal amount of F_a and F_b in T_a is compared to the potential environmental impact of utilization of a technically maximal amount of F_a and F_b in T_b. Thus, if the lesser environmentally valued utilization of a feedstock-technology pairing results in an overall system benefit, the lesser environmentally valued technology is still selected. This results in a given regional system utilizing the overall most environmentally beneficial mix of technology-feedstock pairings as possible.

3. Results

3.1. Regional technology order of preference

For the initial comparison of technology-region-feedstock groupings, all groupings were assessed using both GWP impact as a single score and MED. This allowed for a comparison of feedstock-technology pairings for each region (Figure 2 and Figure 3).

This comparison resulted in a clear relationship between technologies in the technology-region-feedstock groupings for the manure feedstocks, with only a few notable exceptions. In nearly all cases, AD is preferred over AD+PHB, exceptions include Skåne, where MED indicate a preference toward AD+PHB, however in all cases MED also indicated that introducing any of the assessed technologies would induce net damage. Also, in Oregon and Languedoc-Roussillon, GWP shows effective parity between AD and AD+PHB. The MED, however, indicate a strong preference for AD in these cases.

Unlike for the manure feedstocks, the solid residue feedstocks i.e. straw, vine shoots and pomace, do not present as clear a preference. This comparison also presents some of the largest contradictions between MED and GWP interpretation. For example, in Skåne, all AD technologies show significant GWP savings potential while inducing MED. The same can be seen for filler production using straw and vine shoots in Oregon.

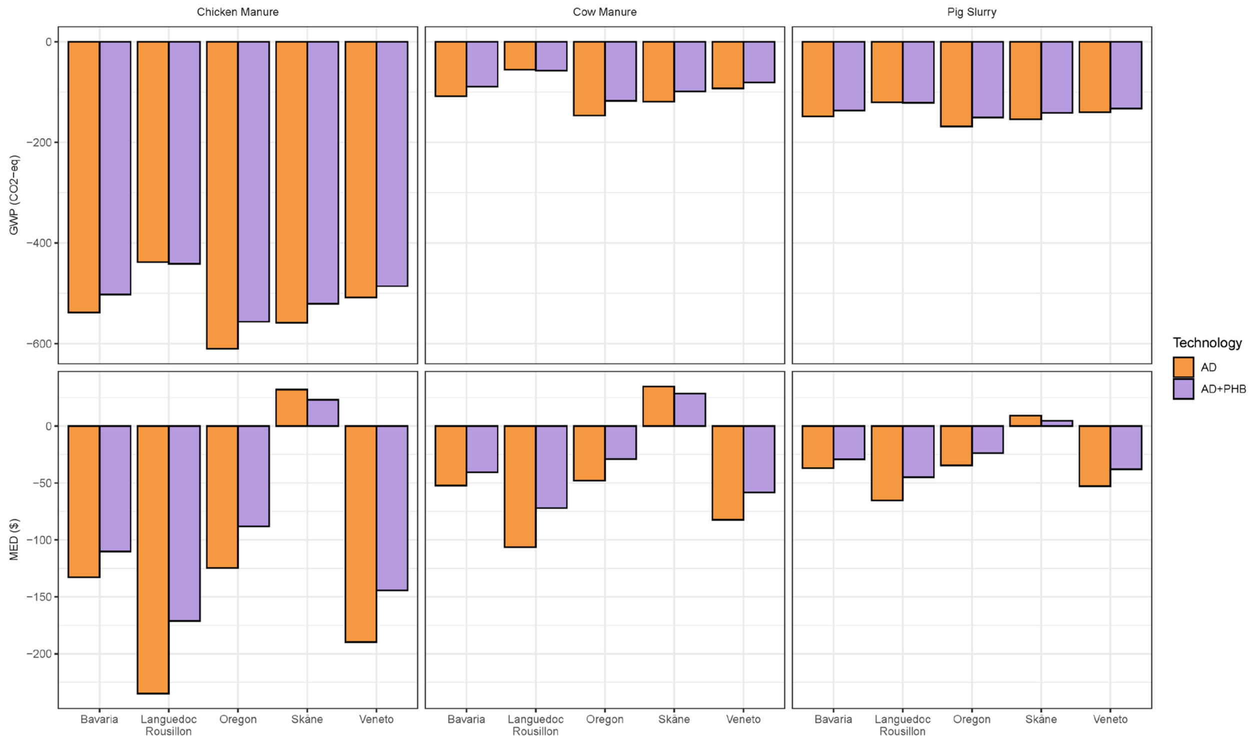

Figure 2: GWP impacts and MED for manure feedstock-region-technology pairings.

When applying the selection metric (Table 3), in most cases within each interpretation method there is little competing interest amongst technologies, insomuch that the preferred technology is either preferred or of equal preference (within a 15% error margin) for all feedstocks in a given region. However when comparing between GWP and MED interpretation of environmental impacts, there is some disharmony. Most notably, this occurs in Oregon, where vine shoots utilized to produce filler induce savings of GWP, but also appear to induce MED. And, following this issue for wine pomace in Oregon, AD is the preferred technology from a MED perspective while filler is the preferred technology from the perspective of GWP. This also occurs for straw in Bavaria, where AD is the preferred technology from a GWP perspective while filler is the preferred technology from a MED perspective.

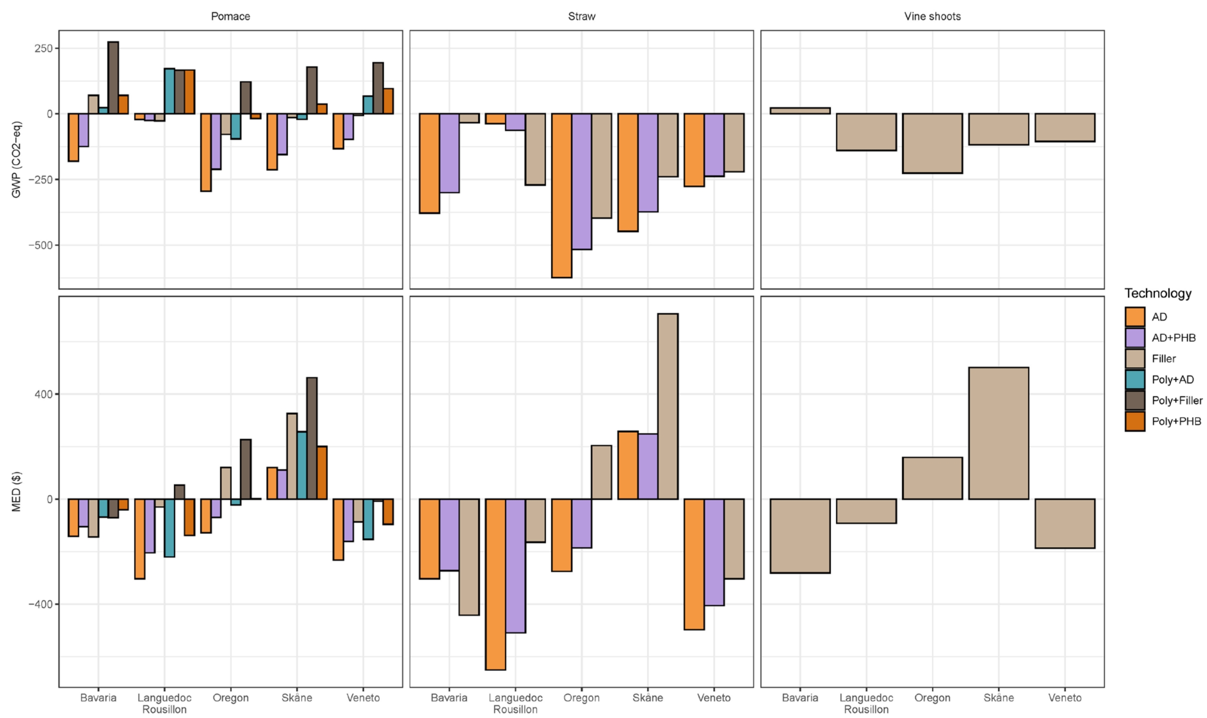

Figure 3: Global warming potential (GWP) impacts and monetized environmental damages (MED) for solid residue feedstock-region-technology pairings

Table 3: region-feedstock first technology preference using both monetized environmental damages (MED) and global warming potential (GWP) environmental impact interpretation. Technologies with an impact or damage preference lower than 15% are shown as equal. Vine shoots shown as either induced impact or savings, as there is no other competing technology for shares of the feedstock.

	Cow Manure		Pig Slurry		Chicken Manure		Grape Pomace						Straw			Vine Shoots
GWP	**AD**	**AD+PHB**	**AD**	**AD+PHB**	**AD**	**AD+PHB**	**AD**	**AD+PHB**	**Filler**	**Poly+AD**	**Poly+PHB**	**Poly+Filler**	**AD**	**AD+PHB**	**Filler**	**Filler**
Skåne	preferred	not preferred	equal	equal	equal	equal	preferred	not preferred	not preferred	not preferred	not preferred	not preferred	preferred	not preferred	not preferred	induced savings
Bavaria	preferred	not preferred	equal	equal	equal	equal	preferred	not preferred	not preferred	not preferred	not preferred	not preferred	preferred	not preferred	not preferred	induced impact
Languedoc Rousillon	equal	equal	equal	equal	equal	equal	not preferred	equal	equal	not preferred	not preferred	not preferred	not preferred	not preferred	preferred	induced savings
Veneto	equal	equal	equal	equal	equal	equal	preferred	not preferred	not preferred	not preferred	not preferred	not preferred	preferred	not preferred	not preferred	induced savings
Oregon	preferred	not preferred	equal	equal	equal	equal	not preferred	not preferred	preferred	not preferred	not preferred	not preferred	preferred	not preferred	not preferred	induced savings
MED																
Skåne	not preferred	preferred	not preferred	preferred	not preferred	preferred	equal	equal	not preferred	not preferred	not preferred	not preferred	equal	equal	not preferred	induced impact
Bavaria	preferred	not preferred	preferred	not preferred	preferred	not preferred	equal	not preferred	equal	not preferred	not preferred	not preferred	not preferred	not preferred	preferred	induced savings
Languedoc Rousillon	preferred	not preferred	preferred	not preferred	preferred	not preferred	preferred	not preferred	not preferred	not preferred	not preferred	not preferred	preferred	not preferred	not preferred	induced savings
Veneto	preferred	not preferred	preferred	not preferred	preferred	not preferred	preferred	not preferred	not preferred	not preferred	not preferred	not preferred	preferred	not preferred	not preferred	induced savings
Oregon	preferred	not preferred	preferred	not preferred	preferred	not preferred	preferred	not preferred	not preferred	not preferred	not preferred	not preferred	preferred	not preferred	not preferred	induced impact

3.2. Regional feedstock availability

After applying the regional technology preferences, it is possible to derive potential feedstock utilization pathways (Table 4). In all cases 100% of the manures is utilized, while for straw, there is a potential for incomplete utilization in AD or AD+PHA due to the technical limitations on the fraction of straw in AD and AD+PHB. In all cases where the straw could not be utilized in AD, it is possible to instead utilize the remainder in filler production, however, this may not result in an environmental benefit, and as such might be removed from the value chain.

Table 4: Feedstock utilization for value-chains. Each color represents a different potential value-chain: yellow: AD, green AD+PHA, orange: polyphenol extraction, and white*: filler production. *value chains marked with an asterisk are able to be added without excluding other value chains*

		Bavaria	**Veneto**	**Languedoc-Roussillon**	**Skåne**	**Oregon**
Area (km^2)		70 550	18 264	27 376	10 939	255 026
Inputs						
Manure (total)	Fraction to AD	100%	100%	100%	100%	100%
	Amount to AD (kton)	27 234	10 051	1 293	2 270	5 457
	Fraction to AD+PHA	100%	100%	100%	100%	100%
	Amount to AD+PHA	27 234	10 051	1 293	2 270	5 457
Straw	Fraction to AD	100%	100%	100%	100%	35%
	Amount to AD (kton)	3 150	381	120	408	963
	Fraction to filler	0%	0%	0%	0%	65%
	Amount to filler (kton)	0	0	0	0	1 794
	Fraction to AD+PHA	85%	100%	100%	69%	20%
	Amount to AD+PHA	2 693	381	120	281	540
	Fraction to filler	15%	0%	0%	31%	80%
	Amount to filler (kton)	457	0	0	127	2 217
Wine pomace	Fraction to AD	100%	100%	100%	100%	100%
	Amount to AD (kton)	0	264	491	0	0
	Fraction to AD+PHA	100%	100%	100%	100%	100%
	Amount to AD+PHA	0	264	491	0	0
	Fraction to filler	0%	0%	0%	0%	0%
	Amount to filler (kton)	0	0	0	0	0
	Fraction to polyphenols	100%	100%	100%	100%	100%
	Amount to polyphenols	0	264	491	0	0
Vine shoots	Fraction to filler	100%	100%	100%	100%	100%
	Amount to filler (kton)	0	131	443	0	0

3.3. Regional application of biorefinery systems for no agricultural waste

Based on the dispersion of feedstock as determined by the regional feedstock availability, the technology value chains were re-evaluated for each region. This resulted in the ability to determine value chains that would provide environmental value for each given region (Table 5). For most regions, introduction of the technology value chains produced either clear environmental benefit or damage, however in some region-technology pairings; there was disagreement between the MED and GWP. In particular, for polyphenol extraction in Veneto, MED indicates induced damages while GWP indicated impact savings. Conversely, for filler production in Skåne and Bavaria, MED indicates induced damage savings while GWP indicated induced impacts. All other technology-region pairings indicate environmental benefit from both GWP and MED perspectives for all technologies, except for in Oregon, where it is indicated that filler production would produce environmental detriment from both a GWP and MED perspective.

Table 5: Impact of technology value chain implementation for Bavaria (DE), Veneto (IT), Languedoc Roussillon (FR), Skåne (SE), and Oregon (US) for both global warming potential (GWP) and monetized environmental damages (MED)

	DE MED	DE GWP	IT MED	IT GWP	FR MED	FR GWP	SE MED	SE GWP	US MED	US GWP
AD	Induced savings	Induced savings	Induced savings	Induced savings	Induced savings	Induced savings	induced impact	Induced savings	Induced savings	Induced savings
AD+PHB	Induced savings	Induced savings	Induced savings	Induced savings	Induced savings	Induced savings	induced impact	Induced savings	Induced savings	Induced savings
Polyphenol-extraction	na	na	induced impact	Induced savings	Induced savings	Induced savings	na	na	na	na
Filler	Induced savings	induced impact	Induced savings	Induced savings	Induced savings	Induced savings	induced impact	induced impact	induced impact	induced impact

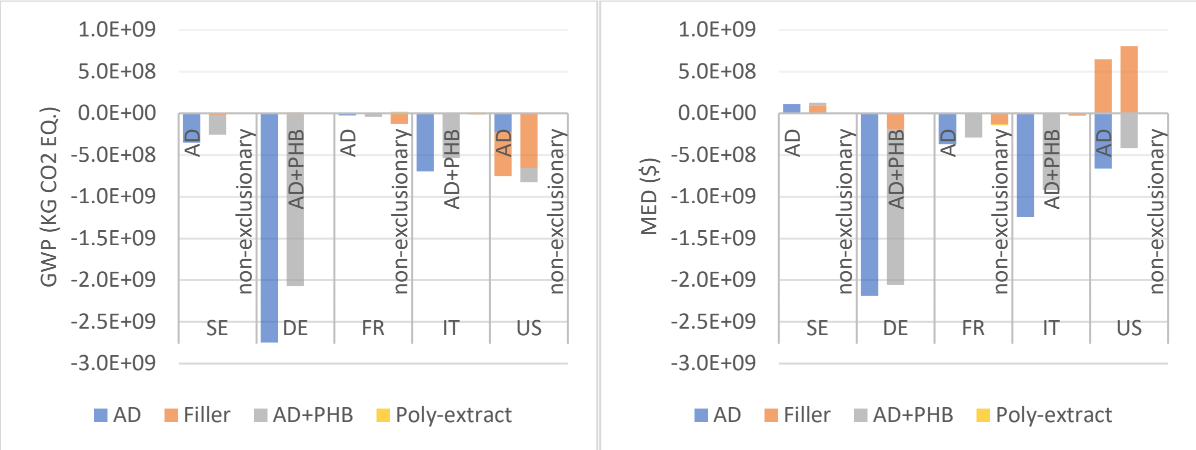

Figure 4: regional value chain implementation environmental impacts for Bavaria (DE), Veneto (IT), Languedoc Roussillon (FR), Skåne (SE), and Oregon (US) for both global warming potential (GWP) and monetized environmental damages (MED), non-exclusionary technologies utilize feedstock that do not preclude the utilization of any other technology in a potential value chain.

4. Discussion

In assessing the concept of no agricultural waste, a large number of smaller assessments were undertaken. When taken together, these assessments offer insight that would otherwise have not been as easily evident. However, because of the large number of assessments, as well as the large number of regions assessed, a number of assumptions were also made. Thus, these insights as well as the limitations of the conclusions that might be drawn are discussed below.

4.1. Feedstock

One of the first major elements that was assessed in order to undertake the evaluation of the various biorefinery value chains across multiple regions was the available feedstock. In this work, a specific group of feedstocks were selected both because they were available in all regions, fit the 2nd generation feedstock criteria, and were compatible with the assessed technologies. This selection is not representative of all available feedstock in the regions, and as such it should be assumed that there is more potential than what is shown here. One potential alternative would have been to select the most viable feedstocks for each region, e.g. those that are most prevalent based on local agricultural practice or those residues that are particularly problematic in terms of e.g. potential for vectoring disease or expense in treatment. However, this approach became too burdensome both in terms of data gathering for the feedstocks and for the impact that said alternative feedstocks would have on the assessed technologies. Furthermore, there were also a number of assumptions made in regard to the feedstock, in particular relative to the ratio of residue to product on the crops as well as the recovery rate for the animal manures. The recovery rate for animal manures is tested in a sensitivity analysis, but in all cases, this would not affect the performance of the technologies in a given region, though it could potentially affect the ratio of manures to crop residues such that the technology value chain shown for the region would no longer be viable. The ratio of manures to solid crop residues is an important aspect, which to some degree, determines the value chain combinations for a given region. As seen in the assessment (Figure 4), the various value chain combinations induce varying degrees of environmental performance, ergo this is a potentially influential aspect for decision making. Overall, though, this is considered to have little effect on the conclusions that might be drawn from this assessment, but should be considered when implementing technologies at a regional scale.

Another element that was not fully addressed in regard to the feedstocks is all potential for synergistic interactions. Thus, there is some potential that there would be greater yields from specific mixes of feedstock that were not captured by the modeled synergistic properties. On the other hand, the potential for various inhibitory factors for AD, such as ammonia nitrogen inhibition from chicken manure, or process failure due to high proportions of straw in the AD mix, were accounted for in the limits for feedstock ratios.

While these present some limitations, there is great inherent value in the ability to make a parallel assessment of the various value chains across a number of regions. Furthermore, this type of assessment provides an overview of the varying environmental profile of various agricultural residues. For example, it confirms once more the benefit of processing manures at least from a GWP perspective as seen in previous studies(Agostini et al., 2015; Styles et al., 2015). That said, the assessment also helps to point out that the environmental profile of the crop-based residues does not always provide environmental benefit in the assessed regions. A case-by-case approach is necessary in order to determine the full potential of each feedstock in its regional context. Thus, the use of this data despite the potential limitations is seen as valuable, in order to provide insight about where regulators and technology developers might focus efforts on developing specific technologies.

4.2. Sensitivity analyses

One of the primary limitations on data used in this assessment was for the manure feedstocks. In particular, the number of days that the animals were housed in a facility where manure could be collected is unknown. Thus, an assumption was made that, as in Sweden, all non-organic cattle are grazed for 120 days per year, however; it is possible that this assumption is not valid for the non-Swedish European regions. To check for sensitivity to this parameter, the feedstocks for these regions were assessed using a secondary assumption that all non-organic cattle were housed with potential for manure collection 365 days per year, which may have, conversely to the originally used assumption, result in an overestimation of available manure. After implementing the secondary assumption, it was observed that for Bavaria, the greater availability of straw would allow for treatment of all straw in AD or AD+PHA, instead of having remainder available for filler production. Overall, this would not otherwise affect the conclusions of the assessment, and as such there was deemed little overall sensitivity to this parameter.

In effect, the use of two interpretation methods, GWP and MED, acts as a sensitivity analysis. Here we do see significant sensitivity to interpretation method. For example, when analyzing cow manure in AD in Skåne, GWP indicates one of the greater reported environmental impact savings, while MED reports induced damages nearly at the same scale as damage reductions reported in other regions. This indicates that, while GWP may be an important issue in current environmental considerations and political discourse, as reported in many cases before (Laurent et al., 2012), using only GWP as a proxy for environmental impacts might lead to conclusions that are not always indicative of the entire environmental profile. This is discussed further in section 4.3.

4.3. Interpretation

In the assessment of the region-feedstock-technology pairings, both GWP and MED were utilized in order to assess if significant burden shift was occurring. In most cases, it was observed that GWP and MED were aligned in terms of technology preferences as well as the indication of environmental benefit or detriment. However, in a few cases this alignment did not hold true. In most cases where this occurred, however, this incongruity should be expected. For example, in Skåne, where the energy grid has largely decarbonized, most AD technologies will perform very well from a GWP perspective. But, while there have been significant reductions in the carbon intensity of the Swedish energy grid over the last several decades, the same cannot be said for all impact categories. Thus, the disagreement between GWP and MED for the selection of AD or AD+PHB was somewhat expected.

These situations do, however leave an issue in regards to the interpretation of the assessment. In this regard, one would typically look to margins and simply calculate respective magnitude of impact in order to make an informed, if essentially political decision. But, in the case of AD in Skåne, there is a further issue, namely that not only is the incongruity in the choice of technology, but there is also a reversal in the evident environmental impact – namely that when looking from a GWP perspective, environmental savings are predicted while when approaching from an MED perspective environmental damages are predicted. Thus, while it can be concluded that from a MED perspective, the decision to implement these technologies in Skåne does not make sense, the decision of whether or not such technologies should be pursued in such a region is ultimately a political one where the reduction of GWP is weighed against the other quantitatively larger impacts.

4.4. Impact of regionality

Another effect that can clearly be seen from the results of this assessment is that of regionality. The effect of the low carbon energy grid in Sweden plays a large role in the potential effects of the implementation of AD technologies. In a similar way, the development of municipal waste treatment plays a large role in whether or not filler materials are of value environmentally, as in regions where there are methods of valorizing the waste plastic apart from recycling, there is opportunity for reduced impacts from the utilization of plastics with filler materials.

Based on the sensitivity to this regionality, it should also be noted that these systems would be equally sensitive to temporally based changes in the background system, which is of particular importance, as in most cases, these technologies represent significant infrastructure that is likely to be in place for many years if constructed. As such, it would be beneficial in any case where recommendations regarding the implementation of these technologies are to be used, that at a minimum a prospective LCA and preferable a dynamic LCA be performed.

4.5. Contribution Analysis

Along with the variations that can be seen from region to region, a contribution analysis can help to shed light on some of the disparity seen between GWP and MED interpretation. For example, for Skåne, where we see one of the greatest differences between GWP and MED, this is particularly true. Based on the contribution analysis, it is easy to see some of the attributes of the Skåne region (Figure 5). In particular, looking at the impact of electricity production, it is easy to see that in terms of GWP, electricity does not contribute in any substantial way. This is due in large part to the significant decarbonization that has occurred in the Swedish energy grid, such that the carbon intensity of electricity production ranges from appx. 3-30 times less than the other assessed regions. However, this reduction in impacts does not hold true for all impacts, such as for ionizing radiation – where impacts are similar or greater than the other assessed regions or water consumption – where the impacts are appx. 10 greater than all other regions except Oregon. And, as such when assessing the system with MED, impacts from electricity inputs outweigh the environmental savings, which come predominantly from reducing the storage of manures, natural gas combustion, and diesel combustion.

Similarly in Oregon for the filler, the energy grid exhibits relatively more important impacts in categories other than global warming potential. Thus when looking at filler production from straw and vine shoots, while only looking at GWP shows environmental benefit, when looking at MED the proportionally greater impacts of electricity provision coupled with proportionally smaller impacts from avoiding the production of plastics lead to an indication of overall environmental damage.

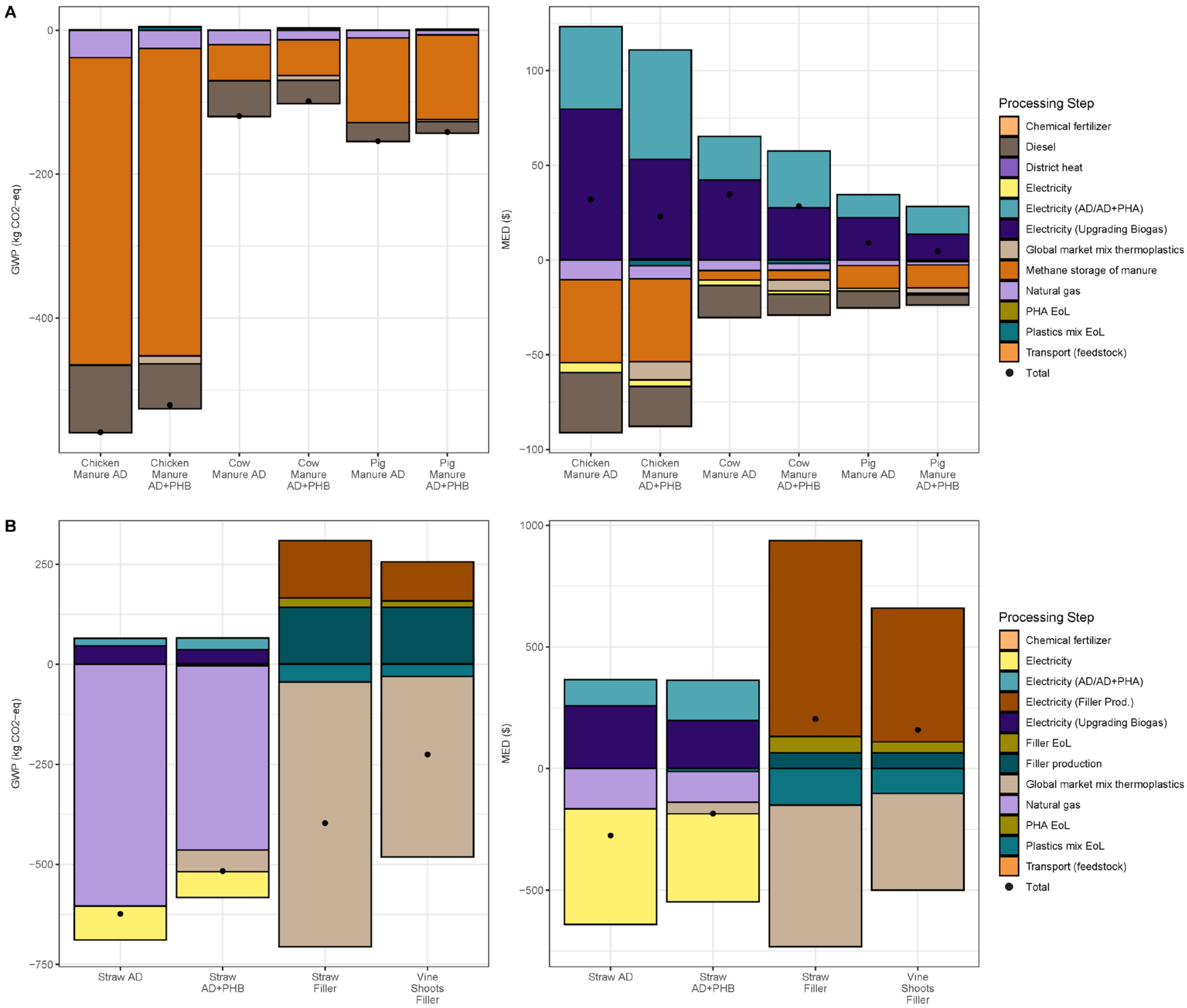

Figure 5 Contribution analysis for impacts (GWP) and damages (MED) from biorefining manures in A) the Skåne region and B) straw and vine shoots in Oregon.

4.6. Potential benefits and drawbacks of no agricultural waste

The bioeconomy is often spoken about in popular culture as a silver bullet for global environmental problems. However, as illustrated in earlier studies as well as here, caution should be used when making the assumption that products derived from bio-wastes are inherently environmentally friendly. As seen in Skåne, there can be tradeoffs even in technologies that appear beneficial for climate change. Thus, care should be taken when approaching the bioeconomy as a solution for environmental issues. A second implication of these findings is that optimization of environmental impacts is necessary from all areas that cause impact, not only from a decarbonization perspective. The evolution of the electricity grid in Skåne, which can be partly attributed to a political will to decarbonize the grid, is shown to trade carbon impacts for other still important impacts leading to damages. From the contribution analysis, it is evident that most of the impacts come from use of electricity, highlighting the need to decarbonize the electricity grid while avoiding burden shifting. Still more crucial, is the need to choose effective measures, when a level of decarbonization in the energy grid as high as Skåne's has been attained. Thus, when no low hanging fruit is available in the target region, these results illustrate the difficult demands on biotechnology developers for designing low energy consuming and high yielding options in order to provide environmental benefits in all areas of environmental impact.

On the other hand, it is also shown here, that when the background systems and available feedstocks align, there is great opportunity for environmental benefit. For example, in Bavaria, assuming an annual carbon footprint per capita in Bavaria of 9.35 tons, the application AD would offset the annual emissions of nearly 300,000 people or a little bit under 2.2 billion dollars of environmental damages.

5. Conclusions

Based on the assessments made on the various biorefinery technologies presented here, several conclusions can be made. The first is that while the bioeconomy is often presented in public discourse as intrinsically environmentally friendly, the reality is much more complicated. In particular, in regions where the energy grid is already optimized for carbon intensity, or as time passes and energy grids become more environmentally optimized, AD becomes less beneficial as a source of electricity production. Consequently, in regions where AD is being contemplated, an assessment including the status quo as well as the energy grid development during a time equivalent to the service life of the AD installation should be made prior to making conclusions regarding the benefits of a specific technology application. Despite this limitation, it can also be concluded that there is great opportunity for environmental impact reductions through the implementation of these technologies in most of the assessed regions. However, care should be taken to assess in which areas i.e. GWP or MED, environmental benefits can come to fruition together with which value chains provide the highest environmental benefit. Thus, this assessment illustrates that, across multiple regions, there is great potential for large-scale implementation of biorefineries as a tool for ameliorating environmental damages.

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PAPER IX

Lessons from combining techno-economic and life cycle assessment–a case study of polyphenol extraction from waste resources. *Publication in conference proceedings Heraklion 2019 – 7th International Conference on Sustainable Solid Waste Management.*

Vega, G. C., Voogt, J., Nilsson, A. E., Sohn, J., Birkved, M., & Olsen, S. I

Lessons from combining techno-economic and life cycle assessment – a case study of polyphenol extraction from waste resources

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Key words: Techno-economic assessment, Life Cycle Assessment, polyphenol extraction, solvent extraction, pressurized liquid extraction

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Abstract

Purpose: To analyze the environmental and economic performance of polyphenol extraction methods being developed within the NoAW project to valorize agricultural residues. And to utilize life cycle and techno-economic assessment as tools for this purpose.

Methods: LCA is applied at an early design stage to obtain a preliminary carbon footprint of the polyphenol extraction methods. The extraction methods tested are solvent extraction and pressurized liquid extraction (PLE). Subsequently, TEA-LCA is applied in simulated industrial conditions, optimized with guidance from literature and the preliminary LCA.

Results: The lab scale results highlight the need to reduce solvent use and maximize yields. The best option selected through the TEA-LCA is PLE, using CO2:EtOH:H2O as solvent with a solvent to dry weight ratio of 5, and 2 extraction steps (PLE-EtOH-5). This is in part due to higher yields for the TEA, and the use of ethanol for the LCA, which is a less environmentally burdensome solvent than acetone.

Conclusions: If the same yields as in lab scale can be attained at the designed industrial scale, then the PLE-EtOH-5 option leads to the highest environmental and economic benefits, despite higher capital expenditure. The LCA at lab scale was useful in pointing out potential environmental hotspots, which served to guide the TEA in order to design a better performing process from both an environmental and economic perspective.

1. Introduction

Biomass demand for the production of bioenergy, biomaterials and biochemicals is estimated to increase by 70-110 % by 2050 compared to 2005 levels [1]. A paradigm shift to renewable sources of production has long been discussed, in the context of circular economy and valorization of biomass waste resources produced through the agricultural value chain. The bioeconomy today is estimated to have a 2.4 € billion annual turnover, which is only expected to increase in the future [2]. Yet, the prefix bio does not guarantee sustainability. For example, growing biomass for biofuels has long been debated, prompting the Renewable Energy Directive [3] at a European level to ensure validity of greenhouse gas reductions claims. In this regard, integration of quantitative sustainability assessment such as life cycle assessment (LCA) and techno-economic (TEA) assessment have been regarded as valuable. Combined TEA-LCA has been applied in many occasions to assess the environmental and economic ramifications of implementing new technologies. Amongst the many of the studies utilizing this method are: the novel use of lignocellulosic material for production of biodiesel from palm oil residues [4], production of biofuels and bioresins [5], and bioblend stocks for the light and heavy-duty transport [6]. More interestingly, TEA-LCA has been used for quantifying and monetize externalities in the form of Disability Adjusted Life Years (DALY) to provide a more complete picture of the financial burdens arising from environmental problems [7], [8]. Recently, combining TEA and LCA has been used to optimize new production routes from an early design phase, such as the integration of wastewater into microalgae production for biodiesel production [9], or the integration of power-to-gas technology of methane and photovoltaics [10]. Combined TEA and LCA lends itself well to finding production hot spots and opportunities for optimization. This is even more relevant when applied to renewable resources such as biomass, which have to be managed sustainably.

Agricultural residues are an increasingly important biomass resource, which continues to be studied to increase maturity level of 2G and 3G production. In this context, the H2020 No Agricultural Waste (NoAW) project is working toward the development of sustainable value added products from agricultural residues, such as biocomposites, biodegradable bioplastics, and others [11]. Among these agricultural residues, wine pomace is a residue rich in polyphenols, which are compounds with high antioxidant value [12]. Polyphenol extraction methods at the laboratory scale can be analyzed using TEA-LCA in order to identify hotspots and potentially environmentally problematic production steps. Therefore, in this study LCA is applied at an early design stage to obtain a preliminary carbon footprint of the polyphenol extraction methods. Subsequently, TEA-LCA is applied in simulated industrial conditions, optimized with guidance from literature and the preliminary LCA. The goal is to obtain a holistic picture of the economic feasibility and possible environmental impacts of each polyphenol extraction method.

2. Methodology

Results of laboratory scale experiments of different methods for the extraction of polyphenols from red grape pomace were evaluated using a combination of LCA and TEA. Based on the preliminary LCA of the laboratory scale experiments, industrial scale processes were designed. The industrial scale processes were thereafter analyzed with both LCA and TEA.

2.1. Polyphenol extraction methods and laboratory experiments

Various polyphenol extraction methods developed within the NoAW project were assessed. The extraction methods include both solvent extraction and pressurized liquid extraction (PLE).

2.1.1. Extraction with acetone – S-AcN

Batch extraction was performed in the laboratory with 75% acetone, 25% water as solvent, with a solvent to dry weight (DW) ratio of 11. Extraction was performed in an air tight vessel at 50°C at atmospheric pressure. The solvent and pomace were kept in contact for 2 hours. After this time the polyphenols were dissolved in the liquid phase from which they could be isolated and obtained as a powder. The polyphenol content was then analyzed. This set up was also tested for 1 and 4 hours.

2.1.2. Extraction with ethanol – S-EtOH

The same procedure as in 2.1.1 was tested with ethanol as solvent. Equal parts ethanol:H_2O were used for the extraction. Extraction times of 1, 2 and 4 hours were tested to observe their influence on yield. The S-EtOH was only examined at industrial scale (section 2.3 and 2.4).

2.1.3. Pressurized liquid extraction with ethanol – PLE-EtOH

Three different options for PLE were studied in the lab. PLE-EtOH-75 with 75% co-solvent composed of equal parts ethanol and water and 25% liquid CO_2. PLE-EtOH-100 is performed without liquid CO_2 and instead there is 100% co-solvent composed of equal parts ethanol and water. The extraction is performed at 80°C and 100 bar. While the third PLE option, PLE-EtOH-oil, is divided into two extraction steps. One with 100% supercritical CO_2 at 350 bar and 80°C for one hour, with a flow of CO_2 of 30g per minute, leading to the production an oily phenolic extract. A second extraction step with the same EtOH:H_2O:CO_2 ratio as applied for PLE-EtOH-75 is performed to obtain polyphenols as dry extract. The solvent flow for the second step was 8g per minute. As this is a continuous set up, both of these steps lead to an extremely high solvent to DW ratio. All extraction operational parameters are presented in Table 1.

All extraction processes listed leave behind the pomace residue, which can be further valorized using different methods not assessed in this study [11].

Table 1 Operational parameters of laboratory experiments.

Scenario Name	**S-AcN**	**PLE-EtOH-75**	**PLE-EtOH-100**	**PLE-EtOH-oil**
Yield (g polyphenol/kg DW)	47	48	44	49
Solvents				
- Water	25%	37.5%	50%	37.5%**
- Ethanol		37.5%	50%	37.5%**
- Acetone	75%			
- CO2		25%		100%*, 25%**
Solvent to DW ratio	11	101	101	583
Stages (no.)	1	1	1	2
Total extraction time (min)	120	30	30	90
Temperature (°C)	50	80	80	80
Pressure (bar)	1	100	100	350*, 100**

*first stage
** second stage

2.2. LCA of laboratory scale experiments

A preliminary LCA was performed on the extraction methods described above, using only the Global Warming potential (GWP) impact category as the environmental indicator. The ReCiPe 2016 Midpoint Hierarchist method [13], which has a 100 year time horizon from point of emission, was used as impact assessment method, supplied by the Ecoinvent 3.4 Database [14]. The functional unit for the LCA is 1 kg of polyphenols assuming equal functionality. The process design software, Superpro designer [15], was used to simulate the polyphenol extraction methods with industrial scale equipment. However, all operating parameters such as temperature, solvent to DW ratio, polyphenol yield, pressure, and extraction times among others, were kept equal to laboratory conditions (Table 1). Simplified flow diagrams with the industrial equipment used are shown in Figure 1 and Figure 2. The polyphenol producing plant is assumed to be placed in Italy and thereby, background processes for Italy from the Ecoinvent database were used as much as possible, e.g. the electricity grid.

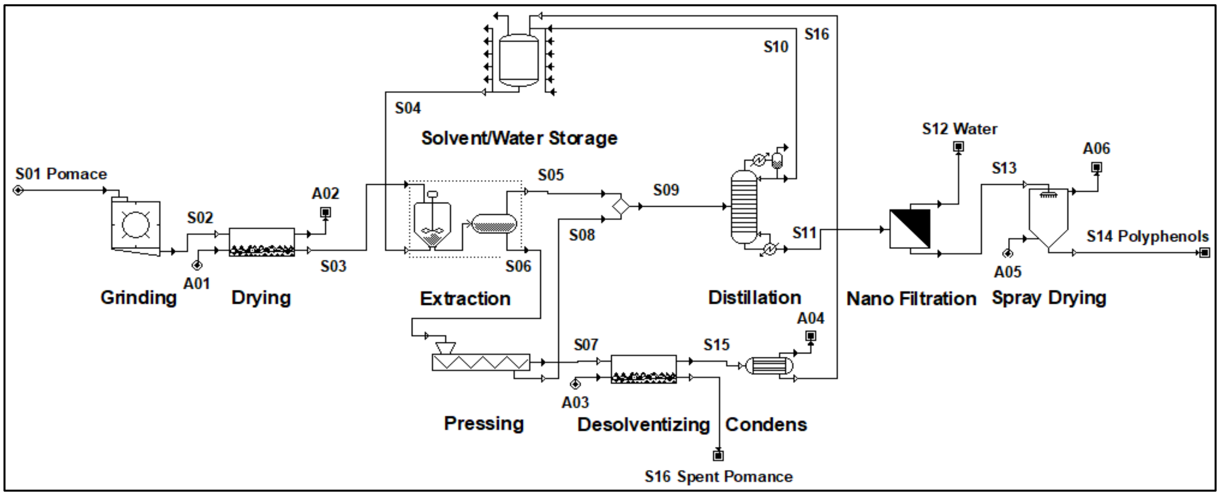

Figure 1 Solvent extraction with either acetone or ethanol at atmospheric pressure. The pomace dryer is optional.

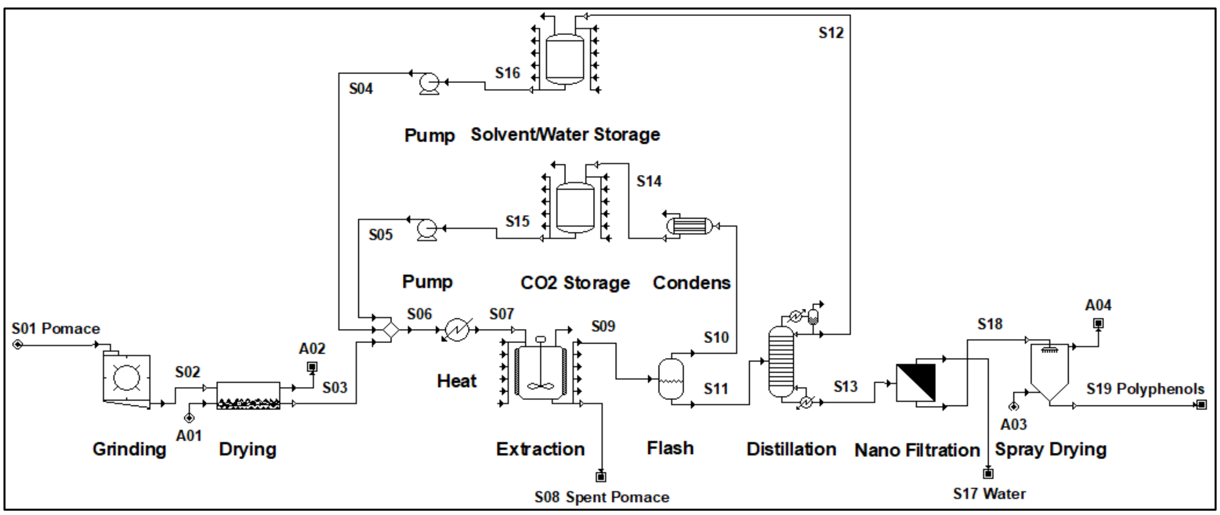

Figure 2 Pressurized liquid extraction with ethanol, water, and supercritical CO_2. The pomace dryer is optional.

2.3. TEA of industrial scale processes

Based on the results of the laboratory scale experiments, the preliminary LCA, and literature [16]–[20], industrial scale processes for solvent extraction and PLE were designed. TEA of the industrial scale processes designed was carried out in order to investigate the economic repercussions of installing a polyphenol extracting plant. The TEA includes Capital Expenditure (CapEx) and Operating Expenditure (OpEx). Assumptions and simplifications were made in order to fill data gaps. Assumptions of economic parameters and estimates of fixed capital costs were based on [15], [21]–[24]. The most important assumptions are reported in Table 2.

Table 2 Parameters for the techno-economic assessment.

Production	8000	h/y
Red pomace	20	kton wet/y
	2500	kg wet/h
	36.2%	DW
Labour related costs	891	k€/y
Plant related costs	10%	of fixed capital/y
Financing costs	10%	of fixed capital/y
Electricity	10%	€/kWh
Steam	25	€/ton
Solvent price		
- Water	0.00	€/kg
- Ethanol	0.80	€/kg
- Acetone	1.20	€/kg
- CO_2	0.50	€/kg
Solvent ΔH evaporation		
- Water	2260	kJ/kg
- Ethanol	841	kJ/kg
- Acetone	539	kJ/kg
- CO_2	380	kJ/kg
Solvent loss	2%	of recycle
Energy solvent recycle	2	x ΔH_{vap}

The labour related costs were assumed to be the same for all processes and are based on: 2 shift positions, an operator salary of k€ 30/y including supervision, direct salary overhead, and general plant overhead. The plant related costs include maintenance, tax, insurance, rent, overhead, environmental charges, and royalties. The financing costs are based on an amortization of the fixed capital costs over 10 years with no interest.

For all processes, a solvent loss of 2% of the solvent in the recycle is assumed. The energy which is required to recycle the solvent is estimated as two times the heat of evaporation. For the recycle of water, acetone, and ethanol, thermal energy is required, while for the recycle of CO_2, electricity is required.

2.4. LCA of industrial scale processes

Following the TEA, a complete accounting LCA was performed on the same systems analyzed for the TEA. The system boundary for the accounting LCA includes all actions carried out in order to obtain 1 kg of polyphenols from when the grape pomace enters the production system to the product leaving the production facility, e.g. all processing steps, such as grinding, drying, adding solvents, filtering, distillation and more (Figure 1 and Figure 2). On the other hand, the "gate-to-gate" LCA does not include end of life of the polyphenols or any transport throughout the life cycle. Furthermore, no allocation is performed on the impacts of polyphenol production, i.e. the entire burden of production is assigned to the main product, the polyphenols. Likewise, no credits are assigned for the production of polyphenols potentially replacing similar products in the market.

The LCA includes all 18 impact categories in ReCiPe 2016 Midpoint (H) methodology. As for the LCA at lab scale, the geographical location of the polyphenol plant is assumed again to be Italy.

To ease interpretation of results, a simple multi-criteria decision assessment (MCDA), was performed. First, results for the 18 impact categories were normalized within each impact category to the worst performing scenario and ranked. Second, normalized results for all impact categories were averaged for each extraction method respectively to obtain a single score per scenario, which was then used to single out the best performing scenario. The average results were compared with normalized Global Warming results in order to assess the possibility of burden shifting between GWP and other environmental impacts (categories).

3. Results

3.1. LCA of laboratory scale experiments

The carbon footprint analysis clearly shows that if laboratory conditions are maintained when implementing a polyphenol extraction plant, then the acetone based solvent extraction method outperforms all other scenarios by a large margin, in terms of global warming potential (GWP). This is largely due to the amounts of solvent used in each scenario, which are lowest for the S-AcN scenario. The large amount of solvent used in the continuous set up for all PLE scenarios results in a very high electricity and heating demand in, for example, electricity for compressing of the system, heating during polyphenol extraction, and heating during distillation to recover the solvents.

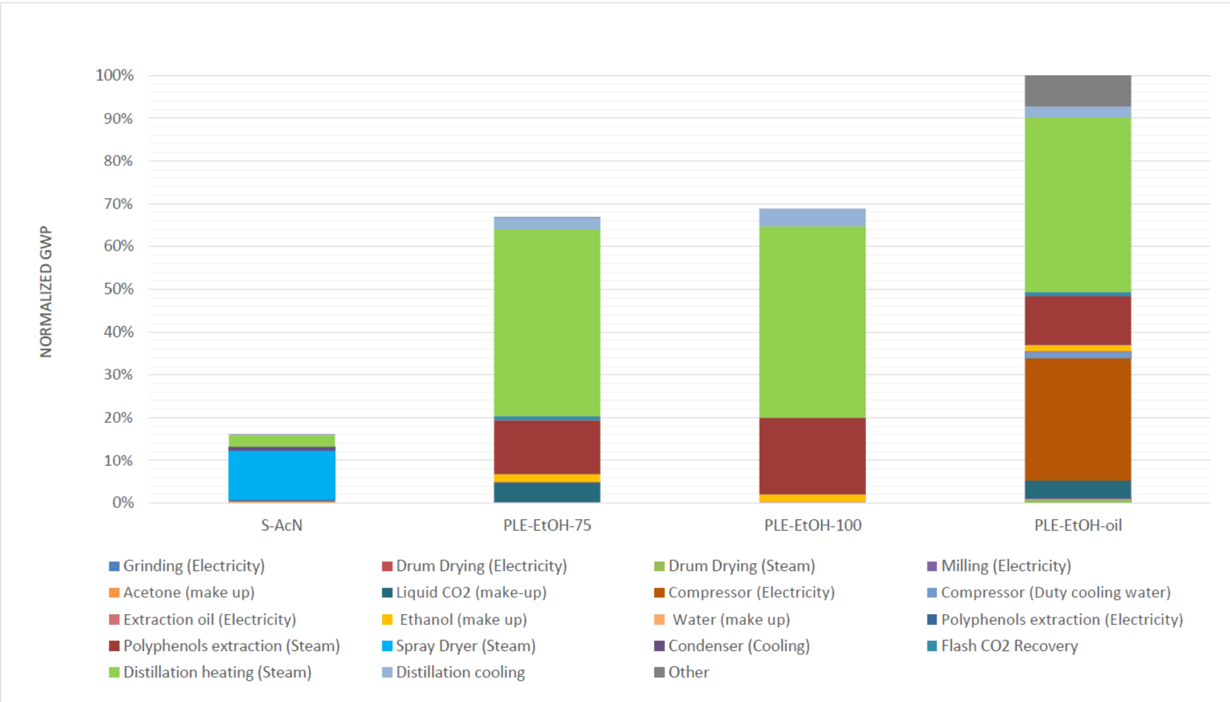

Figure 3 Normalized global warming potential results of polyphenol extraction scenarios at lab scale. Functional unit is 1 kg of polyphenols. Normalization to worst performing scenario PLE-EtOH-oil.

From the preliminary LCA, the importance of keeping the solvent ratio as low as possible is evident. This has a trickle down effect on the energy demand of the whole system. It was also proposed that the contact between solvent and pomace could be increased by changing the set up of the system. Systems with multiple extraction stages and lower solvent to pomace DW ratios were considered in the TEA.

3.2. TEA of industrial scale processes

The TEA focused on optimizing the operational parameters so that it would be economically feasible to implement a polyphenol extraction at industrial scale. Based on laboratory scale experiments and literature

[16]–[20], extraction steps were increased and as a result the solvent to pomace DW ratios decreased. Because water is already present in the pomace, it is necessary to dry the pomace prior to the extraction to maintain a solvent to DW ratio of 2 (S-AcN-2 and S-EtOH-2). Total extraction time was assumed to be 60 minutes for all processes. Equipment was scaled based on the flow sizes and subsequently the purchased equipment costs and fixed capital costs were estimated. The operational parameters and assumed extraction yields are given in Table 3.

Table 3 Operational parameters of designed industrial scale processes.

Scenario Name	**S-AcN-5**	**S-AcN-2**	**S-EtOH-5**	**S-EtOH-2**	**PLE-EtOH-10**	**PLE-EtOH-5**
Yield (g polyphenol/kg DW)	47	47	40	40	79	79
Solvents						
- Water	33%	33%	50%	50%	37.5%	37.5%
- Ethanol			50%	50%	37.5%	37.5%
- Acetone	67%	67%				
- CO2					25%	25%
Solvent to DW ratio	5	2	5	2	10	5
Stages (no.)	2	5	2	5	2	2
Total extraction time (min)	60	60	60	60	60	60
Temperature (°C)	50	50	50	50	80	80
Pressure (bar)	1	1	100	100	100	100
Fixed capital (M€)	4.6	4.1	4.7	4.0	9.4	6.5

The best performing scenario, in economic terms, is PLE-EtOH-5, which also has the highest polyphenol extraction yield. Despite lager fixed capital costs, the costs expressed per kg polyphenol are lower compared to the solvent extraction processes (Figure 4). The second best scenario is S-AcN-2, which has the advantage of a low solvent to DW ratio of 2 and similar cost range for plant related and financing cost. However, the heat demand for S-AcN-2 is larger, because drying of the pomace is required.

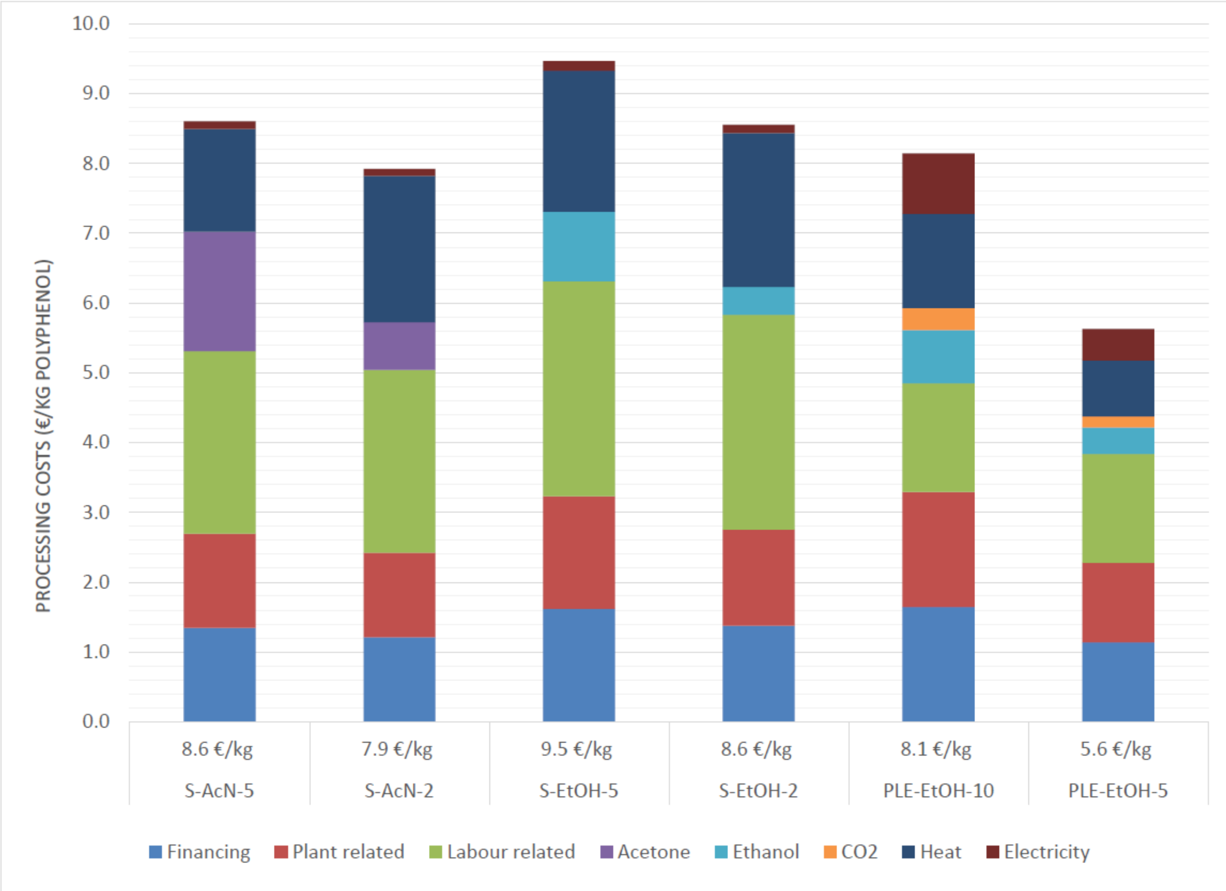

Figure 4 Techno-economic assessment results of optimized polyphenol extraction at industrial scale.

3.3. LCA of optimized industrial scale design

The LCA of optimized operational conditions showed that if seeking to alleviate environmental problems it would be preferable to choose PLE-EthOH-5, that is to say, a pressurized extraction that uses ethanol, water and supercritical CO_2 as solvent, with a solvent ratio of 5 and 2 extraction steps (blue bars, Figure 5). It is noteworthy to say that a solvent extraction using acetone with a solvent ratio of 2 (S-AcN-2) is potentially within the same range of impact when all impact categories for the LCA are equally weighted i.e. all environmental problems encompassed in the LCA are equally valued. If instead, the goal is to reduce global warming at the potential cost of other environmental problems, then the best choice is PLE-EtOH-5. PLE-EtOH-5 is the best performing scenario in terms of GWP. The upper error bar for this scenario represents the worse possible outcome for the scenario, when uncertainty is taken into consideration, which is here called the “GW acceptable value”. As such, scenarios above the dashed line will most likely lead to higher GWP impacts than PLE-EtOH-5. As can be seen in Figure 5, scenario S-AcN-2 just barely falls below the GW acceptable line, and only when taking into consideration a -10% uncertainty.

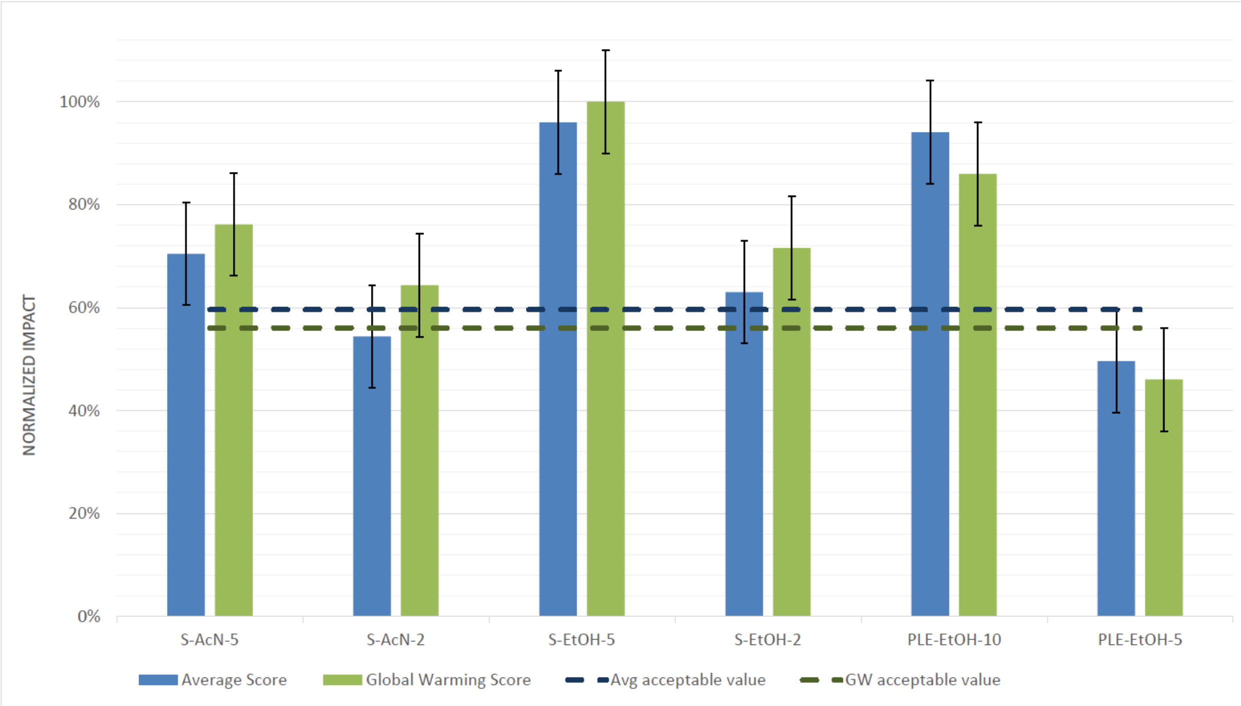

Figure 5 Single score impact results from the full LCA. Single scores are derived by internally normalizing results to the worst performing scenario and averaging all impact categories into a single score (blue bar). While for GWP, internally normalized results for each scenario are shown (green bar). An arbitrary uncertainty value of ±10% is depicted for each single score by the dashed lines, to show distance to the best solution. Error bars also show ±10% uncertainty level.

Results from the TEA align well with the LCA, which points out that, at least in this case, the same parameters that are "expensive" for the environment, are also costly for the investment.

4. Discussion

The preliminary LCA assessment performed on the lab scale emerging technologies can be used in the early design phase, in order to avoid excessive environmental burden later on. By identifying hot spots early on, it is possible to envision adjustments to the production set up, so that the identified hot spots are addressed. In this case, the environmental hot spots coincide well with economic costs, as is shown by the successive TEA-LCA. For both of these assessments, one of the most important parameters was solvent to wine pomace dry weight ratio. High use of solvent leads to high operational costs and increased demand for electricity and heat, which affect the results of both TEA and LCA. On the other hand, higher yields allow more leeway for higher energy consumption. This is observed in the results for PLE-EtOH-5, which has a very high electricity demand, due to the compressed system, but at the same time produces one of the highest yields out of the assessed scenarios. The high yield translates into reductions in the energy demand when looking at the results on a per kilo of product basis.

Results for the TEA showed that increasing the number of extraction steps has consequences for vessel volumes, which can be kept smaller if there is a higher number of extraction steps. In turn, this results in lower fixed capital costs for the extraction. On the other hand, to keep solvent ratios low, it is necessary to add a drying step before mixing the wine pomace, which contains water in itself. The extra drying incurs extra costs for heating, while at the same time saving some costs for material expenditure. These results are mirrored in the LCA, where results benefit from lower solvent use, while impacts are increased due to the extra heating needed. In this regard though, it was clear in the LCA that solvent use, especially if the solvent is acetone, comes with higher impacts than electricity or heat use. This is easily illustrated when looking at the GWP impacts of 1 kg of acetone compared to 1 kg of ethanol or 1 kWh of electricity, as shown in Figure 6, but also when looking at other impact categories (not shown here). From the figure it is possible to visualize that, in terms of the overall LCA assessment, added acetone or ethanol weigh more than added heat or electricity, with acetone being two times more burdensome than ethanol.

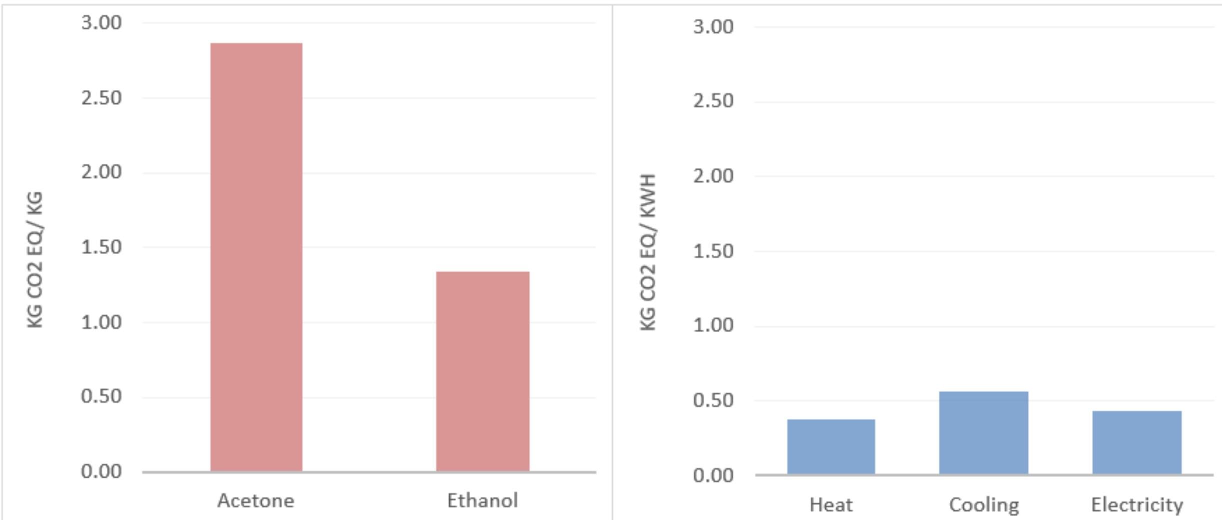

Figure 6 Global warming potential of 1 kg of acetone or ethanol. GWP of 1kWh of cooling, Italian electricity or heating. For illustrative purposes.

In this regard, it is also worth mentioning that the ethanol used for this assessment is of petrochemical origin. However, since the waste being treated is wine pomace, it is quite possible that a biorefinery treating this waste would also produce bioethanol. This is true for distilleries placed in Italy and France, which currently treat wine pomace in order to produce ethanol, bioenergy and food additives, among others.

Furthermore, the TEA in this study considers the processing costs including the financing costs. The market price of the product, the extracted polyphenols, and the market volume are yet to be explored. Once a market price or price range is known, then fixed capital costs and processing costs can be compared to the benefits, and profitability indicators, such as net present value (NPV) and internal rate of return (IRR), can be taken into consideration. A larger investment for more complex technology (PLE instead of solvent extraction) might be justified if the benefits are significantly lager.

Besides the economic (TEA) and environmental (LCA) aspects investigated, it is also useful to consider the technology readiness level (TRL) of the evaluated processes in the future. Solvent extraction, with both acetone and ethanol, is a mature process technology, which is currently implemented at large scale. PLE is a less mature technology for which extra measures might be required for large scale implementation.

5. Conclusion

Polyphenol extraction methods developed in the NoAW H2020 project were assessed using LCA at different maturity levels and with TEA-LCA at industrial scale. The lab scale results highlight the need to reduce solvent use and maximize yields. The best option selected through the TEA-LCA is pressurized liquid extraction, using CO_2:EtOH:H_2O as solvent with a solvent to DW ratio of 5, and 2 extraction steps (PLE-EtOH-5). If the same yields as in lab scale can be attained at industrial scale, then this option leads to the highest environmental and economic benefits, despite higher CAPEX. The most important parameter for optimization indicated by the LCA results is reducing solvent amounts. The most important parameters indicated by the TEA are the polyphenol extraction yield and the solvent to DW ratio. The LCA at lab scale was useful in pointing out potential environmental hotspots, which served to guide the TEA in order to design a better performing process from both an environmental and economic perspective.

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PAPER X

Incorporating Relative Importance: selecting a polyphenol production method for agro-waste treatment in an environmental and economic multi-criteria decision making context. (2019). *Heraklion 2019 – 7th International Conference on Sustainable Solid Waste Management.*

Sohn, J., Vega, G. C., Birkved, M., & Olsen, S.

Incorporating Relative Importance: selecting a polyphenol production method for agro-waste treatment in an environmental and economic multi-criteria decision making context

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Keywords: Multiple Criteria Decision Assessment, Life Cycle Assessment, Decision Support, Environmental-Economic Nexus.
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Abstract

Purpose: The No Agricultural Waste project is faced with selecting the best alternative amongst six extraction methods for polyphenol production used to upgrade agricultural residues.

Methods: In order to complete this, a multiple criteria decision assessment method, Technique for Order of Preference by Similarity to Ideal Solution (TOPSIS), is applied to results for the six extraction methods from techno-economic assessment and life cycle assessment carried out previously in the project. A normalization-based method of relating the weighting applied in the MCDA to the relative importance of environmental impacts in the assessment is applied, and decision support is provided for various levels of weight given to the economic impacts of the system.

Results: One clear ideal alternative, a pressurized liquid extraction method using Ethanol, Water & $SCCO_2$ solvent with a solvent ratio of 5, is specified, along with a second best alternative using acetone and water and a solvent ratio of two. The third best alternative depend on the weight given to economic impacts and the weighting applied amongst environmental impacts.

Conclusions: It is concluded that apart from the ideal alternative and the second ranked alternative, the third ranked alternative depends on the weight given to the economic indicator. Furthermore, the application of the relative importance factor for environmental criteria as a method of deriving weighting reduced the influence of criteria with impacts that are relatively unimportant in absolute terms.

1. Introduction

When policy makers, corporations, or any other actor is faced with the need to choose between alternative solutions to a given problem, there is often a multitude of issues to be taken into account. And, the decision-making context surrounding such a choice can be handled in many ways, from community-based decision making to round table discussions or even executive fiat. However, without a tool for handling fundamentally conflicting information, the results of decision making through discussion can vary wildly and may depend on happenstance and or subjective factors. Since its primary foundation in in the 1950's, Multiple Criteria Decision Analysis (MCDA) has been applied to aid in alleviating these problems by introducing a transparent and repeatable form of decision support [1].

When looking at environmental issues in life cycle assessment (LCA), oftentimes practitioners turn to single indicators such as global warming potential (carbon footprinting), but this poses potential downfalls such as burden shifting (e.g. shifting environmental burdens from carbon emissions to environmental or human toxicity) [2]. In other cases, practitioners turn to endpoint damage modeling, but these have high levels of uncertainty and still leave the decision maker with several categories of environmental damages (e.g. ecosystem health, human heath, and resource availability). Furthermore, neither of these methods can be directly combined with economic indicators. In some cases, LCA practitioners have monetized impacts in order to combine environmental and economic indicators, however these suffer from issues, among others, involving the relationship of internalized and externalized costs [3]. These issues have lead some LCA practitioners to turn to MCDA for providing decision support [4–6].

When applying many types of MCDA, though, there is one element that has a determining effect on decision support, namely weighting. In this paper, MCDA is applied to the decision context of a European Union Horizon 2020 project, No Agricultural Waste (NoAW), choosing between various developed technologies for extracting polyphenols as a means of upgrading agricultural wastes to agricultural co/by-products. A weighting-profile derivation framework is proposed in order to incorporate the relationship between the various environmental impact criteria that are the result of life cycle assessments and an absolute reference point for environmental impacts in order to avoid making a decision based on irrelevant criteria. The criteria from LCA and an economic analysis are then incorporated to provide decision support for selecting a technology for scale-up in the NoAW project.

2. Methodology

a. Definition of the case

The NoAW project will be selecting a technology for polyphenol extraction to undergo further testing at pilot scale, after having developed a number of extraction methods at lab scale. These include both processes using acetone and ethanol as a solvent (Table 1) and are further described in [7]. Amongst these six alternative extraction methods, one must be chosen for upscaling; however, due to the potential for technical issues, a second and third choice method for upscaling should also be chosen. Attributes of the various extraction methods are available in the form of ReCiPe 2016 [8] midpoint environmental impacts and a production cost that is obtained via a techno-economic assessment.

Table 1: Description of assessed alternative extraction methods with ReCiPe 2016 midpoint impacts and production cost shown per kg of gallic acid production [7]

	Solvent Extraction				Pressurized Liquid Extraction		
	Acetone & Water		Ethanol & Water		Ethanol, Water & $SCCO_2$		
	340 ton GA/y		290 ton GA/y		572 ton GA/y		
	solvent ratio: 5	solvent ratio: 2 (dryer required)	solvent ratio: 5	solvent ratio: 2 (dryer required)	solvent ratio: 10	solvent ratio: 5	
Impact	**S-AcN-5**	**S-AcN-2**	**S-EtOH-5**	**S-EtOH-2**	**PLE-EtOH-10**	**PLE-EtOH-5**	Unit
Fine particulate matter formation	2.26E-02	1.93E-02	2.81E-02	2.08E-02	2.62E-02	1.41E-02	kg PM2.5 eq
Fossil resource scarcity	1.13E+01	8.97E+00	1.43E+01	9.87E+00	1.20E+01	6.42E+00	kg oil eq
Freshwater ecotoxicity	3.09E-01	1.77E-01	4.63E-01	2.36E-01	4.38E-01	2.24E-01	kg 1,4-DCB
Freshwater eutrophication	3.47E-03	2.56E-03	5.27E-03	3.21E-03	5.26E-03	2.75E-03	kg P eq
Global warming	3.23E+01	2.73E+01	4.24E+01	3.03E+01	3.64E+01	1.95E+01	kg CO2 eq
Human carcinogenic toxicity	4.24E-01	2.89E-01	5.69E-01	3.40E-01	5.37E-01	2.80E-01	kg 1,4-DCB
Human non-carcinogenic toxicity	8.07E+00	4.77E+00	1.23E+01	6.36E+00	1.16E+01	5.95E+00	kg 1,4-DCB
Ionizing radiation	7.36E-01	7.00E-01	1.05E+00	8.05E-01	1.41E+00	7.48E-01	kBq Co-60 eq
Land use	1.97E-01	2.23E-01	2.93E-01	2.53E-01	3.42E-01	1.85E-01	m2a crop eq
Marine ecotoxicity	4.70E-01	2.85E-01	6.98E-01	3.71E-01	6.51E-01	3.35E-01	kg 1,4-DCB
Marine eutrophication	2.30E-04	1.80E-04	3.40E-04	2.20E-04	4.00E-04	2.10E-04	kg N eq
Mineral resource scarcity	2.82E-02	1.49E-02	4.37E-02	2.09E-02	4.02E-02	2.05E-02	kg Cu eq
Ozone formation, Human health	3.50E-02	2.94E-02	4.25E-02	3.14E-02	3.82E-02	2.05E-02	kg NOx eq
Ozone formation, Terrestrial ecosystems	3.64E-02	3.03E-02	4.42E-02	3.24E-02	3.95E-02	2.12E-02	kg NOx eq
Stratospheric ozone depletion	7.62E-06	6.29E-06	1.09E-05	7.42E-06	1.10E-05	5.80E-06	kg CFC11 eq
Terrestrial acidification	6.05E-02	5.43E-02	7.21E-02	5.70E-02	6.84E-02	3.70E-02	kg SO2 eq
Terrestrial ecotoxicity	4.05E+01	3.57E+01	5.99E+01	4.21E+01	5.24E+01	2.79E+01	kg 1,4-DCB
Water consumption	1.53E-01	8.65E-02	1.69E-01	9.24E-02	2.05E-01	1.06E-01	m3
Production cost	8.6	7.9	9.5	8.6	7	4.9	€

b. Application of MCDA

In order to incorporate the various environmental as well as the economic criteria, the Technique for Order of Preference by Similarity to Ideal Solution (TOPSIS) method of MCDA [9] is used. This is chosen due to its previous application in the context of LCA and because it is one of the most widely applied compensatory methods of MCDA when cardinal indicators are available for all alternatives [10]. This selection is further discussed in section 4.

All midpoint indicators from LCA and production price of the various polyphenol production methods (Table 1) are used as criteria in the application of TOPSIS.

c. Development of Weighting

When applying TOPSIS, there is an inherent application of weighting, even in its default mode, equal weights are applied. This presents a problem because the selection of the ideal alternative is directly related to weighting. Ideally, this process would be completed relative to planetary boundaries [11] using an absolute relationship to impacts from LCA [12]. However, this absolute relationship is not yet well enough understood/developed, nor has it been developed to include all impact categories covered in LCA. As such, an alternative relationship must be established. This poses issues, which are further discussed in section 4.

In this case, normalization factors (NF) [13] are used to derive a relative importance factor (RIF), relating the average value, amongst all of the alternative extraction methods, of each of the midpoint impacts (MI) to the average European's annual environmental impact such that $RIF_i = \overline{MI_\iota}/NF_i$. The relationship between environmental and other criteria, in this case production cost, is then accounted for such that the sum of all weights is equal to 1000. The resultant weighting is then displayed in tabular form to promote full transparency in the assessment (Table 2, Table 3).

3. Results

After applying RIF, weighting strings can be derived for the application of TOPSIS with a range of importance given to economic impact from 0-1000, of 1000 available points distributed in the weighting profile (Table 2). This is also done for equal weights (EW) amongst environmental impacts and the same range of importance of economic impact (Table 3).

Table 2: Weighting strings including RIF for environmental impacts and a range of importance of economics

product production cost	Fine particulate matter formation	Fossil resource scarcity	Freshwater ecotoxicity	Freshwater eutrophication	Global warming	Human carcinogenic toxicity	Human non-carcinogenic toxicity	Ionizing radiation	Land use	Marine ecotoxicity	Marine eutrophication	Mineral resource scarcity	Ozone formation, Human health	Ozone formation, Terrestrial ecosystems	Stratospheric ozone depletion	Terrestrial acidification	Terrestrial ecotoxicity	Water consumption
0	12.83	276.36	183.72	86.75	58.98	59.26	3.93	28.40	0.61	161.97	0.86	0.004	23.98	28.77	2.05	21.34	42.57	7.62
100	11.55	248.72	165.35	78.08	53.08	53.33	3.53	25.56	0.55	145.78	0.77	0.003	21.58	25.90	1.84	19.21	38.31	6.86
200	10.26	221.09	146.97	69.40	47.18	47.41	3.14	22.72	0.49	129.58	0.69	0.003	19.18	23.02	1.64	17.08	34.06	6.10
300	8.98	193.45	128.60	60.73	41.28	41.48	2.75	19.88	0.42	113.38	0.60	0.002	16.78	20.14	1.43	14.94	29.80	5.34
400	7.70	165.82	110.23	52.05	35.39	35.55	2.36	17.04	0.36	97.18	0.51	0.002	14.39	17.26	1.23	12.81	25.54	4.57
500	6.42	138.18	91.86	43.38	29.49	29.63	1.96	14.20	0.30	80.99	0.43	0.002	11.99	14.39	1.02	10.67	21.28	3.81
600	5.13	110.54	73.49	34.70	23.59	23.70	1.57	11.36	0.24	64.79	0.34	0.001	9.59	11.51	0.82	8.54	17.03	3.05
700	3.85	82.91	55.12	26.03	17.69	17.78	1.18	8.52	0.18	48.59	0.26	0.001	7.19	8.63	0.61	6.40	12.77	2.29
800	2.57	55.27	36.74	17.35	11.80	11.85	0.79	5.68	0.12	32.39	0.17	0.001	4.80	5.75	0.41	4.27	8.51	1.52
900	1.28	27.64	18.37	8.68	5.90	5.93	0.39	2.84	0.06	16.20	0.09	0.000	2.40	2.88	0.20	2.13	4.26	0.76
1000	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0

Table 3: Weighting strings including equal weighting for environmental impacts and a range of importance of economics

product production cost	Fine particulate matter formation	Fossil resource scarcity	Freshwater ecotoxicity	Freshwater eutrophication	Global warming	Human carcinogenic toxicity	Human non-carcinogenic toxicity	Ionizing radiation	Land use	Marine ecotoxicity	Marine eutrophication	Mineral resource scarcity	Ozone formation, Human health	Ozone formation, Terrestrial ecosystems	Stratospheric ozone depletion	Terrestrial acidification	Terrestrial ecotoxicity	Water consumption
0	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6	55.6
100	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0	50.0
200	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4	44.4
300	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9	38.9
400	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3	33.3
500	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8	27.8
600	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2	22.2
700	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7	16.7
800	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1	11.1
900	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6	5.6
1000	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0

Applying these weightings to the criteria derived from LCA and techno-economic assessment using TOPSIS, it is possible to provide decision support in the form of a single score indicator of idealness of the various technological alternatives (Figure 1).

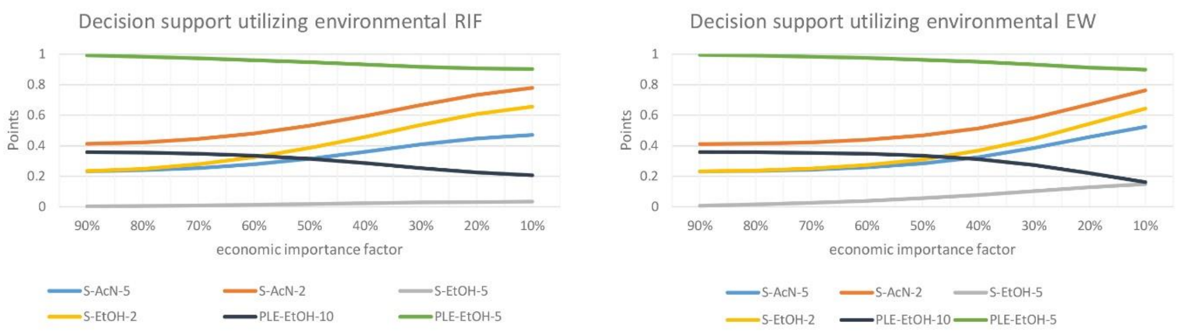

Figure 1: TOPSIS derived single score indicator of idealness (most ideal=1) for both RIF derived environmental weighting and EW environmental weighting amongst a range of EIF

4. Discussion

a. Interpretation of results

Based on the application of TOPSIS, it can be easily concluded that the PLE-EtOH-5 method outperforms all other alternative extraction methods. It is both the best economic performer and the best environmental performer in nearly all impact categories. This results in it being classified as the most ideal solution regardless of weighting. In addition, the S-AcN-2 remains the second ranked method regardless of weighting method. This indicates that these two alternatives exhibit characteristics that consistently perform better than the other alternatives. However, once one moves past the top ranked technologies, and must determine a third ranked technology, the picture becomes far less clear. The PLE-EtOH-10, and S-EtOH-2 alternatives vie for the third rank. S-EtOH-2 outperforms PLE-EtOH-10 environmentally, while PLE-EtOH-10 outperforms S-EtOH-2 economically. This results in a rank reversal as one changes the weight given to the economic criterion.

As can be seen in Table 4, there is significant range in the importance of specific environmental impacts in RIF for the assessed methods. For example, some impacts such as human non-carcinogenic toxicity, marine eutrophication, and land use are insignificant in relative importance, and mineral resource scarcity is almost entirely irrelevant. On the other hand, fossil resource scarcity and freshwater ecotoxicity make up nearly half of weighting applied to environmental impacts due to the scale of their impact compared to the other environmental criteria relative to the average European's environmental impact.

One other element of note is the difference of decision support between 40% and 70% economic importance factor (EIF) for the EW and RIF weighting. When using RIF, at 60% EIF, S-EtOH-2 and PLE-EtOH-10 are ambiguous in terms of ranking between third and fourth. Around 50% EIF, S-EtOH-2 is unambiguously ranked third when using RIF, however; when using EW, PLE-EtOH-10, S-AcN-5, and S-EtOH-2 are all ambiguous in terms of preference. This rank reversal is due to the difference in weighting for certain environmental impact categories where PLE-EtOH-10 performs similarly to S-AcN-5 and S-EtOH-2. However, despite performing similarly in some environmental categories, when the relationship to environmental importance (Table 4) of the magnitude of emissions is accounted for, the similar environmental performance of PLE-EtOH-10 is discounted in some impact categories, as it is irrelevant in relation to the scale of other environmental impacts. And, S-AcN-5 and S-EtOH-2 outperform PLE-EtOH-10 in fossil resource scarcity and marine ecotoxicity which become exaggerated in terms of influence in the decision support using RIF, relative to the decision support when using EW, due to the relative scale of the impacts in absolute terms. Furthermore, the effective removal of impacts without great relative significance by using RIF allows for greater differentiation between S-AcN-5 and S-EtOH-2, as impact categories where they perform relatively similarly, but are not of great consequence, such as mineral resource scarcity or human non-carcinogenic toxicity, are essentially removed from effecting the decision support.

Table 4: Relative weight of environmental impacts between RIF and EW weighting ($RW = W_{RIF}/W_{EW}$)

Fine particulate matter formation	Fossil resource scarcity	Freshwater ecotoxicity	Freshwater eutrophication	Global warming	Human carcinogenic toxicity	Human non-carcinogenic toxicity	Ionizing radiation	Land use	Marine ecotoxicity	Marine eutrophication	Mineral resource scarcity	Ozone formation, Human health	Ozone formation, Terrestrial ecosystems	Stratospheric ozone depletion	Terrestrial acidification	Terrestrial ecotoxicity	Water consumption
0.2309	4.9745	3.3069	1.5616	1.0616	1.0666	0.0707	0.5112	0.0109	2.9155	0.0154	0.0001	0.4316	0.5179	0.0368	0.3842	0.7663	0.1372

b. Alternative weighting methods

Another important element in interpreting the results from RIF weighting is understanding that there is a level of uncertainty in the normalization factors used to derive the RIF, and that the decision to use current emissions as a reference point does not necessarily have a relationship to the severity or consequences of environmental impacts. However, it does provide an indication of the relative importance of an emission, or reduction thereof, to the status quo. If absolute sustainability related factors were available for all relevant impact categories, the application of these instead of normalization factors would be preferable, as they would provide a stronger link to environmental impact.

An alternative to either of these methods would be to derive a RIF weighting from endpoints using e.g. monetization. While this might seem appealing, as there is a stronger connection with environmental damages when using endpoint indicators in LCA, the challenge comes in determining the relative importance of the different damage categories. This relative importance is purely subjective, and as such a specific cultural perspective would be applied to the derivation of the weighting profile. While this could be carried out in a scientific fashion to be representative of a decision maker group, the results would already contain some bias toward certain impacts introduced in the endpoint calculation [4, 6]. This would make the results more challenging to interpret and potentially lead to decision support that in the end does not reflect the true preferences of the decision maker.

c. Alternative MCDA methods

As discussed in the introduction, there are a number of potential alternatives to the use of MCDA. There are also a number of alternative methods of MCDA (other than TOPSIS) that could have been applied. Methods such as those that include preference comparison based on pairwise comparisons such as analytical hierarchy process (AHP) or outranking approaches such as elimination and choice translating reality (ELECTRE) or preference ranking organization method for enrichment evaluation (PROMETHEE). All of these methods include benefits and drawbacks, however, due to the simplicity of application as well as the easy comprehensibility of TOPSIS, it was chosen for this application. In particular, even when faced with a non-expert audience it is easy to describe how TOPSIS functions, including its relationship to weightings used in its application. This was considered a significant benefit, as it greatly increases the transparency of the application of MCDA and reduces the potential for misgivings when relaying results to non-experts.

5. Conclusions

Based on the results of both economic and environmental assessment, it can be concluded that among the tested extraction methods in the NoAW project, it is likely that the PLE-EtOH-5 alternative will perform best. However, should NoAW be unable to proceed with this technology for upscaling, then S-AcN-2 and S-EtOH-2 and PLE-EtOH-10 are all potential alternatives, depending on the importance given to economic performance versus environmental performance. In addition to the demonstrated ability of MCDA to increase the transparency and reproducibility of a decision making process, it can be concluded that the introduction of RIF as a method of deriving a weighting, relative to equal weights, for use in MCDA for LCA can likely reduce the impact of irrelevant and/or subjective criteria on the conclusions drawn from the application of MCDA that include weighting such as TOPSIS.

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