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Ögmundarson, Ólafur

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DTU Management
The Novo Nordisk Foundation Center for Biosustainability

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PhD Thesis
December 2018

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DTU Management Engineering
The Novo Nordisk Foundation Center for Biosustainability
Technical University of Denmark

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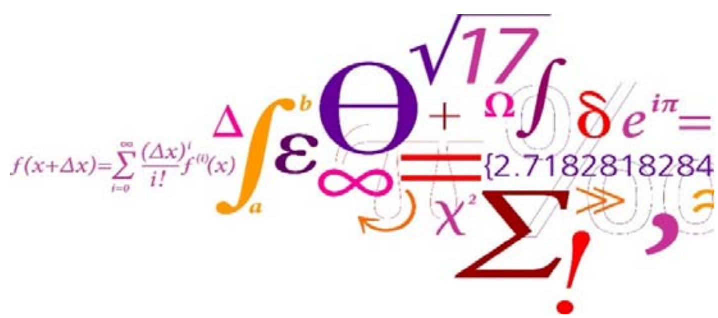

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Date:	31.12.2018
Author:	Ólafur Ögmundarson
Supervisors:	Peter Fantke Markus Herrgard
University:	Technical University of Denmark
Department:	Management, Technology and Economics
Division:	Quantitative Sustainability Assessment www.man.dtu.dk
Department: Address:	Novo Nordisk Foundation Centre for Biosustainability Kemitorvet Building 220 DK-2800 Kgs. Lyngby www.biosustain.dtu.dk
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"Without tech we will never reach the UN Sustainable Development Goals, but today the tech industry isn't sustainability-driven"

Professor Michael Zwicky Hauschild
DTU High Tech Summit 2018

PREFACE

This dissertation, "Life Cycle Assessment of chosen Bio-chemicals and Bio-polymers", is the result of a PhD research project, conducted at the Department of Management Engineering, Division for Quantitative Sustainability Assessments, and the Novo Nordisk Center for Biosustainability at the Technical University of Denmark (DTU Biosustain) from September 2015 to December 2018. The main supervisor was Associate Professor Peter Fantke, and co-supervisor was Professor Markus Herrgard. The project was financed by the EU FP7 project Biorefine 2G (Project No. FP7-613771) and co-funded by the Technical University of Denmark. Three journal manuscripts as first author were prepared by the PhD candidate during the project period and one conference manuscript. In the dissertation, the manuscripts are referred to as listed below, by their roman numerals:

I) Ögmundarson, Ó., Herrgard, M., Förster, J., Hauschild, M.Z., Fantke, Peter. (2019)
Assessing environmental sustainability of bio-based chemicals: State and challenges. Nature Sustainability, *revised version under review*.

II) Ögmundarson, Ó., Sukumara, S., Laurent, A., Fantke, P.
Environmental hotspots of different lactic acid production systems. Manuscript submitted to GCB Bioenergy.

III) Ögmundarson, Ó., Sukumara, S., Herrgard, M., Fantke, P.
Combining economic feasibility and environmental sustainability to optimize performance at early stages. Manuscript under preparation to be submitted to Trends in Biotechnology.

IV) Ögmundarson, Ó., Sukumara, S., Fantke, P. (2018)
Toward a sustainable biochemical industry - Early stage assessments and methodological overlaps between life cycle- and techno-economic assessments of biochemicals.

SETAC Europe 28th Annual Meeting, 2018, Rome. Proceedings of the SETAC Europe 28th Annual Meeting, 2018

In addition, the following manuscripts are under preparation. The first authors laid the foundation of these upcoming publications during their MSc thesis project work, where I had, during my PhD period, the pleasure to co-supervise them:

1) Jonas, T., Stewart, R., Ögmundarson, Ó., Bey, N., Niero, M. *Investigation of the environmental sustainability perception and knowledge of technological characteristics of bio-based plastics*. Manuscript under preparation to be submitted to Journal of Cleaner Production.

2) Tomás-Grasa, E., Ögmundarson, Ó., Gavala, H.N., Sukumara, S. *Commodity chemical production from 3rd generation biomass: A techno-economic assessment on lactic acid production.* Manuscript under preparation to be submitted to Biofuels, Bioproducts & Biorefining.

3) Gonzales, R.G., Owsianiak, M., Ögmundarson, Ó., Fantke, P. *Analyzing toxicity for biochemical-producing organisms*. Manuscript under preparation to be submitted to Environmental Toxicology and Chemistry.

Acknowledgements

It is not an easy task to think back at the three years as a PhD student, and remember all who have supported me and given me inspiration, and if I forget someone, I hope you will forgive me at some point.

Before my journey as a PhD student started, I did a master Environment and Natural Resource Management at the University of Iceland, under the guidance of Professor Brynhildur Davíðsdóttir. I have said this before and now on print, that she is one of the main reasons I sought to do a PhD, and she has been an inspiration to me since I met her the first time, also after she graded the first assignment I submitted to her, which said with large, red letters, “Gather your thoughts”. I hope I have learned that by now.

The journey that this PhD dissertation sums up started when Professor Jochen Förster and Associate professor Peter Fantke hired me as a PhD student in the summer of 2015. After Jochen left DTU Biosustain, Professor Markus Herrgard took over his role. I would sincerely like to thank them all for their endless support, fruitful discussions and comments on my work over the last three years. I would especially like to thank Peter who has been my closest collaborator during the PhD, who with his drive has been of great inspiration to me. Without his support, pushing me when needed, I wouldn´t be writing these words.

I would like to thank the members of the Biorefine 2G consortium and especially Amador Garcia Sancho and Bruno Sommer Ferreira for their collaboration and interesting discussion.

During my PhD period, I have been affiliated both to DTU Management, division of Quantitative Sustainability Assessments (QSA) and DTU Biosustain. It has been a pleasure to be part of both communities, and at DTU Biosustain I would especially like to thank Birte and Susanne for all their help when needed. Mads, thank you too boss. Now to QSA and all the sustainability people that I have learned to know there, that have been my sparring partners, Friday-bar companions, and often those giving me the reason to show up at work by showing endless support, not least over the last months of the PhD. First of all Nienke, thank you for enduring me as an office-mate for more than two years. So many thanks to Benjamin and Alexi for leading the way with a good example when I started at QSA, Philippe for always being the surfer-boy you are, Giovanna for all the runs and coffees and the fruitful discussions on our bio-nerdism. Alexandra, Raphaëlle, Monia, Morten, Serena, Marios, Florence, Niki, Mikolaj, Stig, Yan for all our nice conversations. Alexis, you have been of great inspiration to me, keep up the good work, and last but not least, Christine, Malene and Michael, thank you for your support and that you bought the mocha espresso for the division, it was a life safer.

Former AME group at DTU Biosustain, you will always be my friends.

Sumesh, I could not have done this work without your help and contribution, support and all the good conversations we have had over the last years about LCA and TEA and how we can combine these two methodologies.

Vaka, my beloved wife, thank you for keeping me sane and be there for me when ever needed (especially over the last stressful months). I could never have done this without you, thank you so much, I love you! My kids also own a lot in this work. You have kept me with my two feet on the ground by being there and made me think about other things than work, and "forced" me to have a life, including discussions about NBA and Pokémon, which have been such a relief from sustainability challenges in biotech. You have also been my drive during the PhD, because whenever I have not seen the light at the end of the tunnel, I have thought of you and you have brought me back on track, my beloved Ingimar, Ögmundur, and Una. Helga systir, you have always believed in me, given me support, and shown me another angle of my work that I otherwise would not have thought about. Thank you. And thanks to all the other family members, my in-laws, who have supported me and Vaka and the kids. Last but not least, I would like share a few thoughts about my parents. Shortly before I started my masters studies, my father passed away. When thinking back, I am so grateful that I could share with him that my educational journey had not come to an end, as he was always encouraging me, and others, to learn, learn more and never to settle if one had the urge to learn, and always finish what one started. Before he died, only 62 years old, he always said that he wanted do his PhD after he would go on retirement, and that lesson has taught me not to wait to do the things you want. My mother, a source of endless support, who also passed away, seven years ago, I could never have become who I am without her, takk mamma og pabbi.

To all my other friends not mentioned, and family, thank you too for all your support.

SUMMARY

Modern societies are dependent on fossil-based resources, which are limited, and the processing of those resources has proven to be the main cause of climate change, leading to a global warming of the earth. We feel and see the devastating effects of this on our lives and this will keep on happening into the foreseen future if we do not try to change the way we live and make things. We therefore face the question "what else can we humans use to produce the things we rely on in our everyday life, which today we mostly make from petrochemicals, converted from fossil resources?" Scientists, companies, and people in general, are coming up with new ideas on how to produce the same things from alternative resources, and one of them is that some of the solutions can be found in the bioeconomy. The bioeconomy offers that instead of relying on fossil resources in the production of for example chemicals, they can be produced with microbial fermentation, using renewable resources, i.e. biomass, as feedstocks for the microbes.

Biochemical production is first and foremost market driven, requesting that the producers can make profit out of their business, so there is a lot of interest in the new and fast-growing market for biochemicals. The general notion about biochemicals is that they are by definition environmentally sustainable, because they are bio-based. Scientific literature mirrors this when stating the environmental sustainability of biochemicals, without presenting conclusive results from environmental assessments supporting these statements. Because of the problems, we are facing regarding the earth warming up, now it is time to change this, because the intention is to produce chemicals from renewable, bio-based resources, and actually make them environmentally sustainable. We need to start incorporating environmental sustainability in the development and optimization of biochemicals at a very early stage, by integrating Life Cycle Assessment (LCA) as both a decision support tool when deciding which future biochemicals to produce, and how to produce them. This should be done in parallel when assessing their technical and economic feasibility, given that is already the general practice.

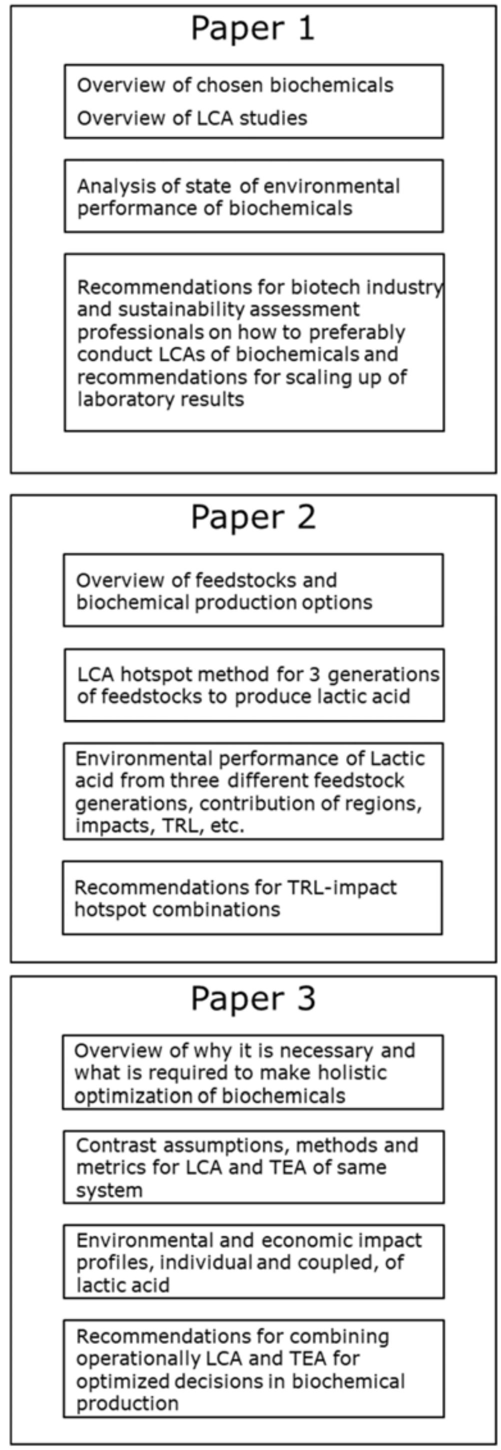
Integrating life cycle assessment in the development of biochemicals will also give us the opportunity, at an early stage of development, to identify environmental hotspots across the whole life cycle of the developed products. Then, we can catch tradeoffs and burden shifting between the assessed environmental impacts, and between the assessed life cycle stages, and instead avoid them in the early stages of development before making large investments to scale up the production, causing unnecessary environmental impacts. Only by doing so, biochemicals become a viable environmentally sustainable alternative. The first steps toward integrating LCA in optimization and decision support

for bio-based chemical production are put forward within a decision support framework, presented in this PhD dissertation.

To address the need of considering environmental sustainability in decision making by integrating LCA as a decision support tool, the PhD project "Life Cycle Assessment of chosen Biochemicals and Bio-polymers" focuses on answering the overarching question "How can the environmental sustainability of biochemicals, including bio-polymers, be consistently and comprehensively quantified?". The conclusion is that in order to optimize biochemicals for environmental sustainability, we should start assessing their performance by applying LCA at an early stage, in combination with the assessment of their technical and economic feasibility, by integrating currently applied Techno-economic assessments (TEA) and LCA together. That will lead to an overarching optimization between environmental and economic impacts.

This dissertation builds on three papers (schematic overview given here to the right). A short description of the papers is as follows. Paper I gives an overview of the state and challenges of five chosen biochemicals, following recommendations of how preferably to conduct LCAs on biochemicals. Paper II assesses the environmental performance of lactic acid production from three different biofeedstock generations and demonstrates how to use the LCA results for optimization of environmental performance and the effects of uncertainty on interpretation of results. Paper III shows how to integrate LCA as a decision support tool, combined with the application of TEA within a combined decision support framework.

Overview of the first author papers prepared during this PhD project period



The structure of the dissertation is as follows. It comprises of six chapters, and each chapter is subdivided into a different number of sections and sub-sections. The first chapter, the introduction, states the background of the PhD dissertation. Then the different feedstock generations and the biorefinery concept are defined, followed by setting the focus of the PhD project.

Chapter two states the overall objectives of the PhD dissertation, followed by three subsections, each giving a short introduction to the objectives of the three papers and research questions that shape the frame of the PhD project.

Chapters three, four, and five are the results of the dissertation, reflecting how LCA is applied in the literature, demonstrate how we should apply LCA as a decision support tool, and then how the integrated decision support framework of LCA is combined the Techno-economic assessment.

In chapter three, the overview of the current status of LCA studies is presented, based on a review of fourteen different biochemicals that have in common that they are all produced through bacterial fermentation. Of the fourteen biochemicals, special focus is set on five fully commercialized biochemicals, namely Lactic acid, Succinic acid, 1,3-Propanediol, 1,4-Butanediol, and 1,5-Pentanediamine. This is done to get an overview of current practices when assessing the environmental sustainability of biochemicals, their state and current challenges. The results of the chapter conclude that we need to assess the environmental sustainability of biochemicals across all life cycle stages, including end-of-life treatment, and assess all impacts. This has to be done in order to address all relevant burden shifting consistently across life cycle stages and to avoid tradeoffs between different environmental impacts. The chapter then provides guidelines of how to improve LCA practices when assessing biochemicals and how to make the biochemicals industry more environmentally sustainable.

In chapter four, I demonstrate how to apply the LCA methodology to identify environmental hotspots when producing lactic acid from three different bio-feedstock generations. There is a special focus on the third feedstock generation, the least developed with the lowest technological readiness level, to demonstrate how to use LCA results to point out process optimization potential, and state how, including the uncertainty of the data, it can affect the interpretation of the results. The inventories for the biorefinery life cycle stage in the LCA are achieved by doing a techno-economic assessment. Firstly, this makes it possible for LCA practitioners to acquire the needed inventories, which often are not available in the literature, and to make the study results more robust. Secondly, as becomes clear in the case of my study, applying TEA also helps scaling up the different feedstock generation processes to the same production volume and makes an assessment like this possible, when access to industrial data is limited.

Chapter five presents an integrated framework how to consistently couple environmental and economic indicators to allow for an overall optimization at early development stages of biochemical production within a decision support framework. The framework is stepwise introduced starting with a discussion on why we need to start looking beyond economic assessments, followed by a short introduction of current application of LCA and TEA, both separately and combined. The last two sections of chapter five describe the structure of the developed integrated framework and the results of a combined LCA and TEA representing the results demonstrating how to couple environmental and economic indicators in an economic single score.

Chapter six concludes the work and addresses future research needs beyond the scope of the present PhD dissertation. The impact of the PhD project is to help understanding how to make biochemicals and derived bio polymers more environmentally sustainable by identifying their most relevant optimization potential (LCA across generations) and integrate environmental aspects into decision processes (integrate LCA and TEA).

In the different sections and sub-sections of this dissertation, some figures and tables are identical to the ones presented in the publications laying the foundations of this dissertation, and they are cited by marking them with the roman numbers of each publication presented in this dissertation summary. If text is reused from the listed publications in the PhD dissertation summary, the same procedure is followed as mentioned before, and it is also clearly stated in the beginning of the chapter. This is done to be transparent about the originality of a text, as is practiced when citing other people's work.[1]

[1] See Guidelines for avoiding plagiarism and self-plagiarism in PhD dissertation writing. DTU PhD Office, November 2018.

DANSK SAMMENFATNING

Moderne samfund er afhængige af fossile ressourcer, som er begrænsede, og behandlingen af disse ressourcer har vist sig at være hovedårsagen til klimaændringer, der fører til global opvarmning af jorden. Vi føler og ser de ødelæggende virkninger af dette på vores liv, og det vil fortsætte ind i fremtiden, hvis vi ikke ændrer den måde, vi lever og producerer tingene på. Vi står derfor over for spørgsmålet: "Hvad kan vi ellers bruge til at producere de ting, vi stoler på i vores hverdagsliv, som vi i dag hovedsagelig fremstiller af petrokemikalier, omdannet fra fossile ressourcer?" Forskere, virksomheder og folk generelt kommer med nye ideer om, hvordan man producerer de samme ting fra alternative ressourcer, og en af dem er, at nogle af løsningerne kan findes i bioøkonomien. Bioøkonomien giver, at de i stedet for at stole på fossile ressourcer i produktionen af for eksempel kemikalier, kan fremstilles ved mikrobiel fermentering ved hjælp af vedvarende resurser, dvs. biomasse som føde for mikroberne.

Den biokemiske produktion er først og fremmest markedsdrevet og anmoder om, at producenterne kan tjene penge ud af deres forretning, så der er stor interesse for det nye og hurtigt voksende marked for biokemiske produkter. Den generelle forestilling om biokemikalier er, at de per definition er miljømæssigt bæredygtige, fordi de er biobaserede. Videnskabelig litteratur afspejler dette, når biokemikaliernes miljømæssige bæredygtighed angives uden at fremlægge afgørende resultater fra miljøvurderinger, der støtter disse udsagn.

Vi står over for et dilemma, men hvis hensigten er at producere kemikalier fra vedvarende biobaserede ressourcer og rent faktisk gøre dem miljømæssigt bæredygtige er det på tide at vi gør noget aktiv til faktisk at gøre dem mere bæredygtige. Vi skal begynde at indarbejde miljømæssig bæredygtighed i udviklingen og optimeringen af biokemiske stoffer på et meget tidligt tidspunkt i deres udvikling, ved at integrere livscyklusvurdering (LCA) som både et beslutningsstøtteværktøj, når vi beslutter, hvilke fremtidige biokemiske stoffer der skal produceres, og hvordan de produceres. Dette skal ske parallelt, når de vurderer deres tekniske og økonomiske gennemførlighed, da det allerede er den generelle praksis.

Integration af livscyklusvurdering i udviklingen af biokemikalier vil også give os mulighed for i et tidligt udviklingsstadium at identificere miljømæssige hot spots over hele produktets livscyklus. Derefter kan vi fange afveje og skiftende byrde mellem de vurderede miljøpåvirkninger og mellem de vurderede livscyklustrin og i stedet undgå dem i de tidlige udviklingsstadier, før store investeringer investeres i at opskære produktionen og forårsage unødige miljøpåvirkninger. Kun ved at gøre det bliver biokemiske stoffer et miljømæssigt bæredygtigt alternativ. De første skridt mod integration af

LCA i optimering og beslutningsstøtte til biobaseret kemisk produktion fremlægges inden for rammerne af et beslutningsstøtteramme, præsenteret i denne Ph.D.-afhandling.

For at imødekomme behovet for at overveje miljømæssig bæredygtighed i beslutningsprocessen ved at integrere LCA som et beslutningsstøtteværktøj, fokuserer Ph.D.-projektet "Livscyklusvurdering af udvalgte biokemikalier og biopolymerer" på at svare på det overordnede spørgsmål "Hvordan kan miljømæssig bæredygtighed af biokemikalier, herunder bio-polymerer, konsekvent og omfattende kvantificeres?" Konklusionen er, at for at optimere biokemikalier for miljømæssig bæredygtighed skal vi begynde at vurdere deres resultater ved at anvende LCA på et tidligt stadium i kombination med vurderingen af deres tekniske og økonomiske gennemførlighed ved at integrere aktuelt anvendte teknoøkonomiske vurderinger (TEA) og LCA koblet. Det vil føre til en overordnet optimering mellem miljømæssige og økonomiske konsekvenser.

Denne afhandling bygger på tre papirer (skematisk oversigt angivet her til højre). En kort beskrivelse af papirerne er som følger. Publikation I giver et oversigt over tilstanden og udfordringerne ved fem udvalgte biokemikalier efter anbefalinger af, hvordan man foretrækker LCA'er på biokemikalier. Publikation II vurderer den miljømæssige ydeevne af mælkesyreproduktion fra tre forskellige bio-råmaterialer generationer og demonstrerer, hvordan man bruger LCA-resultaterne til optimering af miljøpræstationer og virkningerne af usikkerhed om fortolkning af resultater. Publikation III viser, hvordan man integrerer LCA som et beslutningsunderstøttelsesværktøj kombineret med anvendelsen af TEA inden for en kombineret beslutningsstøtteramme.

Afhandlingen er som følgende. Den består af seks kapitler, og hvert kapitel er opdelt i sektioner og underafsnit. Den første kapitel, introduktionen, angiver baggrunden for Ph.D.-afhandlingen. Derefter defineres de forskellige råmaterialegenerationer og bioraffinaderi begrebet, efterfulgt af at sætte fokus på Ph.D.-projektet.

Kapitel 2 angiver de overordnede mål for Ph.D.-afhandlingen, efterfulgt af tre underafsnit, der hver især giver en kort introduktion til målsætningerne for de tre papresse- og forskningsspørgsmål, der danner rammerne for Ph.D.-projektet.

Kapitel 3, 4 og 5 er resultatet af afhandlingen, afspejler hvordan LCA anvendes i litteraturen, viser, hvordan vi skal anvende LCA som et beslutningsstøtteværktøj, og hvordan den integrerede beslutningstilsynsramme for LCA kombineres Teknisk-økonomisk vurdering.

I kapitel 3 er oversigten over LCA-studiens nuværende status præsenteret, baseret på en gennemgang af fjorten forskellige biokemikalier, der har til fælles, at de alle produceres ved bakteriel gæring. Af de fjorten biokemikalier er der fokuseret på fem fuldt kommercialiserede biokemiske stoffer, nemlig mælkesyre, ravsyre, 1,3-propandiol, 1,4-butandiol og 1,5-pentandiamin. Dette er gjort for at få et overblik over nuværende praksis ved vurdering af biokemikaliernes miljøforsinkelighed, deres aktuelle og aktuelle udfordringer. Resultaterne af kapitlet konkluderer, at vi skal vurdere biokemikaliernes miljømæssige bæredygtighed på tværs af alle livscyklusfaser, herunder udtjent behandling og vurdere alle virkninger. Dette skal gøres for at imødegå alle relevante byrdeforskydninger konsekvent over hele livscyklusfasen og for at undgå afvejninger mellem forskellige miljøpåvirkninger. Kapitlet giver derefter retningslinjer for, hvordan man forbedrer LCA-praksis ved vurdering af biokemikalier og hvordan man gør biokemiske industrier mere miljømæssigt bæredygtige.

I kapitel 4 demonstrerer jeg, hvordan man anvender LCA-metoden til at identificere miljømæssige hot spots, når man producerer mælkesyre fra tre forskellige biobrændstofgenerationer. Der er et særligt fokus på den tredje råmaterialegeneration, den mindst udviklede med det laveste teknologiske beredskabsniveau, for at demonstrere, hvordan man bruger LCA-resultater til at pege på procesoptimeringspotentiale og angive, hvorvidt dataenes usikkerhed påvirker fortolkningen af resultaterne. Data til livscyklusfasen for bioraffinaderiet i LCA opnås ved at lave en teknisk-økonomisk vurdering. For det første giver LCA-praktikere mulighed for at erhverve de nødvendige opgørelser, som ofte ikke er tilgængelige i litteraturen, og for at gøre studieresultaterne mere robuste. For det andet, som det fremgår af min undersøgelse, hjælper anvendelsen af TEA også med at opskalere de forskellige råmaterialeproduktionsprocesser til det samme produktionsvolumen og gør en vurdering som denne mulig, når adgangen til industrielle data er begrænset.

Kapitel fem præsenterer en integreret ramme for sammenhængende sammenkobling af miljømæssige og økonomiske indikatorer for at muliggøre en overordnet optimering i de tidlige udviklingsstadier af biokemisk produktion inden for rammerne af beslutningsstøtte. Rammerne introduceres trinvist, begyndende med en diskussion om hvorfor vi skal begynde at se ud over økonomiske vurderinger, efterfulgt af en kort introduktion af løbende anvendelse af LCA og TEA, både separat og kombineret. De sidste to afsnit i kapitel 5 beskriver strukturen i den udviklede integrerede ramme og resultaterne af en kombineret LCA og TEA, der repræsenterer resultaterne, der viser, hvordan man sammenkobler miljømæssige og økonomiske indikatorer i en økonomisk enkeltscore.

Kapitel seks afsluttes med konklusioner og overvejelser om fremtidige forskningsbehov ud over den nuværende Ph.D.-afhandling. Effekten af Ph.D.-projektet er at hjælpe med at forstå, hvordan man gør biokemiske stoffer og afledte biopolymerer mere miljømæssigt bæredygtige ved at identificere deres mest relevante optimeringspotentiale (LCA på tværs af generationer) og integrere miljøaspekter i beslutningsprocesser (integrere LCA og TEA).

I de forskellige afsnit og underafsnit af denne afhandling er nogle figurer og tabeller identiske med dem, der præsenteres i publikationerne, der ligger til grund for denne afhandling, og de er citeret ved at markere dem med de romerske numre af hver publikation, der præsenteres i denne afhandling. Hvis teksten genanvendes fra de angivne publikationer i Ph.D.-afhandlingsoversigten, følges samme procedure som tidligere nævnt, og det fremgår også tydeligt i begyndelsen af kapitlet. Dette er gjort for at være gennemsigtigt om originaliteten af den tekst, som det praktiseres, når man citerer andres arbejde.

ACRONYMS

Acronyms used in the thesis, first time used by full name, then by acronym

Acronyms	Full text
BM	Biomass
BR	Biorefinery
CN	China
CSR	Corporate Social Responsibility
DALY	Disability adjusted life years
DE	Germany
DoE	US Department of Energy
EN	European Standard
EoL	End-of-Life
FU	Functional unit
GSD	Geometric standard deviation
ICE	Iceland
ILCD	International Reference Life Cycle Data System
iLUC	Indirect Land Use Change
ISO	International Organization for Standardization
LA	Lactic acid
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
OECD	The Organisation for Economic Co-operation and Development
PLA	Polylactic acid
PM	Polymerization
TEA	Techno-Economic Assessment
TRL	Technological readiness level
US	United States of America
USD	US Dollars

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1. INTRODUCTION

1.1 BACKGROUND

We, humans, are increasing pressure on the environment with our way of living, resulting in increased global climate changes and degradation of natural resources. One of the main drivers for these catastrophic changes is the intensive use of fossil resources, building the fundament of our consumption. Simultaneously, mainly in the western world, we have reached a higher standard of living than ever before, partially thanks to the use of fossil resources. We therefore have a dilemma on our hands. As fossil resources are non-renewable, "peak-oil" is foreseen in this century[1], and the fact of devastating climate changes, connected to the use of fossil resources[2], leads to a global drive to substitute the non-renewable resources with renewable ones. For this thesis, the interest is in contributing to an improved understanding of the environmental sustainability implications of the transition from fossil-based chemicals to bio-based chemicals.

Bio-based chemicals are perceived as environmentally sustainable because they are made from renewable resources[3], reduce dependency on limited fossil resources, and help "decarbonize" societies[4]. Further, sustainable development within biotechnology makes the discipline one of the keys to overcome products' climate related issues. The shift toward bio-based chemicals and related materials still has some big challenges to overcome before we can claim that they are actually "sustainable". This especially relates to production of biochemicals, the building blocks of bio-based materials, today mostly produced from 1st generation biomass. 1st generation biomass is also known as agricultural crops but the different generations of feedstock dealt with in this thesis are defined further in section 1.2.

Converting biomass to products is not a new thing under the sun, but conversion of biomass to biochemicals has gotten an increased focus since the US Department of Energy (DoE) identified in 2004 a list of 12 building block chemicals which can either be produced through biological or chemical conversions.[5] This work was initialized because of growing concerns about climate change and sustainability issues of petrochemical resource use for chemical production. Since the DoE building block chemicals were identified, many of them are close to being commercialized or have already reached that status. Choi et al. reviewed in 2015 "the current status of biorefinery development for the production of these platform chemicals and their derivatives"[6] without addressing relevant environmental sustainability aspects, such as resources use or chemical emissions along production processes.

Sustainability of the biological chemical conversion of the 12 building block chemicals was studied to some extent more than a decade ago, and in 2007, Hatti-Kaul et al. expressed "the importance of evaluating the environmental impact of bio-based products". They state that "[t]he few studies reported so far have shown that the principle of using renewable feedstock is not necessarily favorable in all situations and for all environmental aspects".[7] For assessing environmental sustainability, life cycle assessment (LCA) is the most widely used methodology. By applying LCA we can assess the environmental impacts of processes and products from their cradle, including excavation for materials used for production, to the disposal stage, e.g. from recycling or incineration, or what is called from cradle-to-grave in LCA terminology.[8]

Since Hatti-Kaul et al. published their results, some environmental assessments of bio-chemicals have been published[9–11], indicating there are environmental benefits included in the shift from petro- to biochemical production. Nevertheless, one should ask how environmentally sustainable biochemicals and derived products really are? For example, the results of Jeswani et al.[12] suggest that the production of ethanol from chosen feedstocks "offers significant savings in eight out of 12 environmental impacts when the system is credited for the avoided impacts from producing the co-products from fossil resources".[12] Smidt et al. also show that "[b]io-based succinic acid outperforms fossil-based succinic acid in the impact categories Global Warming and Resource Depletion. Bio-based succinic acid shows a higher impact in other impact categories, such as Dust and Particulate Matter and Land Use".[10] A somewhat similar picture is drawn when we look at environmental assessments of lactic acid (LA) and derived polylactic acid (PLA) production. Results from Morales et al. and Daful et al. show LA performing environmentally better than functionally equivalent fossil based monomers[13,14], and e.g. Vink et al., Adom et al., and Patel et al. as well when polymerization of the monomers is included in the assessment[15–17].

Despite this overall positive picture of the environmental performance of biochemicals and derived products, there are still many challenges to overcome before we can claim that bio-based materials and other biotechnological products, making use of bio-based chemicals derived from renewable resources, can fully perform favorably when compared to petrochemical-based polymers and other products.[18] The first challenge that we need to overcome is the large variation in in- and excluded environmental impacts and life cycle stages across LCA studies when assessing biochemicals. Despite this being a well know problem within LCA, it is especially relevant to assess all relevant impact categories for biochemicals and derived products because of the possible trade-offs between impacts categories related to different biomass utilization. Also, by not assessing all life cycle stages from cradle-to-grave of bio-based products, LCA studies reveal that they do not give a fair picture of the assessed environmental sustainability.[19] Studies' complexity can be blamed[20], but the eminent danger of seeing past the burden shifting should be avoided, by always including all life cycle stages when assessing bio-based products and thereby support life cycle thinking.[4,19] As an example, not assessing the growing of biomass when assessing the environmental impacts of biochemicals, and only the actual production stage, would lead to a situation where we claim sustainability of our product without having included pesticide impacts in the environmental impacts of the product.

One could claim that environmental performance of bio-based products is not relevant, or that they are by default sustainable because they are derived from renewable resources, or only what matters is that their production is economically feasible, this might only be right when production volumes are meager. But as global demand and production capacity is on the rise we cannot keep on overlooking the inconsistency in the environmental performance of bio-based chemicals when they are projected to reach up to 22% market share by 2025[21] and derived bio-based polymers[22] and bio- based plastics marked is growing fast with ever-increasing demand[23].

The main goal of the present PhD project is hence to address the urgent question "[h]ow can the environmental sustainability of bio-based chemicals, applied in biotechnologies, including bio-polymers, be consistently and comprehensively quantified, optimized, and ultimately included in decisions related to biochemical production?" Specific aims are to identify current gaps and issues, to quantitatively assess the environmental sustainability of products containing bio-based chemicals, specifically using lactic acid and derived polylactic acid production as case studies, to develop a consistent methodology to fill current gaps and solve existing issues for analyzing sustainability of bio-based chemicals. This will result in an operational assessment tool, along with a set of recommendations of how we can consistently optimize the environmental performance of bio-based products, and when fully optimized, compare the bio-based products to conventional ones.

Before presenting a more detailed overview of the PhD objectives and research questions, the methodologies applied, results, discussion and conclusions, I will briefly set the scene and give an overview of the cornerstones of biochemical production. This means giving an overview of the renewable resources, i.e. feedstocks, which are converted from raw materials, in a biorefinery, to different categories of biochemicals.

1.2 Feedstocks, definition of generations

Renewable biomasses are the core of producing biochemicals and the most common group of feedstocks used today for that purpose are agricultural crops, also defined as 1st generation feedstocks (see Figure 1). For the definition of different feedstock generations I lean on to how they are defined by Yang et al.[24]

1st Generation

Feedstock:

- Agricultural crops (with/without pretreatment)
- World production 891.1 million metric tons/annually[1]
- Of which 42% in USA[1], accounting for 42%[2] of US N fertilizer use

TRL 8-9:

- Mature commercial markets
- Well-understood technologies

Pros:

- Easily fermentable sugar rich feedstocks

Cons:

- Competition with edible food
- Impact on food prices

2nd Generation

Feedstock:

- Lignocellulosic biomass (with pretreatment)
- USA production 108.9 million metric tons/annually[3]

TRL 4-5:

- Immature commercial markets
- Partly understood technologies for commodity chemical production

Pros:

- Non-food biomass
- Abundant availability for a low price

Cons:

- Feedstock conversion into easy fermentable sugars

3rd Generation

Feedstock:

- Macroalgae biomass (with pretreatment)
- Industrial use, minimum 8 million tons (numbers since 2003)[4]

TRL 1-2:

- Not commercialized
- Poorly understood technologies for commodity chemical production

Pros:

- Not grown on land
- Bioremediation of feedstock in nature

Cons:

- Biomass availability, and for a reasonable price

Figure 1 Overview of the different feedstock generations, their TRL (Technological readiness level) when it comes to commodity biochemical production. For definition of feedstock generations, see Yang et al.[24] 1)See reference[25], 2)See reference[26], 3)See reference[27], 4)See reference[28]. *(Figure is from Paper II).*

Generally, 1st generation biomass is relatively easy to ferment and little or no pretreatment is needed. As mentioned before, utilizing 1st generation biomass does, though, not come without complications, such as increasing food prices based on land competition with edible food.[29] Therefore, over the last years, there has been increased interest in non-food biomass, leading to the development of the 2nd generation biomass, for production of biochemicals. The challenges of utilizing 2nd generation biomass are that pretreatment is needed to break down the fiber rich biomass to smaller fractions, and to separate the lignin from the rest of the biomass resulting in high cost. Separating the lignin is necessary because of its toxic effects toward the bacteria fermenting the biomass resulting in high costs[30–32], and the relative low fermentable sugars content.[33]

The search for alternative feedstocks to 1st generation biomass is therefore ongoing, and macroalgae are getting more and more attention as 3rd generation biomass for biorefinery applications. The absence of lignin[32] in macroalgae makes it a feasible feedstock as well as their potential to reduce the environmental impacts of biochemicals in comparison to petrochemicals by avoiding any biomass growth related impacts. Other advantages of utilizing macroalgae for biochemical production are their potential to reduce effects from fish aquaculture in integrated multi-trophic aquaculture (IMTA) systems[34] providing bio-mitigation benefits.[35] Another identified benefit of macroalgae is its potential bioremediation efficiency, for example to control eutrophication[36] and for bioaccumulation of marine pollutants to clean up contaminated areas.[37,38]

In the present thesis, macroalgae are explored in more detail as potential feedstock for biochemicals production, with focus is on the *Laminaria* genus, a brown algae that grows among others close to shore around the Atlantic Ocean.[39,40]

However, utilizing macroalgae as feedstock for biochemical production also faces different challenges. The hurdles include seasonal and spatial variation of the carbon content and dry matter content of the different macroalgae species calling for new development of storage technologies,[41,42] and currently high production costs of possible large scale cultivation options of macroalgae[43]. Another challenge when assessing the potential of an emerging feedstock like macroalgae, especially if it is grown in waters with high concentrations of different chemical contaminants like persistent organic pollutants or toxic metals, is the potential of such contaminants to accumulate in the different macroalgae species. Such contaminant residues in macroalgae can induce possible toxicity-related effects on the microorganisms used to convert the macroalgae biomass to commodity chemicals[38,44], which can lead to decreased microorganism productivity and related increased fermentation costs. Reducing any potentially harmful chemical contaminant content in macroalgae harvested as feedstock for biochemicals production would likewise increase costs as function of the utilized contaminant removal technology. Such impacts on the production system itself, however, are not considered in the present study where a first focus is on the assessment of potential effects on the (natural) environment, but might be relevant when assessing the overall performance of biochemical production beyond environmental impacts. This is especially important for harvesting macroalgae as feedstock for bacterial fermentation, which might be cultivated with the primary purpose of reducing chemical toxicity loads in marine waters.[45]

1.3 THE BIOREFINERY, DEFINITION AND GENERAL CONCEPT

The vast majority of energy and materials we use every day are fossil based and are produced from materials converted from fossil fuels in a refinery. The different products from the fossil fuel refinery are produced by separating the feedstock (oil) into usable fractions, which are then converted into products. The biorefinery concept follows the same principles, but instead of fossil fuels, it utilizes renewable feedstocks for the conversion of feedstocks to products.[46] Utilization of bio resources is one of the cornerstones for the establishment of the future bio-based economy. The economy needs to cut our dependency on fossil resources.[47]

Biorefining is defined as a "synergetic processing of biomass into a spectrum of marketable food & feed ingredients, products (chemicals, materials) and energy (fuels, power, heat)".[48] The general concept of the biorefinery, per definition, is to make marketable products of different sorts from different bio feedstocks (see section 1.2). Today´s biorefineries mostly utilize 1st generation biomass, also known as agricultural crops, which are fermentable without any pretreatment, because of the easily extractable sugars they contain.[24] Linking biorefining and sustainability assessments is not new but is still an emerging field that could help for example research centers, like DTU Biosustain, to optimize the environmental sustainability potential of the biochemicals the develop.

1.4 FOCUS OF THIS PHD PROJECT

The variety of biochemicals produced in biorefineries is large, e.g. bulk/commodity chemicals, polymers, food/feed, and pharmaceuticals.[49] The focus in this thesis is on commodity chemicals, produced by bacterial fermentation, because this PhD project is conducted in the frame of the project Biorefine 2G, which had the aim of developing commodity chemicals from cell-factories. Another reason for this focus is that because this project being co-supervised from DTU Biosustain, it has the vision to develop a decision support framework helping to make more environmentally sustainable biochemicals and bio-pharmaceuticals laying the foundation of a future sustainable lifestyle. With the approach presented in my PhD, we can assess the environmental sustainability of new biochemical products at an early stage to make the new products minimizing their environmental impacts. Another reason for the focus on commodity chemicals is that today they are produced in large quantities (see Table 1) so minimizing their environmental impacts can have large environmental benefits.

More than 10 years ago, the US Department of Energy (DOE) proposed a list of 12 bio-based chemicals as potential substitutes for some of the current fossil-based chemical building blocks on the market, using a techno-economic analysis[5]. The intention was not to directly replace particular intermediates in the chemical industry, but rather use the proposed chemicals as new intermediates for functionally equivalent downstream products, such as packaging materials. Increased use of renewable resources and sustainability of bio-based industrial products were among DOE's major motivations behind establishing this list.[50] Two chemicals were added and five removed in an update of the original DOE list in 2010, mainly related to shifts in research and development in the biochemical industry.[51] The current level of commercialization of the chemicals on the updated DOE list ranges from laboratory scale to full commercial production (see Table 1).[6,52]

The general trend within the literature assessing environmental sustainability of the DOE chemicals and in addition 1,3-Propanediol and 1,4-Butanediol (included because of high interest by industry), is that a surprisingly low number of LCA studies is available. Those available show inconsistencies in terms of a narrow look at environmental impacts, and a limited coverage of the different stages of biochemical life cycles. Moreover, results of the few available LCA studies for bio-based products vary widely and give in part contradictory conclusions regarding environmental performance. Sustainability claims for bio-based chemicals are often exclusively based on reduced global warming impacts compared to fossil-based chemicals, whereas other impacts, such as land use from bio-feedstock production, are largely ignored.[18,19] To move towards truly sustainable biochemicals, a broader range of impacts and life cycle stages need to be covered, to identify tradeoffs and help avoid burden shifting from one impact category or life cycle stage to another.

As presented in the results section of this thesis, I will demonstrate how to use LCA to analyze the environmental sustainability of biochemicals in the early stages of development, assess the potential environmental impacts of future biochemicals, and how that assessment could go hand in hand with the economic assessment anyway conducted. In support of the development of biochemicals with improved environmental sustainability performance, I evaluate studies applying life-cycle assessment (LCA) to 14 listed bio-based chemicals produced by means of microbial fermentation, excluding biofuels and derived products. The reason for the scope is firstly the general focus of the Biorefine 2G project on development of new microbial processes for producing dicarboxylic acids and DTU Biosustain on microbial fermentation processes. Secondly, the reason for excluding biofuels and their bi-products (glycerol) is that there is extensive literature covering biofuel production and that glycerol is not relevant for microbial fermentation processes.[53,54]

2. Objectives and research questions of the PhD

The overarching goal of this PhD project is to address the question, "[h]ow can the environmental sustainability of bio-based chemicals applied in biotechnologies, including bio-polymers, be consistently and comprehensively quantified, optimized, and ultimately included in decisions related to biochemical production?" To achieve this goal, I defined three objectives of how to consequently assess the environmental sustainability of bio-based chemicals at an early stage and use the results for decision support, when assessing which biochemicals should be further developed. This included looking in depth into which level we need to bring the LCA to, for it to becoming a vital decision support tool, as well as how to incorporate LCA into already integrated decision support analysis like techno-economic assessments. To reach the objectives, the PhD project was divided into three different sections, each contributing to reaching the objectives.

The first specific objective is to analyze and systematically identify the environmental performance of selected biochemicals that have been identified as promising substitutes for petrochemicals, with special focus on commercialized biochemicals that have proven to be market fit.

The second specific objective is to demonstrate with an LCA how to systematically identify and increase sustainability of biochemicals, as an example by applying hot-spot analysis of lactic acid production from three different feedstock generations. The data for the biorefinery life cycle stage is generated by simulating the processes in a techno-economic assessment.

The third specific objective is to, based on the experience gathered in applying LCA and TEA simultaneously, and based on combining the results of the two assessments, design an operational framework to consistently integrate both environmental and economic performance in the design and optimization of new biochemicals. The framework should also be applied at an early stage as a decision support tool for targeting chemical selection.

2.1 Exploring state-of-art in assessing environmental performance of biochemicals

To understand the state of art and identify current environmental sustainability challenges of biochemicals and biotechnology in general, the first focus point of this PhD was to analyze the landscape of LCA studies applied to biochemicals and will be addressed in Chapter 3. To narrow the scope from all biochemical types (e.g. biofuels, commodity chemicals, fine chemicals, and food/feed) the focus was set on the top eleven chemicals proposed by the United States Department of Energy (DOE) in 2004 and 2010 because of their potential to substitute some of the current fossil-based chemical building blocks on the market. Because the DOE chemicals do not catch all relevant biochemicals today, current commercialized biochemicals were also included in the analysis bringing the total number of assessed biochemicals to fourteen. For this part, I applied the following research questions (for overview of analyzed chemicals see Table 1):

(1) Which are the main methodological choices to make when assessing environmental sustainability of bio-based chemicals?

(2) What are the main conclusions on environmental sustainability found in published LCA studies on commercialized bio-based chemicals?

(3) How can we improve the use of LCA for bio-based chemicals, to help striving towards a viable and sustainable future for the biochemical industry, also considering the role of public perception?

2.2 ENVIRONMENTAL PERFORMANCE ASSESSMENT OF A SPECIFIC BIOCHEMICAL PRODUCTION SYSTEM USING DIFFERENT FEEDSTOCKS

Existing LCA studies performed on biochemicals show some trends and limitations. When biochemicals and derived products are compared to functionally equivalent fossil-based products, LCA results show that in some impact categories biochemicals perform better and in other categories they perform worse than their fossil-based counterparts.[18,19] However, several studies only focus on assessing global warming impacts and do not include other relevant environmental impacts, thus potentially overlooking burden shifting from one environmental impact to another. There is a large variation in the coverage of life cycle stages across studies, where too may result in burden-shifting from one life cycle stage to another, if a full life cycle perspective is not adopted.[19] These trends and limitations suggest that there is a strong need for a more comprehensive overview of the differences between feedstocks, life cycle stages and impacts, to identify optimization potentials of biochemical production, in terms of environmental performances from conventional and emerging feedstocks.[19]

The scope of Chapter 4 is to identify environmental hotspots within production of LA from three different bio-based feedstocks.

To address this need, I aim in the present study to:

(1) Consistently define life cycles of biochemical product systems across bio-feedstock generations, focusing as an illustrative example on lactic acid as an important building block chemical.[55,56]

(2) Characterize the environmental performance of lactic acid production systems with a full life cycle assessment.

(3) Discuss related environmental hotspots and their potential drivers. That includes showing how hotspot results can be used to inform technology system design, identify optimization potential of future processes, and operationalize decision support.

2.3 OPERATIONAL FRAMEWORK FOR CONSISTENTLY COMBINING ENVIRONMENTAL AND ECONOMIC PERFORMANCE ASSESSMENTS APPLIED IN THE DEVELOPMENT OF NEW BIOCHEMICALS

The right thing to do is to transform our fossil-based economy to a bioeconomy to make us less dependent on fossil fuels. In our transition to the bioeconomy[57,58] industrial biotechnology plays a key role, but there are still hurdles to overcome. In relation to biochemicals, the obstacles are related to the fact that despite being produced from renewable resources, they are not environmentally benign.[18,19] But how can we make them more environmentally sustainable? In Chapter 5 of this thesis, an operational framework is presented, that combines life cycle assessments and techno-economic assessment. This is necessary because if we want to be able to state that our future biochemicals are both environmentally and economically sustainable, we need to combine the two methodologies consistently from an early stage, as decision support methodologies, to address future production challenges as soon as they can be identified.

3. FROM CURRENT PRACTICE TO RECOMMENDATIONS FOR LIFE CYCLE ASSESSMENT APPLIED TO BIOCHEMICALS

This chapter of the thesis will present the results of the PhD project with focus on what is needed to, and how we can assess the environmental sustainability of biochemicals in a more consistent way.

The chapter is divided into three sub-sections. Each section starts with a short introduction, followed by an overview of the applied methodology to create the results (might not always be needed), followed by the results and a short discussion. If *From Paper XX* is stated in the beginning of the section, which means it has been taken directly from the relevant paper.

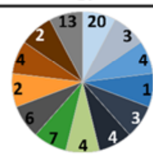
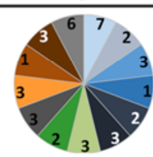
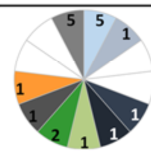
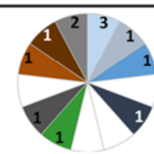
3.1 CURRENT STATE OF ASSESSING ENVIRONMENTAL SUSTAINABILITY OF SELECTED BIOCHEMICALS

(This section is based on the result presented in *Paper I*)

The environmental sustainability of bio-based technologies and products is largely unassessed and, hence, unclear in relation to conventional technologies and products. The work started by identifying limitations in present environmental sustainability assessment methods with respect to bio-based chemicals and related biotechnologies and products. Special focus was set on the scope beyond bio-based chemicals to identify hotspots and focus areas for improvement in bio-chemical production, with focus on products and processes developed including microbial fermentation

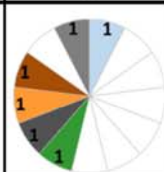
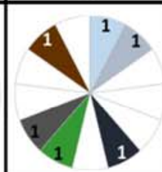
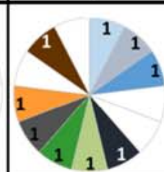
To find the literature I systematically searched Scopus and Google Scholar for biochemical name synonyms as listed in PubChem[59] along with "sustainability" or "LCA" and "life-cycle assessment" or "Foot Print" and "Footprint". In total I found 43 environmental sustainability assessment studies published between 2003 and 2018 that matched these search criteria (last search conducted on 30.06.2018). Table 1a and b summarize the characteristics of the chosen DOE-listed chemicals, e.g. world production volumes, identified main applications, the number of scientific studies found assessing their environmental sustainability by conducting LCA, and state of commercialization.

Table 1a Main characteristics of bio-based chemicals identified by the US DOE in 2004 and 2010 (Lactic acid and Succinic acid) and three other commercialized biochemicals that were not on the DOE lists (1,3-Propanediol, 1,4-Butanediol, 1,5-Pentadiamine) and respective number of evaluated environmental sustainability assessment studies (LCA studies). For pie chart legends, see Figure 2.

CAS Number		50-21-5	110-15-6	504-63-2	1070-70-8	462-94-2
Chemical		Lactic acid	Succinic acid	1,3-Propanediol	1,4-Butanediol	1,5-Pentanediamine
World production in kt/yr (Year)	Fossil-based	n/a	76 (2015)[17]	n/a	2500 (2015)[17]	n/a
	Bio-based	472 (2015)[17]	38 (2015)[17]	128 (2015)[17]	3 (2015)[17]	50[18,19]
Main current application		Food supplement, (Poly)Lactic Acid	Food supplement, Pigment, Resin	Plastics, Cosmetics, Cleaning products	Plastics, Fibers	Nylon, Chemical intermediate
Published LCA studies		20[i]	8[ii]	5	3	0
Impact categories covered by given number of studies according to the requirements of ISO14040 and EN16760 standards[iii]						-
Production with fermentation from renewable biomass - State of commercialization		Commercialized[11]	Commercialized[11,20]	Commercialization[11,20]	Commercialized[11,20]	Commercialized[18]
Availability of inventory data		Production process data in LCI database Ecoinvent[21]	Production process data in LCI database Ecoinvent[21]	No data in LCI database	No data in LCI database	No data in LCI database
Limitations of available LCA's with focus on ass		Variation in assessed life cycle stages: 2 studies assess stages from resource extraction to acid production. 11 include polymerization and 11 assess the whole life cycle	Few studies available			No LCA studies publicly available Nothing about environmental performance is known
Opportunities for applying LCA		Conduct and publish more studies identifying hotspots and burden shifting within the life cycles. More focused research leads to increased sustainability of bio-based substances				Apply- and publish LCA studies on products and processes

If quantities are not specifically cited, information come from Amador Garcia Sancho (AIMPLAS) ii) Global Warming (GW) & Energy Demand (ED) impact results are only retrievable from Morales et al.[13] iii) One of the studies is not an LCA[11] iv) GW impact only retrievable from Gaudreault et al.[60] v) 27 follow ISO standard 4 do not follow ISO standard vi) Estimated production volume

Table 1b Main characteristics of bio-based chemicals identified by the US DOE in 2004 and 2010 (marked with bold blue letters) and three other commercialized biochemicals that were not on the DOE lists (marked with orange italic letters) and respective number of evaluated environmental sustainability assessment studies (LCA studies). For pie chart legends, see Figure 2

CAS Number		110-00-9	97-65-4	123-76-2	617-65-2	78-79-5	110-17-8	6915-15-7	617-45-8	87-73-0
Chemical		2,5-Furan dicarboxylic acid	Itaconic acid	Levulinic acid	Glutamic acid	3-Hydroxypropionic acid	Fumaric acid	Malic acid	Aspartic acid	Glucaric acid
World production in kt/yr	Fossil-based	n/a	n/a	n/a	n/a	n/a	101.3 (2014)	>70 (2017)	39.3 (2014)	0.05 (2011)
	Bio-based	0.045 (2015)[17]	41.4 (2015)[17]	3 (2015)[17]	>1500 (2014)	0.04 (2015)[17]	n/a	n/a	n/a	n/a
Main current application		PET, Plasticizer, Polyester polyols	Detergents, Latex, Polyester resins	Cosmetics, Pharmaceutical, Plasticizer	Food supplements, Pharmaceutical, Buffer	Paints, adhesives, diapers, detergents	Alkyd resins, Polyester resins, Pharmaceutical	Food ingredient, Personal care, Pharmaceutical	Aspartame, Latex, Pharmaceutical	Food Ingredient, Detergents
Published LCA studies		2[iv]	1	1	1	2	-	-	-	-
Impact categories covered by given number of studies according to the requirements of ISO14040 and EN16760 standards[v]							-	-	-	-
Production with fermentation from renewable biomass - State of commercialization		Preparing commercialization [11,18]	Commercialized [11,18]	Preparing commercialization [11,18]	Commercialized [11,19]	Preparing commercialization [11]	Not commercialized	Not commercialized	Not commercialized	Preparing commercialization[18]
Availability of inventory data		No data in LCI databases	No data in LCI databases	No data in LCI databases	No data in LCI databases	No data in LCI databases	No data in LCI databases	No data in LCI databases	No data in LCI databases	No data in LCI databases
Limitations of available LCA´s with focus on assessing environmental impacts of bio-based chemicals		Between one and two studies are available which makes it hard to draw any conclusions on the actual sustainability of the products or processes					No LCA studies publicly available Nothing about environmental performance is known			
Opportunities for applying LCA		Conduct and publish more studies identifying hotspots and burden shifting within the life cycles. More focused research leads to increased sustainability of bio-based substances					Apply- and publish LCA studies on products and processes			

Special focus was on the following five, because of their commercialization status, 1) Lactic acid: e.g. Cargill (U.S.), 2) Succinic acid: e.g. BioAmber (Canada), Succinity (Spain), 3) 1,3-Propanediol: e.g. DuPont, Tate & Lyle (U.S.), 4) 1,4-Butanediol: e.g. BioAmber (Canada), 5) 1,5-Pentanediamine (also known as Cadaverine): e.g. BASF (China)

When analyzing the status of the LCA literature presented in Table 1 for the fourteen analyzed biochemicals it has given the opportunity to make the following conclusions. There are few studies available to back up the sustainability claims of the commercialized biochemicals, when compared to conventional petrochemicals and derived products. Of the assessed impact categories, the studies have that in common that all of them assess global warming impacts, but other impact categories are less covered. It is unclear how biochemicals can sometimes perform better environmentally and sometimes worse than the petrochemicals. To investigate that I looked in depth into the identified LCAs and contrasted what impact categories and life cycle stages are covered, leading to the identification of unassessed trade-offs and hence, the results of the biochemical performance compared to petrochemicals, might be biased in both directions.

Other impact categories, identified when analyzing the existing LCA literature on biochemicals as highly relevant, land use and water use, indirect land use change, (increased demand for 1st generation crops), eutrophication (fertilizer use) and ecotoxicity (pesticide use) during feedstock production, and energy and water use in biorefineries, are much less presented.

Table 2 shows the published performance of biochemicals compared to petrochemicals and the factors of difference in environmental performance between the chemical groups.

It is unclear how biochemicals can sometimes perform better environmentally and sometimes worse than the petrochemicals. To investigate that I looked in depth into the identified LCAs and contrasted what impact categories and life cycle stages are covered, leading to the identification of unassessed trade-offs and hence, the results of the biochemical performance compared to petrochemicals, might be biased in both directions.

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Table 2 Environmental impact ratios expressed as factors of difference between bio-based and fossil-based chemicals (color range) and study count (numbers) for different chemical-process-impact combinations. Different colors within a single combination (e.g. global warming (GW) impacts associated with acid production of succinic acid) indicate that multiple scenarios in a single study (i.e. study count n=1) or results of multiple studies (i.e. study count n>1) show different impact ratios for the same chemical-process-impact combination. This variability is plotted as color range. (*Table is an extended version of Table 1 in Paper I*).

	Lactic acid			Succinic acid			1,3 Propanediol		1,4 Butanediol		1,5 Pentadiamine	2,5-furandicarboxylic acid (ii,iii)	Glutamic acid	Itaconic acid
	Assessed Life-cycle stages													
Impact category*	acid production	polymerization	end-of-life scenario	acid production	polymerization	end-of-life scenario	diol production	end-of-life scenario	diol production	end-of-life scenario	amine production	end-of-life scenario	acid production	acid production
Global Warming	2[i]	11	11	4	1	2	4	2	1	2		1	1	1
Ozone Formation	1	1	1	1		1	1		1				1	
Ozone Depletion	1	1	2	1		1			1				1	
Ionizing Radiation	1													
Particulate matter	1	1	1	1			1		1					
Human Toxicity	1	2	1	1		1	1						1	
Ecotoxicity	1	1	2	1		1	1						1	
Acidification	1	4	3			1	2		1				1	1
Eutrophication	1	3	3	1		1	1		1				1	1
Land Use	1		1			1	1	1					1	1
Water Use	1	3		1					1					1
Resource Use	1		1	1		1			1				1	
Energy Demand		9	3	3	1	1	4	2		2		1		1

Factor of difference: >10, 10, 2 — bio-based better; 1 — bio ~ fossil; 2, 10, >10 — fossil-based better; no data

i) In Morales et al., different production scenarios for lactic acid show minimum 10 times higher global warming impacts than in Daful et al.[14]. ii) Isola et al_2017[61], no comparison to a fossil-based substance with an equivalent function. Comparison between life cycle impacts on different life cycle stage of production of 2,5 FDCA. iii) Eerhart et al_2012[62], land use is calculated in relation to GWP and therefore not quantifiable in an LCA context.

There is large variation in the assessed life cycle stages presented in the analyzed studies. For succinic acid, Figure 2 shows the variation between assessed life cycle stages. Despite being ruled as problematic to always assess all life cycle stages[15] if they are not all assessed, we might overlook the possible burden shifting between the life cycle stages.

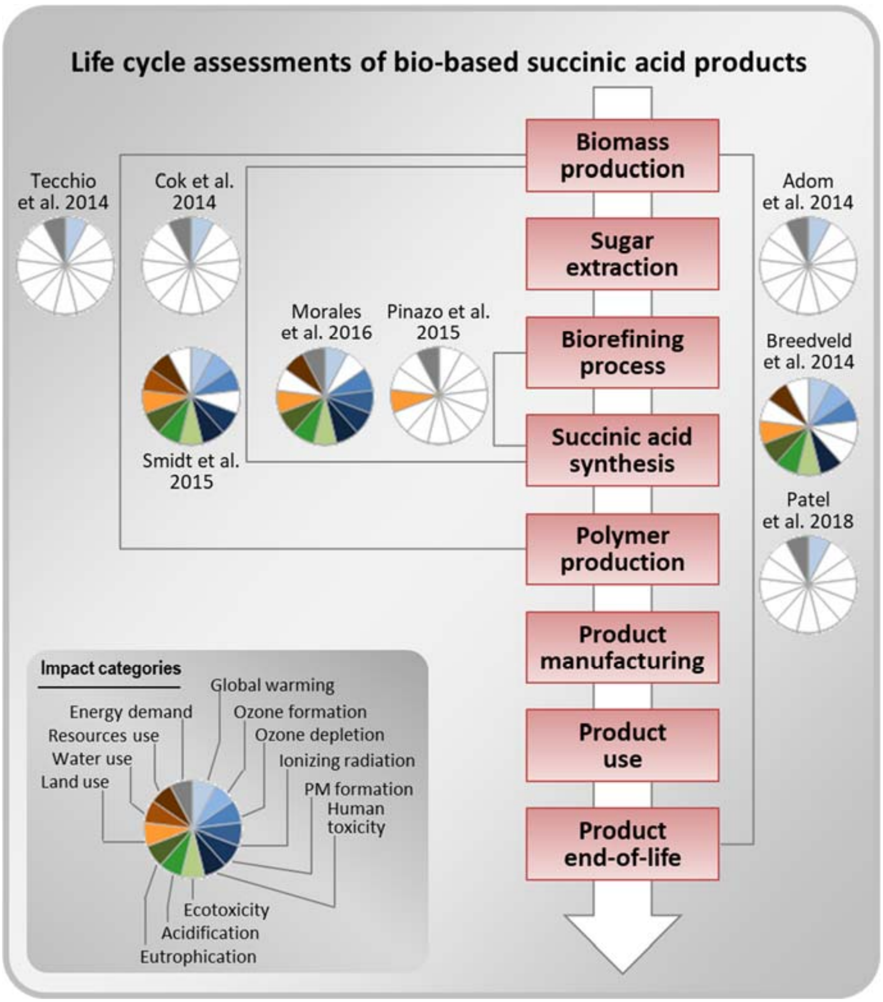


Figure 2 Overview of life cycle stages covered and impact categories considered in seven life cycle assessments of succinic acid production (*From Paper* I).

These conclusions lead to the statement that environmental sustainability claims of bio-chemicals are on thin ice because we

a) base our conclusions on few and in application limited studies, and
b) we might be overseeing trade-offs between the assessed and non-assessed impact categories, and
c) burden shifting when not assessing all life cycle stages.

3.2 HOW CAN WE IMPROVE LCA PRACTICES, BOTH AT INDUSTRY LEVEL AND AT LCA PRACTITIONER LEVEL WHEN ASSESSING THE ENVIRONMENTAL SUSTAINABILITY OF BIOCHEMICALS?

There is no simple answer to this question, but this section focuses on recommendation for improvements of LCA practices when assessing biochemicals.

A strength of LCA is its broad coverage of impact categories, ensuring that relevant impacts are reflected in the results. Generally will still though face the challenge how to communicate the variable of results to relevant stakeholders. When faced with LCA results, that will often require from our side some sort of aggregation of the results across impact categories, based on normalization and weighting of the impact scores.[8] It can also require us to translate our LCA results into common metrics representing damages to natural ecosystems (e.g. species loss) or human health (disability-adjusted life years).[63]

3.2.1 IMPROVING LCA PRACTICE FOR BIOCHEMICALS

Parts directly taken from *Paper I* are marked with "…", but the order of the paragraphs has been changed compared to the reference:
Ögmundarson, Ó., Herrgard, M. J., Forster, J., Hauschild, M. Z. & Fantke, P. *Assessing environmental sustainability of bio-based chemicals: State and challenges*. Nature Sustainability. Revised version under review.

"The analysis of existing LCA studies on biochemicals revealed that the most relevant impact categories are global warming (in many studies the only assessed category), land use and water use, eutrophication (fertilizer use) and ecotoxicity (pesticide use) during feedstock production, and energy use in biorefineries. The most relevant and variable life cycle stage is feedstock production, where a potentially very important modelling aspect is the impacts from indirect land use changes (iLUC) representing those changes in land use that may result from expansions in cropland induced by an increased demand for crops due to increases in biochemical (or biofuel) production. Biochemical processing has significant potential for sustainability optimization that becomes even more important during upscaling from laboratory to market scale, where the biochemicals industry will still need further innovation for process maturation. Finally, end-of-life treatment is relevant, as biodegradable chemicals are often claimed to be CO_2 emission neutral, but methane emissions from landfilling can offset these benefits."

“The large variation in included impacts and life cycle stages across LCA studies reflects current challenges when assessing biochemicals. Each studied system is unique in features and components, rendering it difficult to compare it with functionally equivalent systems or processes. This well-known problem, however, is not unique to biochemicals but applies to many product systems.[64–66]”

Ensuring coverage of all relevant system components and environmental impacts

“For improving LCA practice for biochemicals, we emphasize the key components to be included in each study, such as all life cycle stages, including end-of-life scenarios, and all impact categories. Indeed, it is an ISO requirement that all life cycle stages should be included in an LCA[15] to uncover possible burden shifting along product life cycles, such as environmental benefits or impacts related to certain end-of-life treatments. Below, we detail the needed adaptations of LCA for the biochemicals industry to allow giving a relevant impression of the environmental sustainability, including to adhere to existing assessment standards and available practical guidance, and to address the need to estimate currently missing data.”

“Because of the special nature of bio-based chemicals originating from biotic resources, all impact categories assessing impacts occurring in the growing phase of the biomass should by default be included in related LCA studies. For end-of-life scenarios, it is especially important to include the impact categories that address possible toxicity-related effects of waste treatment, including ecotoxicity and human toxicity, and to model potential landfill emissions of methane, a strong greenhouse gas. Spatial variability may have an important influence on the LCA results, and it should be considered if data and models are available, in particular for locally variable impact categories like freshwater use, eutrophication and ecotoxicity.“

“When assessing end-of-life scenarios, the most representative setups for relevant product applications should be included, as environmental impacts can vary greatly between disposal methods.[64,67] If end-of-life scenarios are not considered, it is still important to outline applicable scenarios, stating if relevant whether products are compostable, biodegradable under environmentally relevant conditions, or recyclable.”

Adherence to existing standards and guidelines to increase cross-study comparability

“Inconsistent application of well-defined guidelines yields highly variable LCA results even when the same impact categories are considered.[68] To avoid such issues and to strengthen the credibility of LCA results for biochemicals, we strongly suggest that future studies follow the ISO 14040 standards (International Organization for Standardization) series and the US-Environmental Protection Agency LCA principles and practices.[69] Furthermore, for making LCA on bio-based chemicals much more representative, we recommend to follow the special standard EN 16760:2015 (European Standard) for LCA on bio-based products. This standard builds on the ISO for guidance concerning the general LCA methodology, but gives for example explicit guidance modelling of agriculture, forestry and aquaculture systems, which are recognized to have relevant environmental impacts in bio-based production systems.[70]"

Estimation of missing data especially for early-stage technologies

"In the absence of real-world data, which is often the case for lab-scale production processes, reference process data, default optimization potentials, and relevant scale-up mechanisms should be considered for a first hot-spot screening. Data then need to be systematically provided for hot-spot processes and related impacts."

"We recommend more specifically the following: For modelling feedstocks, focus should be on impacts from emissions of pesticides, nutrients, and use of water and land, which may be estimated based on generic database values adapted from actual practices. For addressing geographic differentiation, chemical emissions and resource uses, inventory-modelling needs to be performed for the specific processes of the life cycle (possibly based on modification of generic inventory database processes and using local grid mix for electricity). In the impact assessment part, spatially differentiated methods are available for all non-global impact categories, which means that impact assessment research is already focused on strengthening the available methods, for example addressing spatial differentiation of human toxicity and ecotoxicity life cycle impacts.[71,72] For production efficiency, specific data should be available for the studied system and upscaling and learning may be relevant to consider when comparing new and immature technologies to conventional alternatives, depending on the scale and maturity of the processes included. For the impact assessment, we can also *a priori* identify the relevant impact categories when we know the specificities of the bio-based chemical life cycle and the conventional chemical that we want to compare. Normally, they are found among climate change (CO_2, N_2O, and CH_4 related to agriculture and energy systems), eutrophication (nutrients from agriculture), ecotoxicity (pesticides from agriculture and from the production of bio-based chemical and conventional alternative), water use (from agriculture if water is critical in the concerned region) and land use (agriculture again)."

3.2.2 PUBLIC PERCEPTION AND EXPECTED BENEFITS OF BIO-BASED PLASTICS BY COMPANIES AND OF THE APPLICATION OF LCA TO ASSESS ENVIRONMENTAL SUSTAINABILITY CLAIMS

When sustainability is on the agenda, the social dimension, despite being one of three pillars of sustainability, is often not addressed. There is need to include LCA as part of the decision support framework in a bio-based economy, and in order to understand what possible challenges and benefits are, I studies the public perception in addition to improving the LCA practice as such. Both aspects are then inputs to improve current practice in the bioeconomy to consider environmental sustainability aspects. Despite not falling within the focus of my PhD project, I had the privilege to participate in a masters project of Tim Jonas, studying university students' perceptions of bio-based plastics, and companies' uptake of environmental sustainability of bio-based plastics.[73]

The survey conducted to explore the perception of university students in Denmark and Colombia to bio-based plastics was distributed online through social media and by direct contact in different classes at the Technical University of Denmark (DTU) and Escuela de Administración, Finanzas e Instituto Tecnológico University, Medellín. This part of the MSc thesis is the foundation of a manuscript under preparation to be submitted to the Journal of Cleaner Production.[74]

Figure 3 shows the results of the qualitative public perception survey where the students were asked about their perception of environmental sustainability of bio-based plastics. 73% of the students agree, or strongly agree to that bio-based plastics are environmentally sustainable. These results are positive for everyone working in biotechnology and show that the students asked have a positive perception of bio-based products.

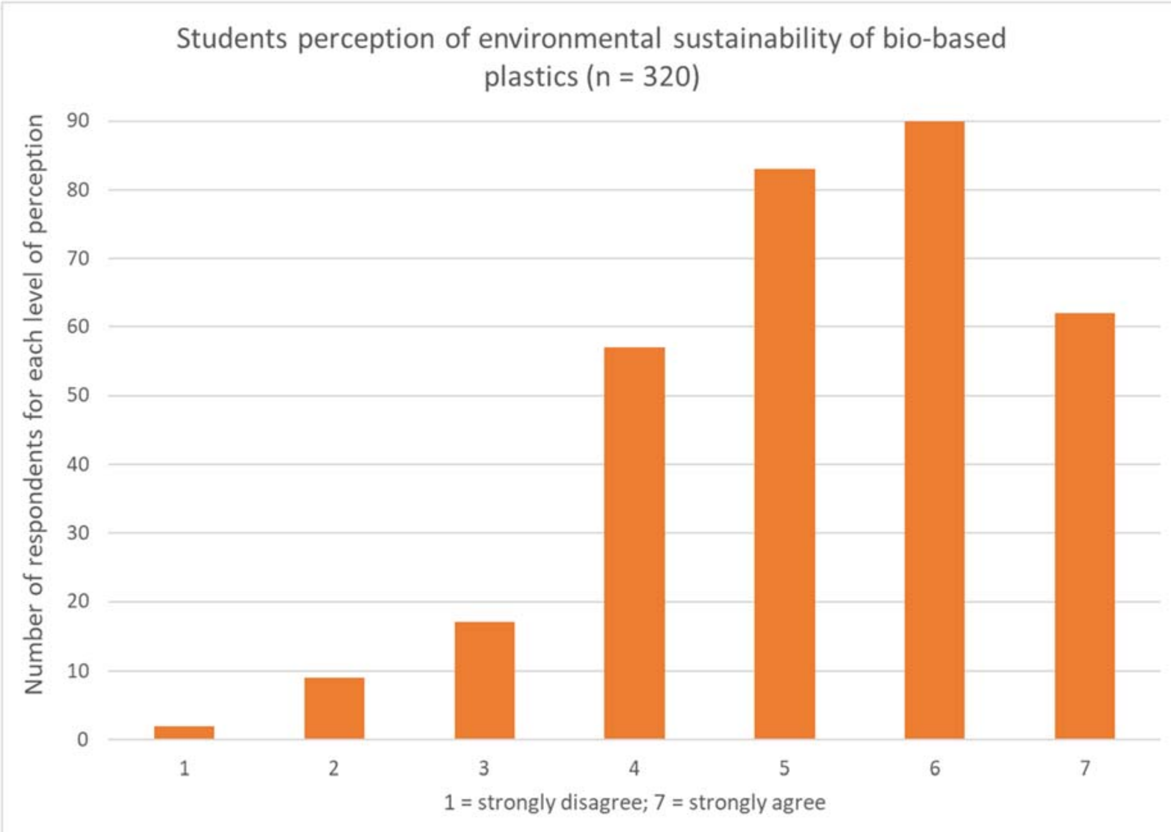


Figure 3 Number of respondents (n = 320) for each level of perception of environmental sustainability performance of bio-based plastics and perception of their environmental sustainability performance associated with production.

When corporate social responsibility reports (CSRs) were analyzed (n = 81), demonstrated in Figure 3, from companies that either produce the bio-based plastics (or their bio-based building blocks) (61%) or users of bio-based plastics for packaging or other applications (39%), a different and somewhat less optimistic view on the environmental sustainability performance of bio-based plastics is presented.

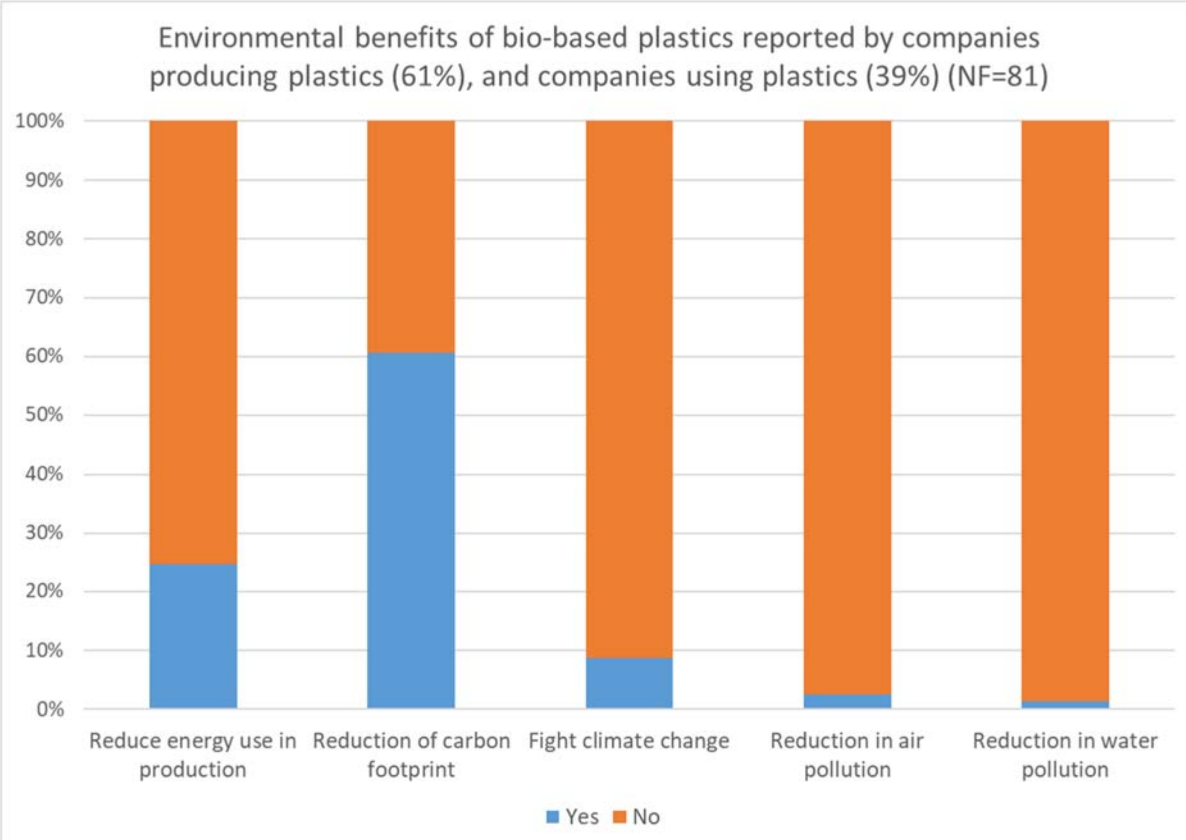


Figure 4 Companies reporting on environmental benefits of bio-based plastics.

The analyzed CSRs only report the perception of the related companies, but still show an interesting trend, presented in Figure 4. Less than 25% and down to 1% report that they see benefits of bio-based plastics when compared to fossil-based plastics, on reducing energy use in production (including production of bioplastics), and reducing air and water pollution. It was not stated, why this perception was stated in the reports. Production and use of bioplastics is also not perceived as contributing to fight climate change. On the other hand, bio-based plastics are stated to contribute to reduction of products' carbon footprints, by 61% of the CSRs.

The CSR reports were also analyzed for how the companies assess the environmental benefits of using bio-based plastics. Interestingly, only 35% of the companies apply LCA to back up their environmental sustainability claims, as presented in Figure 5. It was not stated in the 65% of the CSR reports how the companies assess their environmental sustainability claims. These results show a higher occurrence of LCA stated as the tool used to benchmark environmental sustainability, than was presented Stewart et al.[75] This difference in results between the two studies can be explained by the sectoral variation in applying LCA as stated by Stewart et al.

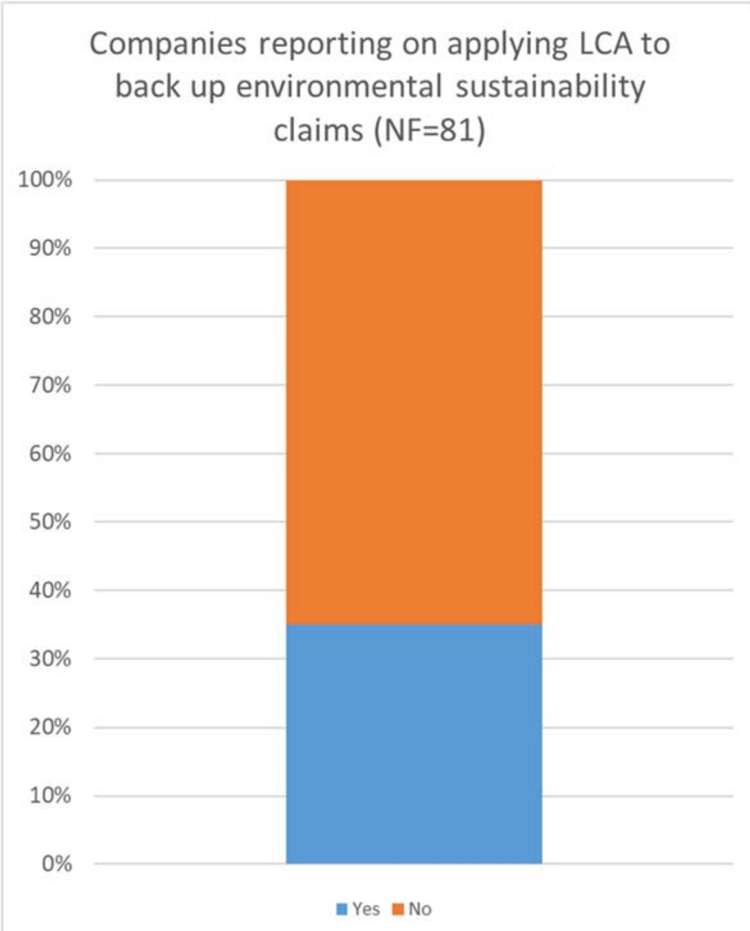


Figure 5 Percentage of companies stating in their CSR reports that they apply LCA to back up their environmental sustainability claims.

The positive perception of university students toward bio-based plastics, the less positive perception reported in CSR reports and the fact that companies rely on other methods than LCA to back up their environmental sustainability claims, or lack thereof, shows the need to systematically assess environmental sustainability of biochemicals and their derived products like bio-based plastics. If we do not back up environmental sustainability claims with quantitative assessments, we might end up producing bio-based plastics not fulfilling the expectations of customers, and companies might miss on reduction of impacts just because they do not apply the right assessment methods, or no one at all.

3.2.3 TOWARD A SUSTAINABLE BIOCHEMICAL INDUSTRY

Parts directly taken from *Paper I* are marked with "…", but the order of the paragraphs has been changed compared to the reference:
Ögmundarson, Ó., Herrgard, M. J., Forster, J., Hauschild, M. Z. & Fantke, P. *Assessing environmental sustainability of bio-based chemicals: State and challenges.* Nature Sustainability. Revised version under Review.

“We identified several sustainability challenges for the biochemicals industry that require additional development efforts. Bio-based chemicals can show lower or higher global warming impacts compared to fossil-based chemicals, and often show higher impacts in other categories, such as land use. LCA is a useful tool to identify hotspots in environmental sustainability profiles of bio-based chemicals.[76] Significant additional research and development efforts are required mainly regarding feedstock production, biorefining and product recycling, for further improving the overall sustainability of bio-based products.”

“When assessing opportunities using lignocellulosic biomass, macro- and microalgae as next generation feedstock, main challenges are related to data availability and accessibility, as well as targeting sustainability-related hotspots in biochemicals production that may differ between feedstock generations. We need methods for effectively scaling up laboratory data to being more representative for commercial scale production and to better reflect on the optimization potential of bio-based chemicals, as various production processes are currently still immature. This work may be inspired by comparisons of efficiencies and emissions for lab scale process and commercial full scale processes for other similar biotech chemicals and materials. It is further possible to define minimum fermentation yield performance and productivity that would be required to become commercially viable, or to soft-link process simulation with LCA, enabling plant-wide design by scaling up lab-scale technologies using scaling factors.[77]”

“At the early stages of biorefinery development, feasibility studies should include at least screening-level LCA to identify major hotspots in the product system proper. For assessments where the purpose is to investigate the consequences at societal scale of a change towards 1st generation bio-based chemicals, the LCA should aim to model the consequences at societal scale, and further modelling efforts are required to address the indirect land use change impacts. As an example, an increased demand for corn to produce bio-based chemicals in the United States, may lead to expansion of the corn production to other regions to meet overall greater demand. This may eventually induce conversion of natural areas into farmed land causing environmental impacts that are potentially large[78] but typically not considered in LCA of individual products and materials as reported in this study. Finally, the ‘wicked nature of sustainability’[79] calls for considering consumer preferences to a higher degree[80], since traditional methods dealing with optimization problems might not be sufficient and application of multidisciplinary approaches are necessary to boost the sustainability of bio-based products.”

"In perspective, we observe that socio-economic aspects including population, transportation, and the use of primary energy, water, fertilizers and biotic and abiotic resources grow rapidly over the last decades.[81] These aspects drive increasing impacts on global warming, ocean acidification, eutrophication, stratospheric ozone depletion, and impacts on humans and ecosystems from chemical emissions, and on depletion or degradation of land, water, fossil and other resources. Some of these trends already exceed our earth's capacity for sustaining the current socio-economic development. Hence, just ever being "more sustainable" is not enough, especially when consumption increases globally[2,82]. The biochemicals industry needs to explore how innovation can contribute to being sustainable in absolute terms based on the capacity of sustaining our biophysical earth systems, while meeting the growing needs for viable bulk chemicals in today's and future societies. For LCA practitioners, this means that there is no excuse not to look at all relevant impacts and include all life cycle stages to fully supporting a comprehensive improvement of biochemicals' environmental performance. For biotechnology developers, this means to better integrate LCA as a tool that can quantitatively support a truly sustainable development of biochemicals instead of relying on partially justifiable sustainability claims such as reduction of CO_2 emissions in the chemical production phase alone compared to a petrochemical alternative."

In the next chapter, based on the deficits and recommendations provided in this chapter, I showcase to set a standard of what to include and how to do an LCA on a biochemical, with focus on lactic acid.

4. Life cycle assessment of lactic acid produced from different feedstocks

The following chapter will give an overview of what we can learn from applying LCA to identify environmental hotspots in the production of lactic acid from three feedstock generations and how it can help explore environmental optimization potential at a very early stage.

The chapter is divided into three sub-sections. Each section starts with a short introduction, followed by an overview of the applied methodology to create the results (might not always be needed), followed by the results and a short discussion. The phrasing *From Paper XX* in the beginning of a section means it has been taken directly from the relevant paper.

4.1 LCA of lactic Acid production from three generations of feedstocks

This section is based on Paper II and Paper IV and the lessons learned here are how to assess and identify, with hot-spot analysis, potential process optimization of established and future feedstocks in biochemical production, considering the different maturity of the feedstock processes and their uncertainty. This section starts with a short description of the LCA methodology, followed by the results from the study.

For identifying the different environmental impact hotspots within each biochemical product system based on either 1st, 2nd or 3rd generation biomass as feedstock, in support of optimizing biochemical production at each of the considered TRLs, our LCA study follows the International Reference Life Cycle Data System (ILCD) Handbook for LCA[83],the EN 16760:2015 standard for bio-based products[70], and the ISO 14040 and 14044.[84,85]

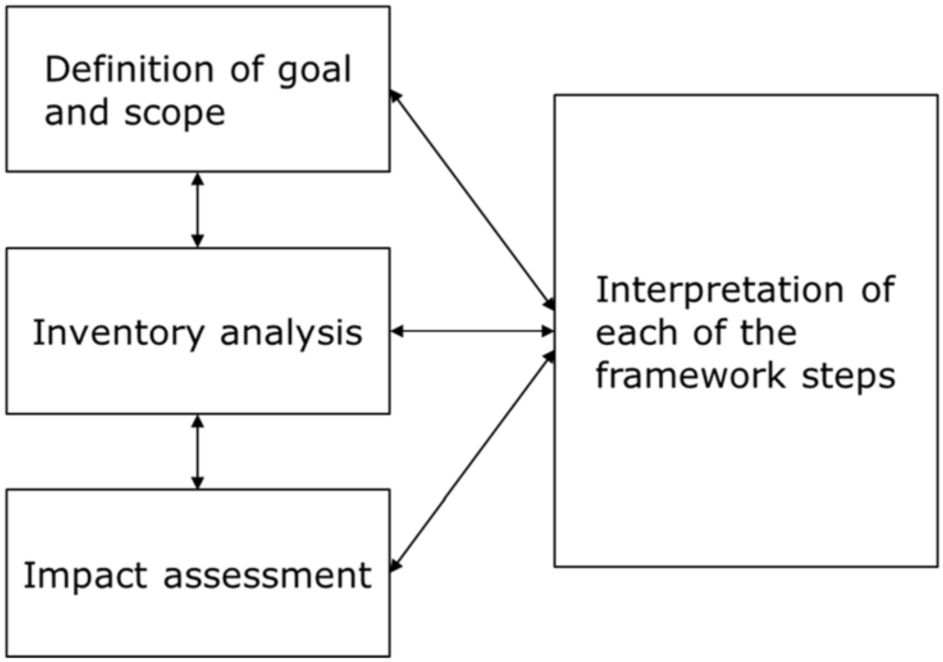


Figure 6 Life cycle assessment framework adapted from the ILCD handbook.[86]

When complying to the ISO 14040[84], 14044[85], and the ILCD [86], the LCA is performed through four different phases (Figure 6). In the first step, the goal and scope of the study is defined and developed, considering the intended application. The object of the study is defined by the functional unit (FU). Besides describing the functional unit, the goal and scope refer to the overall approach used to determine the system boundaries. The system boundaries establish which unit processes are included in the study and have to represent the goal of the study.

The second step in the LCA is the inventory analysis, which implies data collection and modelling of the product system. Further, it involves data description and verification. At this phase in the LCA, all data are included regarding inputs, energy consumption, emissions, etc. that are relevant for all the unit processes in the system boundaries. The system boundaries define the product system. The data has to be related to the functional unit.

The third phase in the LCA is the life cycle impact assessment, which aims to assess the contribution to impact categories such as climate change and ecotoxicity. Characterization is the first step within this phase, where the impact potentials are estimated based on the LCI results.

The fourth and last stage is the interpretation where analyses of the major contributions are performed, as well as a sensitivity and uncertainty analysis. This step allows drawing a conclusion whether the objectives in the goal and scope can be fulfilled.

4.1.1 SCOPE DEFINITION

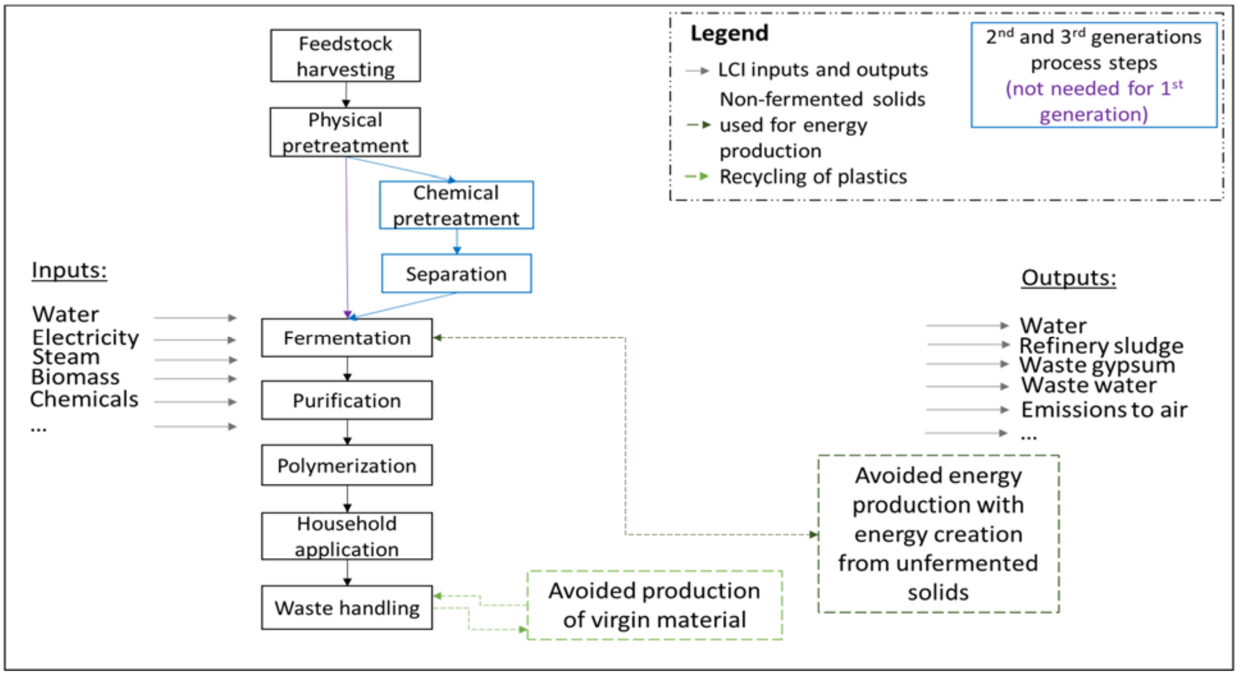


Figure 7 Process flow diagram – Utilization of three generations of feedstocks, corn (1st generation), corn-stover (2nd generation) and macroalgae (3rd generation). (*Figure from Paper II*).

The assessment of LA production from three different generations of feedstocks (for overview, see section 1.2) is a cradle-to-grave study, as demonstrated in Figure 7, which includes the biomass life cycle stage, consisting of the assessment of 1st generation feedstock cultivation, 2nd generation feedstock fertilizer value, and 3rd generation feedstock harvesting. The second life cycle stage is the biorefinery, the third is polymerization and fourth and last life cycle stage is the assessment of end-of-life.

The functional unit, which reflects the systems function and is the basis of the assessment[84], is defined as "the production and use of 1 kg of lactic acid, with 99.9% purity, for household packaging application in the United States". By household packaging application, we mean food packaging, which follows the waste streams of household waste.

When deciding the scope of the study, applying system expansion was identified as the most appropriate methodological approach when applying hot-spot analysis to assess the optimization potential of biochemicals production from different feedstock sources. When assessing literature assessing biochemical production from feedstocks like corn-stover, as an example, applying economic allocation was the primary choice[87] despite that the market value for the co-product (2nd generation biomass) is often low, leading to small or even no impacts allocated to the biomass[88]. Another issue is that often, for 2nd generation biomass, it can be a valuable source of nutrients for the fields if left on them, such as in the case of corn-stover. It is therefore a crucial point to assess the fertilizer value to that kind of feedstock.[89]

4.1.2 Inventory analysis

Acquiring detailed and reliable inventory data is not easy and is often the most time consuming part of conducting an LCA, but the quality of our work is fully dependent on the detail level of the data. If the available databases for life cycle inventories, like ecoinvent, do not include the needed background processes to populate the data, we need to have access or populate the needed energy and mass balances by other means. An example of this is the case when assessing non-developed processes only explored in laboratories, like utilizing macroalgae as feedstock for biorefineries. The first option to collect the needed data is to get it from industry, through measuring on site, and that is the most accurate source, but this option is often not possible for LCA practitioners. The second option is to get the data from literature and the third option is to first simulate the e.g. biorefinery process with different software's, like AspenPlus©.

For this study, process simulation was identified as the most suitable tool. Before simulation is executed, the biorefinery process needs to be conceptualized, mostly based on scientific data or relevant patents for process configurations. Actual real-time data from industry is favored, but that is often not the case and then we need to rely on available data.

By integrating process simulation in LCAs of biorefineries, we overcome a large hurdle of data availability and accessibility. When assessing environmental impacts of biochemicals at an early stage, if available, we need to effectively scale up laboratory data to be more representative for commercial scale production and to better reflect on the optimization potential of bio-based chemicals, as various production processes are currently still immature. This can be done by integrating LCA with chemical process simulation that accounts for the increase in capacity of the process under scrutiny.[77,90,91]

The process simulations providing the energy and mass balances for the LCA study, which is part of this PhD, were conducted by Sumesh Sukumara (Researcher at DTU Biosustain), providing the inventory for the 1st and 2nd generation biomass processes. Elena Tomás Grasa (MSc thesis[92]), provided the 3rd generation biomass process, with me contributing to conceptual design and data interpretations. This work will be presented in a paper manuscript that is being written[93], and I co-author, and is also described in the supplementary information to Paper II.

Another way to get access to available inventory data is to rely on other databases that provide sufficient energy and mass balances to build the life cycle inventory (LCI), like the IHS Markit© database.[94] Getting access to these kinds of databases is though often very expensive and they often limit what data information can be revealed in for example scientific studies. In the LCA part of this PhD we had access to the IHS Markit© database and used their energy and mass balances for the polymerization life cycle stage, and also for evaluating the results we got from the process simulations of the different biorefinery feedstock generation processes. When relying on databases, like the IHS Markit©, it can open up for questions about how certain they are, and the short answer is that you cannot be absolutely sure about precisely that. That is therefore expressed in the uncertainty analysis with the proper uncertainty value.

Despite being ruled as infeasible to include end-of-life options for all LCA studies[20], as benchmarked in section 3.2.1, if we want to assess the overall environmental impacts we need to assess all life cycle stages. It is right, still, that assessing for example end-of-life scenarios is not always easy. For this study I took the country specific municipal waste treatment average, as given by the OECD Stats,[95] and used that information to model the End-of-Life (EoL). Using average data, like I did, can be misleading in the sense that it does not give specific geographical results if there are differences for example between states in the US on how they treat their municipal waste. This methodological approach still gives the opportunity to assess the scenario sensitivity of the end-of-life stage, and for example in the case of PLA, if recycling has a positive effect and then to what extent.

4.1.3 Life cycle impact assessment at midpoint

In this section, as a first step, lessons learned from the life cycle assessment with a hotspot analysis are shown and discussed, addressing general trends between the different feedstock generations, across life cycle stages, at midpoint. Secondly, we present the main observations gathered and how the different results can provide recommendations for process optimization to professionals working with the different feedstock generations. All figures presented in this section are taken from Paper II, and parts of text too (marked with *From Paper II*).

When it comes to the impacts assessed in the LCA, we need to have a clear vision of how we are going to use the results, and that defines which results we need. When using hotspot analysis for process optimization, if one impact category is more of interest than another, we can assess the impact at midpoint (see Table 3) as covered for example in the ReCiPe 2016 impact assessment method.[63] The methodological choice of ReCiPe 2016 is that it also allows impacts at damage level assessed, as will be demonstrated, and is of interest when optimizing for environmental impacts across life cycle impacts. The interest in impacts at damage level is because if we want to optimize across impact categories, we need to translate the LCA results to that level.

Table 3a, b, and c. Midpoint environmental impact results for lactic acid production from three feedstock generations, including Monte Carlo uncertainty analysis, TRL and hotspots (expressed as % contribution of life cycle stages, BM: Biomass, BR: Biorefinery, PM: Polymerization, and EoL: End-of-life. (*Table 3 from Paper II*).

3.a		TRL 8-9	Lactic acid from corn Life cycle stages			
Impact categories	Unit	Tot. Res. (2.5^{th} – 97.5^{th}%)	BM	BR	PM	EoL
Global warming	kg CO2 eq	4.2 (1.3 – 4.8)	47.5%	59.1%	1.8%	-8.5%
Stratospheric ozone depletion	kg CFC11 eq	2x10^{-5} (8.4x10^{-06} – 1.6x10^{-05})	95.8%	3.4%	0.1%	0.6%
Ionizing radiation	kBq Co-60 eq	-0.22 (-1.2 – 0.015)	14.9%	-121.6%	3.6%	3.1%
Ozone formation, Human health	kg NOx eq	7.1x10-^{3} (4.4x10^{-3} – 8.9x10^{-3})	37.7%	75.8%	2.4%	-15.8%
Fine particulate matter exposure	kg PM2.5 eq	1.8x10^{-2} (5.8x10^{-3} – 3.6x10^{-2})	18.2%	82.8%	1.0%	-1.9%
Ozone formation, Terrestrial ecosystems	kg NOx eq	7.3x10^{-3} (4.5x10^{-3} – 9.1x10^{-3})	37.6%	77.3%	2.7%	-17.5%
Terrestrial acidification	kg SO2 eq	0.077 (0.037 – 0.13)	20.3%	81.2%	0.4%	-1.8%
Freshwater eutrophication	kg P eq	5.2x10^{-5} (-3.1x10^{-3} – 1.7x10^{-3})	978.6%	-1057.4%	120.0%	58.8%
Marine eutrophication	kg N eq	5.6x10^{-3} (1.8x10^{-3} – 3.5x10^{-3})	84.8%	9.3%	0.1%	5.9%
Terrestrial ecotoxicity	kg 1,4-DCB	12 (7.1 – 19)	20.0%	72.8%	10.8%	-3.6%
Freshwater ecotoxicity	kg 1,4-DCB	0.12 (-0.055 – 0.35)	25.1%	-6.1%	3.6%	77.4%
Marine ecotoxicity	kg 1,4-DCB	0.17 (-0.064 – 0.49)	19.8%	2.9%	4.1%	73.2%
Human carcinogenic toxicity	kg 1,4-DCB	0.048 (-0.19 – 0.24)	73.7%	34.4%	10.0%	-18.1%
Human non-carcinogenic toxicity	kg 1,4-DCB	4.3 (-0.041 – 13)	7.6%	37.7%	4.9%	49.8%
Land use	m2a crop eq	1.4 (0.99 – 1.9)	93.5%	6.5%	0.1%	0.0%
Mineral resource scarcity	kg Cu eq	0.012 (7.8x10^{-03} – 1.8x10^{-02})	33.6%	59.0%	11.8%	-4.4%
Fossil resource scarcity	kg oil eq	0.65 (6.4x10^{-03} – 1.1)	36.6%	137.8%	2.7%	-77.1%
Water consumption	m3	0.43 (-2.0 – 2.8)	75.1%	26.2%	0.6%	-1.9%

3.b

Impact categories	Unit	TRL 4-5 Tot. Res. (2.5th – 97.5th%)	Lactic acid from corn stover Life cycle stages BM	BR	PM	EoL
Global warming	kg CO2 eq	7.9 (6.0 – 9.2)	36.7%	66.8%	1.0%	-4.5%
Stratospheric ozone depletion	kg CFC11 eq	$3.3x10^{-6}$ ($2.6x10^{-6}$ – $4.0x10^{-6}$)	40.7%	54.7%	0.9%	3.7%
Ionizing radiation	kBq Co-60 eq	0.3 (0.038 – 1.3)	0.0%	95.2%	2.6%	2.2%
Ozone formation, Human health	kg NOx eq	$1.2x10^{-2}$ ($9.7x10^{-3}$ – $1.4x10^{-2}$)	47.7%	60.4%	1.4%	-9.5%
Fine particulate matter exposure	kg PM2.5 eq	0.024 (0.015 – 0.035)	22.0%	78.7%	0.7%	-1.4%
Ozone formation, Terrestrial ecosystems	kg NOx eq	$1.2x10^{-2}$ ($9.9x10^{-3}$ – $1.5x10^{-2}$)	47.4%	61.5%	1.6%	-10.5%
Terrestrial acidification	kg SO2 eq	0.064 (0.038 – 0.11)	16.2%	85.5%	0.4%	-2.2%
Freshwater eutrophication	kg P eq	$3.5x10^{-3}$ ($1.3x10^{-3}$ – $6.6x10^{-3}$)	35.3%	62.0%	1.8%	0.9%
Marine eutrophication	kg N eq	$1.3x10^{-3}$ ($8.7x10^{-4}$ – $1.9x10^{-3}$)	12.1%	61.5%	0.3%	26.1%
Terrestrial ecotoxicity	kg 1,4-DCB	22 (14 – 37)	42.1%	53.9%	6.0%	-2.0%
Freshwater ecotoxicity	kg 1,4-DCB	0.33 (0.16 – 0.69)	25.8%	45.3%	1.3%	27.6%
Marine ecotoxicity	kg 1,4-DCB	0.46 (0.22 – 0.95)	26.3%	45.6%	1.5%	26.6%
Human carcinogenic toxicity	kg 1,4-DCB	0.29 (0.095 – 0.82)	38.3%	63.1%	1.7%	-3.0%
Human non-carcinogenic toxicity	kg 1,4-DCB	9.5 (3.7 – 23)	27.0%	47.9%	2.2%	22.9%
Land use	m2a crop eq	0.17 (0.17 – 0.33)	0.0%	99.8%	0.5%	-0.4%
Mineral resource scarcity	kg Cu eq	0.011 (0.011 – 0.031)	0.0%	91.6%	13.4%	-5.0%
Fossil resource scarcity	kg oil eq	1.8 (1.2 – 2.2)	39.5%	87.8%	1.0%	-28.3%
Water consumption	m3	0.15 (-3.6 – 3.2)	13.5%	90.1%	1.6%	-5.2%

3.c

Impact categories	Unit	TRL 2-3 Tot. Res. (2.5th – 97.5th%)	Lactic acid from macroalgae Life cycle stages BM	BR	PM	EoL
Global warming	kg CO2 eq	11 (7.14 – 15.2)	50.9%	51.6%	0.7%	-3.2%
Stratospheric ozone depletion	kg CFC11 eq	$5.8x10^{-6}$ ($3.6x10^{-6}$ – $1.0x10^{-5}$)	51.4%	46.1%	0.5%	2.0%
Ionizing radiation	kBq Co-60 eq	0.27 (-0.13 – 2.7)	45.4%	51.8%	1.5%	1.3%
Ozone formation, Human health	kg NOx eq	0.015 (0.011 – 0.022)	54.5%	51.6%	1.1%	-7.2%
Fine particulate matter exposure	kg PM2.5 eq	0.02 ($8.6x10^{-03}$ – $3.2x10^{-02}$)	50.1%	50.8%	0.9%	-1.7%
Ozone formation, Terrestrial ecosystems	kg NOx eq	0.016 (0.011 – 0.022)	54.9%	51.9%	1.2%	-8.0%
Terrestrial acidification	kg SO2 eq	0.045 (0.032 – 0.061)	51.1%	51.4%	0.6%	-3.1%
Freshwater eutrophication	kg P eq	$3.0x10^{-3}$ ($9.0x10^{-5}$ – $1.0x10^{-2}$)	43.6%	53.8%	1.8%	0.9%
Marine eutrophication	kg N eq	$1.1x10^{-3}$ ($7.3x10^{-4}$ – $2.0x10^{-3}$)	9.2%	62.9%	0.3%	27.6%
Terrestrial ecotoxicity	kg 1,4-DCB	29 (17 – 73)	41.9%	55.4%	4.0%	-1.3%
Freshwater ecotoxicity	kg 1,4-DCB	0.3 (0.078 – 0.74)	28.3%	42.3%	1.3%	28.1%
Marine ecotoxicity	kg 1,4-DCB	0.42 (0.13 – 1)	28.8%	42.9%	1.5%	26.8%
Human carcinogenic toxicity	kg 1,4-DCB	0.26 (0.096 – 1.2)	55.6%	45.5%	1.4%	-2.5%
Human non-carcinogenic toxicity	kg 1,4-DCB	7.1 (2.9 – 22)	24.7%	46.8%	2.5%	26.0%
Land use	m2a crop eq	1.1 (0.74 – 1.4)	86.4%	13.6%	0.1%	-0.1%
Mineral resource scarcity	kg Cu eq	0.022 (0.013 – 0.047)	51.7%	44.5%	6.1%	-2.3%
Fossil resource scarcity	kg oil eq	3.1 (1.9 – 4.6)	61.6%	53.9%	0.6%	-16.1%
Water consumption	m3	0.46 (-2.64 – 3.2)	43.0%	58.2%	0.5%	-1.7%

However, before we start looking into optimization potential of individual impact categories or across impact categories, I would like to start with addressing the trends illustrated in Table 3. First, when we look at the different feedstock processes, we can see that they are all three at different technical readiness levels (TRLs), which means that they are not optimized to the same extent. LA from corn is a commercialized process, so for optimization the challenges are more technical, while LA from corn-stover has passed its early stages of development and LA from macroalgae is still only at laboratory scale, which still gives optimization potential in the early stages of development because of the under-developed processes. This reflects the high uncertainty also shown in Table 3. The different TRLs means we cannot compare the results across feedstock generations, but the optimized process still sets the threshold, which the two other feedstock processes need to reach as a minimum to become of interest to biochemical producers.

When assessing hotspots across life cycle stages of the three feedstock processes, there are certain observable trends, but they are all mostly affected by high energy utility use and where it is used in the different life cycle stages. In the 1st and 2nd generation feedstock processes, most energy utility inputs happen in the biorefinery stage, while in the 3rd generation feedstock process, energy utilities have the highest effect in the biomass life cycle stage. This is because drying of the biomass is considered the best way to reduce the high water content of the biomass, leading to that energy utility inputs account for up to 86% of the related impacts. Table 4a and b shows the changes in hotspots across life cycle stages, given that drying is not a necessary pre-treatment step to make the macroalgae more suitable for fermentation. This option is being explored in the research project ThermoFactories by Elleke Fenna Bosma, Postdoc at DTU Biosustain, and the preliminary results show positive signs that this could also be a preferable step regarding biomass yields.

Table 4a and b. Changes in hotspots across life cycle stages if drying of macroalgae is not needed and the decrease of all environmental impacts effected by the energy utility reduction.

4.a

		TRL 2-3	Lactic acid from macroalgae Life cycle stages			
Impact categories	Unit	Tot.Res.	BM	BR	PM	EoL
Global warming	kg CO2 eq	1.1E+01	50.9%	51.6%	0.7%	-3.2%
Stratospheric ozone depletion	kg CFC11 eq	6.1E-06	51.4%	46.1%	0.5%	2.0%
Ionizing radiation	kBq Co-60 eq	5.2E-01	45.4%	51.8%	1.5%	1.3%
Ozone formation, Human health	kg NOx eq	1.6E-02	54.5%	51.6%	1.1%	-7.2%
Fine particulate matter exposure	kg PM2.5 eq	2.0E-02	50.1%	50.8%	0.9%	-1.7%
Ozone formation, Terrestrial ecosystems	kg NOx eq	1.6E-02	54.9%	51.9%	1.2%	-8.0%
Terrestrial acidification	kg SO2 eq	4.5E-02	51.1%	51.4%	0.6%	-3.1%
Freshwater eutrophication	kg P eq	3.6E-03	43.6%	53.8%	1.8%	0.9%
Marine eutrophication	kg N eq	1.2E-03	9.2%	62.9%	0.3%	27.6%
Terrestrial ecotoxicity	kg 1,4-DCB	3.2E+01	41.9%	55.4%	4.0%	-1.3%
Freshwater ecotoxicity	kg 1,4-DCB	3.3E-01	28.3%	42.3%	1.3%	28.1%
Marine ecotoxicity	kg 1,4-DCB	4.6E-01	28.8%	42.9%	1.5%	26.8%
Human carcinogenic toxicity	kg 1,4-DCB	3.5E-01	55.6%	45.5%	1.4%	-2.5%
Human non-carcinogenic toxicity	kg 1,4-DCB	8.3E+00	24.7%	46.8%	2.5%	26.0%
Land use	m2a crop eq	1.0E+00	86.4%	13.6%	0.1%	-0.1%
Mineral resource scarcity	kg Cu eq	2.4E-02	51.7%	44.5%	6.1%	-2.3%
Fossil resource scarcity	kg oil eq	3.1E+00	61.6%	53.9%	0.6%	-16.1%
Water consumption	m3	4.6E-01	43.0%	58.2%	0.5%	-1.7%

4.b

		TRL 2-3	Lactic acid from macroalgae WO Drying Life cycle stages			
Impact categories	Unit	Tot.Res.	BM	BR	PM	EoL
Global warming	kg CO2 eq	5.6E+00	2.6%	102.4%	1.4%	-6.4%
Stratospheric ozone depletion	kg CFC11 eq	3.1E-06	2.8%	92.2%	1.0%	4.0%
Ionizing radiation	kBq Co-60 eq	2.9E-01	1.2%	93.7%	2.7%	2.3%
Ozone formation, Human health	kg NOx eq	7.2E-03	2.0%	111.3%	2.3%	-15.6%
Fine particulate matter exposure	kg PM2.5 eq	1.0E-02	1.0%	100.7%	1.8%	-3.4%
Ozone formation, Terrestrial ecosystems	kg NOx eq	7.3E-03	2.1%	112.7%	2.7%	-17.4%
Terrestrial acidification	kg SO2 eq	2.3E-02	1.3%	103.6%	1.2%	-6.2%
Freshwater eutrophication	kg P eq	2.0E-03	0.2%	95.2%	3.1%	1.5%
Marine eutrophication	kg N eq	1.1E-03	0.0%	69.3%	0.3%	30.4%
Terrestrial ecotoxicity	kg 1,4-DCB	1.9E+01	0.1%	95.3%	6.9%	-2.3%
Freshwater ecotoxicity	kg 1,4-DCB	2.3E-01	0.1%	58.9%	1.8%	39.2%
Marine ecotoxicity	kg 1,4-DCB	3.3E-01	0.2%	60.2%	2.1%	37.5%
Human carcinogenic toxicity	kg 1,4-DCB	1.6E-01	0.5%	102.0%	3.0%	-5.5%
Human non-carcinogenic toxicity	kg 1,4-DCB	6.3E+00	0.2%	62.0%	3.4%	34.4%
Land use	m2a crop eq	1.4E-01	0.3%	99.5%	0.6%	-0.4%
Mineral resource scarcity	kg Cu eq	1.1E-02	0.6%	91.5%	12.5%	-4.7%
Fossil resource scarcity	kg oil eq	1.3E+00	9.3%	127.4%	1.3%	-38.0%
Water consumption	m3	2.6E-01	0.0%	102.1%	0.9%	-3.0%

As described in detail in section 3.2.1, when assessing the environmental sustainability of biochemicals we should pay special attention to global warming, land use and water use, eutrophication, ecotoxicity, and indirect land use change, especially when assessing 1st generation biomass[19] (see Table 5) because of the farming practices of the feedstock.

A general discussion point for all the result, impacts are highly influenced by whether the feedstock is grown, collected or harvested. Effects such as terrestrial acidification and marine eutrophication are highly influenced by fertilizer and or pesticide use, leading to environmental hotspots shifts between the different feedstock processes. However, because of different units, we cannot compare changes in hotspots across impact categories because they are expressed in different units. That results in, if we want to compare hotspots across impact categories, the need to translate the LCI results to area of protection. By doing that we can identify environmental hotspots across impact categories.

Table 5 Global warming impacts (kg CO2 eq.) of lactic acid production from 3 generations of biomass, from cradle-to-grave. (*From Paper II)*.

	Unit	Global warming	Global warming without iLUC	Biogenic carbon storage	Net biogenic carbon emissions
Lactic acid from corn	**kg CO2 eq**	**6.47E+00**	**4.72E+00**	**2.55E+00**	**3.91E+00**
Lactic acid from corn-stover	**kg CO2 eq**	**8.96E+00**	**-**	**2.91E+00**	**6.05E+00**
Lactic acid from macroalgae	**kg CO2 eq**	**1.10E+01**	**-**	**4.51E+00**	**6.52E+00**

One more thing before going to the LCA results at damage level, assessing iLUC is not general practice in the LCA literature of biochemicals. That fact makes this LCA the first to include these impacts when assessing the environmental impacts of a commodity chemical, and show how much including the iLUC in the 1st generation feedstock process affects the overall impact of the process. The LCA includes the biophysical indirect land-use change (iLUC) attributional model developed by Schmidt et al.[78] iLUC contributes to the LCA results through LCI results, where intensification of already farmed land (to meet increased demand for crops) contribute to relevant environmental impacts through increased nitrogen fertilizer use and transformation from secondary forests to croplands contributes to CO_2 emissions[78] (see Figure 8). When assessing iLUC impacts, indirect impacts are considered in the decisions for optimization of biochemicals produced from 1st generation biomass and effect decisions when exploring alternative feedstocks for biochemical production.

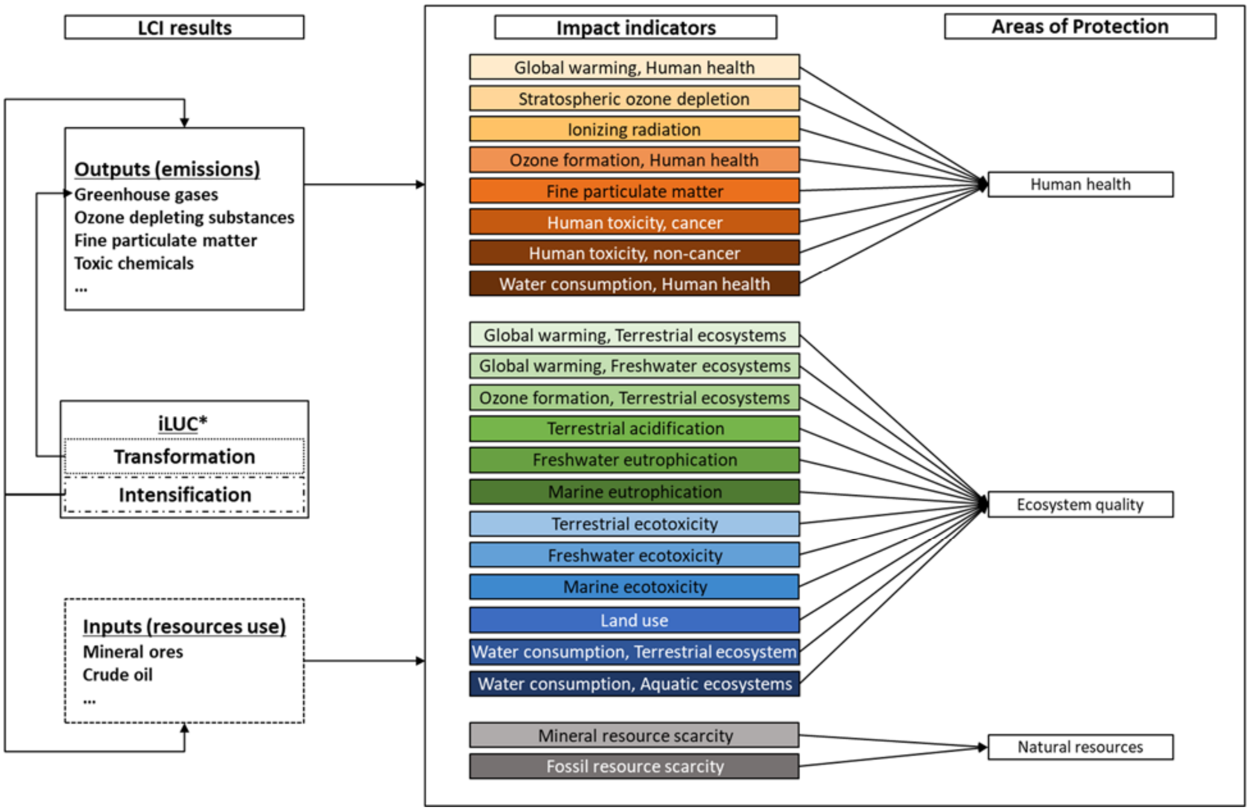


Figure 8 Overall indirect land use change effects on impact indicators at damage level. *(From Paper II)*

4.2 OPPORTUNITIES FROM APPLYING LCA FOR THE BIO-BASED INDUSTRY WITH SPECIAL FOCUS ON MACROALGAE

Parts of this section are taken directly taken from *Paper II* and are marked with "…": Ögmundarson, Ó., Sukumara, S., Laurent, A. & Fantke, P. *Environmental hotspots of different lactic acid production systems*. Energy and Environmental Science - To be Submitted.

When assessing hotspots across feedstock generations, the biggest limitation is the difference in the technical readiness level of the assessed processes. To overcome this limitation we need to bring all processes to the same level for assessing their environmental impacts, and we do that by systematically scaling all assessed processes to a full manufacturing scale with the end in mind, meaning making realistic scenarios (e.g. in this case assume production capacity of biorefinery close to market leading producer). That also embodies being persistent on level of details of all assessed processes.[96] For early stage environmental assessments of processes with low TRL, there is high optimization potential, while processes with a high TRL have lower optimization potential and commercialized processes have the least potential to lower their environmental impacts. Still, by seeing how a process with high TRL performs, gives the benchmark for how well future processes need to perform to become valid. For generating the needed inventory for the three-feedstock processes assessed in this study, based on feedstock reactions and chemical engineering, mass flows, energy balances, and process flow diagrams were generated with techno-economic assessment (see section 4.1.2).

Despite the TRL and from that point on optimize the processes, based on the different results. If one wants to compare processes across TRLs, the interpretation of the results must be done with precaution and the different TRLs must be stated. Given that uncertainty of process mass flows increases with lower TRLs, this should be reflected in an uncertainty analysis as presented in Figure 9.

If we look specifically at the optimization potential of the macroalgae feedstock process, based on the results for human health and ecosystem quality, uncertainty ranges across generations are overlapping to the extent that with current uncertainty it is difficult to identify the most relevant optimization potential in macroalgae scenario. This is different for natural resources, where macroalgae has high potential to reduce impacts, as compared to corn and corn stover, by 59-83%. As mentioned before, drying of macroalgae is the most contributing input to resources impacts for macroalgae, and here is therefore the highest optimization potential for feedstock process. Therefore, we also assessed if drying of the biomass was not needed before fermentation and that would reduce the environmental impacts of the 3rd generation process drastically. Despite the data uncertainty, it has the potential to bring the environmental impacts close to the assessed impacts of the 1st generation process.

When we consequently want to explore the environmental optimization potential of biochemicals, we can do that by assessing their environmental hotspots

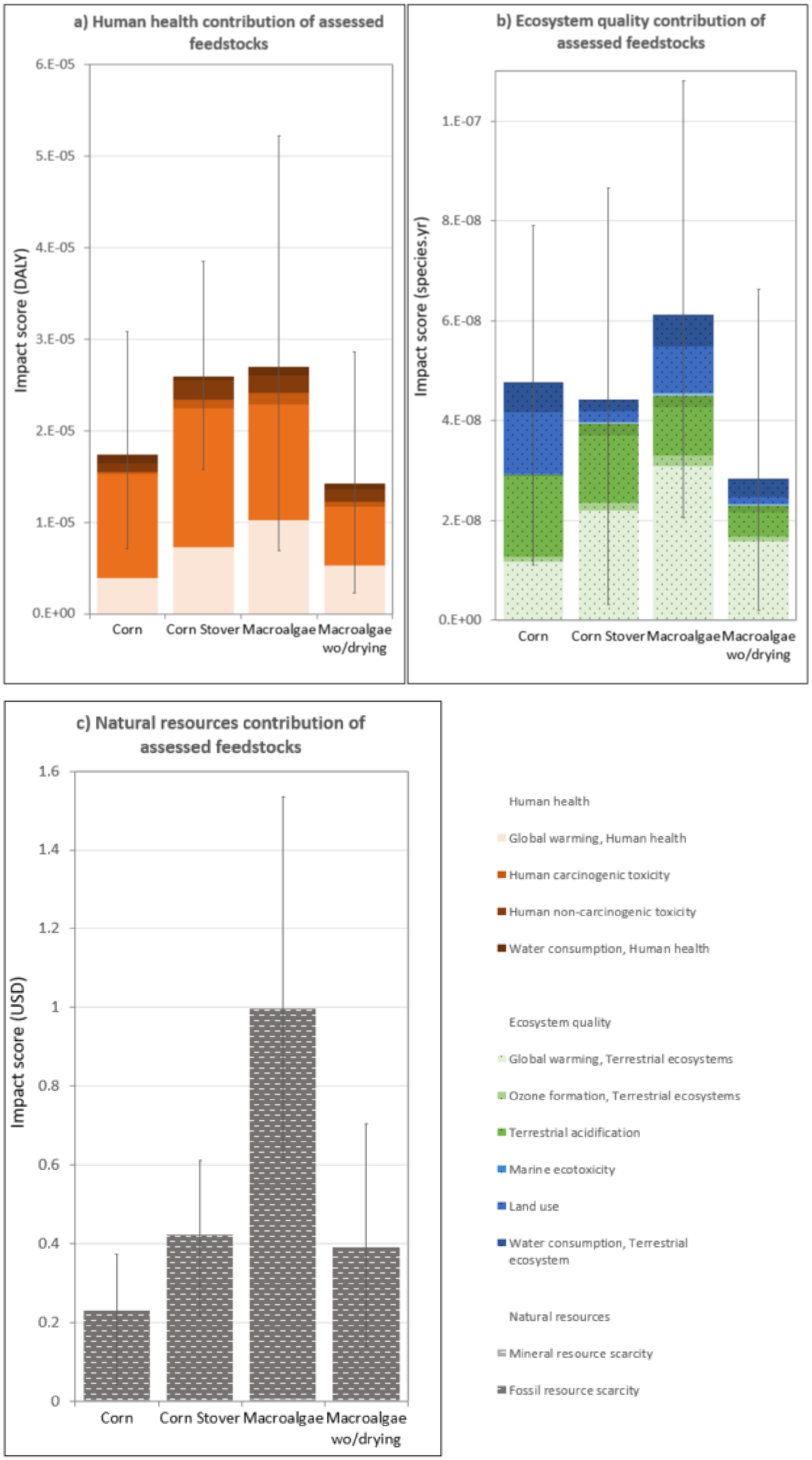


Figure 9a, b, and c. Actual LCA results at damage level including uncertainty ranges for the different generations of feedstock processes. In addition, to assess the optimization potential of not drying the 3rd generation biomass, the figure presents the results and uncertainty range for that scenario. *(From Paper II).*

Another way to assess the optimization potential of LCA from macroalgae is to change the assessed scenarios, as presented in Figure 10.

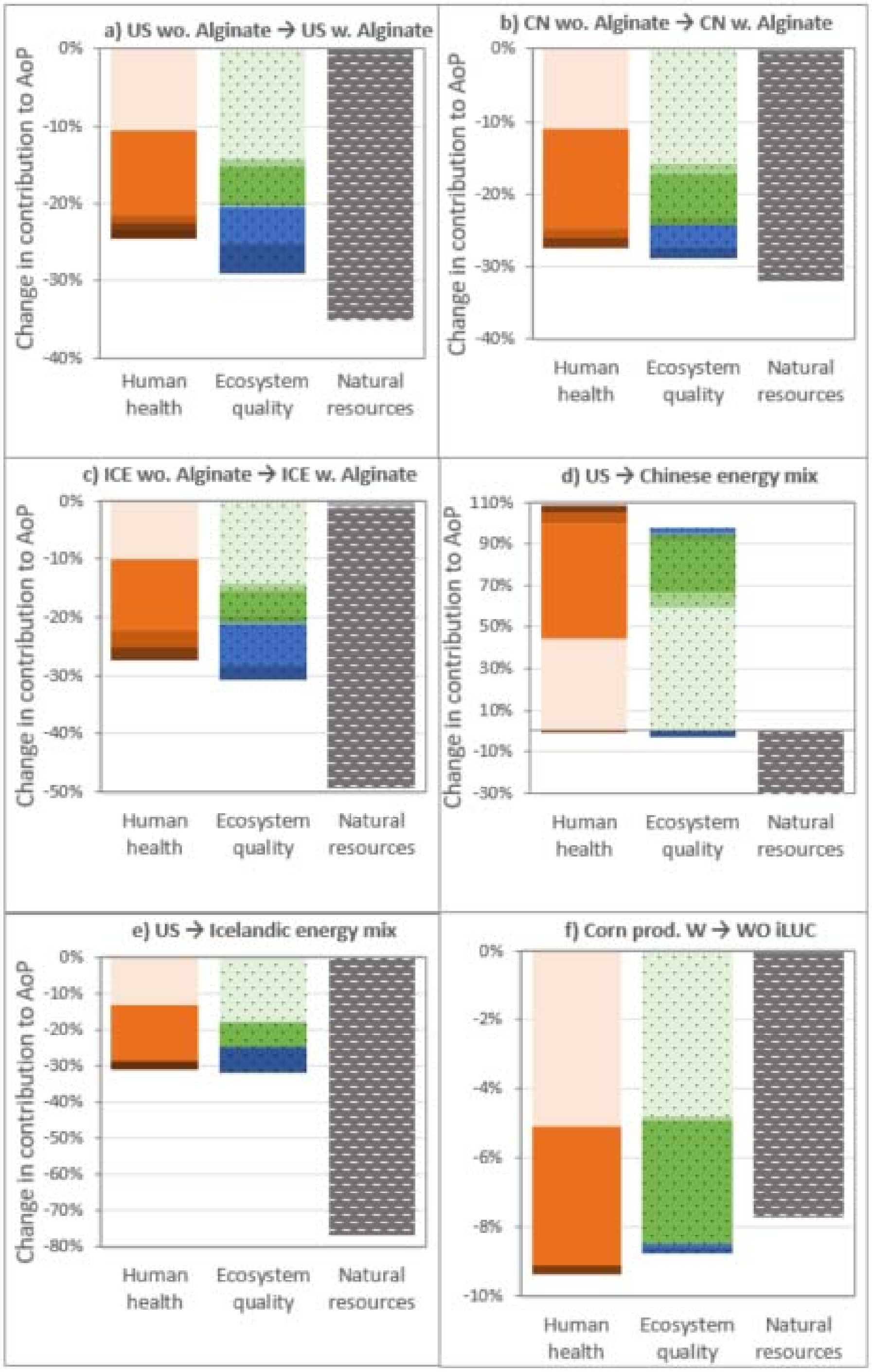


Figure 10 Changes in environmental hotspots at area of protection when assessing the future scenario of fermenting alginate for lactic acid production from macroalgae (7a-7c). Sensitivity scenarios assessing changes in environmental damages when changing geographical to China and Iceland respectively (7d and 7e). Sensitivity scenario showing reduction in impacts when excluding iLUC damages (7f). (*From Paper II*).

When using macroalgae for biorefinery production, only parts of the available carbons are useable for fermentation purposes, namely cellulose, laminaria and mannitol. "Alginate, that accounts for about 30% of the carbohydrates composition of the macroalgae[42] cannot be fermented (and is hence not included in the base case scenario Figure 10c) with current technology. To understand what the environmental gains would be if alginate was fermentable, this scenario was modeled" and the results are presented in Figure 10a. Fermenting alginate would yield an overall reduction of environmental impacts by 38%. "The highest reduction in impacts affecting damages on human health are global warming (15%) and fine particulate matter exposure (19%); both related to the reduction in biomass needed per kg product, resulting in less energy needed for drying of biomass. The lower demand for biomass also drives the reduction in damages on ecosystem quality, where the highest reduction is associated with global warming impacts on terrestrial ecosystems (18%) and land use impacts (9%). For natural resources, fossil resource scarcity is reduced by 38% because of decreased biomass demand."

In addition, increased feedstock yield when fermenting the alginate per FU results in that the country with the more impacting energy mix—in our study this is China (CN)—sees a higher benefit in the reduction of energy-related impacts. This applies to for example global warming impacts on terrestrial ecosystems and terrestrial acidification (Figure 10c).

"Production location also influences the optimization potential of the macroalgae feedstock process. Changing the location of the LA production from USA to CN, we see a drastic increase in damages on human health (108%) and on ecosystem quality (95%). The composition of the energy inputs causes these changes. Damages on natural resources on the other hand are reduced by 30%. The reason for this is that the country-specific ecoinvent processes chosen for modeling the background of the heat-mix production composition in the US cause higher impacts related to fossil resources scarcity than the background processes chosen from ecoinvent to model the CN heat-mix production distribution" (see Figure 10e and f).

"Since the energy inputs have the single most dominating impact on the production process of LA from macroalgae, we evaluated if the results change when the considered energy mix has a different composition. The trend in reduction of damages is only the same for damages to natural resources. Fossil resource scarcity would be reduced by 76,5% in Iceland (ICE) and by 51% in CN. For CN this might come as a surprise, but occur because of lack of specific CN energy processes in the ecoinvent database and therefore average world processes were selected."

“For both human health and ecosystem quality damages, there is an opposite trend for CN and ICE when compared to the base case. For ICE, that gets its energy to 87% from renewable resources[97], we see a reduction in damages to human health (31%) and ecosystem quality (33%), compared to damage increase for human health (130%) and ecosystem quality (99%). USA and CN rely to a great extent on fossil resources in their energy mix.[98,99] Geothermal energy conversion in Iceland emits large quantities of fine particulate matter, which explains the high related reduction presented in Figure 10b. Reduction in damages on ecosystem quality differs on the other hand somewhat as compared to the US and CN. This mostly relates to the fact that hydropower energy conversion demands large areas resulting in the land used savings for ICE.”

There is an opportunity to align LCA input data with data that are used already in TEAs for biochemicals, and if same data/assumptions are used in both, that LCA results can be included as valuable elements in decisions support instead of only relying on TEA to move toward innovation/sustainability. However, how much LCA hotspots trade off against TEA hotspots remains to be further investigated. Nevertheless, our study lays the foundation of using TEA data in LCA assessments to explore optimization potential at a very early stage, in this case when fermentation bacteria are still being modified for optimal performance, and thereby include environmental sustainability as proxy/goal/standard for process optimization for biochemicals. An example is the optimization potential of not needing to dry the macroalgae biomass before fermentation.

The limitations of this work is that it relies on process simulations, using the best available data for building the conceptual process flow diagram followed by choosing potential upstream and downstream process figurations for all three feedstock generations. This is done to simplify the process simulations. In practice, assuming for example the same downstream process might not be feasible, but because applying LCA and TEA combined is an iterative process, changes in the process flow would be built in, in the next iteration immediately reflecting the changes in the results.

The way forward is to quantitatively assess the environmental and economic potential of future biochemicals at an early stage, for target compound selection, to not wait, as generally is the case for applying LCA and TEA, until the scale-up or commercialization stage. The reason is because “this end-of-pipe approach severely limits our capacity for producing bio-based chemicals that are both economically viable and environmentally benign.”[100]

5. A FRAMEWORK TO INCORPORATE ENVIRONMENTAL SUSTAINABILITY IN DECISION MAKING IN BIOTECHNOLOGY

This chapter will present a framework on how we can incorporate environmental sustainability in decision making in the development of biochemicals at an early stage. This framework will help decision makers to decide which future environmentally sustainable biochemicals and associated technologies to develop.

The chapter is divided into four sub-sections. It starts with stating the problem of how target chemicals are selected today. Following is a short state-of-the art section. Then the methods applied for this study are presented, followed by the results and a short discussion. This chapter is linked to Paper III.

Before going into details of how to incorporate environmental sustainability in decision-making in biotechnology, it needs to be stated that the first step of the early stage assessment framework is to select target compounds based on market potential of the biochemical of interest. This step does not include LCA or TEA and builds solely on market analysis identifying future marketable biochemicals. After identification of target chemical, which shows positive market potential, and before any strain optimization takes place, one should do an early stage LCA and TEA to identify technical, economic, and environmental challenges before any time and money has been invested in strain development and scaling up. For the early stage assessment aligning LCA and TEA methodology is needed.

5.1 WHY DO WE NEED TO LOOK BEYOND ECONOMIC ASSESSMENTS?

Since before the early 1980s, the petrochemical industry has applied technical and economic assessments to identify petrochemicals that show increased market potential and to optimize their production processes.[101] In 1981, the same methods were applied to identify the technical and economic potential of production processes for biochemicals.[102] The development of biochemicals is economically and market driven and as presented in Table 1, there have been some success stories demonstrating that biochemicals can substitute, or compete, on the same markets as petrochemicals as drop-in chemicals. Economic viability of biochemicals still needs to be increased to be fully competitive with petrochemicals at market level.[103,104] This problem is compounded by the lack of consistently demonstrated environmental advantages of biochemicals in comparison to petrochemicals, as demonstrated in Chapter 3.

In the transition to a viable bioeconomy, economic improvement cannot be at the expense of the potential increase of environmental problems and vice versa, for biochemical production, if we want them to replace petrochemicals in the long term and even outcompete them. To identify all potential trade-offs between economic and environmental aspects, we need a framework to combine assessing the economic potential of biochemicals with techno-economic assessments, as well as assessing the environmental potential with life cycle assessments in a systematic way. Before such a framework can be built, inconsistencies between TEA and LCA in different assumptions, system boundaries, assessment basis, models, and data, need to be resolved. Once these inconsistencies are identified and resolved, a coherent framework combining TEA and LCA can be developed.

5.2 Short overview of Current application of TEA, LCA, and combined TEA and LCA

When TEA is applied independently applied to biochemicals, as an example demonstrated in the scientific literature, it is used to assess the technical and economic feasibility of a proposed process configurations[e.g.105,106] and for comparing different processes that could be of interest.[e.g.107,108]

When LCA is applied separately to biochemicals it is used to compare the environmental performance of different biochemical production processes.[e.g.15,87]

When presented in the scientific literature, the application of LCA and TEA combined, the methodologies, as an example, are used for optimizing processes[e.g.109] and to assess different process optimizations.[e.g.110,111] Despite literature demonstrating how to use LCA and TEA combined at an early stage of design and process development, the idea of using them as decision support tools for target compound selection, at a stage earlier than process optimization and development, has not been explored. To perform a combined assessment of LCA and TEA possible we need to translate the individual results from both methods to a combined monetary score.

5.3 HOW TO CONSISTENTLY COMBINE AND ALIGN LCA AND TEA FOR COMBINED RESULT INTERPRETATION

Before carrying out any assessment, we need to align the two assessment methods. For this step, I recommend the use of the LCA standardized methodology to provide a basis for both the TEA and the LCA, as presented in the ISO 14040[84] and ISO 14044[85] standards, because the LCA methodology has been ISO standardized to give the methodology extra reliability and robustness, while TEA methodologies have not been standardized.

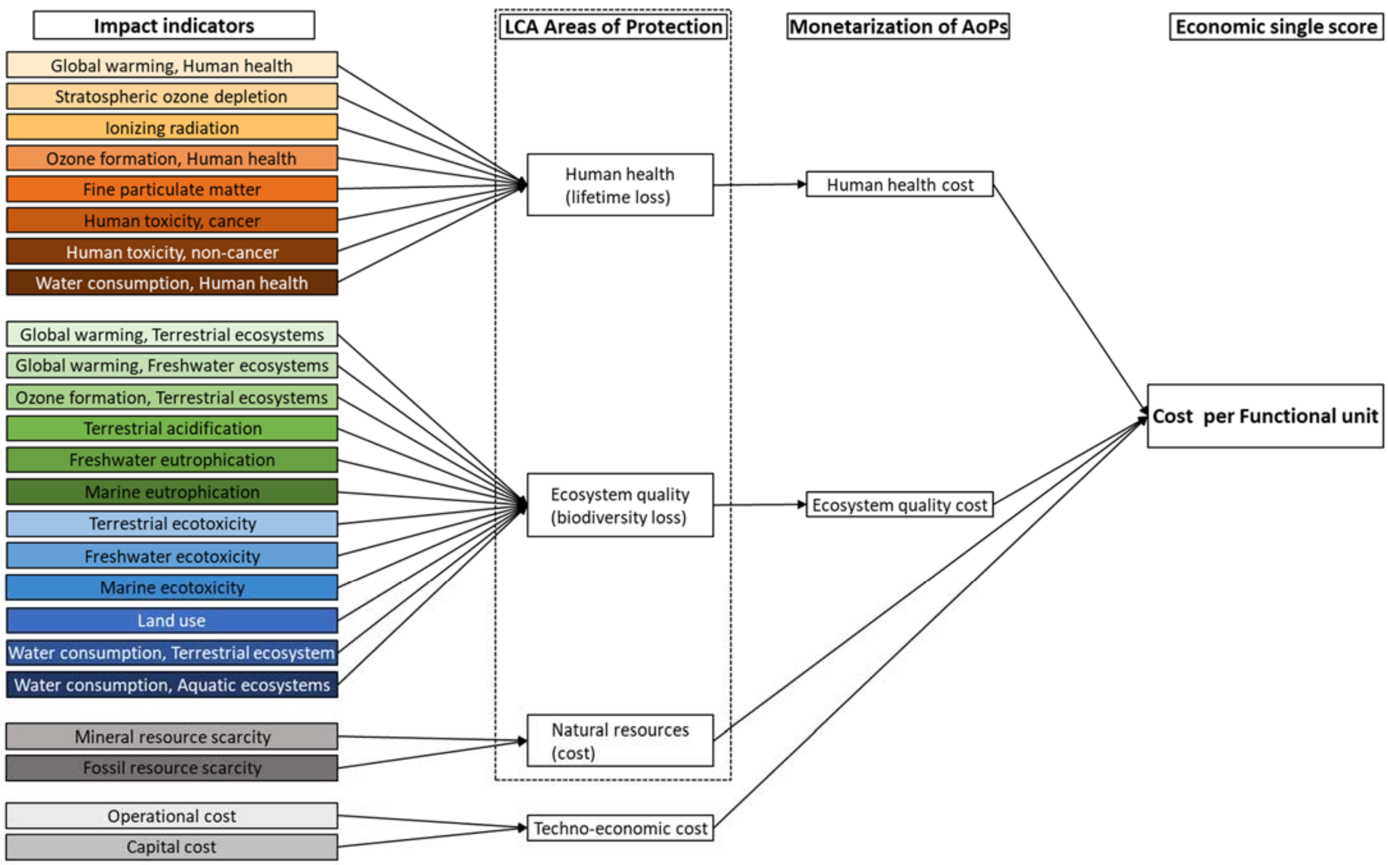


Figure 11 From impact indicators in LCA, to LCA areas of protection (AoP) translated to monetarization of AoPs combined with TEA for an to economic single score. TEA has similar refinement as LCA, but as the focus of the PhD project is on the LCA methodology, the figure is meant to show how the LCA needs to be aligned with the TEA which results are already expressed in monetary cost. *(Adapted version from paper III).*

For an operational assessment framework, alignment of both LCA and TEA practices is needed. First is to decide the goal of the study, second is to define the objectives of the combined studies. This decision will shape the whole assessment structure. Next step is setting the scope, deciding the common functional basis assessed, and defining the product system and boundaries of the study. That includes deciding what is assessed and what is not. This lays the foundation of the harmonized metric to apply the LCA and the TEA together for combined results interpretation, as is presented in Figure 11.

5.3.1 Application and role of Techno-economic assessment in the framework

TEA begins with setting up a conceptual process flow diagram, based on fermentation parameters and the knowhow of by-products formed while producing the target compound as presented in Figure 12. This information must be always backed up by in-house experimental data or must be obtained from the literature. The subsequent step is to identify potential upstream and downstream process configurations. Among the several possible process configurations, one is selected based on separation efficiency and robustness, with varying real feedstock composition and process parameters. Based on these findings, a baseline process model is consolidated, acting as a scaffold, over which several simulations are performed for further optimization. Plant size plays a significant role because the process simulation is based on the plant capacity and the software scales all processes to meet the plant capacity.[77,90,91]

Eventually, the TEA is performed by incorporating the real-world monetary values to the parameters, such as, feedstock, energy utilities, materials and consumables, which are necessary to run the plant efficiently. Appending this analysis is also insights into the long-term economic impacts of producing the assessed biochemical at the given plant capacity.

5.3.2 Application and role of Life cycle assessment in the framework

For securing the consistency of the assessed data within the assessment framework, the inventory from the TEA is the used for modelling the LCA for the biorefinery life cycle stage. The first step required is to adapt the TEA mass flows, by normalizing the inventory based on the functional unit decided for LCA study (see Figure 12). This is necessary because the TEA results are given in mass or energy over time (e.g. per hour, per annum, defining the rate of production and consumption). The most convenient way is to rely on already existing LCA inventories, but for processes for which inventories are not available, the practitioner needs to develop and rely on software tolls to get reasonable estimates for process parameters based on prior experience, encoded into the proprietary databases that come with the software tools. The LCA model is simulated and the results should be translated to damage level results (see Figure 11).

As a next step to combine LCA and TEA under the same decision support framework, we need to be able to combine the results from the two assessment methodologies as demonstrated in Figure 11 with an economic single score that can be understood without in depth knowledge of either LCA or TEA. The figure is an extended version of Figure 8 from section 4.1.3, with the inclusion of the TEA "impact indicators". For the TEA results that is straight forward as the assessment provides monetary values as a result.

The next step needed is to translate the LCA results into an economic single score. That is done by translating the life cycle impacts to the different areas of protection (AoPs) in ReCiPe 2016 where different impact indicators contribute to human health expressed in lifetime lost (DALY), different impact indicators contribute to ecosystem quality expressed in biodiversity loss (species.yr), and natural resources expressed in US dollars (USD).

5.3.3 COMBINED APPLICATION OF TEA AND LCA, AND MONETARIZATION OF ENVIRONMENTAL IMPACTS

This section is directly taken from *Paper III* , reference:
Ögmundarson, Ó., Sukumara, S., Herrgard, M. & Fantke, P. *Combining economic feasibility and environmental sustainability to optimize performance at early stages.* Trends in Biotechnology. To be submitted.

Combining the TEA results and natural resources results from the LCA is easy as they are both assessed using a monetary score. DALYs and species.yr results are on the other hand by default not expressed as monetary values. The next step in the framework therefore requires monetarizing DALYs and species.yr, which allows combining the results into a single economic score, or cost per functional unit. The economic single score then reflects the real cost of producing products and should be used for identifying environmental and monetary hotspots and tradeoffs between these two pillars of sustainability and is demonstrated in Figure 11 and Figure 12.

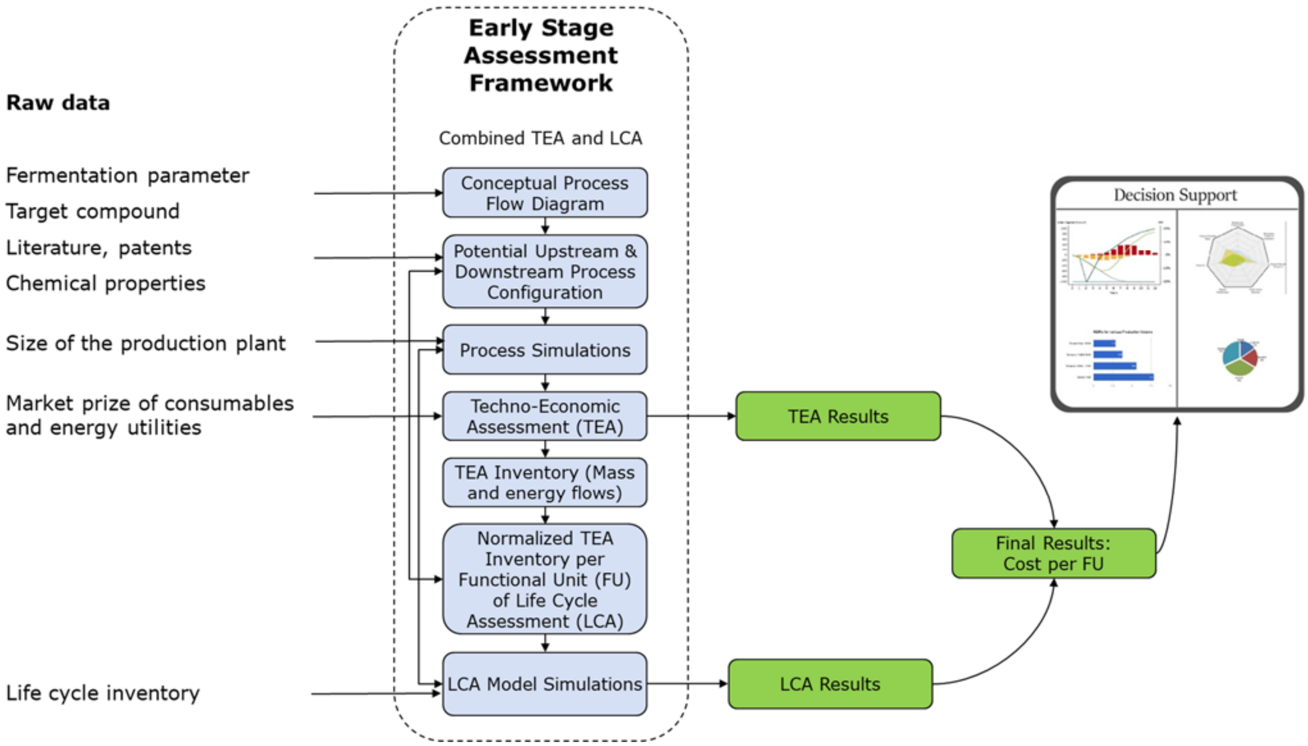


Figure 12 Early stage assessment framework for applying LCA and TEA as a decision support tool in biotechnology. *(From Paper III).*

The publication "Evaluating the monetary values of greenhouse gases emissions in Life Cycle Impact Assessment" by Dong et al. (2018)[112] gives a good overview of the monetizing values that have been presented in the scientific literature, and based on that I then calculated the average monetary values for DALYs, which is 100.000$. For species.yr I used the value provided by Weidema (2009)[113], which is 65.000$.

Monetarization of environmental impacts is not new[113–115], but to my knowledge, it has never been adapted and presented before via an economic single score to combine LCA and TEA results. By doing this, for the first time, I exploit the potential of both LCA and TEA to identify tradeoffs between the results of both methodologies, which makes it possible to incorporate them as a decision support tool in biotechnology.

5.4 THE RESULTS FROM THE EARLY STAGE ASSESSMENT FRAMEWORK, COMBINED LCA AND TEA

This section is directly taken from *Paper III* , reference:

Ögmundarson, Ó., Sukumara, S., Herrgard, M. & Fantke, P. *Combining economic feasibility and environmental sustainability to optimize performance at early stages.* Trends in Biotechnology. To be submitted.

The results presented in this section are based on the LCA results presented in Chapter 0. The functional unit of the study was "the production and use of 1 kg of lactic acid, with 99.9% purity, for household packaging application in the United States". For the LCA study the system boundaries were from cradle-to-grave, but the TEA assessed the system from cradle-to-gate (polymerization included). This is because it was not possible to get monetary values for the waste scenario for the TEA as this is highly dependent on country/region specific waste handling processes and costs are highly dependent on regulations and societal norms[116].

Table 6 Monetarization of areas of protection and economic single score per functional unit, for 1 kg of PLA, in $. Monetarization values for every DALY is 100.000$ based on average value from Dong et al.[112] and species.yr is 65.000$ based on Weidema[113]. TEA results for the 1st and 2nd generation feedstock processes were done by Sumesh Sukumara[93] and Elena Tomás Grasa did the 3rd generation feedstock process simulation.[92,93] *(From Paper III).*

1st generation LCA results			**TEA results**		**Total economic single score**	
DALY	1.74	$				
Species.year	0.003	$				
USD	0.23	$				
Total	**1.97**	$ Cost per functional unit	**3**	$ Cost per functional unit	**4.97**	$ Cost per functional unit
2nd generation LCA results			**TEA results**			
DALY	2.59	$				
Species.year	0.003	$				
USD	0.42	$				
Total	**3.02**	$ Cost per functional unit	**2.14**	$ Cost per functional unit	**5.16**	$ Cost per functional unit
3rd generation LCA results			**TEA results**			
DALY	2.70	$				
Species.year	0.004	$				
USD	1.00	$				
Total	**3.70**	$ Cost per functional unit	**4.5**	$ Cost per functional unit	**8.20**	$ Cost per functional unit
3rd generation LCA results without drying			**TEA results**			
DALY	1.42	$				
Species.year	0.002	$				
USD	0.003	$				
Total	**1.43**	$ Cost per functional unit	**4.21**	$ Cost per functional unit	**5.64**	$ Cost per functional unit

When looking at the TEA results for LA from corn (1st generation) and comparing them to the LA from corn stover (2nd generation), the monetary cost of the 2nd generation is only higher by 2%. In spite of lower overall yield, the feedstock cost for the 2nd generation process is one fourth compared to that of the 1st generation. This calculation highly relies on the monetary value assigned to a unit of corn stover[117] which will increase significantly with the size of the plant due to the supply chain dynamics and low fraction of fermentable sugars present in 2nd generation feedstock as demonstrated in Sukumara et al 2014.[118] Also, despite the 2nd generation process is a factor of 3.6 more energy demanding compared to the 1st generation, it does not show in the TEA results, except by 0.03$ per kg LA. This is despite the currently lower level of optimization of the 2nd generation process and lower TRL, and despite the physical composition of the 2nd generation biomass (more fiber rich) requiring a more intense separation process demanding higher chemical concentrations and more intensive energy use.

As demonstrated in Table 6, the economic single score of the LCA results are 25% higher for the 2nd generation process than for the 1st generation process. This is due to that the energy use in the 2nd generation feedstock process is 3.6 times higher, compared to the 1st generation feedstock processes. The increased energy consumption is e.g. because of separation of the fiber rich biomass that in this study is done by steam explosion, which is not needed in the pretreatment of the 1st generation biomass. The higher energy use is mostly visible for the DALY and US Dollars (USD) results because the modeled energy is 88%[98] from fossil resources which environmental impacts contribute mostly to human health (DALYs) and environmental resource use (USD).

When analyzing the results in further detail, we can see that for both the 1st and 2nd generation processes, environmental impacts are the highest expressed in DALYs. For the 1st generation the highest impacts come from indirect land use change (iLUC). This is related to increased global warming impacts due to increased demand for arable land[78]. The highest environmental impacts for the 2nd generation process are also global warming impacts, but from a different source, namely high energy demand of the biorefinery stage. This is due to intensive energy use in the pretreatment stage.

In the future, 3rd generation biomass could become a viable biomass, and with the framework presented here, we can assess the potential of producing LA from the biomass and analyze which environmental hotspots could be optimized to make the process more compatible, compared to the 1st generation feedstock process.

First, by looking at the TEA results for the 3rd generation process, the feedstock cost accounts for almost 50% of the total cost. Energy utilities stand for about 20% of the total cost with drying, and the process without drying has under 10% lower total cost than the process with drying. This is despite the fact that not drying the biomass results in a cut down on steam use of more than 100 MJ per kg product. The reason for the low effect on the total cost per functional unit is that steam bears a low price, and despite high amounts used the steam therefore has a minimal impact.

On the other hand, when analyzing the reduction in steam use in the LCA results, $ cost per functional unit, the benefits of reducing steam usage and thereby lowering the environmental impacts of 1 kg LA from macroalgae by more than 40% becomes evident. With the economic single score based on both LCA and TEA results for the macroalgae processes, we identify tradeoffs between drying and not drying the biomass that only assessing the TEA would not have revealed stating the benefits of incorporating environmental and economic aspects within one framework. This framework makes it possible to identify tradeoffs that only assessing either the economic or environmental aspects would not identify, which could lead to unnecessary environmental impacts that could have been avoided from the earliest stages of development of future biochemical production processes.

5.4.1 UNCERTAINTY AND INTERPRETATION OF RESULTS

Depending on the considered data sources, uncertainty of the input parameters used in Life Cycle Inventory (LCI) can have considerable effects on the interpretation of the results. That includes uncertainties of the life cycle inventories used for the foreground processes, as demonstrated e.g. in Owsianiak et al.[119] For my LCA study, I followed their procedure, estimating uncertainty factors for the respective process by using the Pedigree matrix approach.[120] Uncertainties associated with characterization results as outcome of the Life Cycle Impact Assessment in my study are not included, since uncertainty is currently not reported along with characterization factors in any existing LCIA method. Acknowledging that characterization factors can come with considerable uncertainty associated with various aspects in the modeling of the various impact pathways[121], this is a current gap, which needs to be addressed in future research.

In the present study, LCI-related data uncertainty is obtained as described in the following. Input parameter related squared geometric standard deviations, GSD_x^2, which are used for log-normally distributed input parameters x, are obtained from a combination of uncertainty factors for base uncertainty of x, U_{base}, and for uncertainty associated with different quality criteria c, U_c, according to the Pedigree matrix approach (Equation 1):

$$\mathrm{GSD}_x^2 = \exp\left(\sqrt{(\ln U_{\text{base}})^2 + \sum_{c=1}^{n}(\ln U_c)^2}\right)$$
(Equation 1)

Such uncertainty factors are assigned if the used process, e.g. from the ecoinvent LCI database, does not carry any pre-assigned uncertainty value. For calculating the uncertainty value, inputs and outputs of each life cycle stage were evaluated based on $n = 5$ criteria c (reliability, completeness, temporal correlation, geographical correlation, and further technological correlation), each assigning an uncertainty factor expressing the quality of the input parameter, which are finally combined to derive the uncertainty of the resulting LCI flow.

Such uncertainty factors are assigned if the used process, e.g. from the ecoinvent LCI database, does not carry any pre-assigned uncertainty value. For calculating the uncertainty value, inputs and outputs of each life cycle stage were evaluated based on n = 5 criteria c (reliability, completeness, temporal correlation, geographical correlation, and further technological correlation), each assigning an uncertainty factor expressing the quality of the input parameter, which are finally combined to derive the uncertainty of the resulting LCI flow.

Based on Equation 1 assigned a separate uncertainty factor for each of the three biorefinery feedstock processes as demonstrated in Table 7. The same procedure was used for the biomass feedstock processes. For each feedstock generation, I calculated separate uncertainty factors based on the differences in underlying available data.

Table 7 Uncertainty factors and geometric standard deviation for each of the biorefinery life cycle stages for calculating the uncertainty. *(From supplementary information Paper II).*

Biorefinery								
1st generation	Flows and emissions	U1	U2	U3	U4	U5	Ub	Geometric standard deviation
		1.05	1.02	1.03	1.001	1.05	1.05	1.096
2nd generation	Flows and emissions	U1	U2	U3	U4	U5	Ub	Geometric standard deviation
		1.1	1.05	1.03	1.001	1.5	1.05	1.527
3rd generation	Flows and emissions	U1	U2	U3	U4	U5	Ub	Geometric standard deviation
		1.2	1.1	1.03	1.001	2	1.05	2.065

For the life cycle stage 'polymerization', I calculated a generic uncertainty factor across all considered feedstock generations; and the same applies to the 'end-of-life treatment of waste' stage. The reason for this is that these two life cycle stages were not changed for each of the feedstock generations processes.

The squared geometric standard deviation was then assigned to each of the processes in SimaPro and the uncertainty ranges were calculated with Monte Carlo simulations, using 10.000 iterations of each cradle-to-grave scenario. All considered parameters were included in the uncertainty analysis.

The learnings that we can draw from the uncertainty results presented in this PhD study are that the overlap of uncertainties results in that with the given assumptions and data quality, no definitive conclusion can be made about possible optimization potentials for the different feedstock generations. However, trends can be observed, in particular that the performances of the 3rd generation lactic acid production (i.e. using macroalgae as feedstock) can be dramatically improved relative to the other 2 generations (i.e. corn and corn stover) if the drying process is drastically optimized or removed. This requires that the microorganisms used for fermentation need to be capable of breaking down the feedstock biomass without the pre-treatment (i.e. drying) that otherwise would make the sugars in the biomass more easily available. Simulations without drying show that macroalgae scenarios perform better than the other 2 generations, with the exception of natural resources, where the same level of impacts as for the 2nd generation can be reached. This can be observed in Figure 9, showing that not drying the biomass can reduce impacts around 50% across areas of protection.

To demonstrate the difference in uncertainty between the assessed feedstock processes,

Figure 13, Figure 14, Figure 15, and Figure 16 show the correlation of uncertainty between the assessed processes at midpoint level using ReCiPe 2016 as LCIA method. When at least 95% of all 10.000 Monte Carlo runs were in favor of a feedstock process, statistical significance was assumed.

Figure 13 A) LA from corn, B) LA from corn-stover. When at least 95% of results favor either process, results are assumed statistically significant.

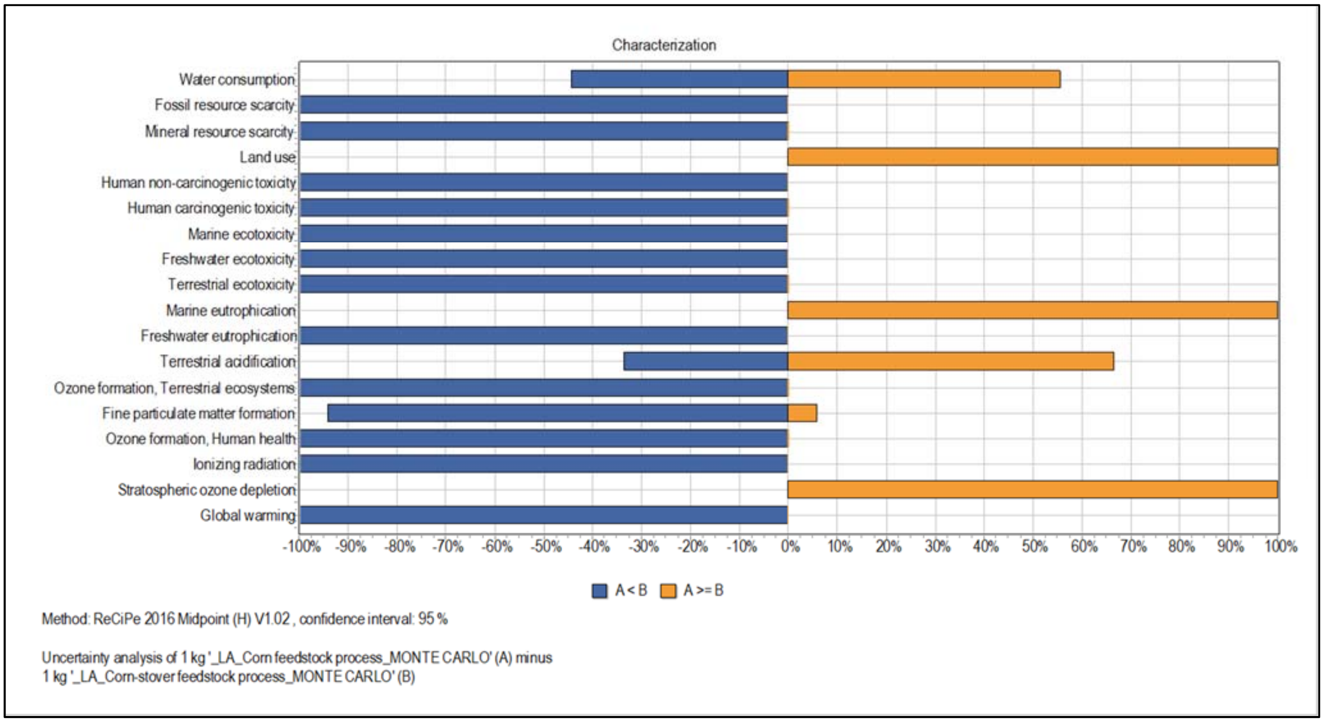
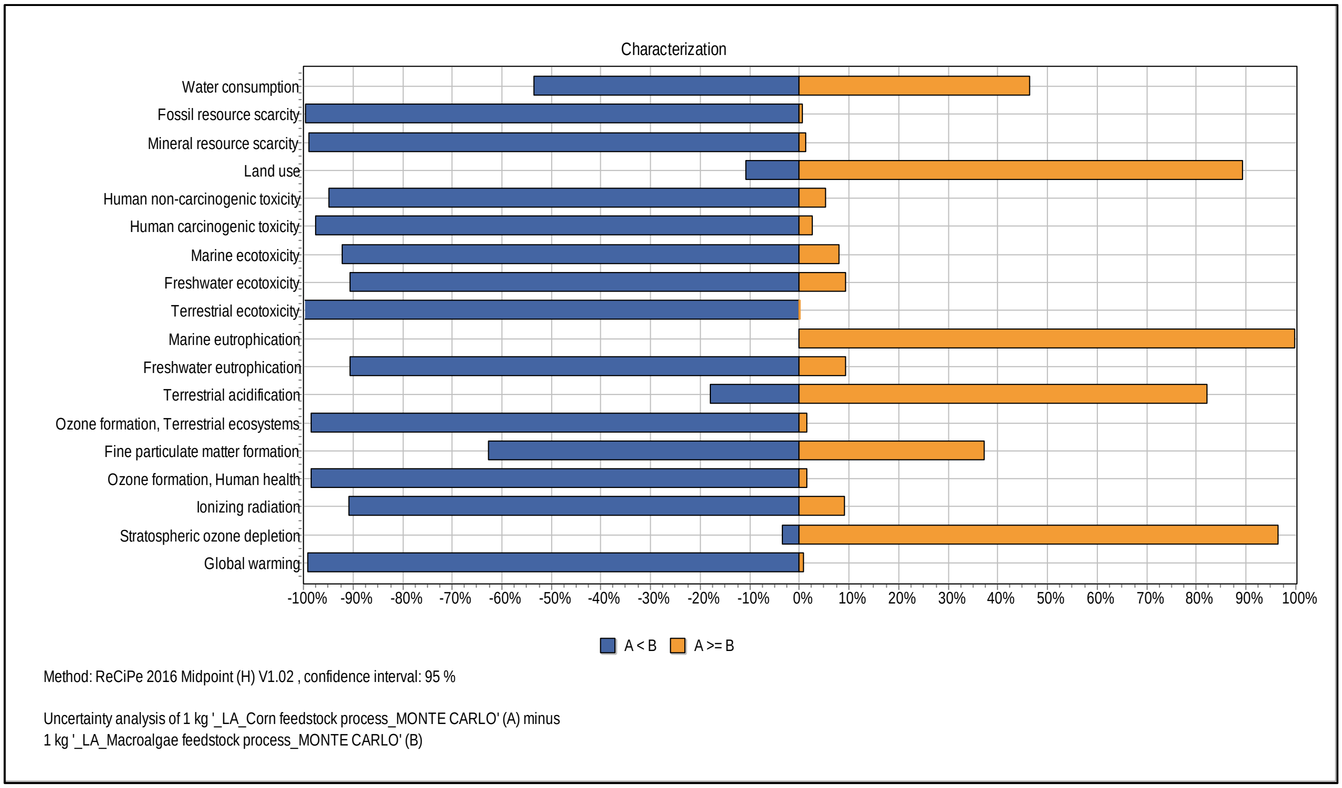


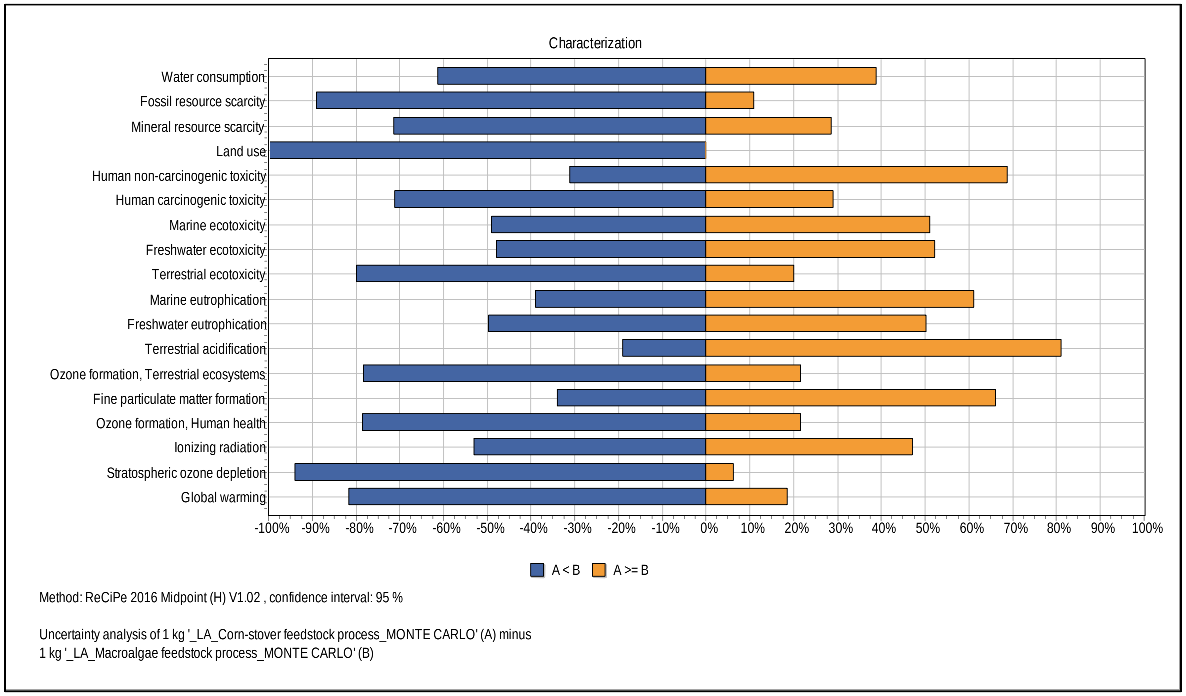
Figure 13 shows that the results of assessed impact categories, when comparing the corn to the corn-stover feedstock process, the results are in favor of the corn process. The impact categories where the corn feedstock process does have at least 95% of the Monte Carlo runs in favor is for Land use, Marine eutrophication, and Stratospheric ozone formation—these are all impact categories affected by the growing of the corn biomass.

Figure 14 A) LA from corn, B) LA from macroalgae. When at least 95% of results favor either process, results are assumed statistically significant.



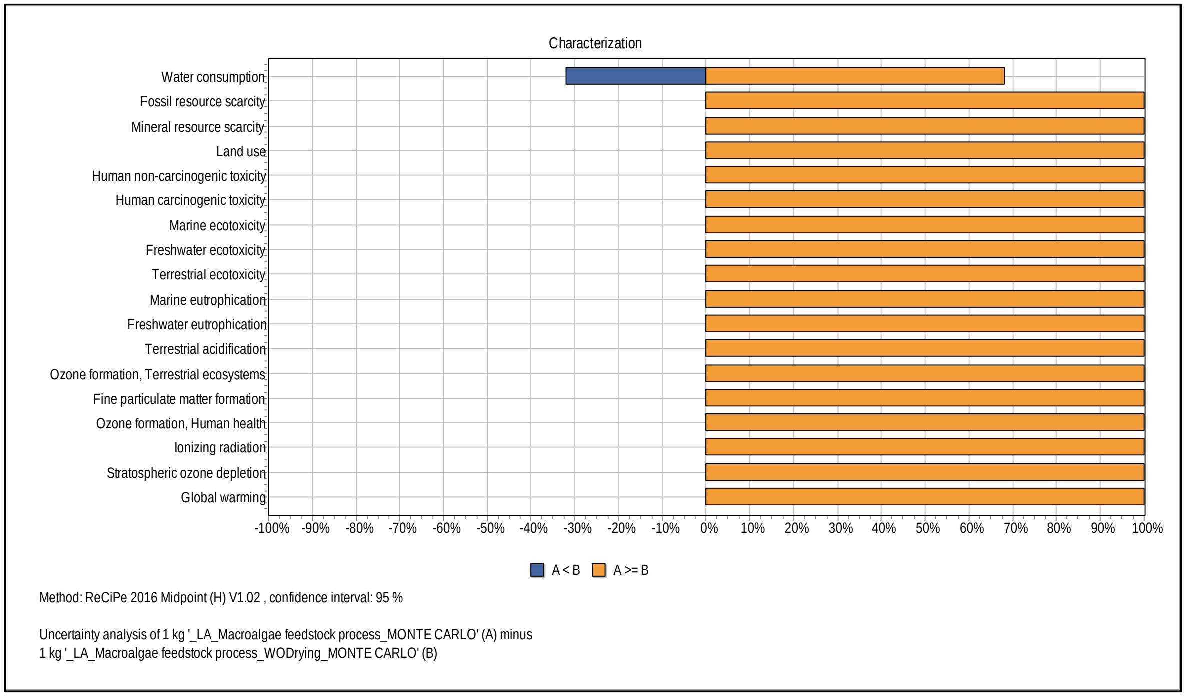
When looking at the comparison between lactic acid produced from corn process and lactic acid produced from macroalgae process, Figure 14 shows that there are more impact categories favorable for the corn process than for the macroalgae process. It is only Marine eutrophication, and Stratospheric ozone depletion that are in favor of the macroalgae process. These impact categories are dominated by the growing of the corn feedstock biomass as evaluated underlying LCI process.

Figure 15 A) LA from corn-stover, B) LA from macroalgae. When at least 95% of results favor either process, results are assumed statistically significant.



When comparing the corn-stover to the macroalgae feedstock process, Figure 15 shows that only Land use and Stratospheric ozone depletion show statistical significance favoring one of the two compared feedstock generations, namely the corn-stover feedstock process. All other impact categories show no significant favor to either of the two compared feedstock processes. This is mainly because both feedstock processes are highly energy intensive, causing the uncertainty factors to assign high uncertainty to both feedstock processes.

Figure 16 A) LA from macroalgae, B) LA from macroalgae without drying. When at least 95% of results favour either process, results are assumed statistically significant.



LA production from Macroalgae without drying is favourable over LA production from macroalgae including drying, for all impact categories except water consumption. Despite reducing water consumption by 57% when not drying the biomass, washing is still required demanding large quantities of water resulting in high uncertainty of both feedstock processes.

These results demonstrate that considering uncertainty in LCA results helps understanding where differences between feedstock generation systems are significant and where LCI data improvement and refinement efforts should be focused.

When looking at the different results presented in Table 6, keeping in mind the large uncertainties it is hard to state if the results of the three different feedstock processes are significantly different. In this context, it is relevant to state the need to increase data quality to get more precise results (to reduce uncertainty) and to work on reducing the highest contributors to overall impacts in 2nd and 3rd generation, in order to become competitive with higher TRL processes in 1st generation. Still, based on the results, the 3rd generation feedstock process without drying, already becomes competitive with the 1st generation feedstock process.

To summarize, developing data and models both for LCA and TEA, that build on the same system boundaries, are expressed in the same metrics and assumptions, and are consistent in terms of a single mass balance will facilitate a combined and consistent assessment framework. With such a combined LCA-TEA framework, we can identify both environmental, and techno-economic hotspots and tradeoffs between the two, that will ultimately help taking decisions on which biochemicals should be developed and where to concentrate process optimization for boosting the overall optimal performance of future biotechnologies. For the biotechnology industry, this framework opens up for including environmental sustainability to continue increasing sustainability-driven innovation, i.e. not only to do the right thing (i.e. producing bio-based), but also to do it the right way (i.e. producing bio-based with minimal environmental as well as economic impacts).

6. CONCLUSIONS AND RECOMMENDATIONS FOR FUTURE RESEARCH

6.1 GENERAL CONCLUSIONS

The main goal of this PhD project was to address the overarching question, "How can the environmental sustainability of bio-based chemicals applied in biotechnologies, including bio-polymers, be consistently and comprehensively quantified, optimized, and ultimately included in decisions related to biochemical production?"

By addressing this question, the present thesis supports an important and desirable global trend that in the future, we will have to reach the point when environmental sustainability is among the main drivers of countries' and companies' innovation, and not only economic growth. This would result in the application of LCA as an effective tool supporting process optimization based on a comprehensive set of indicators and covering the entire life cycle, instead of using it as a marketing tool for selected applications based on a limited set of indicators and life cycle stages. We have not yet reached this point, but environmental sustainability is getting more and more attention despite setbacks, e.g. in the form of political leaders that deny changes in our earth's climate.

The fact that environmental consciousness is rising calls for action in all layers of society, from the public to companies and governments. On the company level, this means that we need to incorporate environmental sustainability in decision making, at a very early stage. That is to optimize our future products, if we actually want to reach the point of making environmental sustainability a driver and not only a drag-along thing only a few people take seriously, and also to live up the expectations of future buyers toward sustainability claims of our products.

Incorporating sustainability does not only include future or possible reductions in global warming impacts, but requires assessing all possible environmental impacts caused by a product or process. It also requires that all life cycle stages are assessed, despite not being always "under control" of the assessors. That is because if we do not look at environmental sustainability holistically, we might miss the benefits or face increased impacts of our actions further up or down in the value chain that could influence our actions at the time of the assessment. Then the question arises, what does it require for coupling environmental sustainability and TEA as a decision support tool in for example the chemical industry?

When I started looking into this, it became evident that the chemical industry and scientists researching chemicals do apply techno-economic assessments at an early stage to identify economic and technical feasibility of new products and processes, and LCA, to some extent. Scientific literature, though, showed that when applied together it was done mostly to assess the potential benefits that could be gained from innovative technologies when compared with existing ones. The combined application of both methodologies has not been used as a decision support tool when dealing with the question if resources should be invested in, in the process of developing a new chemical further, based on TEA and LCA results, right after proof of concept. When doing so, it can lead to tremendous gains for society as a whole, because by optimizing future products at an early stage, we can optimize the process for future environmental emissions, combined with securing the highest return on investment assessed with the TEA. Methodological overlaps between LCA and TEA requires practitioners to adapt certain steps, like build their models within the same boundary, and when exchanging the data, the TEA data needs for example to be normalized per functional unit to incorporate it in the LCA.

In my opinion, based on the experience gained by working with TEA and LCA data and methods, arguments for not doing both assessments in parallel are weak, precisely because of the gains acquired by the holistic optimization framework presented in this thesis. By consistently combining LCA and TEA in a common decision support framework, we can effectively consider both environmental and economic sustainability at the same time to optimize the overall performance of existing and newly developed biochemical production systems. With such a framework, the biochemicals industry would have a tool at hand, based on which actual sustainability can be reached and related claims be scientifically justified.

6.2 CONCLUSIVE REMARKS ON THE STATED THESIS OBJECTIVES AND RESEARCH QUESTIONS

To address the main thesis question, three objectives were defined, each with its own specific research questions. Conclusions for each of the objectives are presented in the following, detailing how the specific research questions have been answered.

Objective 1 of the thesis was to identify the environmental performance of selected biochemicals that have been identified as promising substitutes for petrochemicals. Main research questions for this objective were: (1) What are the main conclusions on environmental sustainability found in published LCA studies on commercialized bio-based chemicals? (2) Which are the main methodological choices to make when assessing environmental sustainability of bio-based chemicals? (3) How can we improve the use of LCA for bio-based chemicals, to help striving towards a viable and sustainable future for the biochemical industry, also considering the role of public perception?

In conclusion, I found that the environmental sustainability claims for the chosen group of chemicals are questionable because, a) they are generally based on results of a few number of publicly available studies, b) the LCAs followed in the considered studies do not consistently assess the whole life cycle of the products possibly resulting in that burden shifting between life cycle stages are overseen, and c) the assessed studies do not consistently assess all relevant impact categories. All assess global warming, but other relevant impact categories like ecotoxicity and eutrophication are not covered in the same extent.

For assessing the environmental sustainability of bio-based chemicals, LCA is the most suitable methodology, to date. That is both for comparison studies that compare products to back up sustainability claims, and also when the intention is to apply LCA as a process optimization tool, assessing each stage of products life cycle in given detail, with an environmentally sustainable end in mind. To do so, the LCA needs to assess the whole life cycle of the product to make sure possible burden shifting between life cycle stages can be identified and optimized for. An example can be plastic formulation affecting the end-of-life treatment of the product.

Another methodological choice is to assess (at least) the most relevant impact categories. As most relevant impact categories, land use, indirect land use change, water use, eutrophication (due to fertilizer use), and ecotoxicity (due to pesticide use) during feedstock production, and energy and water use in biorefineries, have been identified. When assessing end-of-life scenarios, it is furthermore necessary to include toxicity-related impacts, including ecotoxicity and human toxicity related to potential emissions from landfills and potential environmental savings when recycling the wasted products.

Adherence to existing standards is necessary, to secure that assessments are done in the right way making them comparable, which sometimes can be hard when authors deflect from the given standards.

Objective 2 of the thesis was to demonstrate with an LCA how to systematically identify and increase sustainability of biochemicals. Main research questions for this objective were: (1) How can we consistently define life cycles of biochemical product systems across bio-feedstock generations, focusing as an illustrative example on lactic acid as an important building block chemical? (2) How can we characterize the environmental performance of lactic acid production systems with a full life cycle assessment? (3) How to discuss related environmental hotspots and their potential drivers? That includes showing how hotspot results can be used to inform technology system design, identify optimization potential of future processes, and operationalize decision support.

In conclusion, when assessing environmental performance of biochemicals produced from different feedstock generations, differences in TRL of the bio-feedstock generations, associated differences in impact hotspots for each generation, and characteristics of different locations need to be accounted for. Considering the TRL is relevant, because chemical production from 1st generation feedstocks has a higher TRL than chemicals production from 2nd and even 3rd generation. To even out the differences in TRLs between the feedstock generations, for each of the processes a conceptual process flow diagram model is needed in techno-economic assessments. The production capacity of all feedstock processes needs to be set to the same level for simulating the processes. This allows to create the necessary but often missing inventories for LCA practitioners assessing biorefinery processes. The available inventory then makes it possible to apply LCA to identify different hotspots across feedstock generations. Both in the TEA, and LCA, the uncertainty of the mass- and energy-flow data needs to be assessed and incorporated in the results, affecting their interpretation.

I have demonstrated that applying hotspot analysis to identify possible optimization potential across feedstock generations, using lactic acid production as example, is an effective way to characterize the environmental performance of biochemical production systems. At midpoint level, we can identify tradeoffs between life cycle stages, concluding how important it is to include all of them when doing an LCA to avoid that optimizing on one stage of the life cycle does not affect the environmental performance of other life cycle stages. While the environmental performance from the first generation (highest TRL) bio-feedstock seems to be best, my results indicate well that focusing on a single process (namely drying of biomass) can bring even the third generation (lowest TRL) system in the same performance range.

In the discussion of relevant hotspots it is very important, especially for energy intensive processes like biochemical production, to demonstrate the possible optimization potential related to the energy sources and geographical specificities. Therefore it is highly recommended to take production location into account in any future LCA assessments of biochemicals to demonstrate the necessity to consider energy sources, preferably non-fossil based, to avoid making future biorefineries as dependent on fossil based resources as the petrochemical industry.

Objective 3 of this thesis was to design an operational framework to consistently integrate both environmental and economic performance in the design and optimization of new biochemicals. Main research question for this objective was: How can we systematically, in an operational framework, integrate life cycle assessments and techno-economic assessment results consistently from an early stage, as decision support methodologies, with environmental- and economic sustainability of future biochemicals in mind?

In conclusion, integrating LCA and TEA requires the alignment of methodologies of the two methods. Because the LCA method has been ISO standardized and the TEA not, I recommend to follow the LCA standards in the fundamental structure of the framework. This is to give the results from the combined application of both methods extra reliability and robustness. When following the ISO standards 14040 and 14044, the first thing to do is decide the goal of the study, second is to define the objectives of the combined studies. This decision will shape the whole assessment structure. For a consistent combination, the setting the scope, deciding the common functional basis assessed, and defining the product system and boundaries of the study need to be aligned in both LCA and TEA. This lays the foundation for using harmonized metrics and for combined results interpretation. This requires, as demonstrated in this thesis, to bring all indicators from both methods to the same unit, for which monetization is suitable. Monetary units are something people can relate to, making it easier to convey the message of environmental sustainability, to those who are not experts or have little knowledge in the field of life cycle assessments. It also enables to identify trade-offs between the results of LCA and TEA, like in the case of environmental costs of high energy use and low monetary costs of the steam used when drying the alginate biomass.

With a combined LCA-TEA framework, we can see how changes on in process design or use of alternative consumables or energy utilities affect the environmental performance of the product, making it possible to optimize it simultaneously with respect to both environmental sustainability as well as technical and economic feasibility. This ultimately helps to lead to decisions at an early stage in product and process design, where we can identify the most environmentally sustainable product, without jeopardizing the economic drivers of a viable bioeconomy.

6.3 RECOMMENDATIONS FOR FUTURE RESEARCH

During the process of the PhD period that this thesis collects in one document, numerous future research requirements were identified, and are listed below.

Having developed the integrated decision support framework in Chapter 5, the next step is to identify the possible things that need to be adapted to make the framework more streamlined for application in a working environment. This can only be done by routinely applying it on biochemicals under development. Applicability of the framework to other chemical types than commodity chemicals (like lactic acid) needs to be tested.

The framework has been tested on a single product biorefinery setup. For applying it on biorefinery setups like the cascading biorefinery,[47] with up to multiple product outputs, also needs to be tested and adapted as needed.

In this PhD project, the framework built is based on two quantitative methodologies relying on process simulations, namely LCA and TEA. To advance the framework there is need to be able to integrate data from other research fields. That requires that data from for example fermentation, downstream processes, pre-treatment, market analysis (price supply and demand), and physical property data. needs to be aligned within the LCA/TEA framework. How to standardize the data is the first challenge, and calls for developing the framework further, increasing the frameworks efficiency and accuracy to decreases uncertainty of results.

Depending on the biochemical produced, life cycle stages like downstream processing (purification) can be environmentally impactful. For example for bioplastic applications, purity of building blocks needs to be 99.9% because traditionally this is easiest way to convert the chemicals to plastics. It would be relevant to look into the trade-offs between lower purity of the building block and additives/stabilizers needed to possibly compensate for lower purity.

Bioremediation of macroalgae and assigning economic values to ecosystem services of macroalgae by applying ecological economics is required to be integrated in the decision support framework. This is an aspect not considered today, but would help assign, if any, the external cost/benefits of using macroalgae as a feedstock for biorefineries. This could be integrated with assessing the socio-economic impacts of utilizing alternative feedstocks' for biochemical production, locally and globally.

Life Cycle Impact Assessment (LCIA) uncertainties are currently not usually included in uncertainty calculations of LCA study results. However, LCIA related uncertainty should be reported along with characterization factors in any existing LCIA method, in order to derive information on how this uncertainty would affect the overall uncertainty of LCA studies on biochemicals. It would be relevant to see if and how uncertainties of LCIA would affect the results of LCAs of biochemicals, because the different impact categories do not all share the same types and magnitudes of uncertainty.

Overall, the presented thesis provides a valuable starting point for assessing the environmental sustainability of biochemical production systems and for integrating environmental sustainability into the early-stage decision process for designing and developing future biochemicals.

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PAPERS

PAPER I

Assessing environmental sustainability of bio-based chemicals: State and challenges

Ólafur Ögmundarson[1,2], Markus J. Herrgård[1], Jochen Förster[1,#], Michael Z. Hauschild[2] & Peter Fantke[2*]

[1] The Novo Nordisk Foundation Center for Biosustainability, Technical University of Denmark, Kgs. Lyngby, Denmark

[2] Division for Quantitative Sustainability Assessment, Department of Management Engineering, Technical University of Denmark, Kgs. Lyngby, Denmark

[#] Current Address Carlsberg Research Laboratory, Carlsberg A/S, Copenhagen, Denmark

*Corresponding author: Address: Diplomvej 371/214, 2800 Kgs. Lyngby, Denmark, Tel.: +45 45254452, fax: +45 45933435. E-mail: pefan@dtu.dk

Abstract

Using renewable resources for producing biochemicals and related products is a key driver for moving the global sustainability agenda forward. Life cycle assessment (LCA) is a standardized tool for quantitatively assessing environmental sustainability of products along their life cycles. We analyzed the landscape of LCA studies applied to currently commercialized commodity biochemicals, namely lactic acid, succinic acid, 1,3-propanediol, 1,4-butanediol, and 1,5-pentanediamine. For these biochemicals, the very few published LCA studies available show inconsistencies in terms of a narrow look at environmental impacts and coverage of biochemical life cycle stages. LCA results vary widely and give in part contradictory conclusions regarding environmental performance. Sustainability claims for bio-based chemicals are often exclusively based on reduced global warming impacts compared to fossil-based chemicals, whereas other impacts, such as land use from bio-feedstock production, are largely ignored. To move towards truly sustainable biochemicals, we recommend that LCA practitioners include a broader range of impacts and life cycle stages, adhere to existing standards and guidance, and address the need to estimate currently missing data. For the biochemicals industry, we recommend to systematically use LCA to direct research and identify impact hotspots, and to make scale-up data on process performance available. With that, it will be possible to promote biotechnology as significant contributor to solving environmental sustainability problems.

Keywords: Environmental impacts; Biotechnology; Commodity chemicals; Lactic acid, Succinic acid, 1,3-Propanediol, 1,4-Butanedio, and 1,5- Pentanediamine; Life cycle assessment; Life cycle stages; Environmental trade-offs

Introduction – the role of biochemicals for achieving environmental sustainability

Chemicals are an essential part of our every-day goods. In the United States (US), 96% of chemical sales are still fossil-based, while only 4% are bio-based[1]. This continuous dependency on processing fossil resources is a major contributor to greenhouse gas emissions driving global warming impacts[2]. Fossil-based chemical production is furthermore very energy demanding, accounting for roughly 20% of the total energy used by industry[3]. Significant investments support exploring renewable 'bio-based' resources as new ways of producing chemicals, which have been reported to cause less global warming than their fossil-based counterparts[4]. This picture, however, is strongly influenced by the covered processes and choice of end-of-life treatment, where global warming impacts from bio-based chemicals can also exceed those from fossil-based chemicals when for example moving from composting to landfilling without energy recovery as end-of-life process[5,6].

Fighting fossil resources depletion and global warming are the main drivers to shift globally from pure fossil-based to bio-based products. Industry and academia have hence jointly taken on the challenge to develop bio-based processes for chemical production, and bio-based chemicals are projected to take up to 22% market share by 2025[7].

Using non-fossil resources for chemical production comes, however, with its own challenges for environmental sustainability. Feedstock selection, shifting from laboratory to commercial-scale production, and end-of-life treatment of bio-based products may all introduce sustainability tradeoffs[8]. To minimize such tradeoffs and move the biochemical industry to becoming truly more environmentally sustainable than the fossil-based chemical industry, it is crucial to systematically identify and address challenges related to environmental sustainability.

More than 10 years ago, the US Department of Energy (DOE) proposed a list of 12 bio-based chemicals as potential substitutes for some of the current fossil-based chemical building blocks on the market, using a techno-economic analysis[9]. The intention was not to directly replace particular intermediates in the chemical industry, but rather use the proposed chemicals as new intermediates for functionally equivalent downstream products, such as packaging materials. Increased use of renewable resources and environmental sustainability of bio-based industrial products were among DOE's major motivations behind establishing this list[10]. Two chemicals were added and five removed in an update of the original DOE list in 2010, mainly related to shifts in research and development in the biochemical industry[11]. The current level of commercialization of the chemicals on the updated DOE list ranges from laboratory scale to full commercial production,[12,13], with microbial fermentation as key process for using bio-based feedstocks in the chemical industry[13]. As the DOE list was not developed based on a specific set of criteria, we systematically selected those biochemicals that are currently highly relevant for the global community. As a result, we focused on studies assessing the environmental performance of commercially available commodity chemicals produced from bio-feedstocks through microbial fermentation as well as assessing the environmental performance of functionally equivalent petrochemicals. We thus analyzed studies applying environmental life cycle assessment (LCA) as a standardized method[14] widely used to assess the environmental sustainability performance of products and services. LCA aims at capturing all relevant environmental impacts occurring along product life cycles from raw material extraction ('cradle') and manufacturing to end-of-life ('grave'), and helps pinpointing hotspots in e.g. production processes (see Box 1 for related definitions). It is a powerful tool for identifying tradeoffs between life cycle stages and to avoid burden shifting from impacts on, for example, global warming or ecotoxicity[15]. We focused on biochemicals that have been fully commercialized to harvest maximum information on reported environmental performance, and exclude biochemicals that are made by chemical conversion from bio-based feedstock (e.g. Monoethylene glycol), or are not primarily used directly as monomers derived from microbial fermentation for polymerization (e.g. ethanol and glycerol). Reviews on LCA studies for using ethanol and glycerol in biofuel production are found elsewhere[16,17]. With that, our list focuses on the following commercialized biochemicals, for which we could identify at least one company that produces and sells these chemicals:

1) Lactic acid: e.g. Cargill (U.S.)

2) Succinic acid: e.g. BioAmber (Canada), Succinity (Spain)

3) 1,3-Propanediol: e.g. DuPont, Tate & Lyle (U.S.)

4) 1,4-Butanediol: e.g. BioAmber (Canada)

5) 1,5-Pentanediamine (also known as Cadaverine): e.g. BASF (China)

In support of the development of biochemicals with optimal environmental sustainability performance, we also evaluated studies applying LCA to nine DOE listed bio-based chemicals produced by means of microbial fermentation that are not yet commercialized. With our study, we seek answers to three questions: (1) Which are the main methodological choices to make when assessing environmental sustainability of bio-based chemicals? (2) What are the main conclusions from published LCA studies on commercialized bio-based chemicals? (3) How can we improve the use of LCA for bio-based chemicals, to help striving towards a viable and sustainable future for the biochemical industry? We provide specific recommendations for improving future LCA practice, and highlight opportunities and constraints in shifting from fossil-based to bio-based chemicals.

Box 1 Environmental sustainability assessment terminology.

Life cycle assessment (LCA). ISO-standardized method to quantify environmental impacts from inputs (resources used) and outputs (chemical emissions) along the life cycle of one or more defined product or service systems on a common functional basis. LCA consists of four iterative methodological phases, namely goal and scope definition, life cycle inventory analysis, life cycle impact assessment, and interpretation.
Life cycle stages. Stages of product or service life cycles, which usually include raw materials extraction, manufacturing, use, and end-of-life.

Life cycle inventory (LCI) analysis. Phase of LCA quantifying life cycle inputs and outputs for product or service systems as flows from or toward the natural environment.

Life cycle impact assessment (LCIA). Phase of LCA characterizing life cycle inputs and outputs of product or service systems in terms of the magnitude and significance of their potential impacts on human health, ecosystem quality, and natural resources.

Impact category. Class of impacts that represent an environmental issue of concern. Examples of impact categories are global warming, ozone depletion, human toxicity, ecotoxicity, land use, water use, and resources use, to which product system life cycle inputs and outputs may be assigned.

Cradle-to-gate. LCA where the product system is defined from raw materials extraction ('cradle') to factory gate, i.e. not all life cycle stages are covered.

Cradle-to-grave. LCA where the product system is defined from raw materials extraction ('cradle') to end-of-life ('grave'), i.e. all life cycle stages are covered.

End-of-life. Life cycle stage representing the end of the product's use. May include processes like reuse, recycling, chemical and energy recovery, incineration, landfilling, wastewater treatment, and release of bio-based products in nature.

Current state of commercialized commodity biochemicals

We systematically searched Scopus and Google Scholar for biochemical name synonyms as listed in PubChem[18] along with "sustainability" or "LCA" and "life cycle assessment" or "Foot Print" and "Footprint". We found 36 environmental sustainability assessment studies published between 2003 and 2018 that matched these search criteria (searches conducted until 28.02.2018). Table 1 summarizes market information and results from these studies conducted for the commercialized biochemicals.

Table 1 Main characteristics and life cycle assessment studies available for currently commercialized biochemicals.

CAS Number		50-21-5	110-15-6	504-63-2	1070-70-8	462-94-2
Chemical		Lactic acid	Succinic acid	1,3-Propanediol	1,4-Butanediol	1,5-Pentanediamine
World production in kt/yr (Year)	Fossil-based	n/a	76 (2015)[19]	n/a	2500 (2015)[19]	n/a
	Bio-based	472 (2015)[19]	38 (2015)[19]	128 (2015)[19]	3 (2015)[19]	50[20,21,(i)]
Main current application		Food supplement, (Poly)Lactic Acid	Food supplement, Pigment, Resin	Plastics, Cosmetics, Cleaning products	Plastics, Fibers	Nylon, Chemical intermediate
Number of published LCA studies per biochemical*		20[(ii)]	8[(iii)]	5	3	—
Number of published biochemical LCA studies addressing different impact categories according to the requirements of ISO14040 and EN16760 standards[(iv)]	Global warming	20	7	5	3	—
	Ozone formation	3	2	1	1	—
	Ozone depletion	4	3	—	1	—
	Ionizing radiation	1	1	—	—	—
	Particle formation	3	2	1	1	—
	Human toxicity	4	3	1	—	—
	Ecotoxicity	4	3	1	—	—
	Acidification	7	2	2	1	—
	Eutrophication	6	3	1	1	—
	Land use	2	3	1	—	—
	Water use	4	1	—	1	—
	Resources use	2	3	—	1	—
	Energy demand	13	6	5	2	—
Production with fermentation from renewable biomass - State of commercialization		Commercialized[12]	Commercialized[12,22]	Commercialization[12,22]	Commercialized[12,22]	Commercialized[20]
Availability of inventory data for biobased production routes		Production process data in LCI database ecoinvent[23]	No data in LCI databases	No data in LCI databases	No data in LCI databases	No data in LCI databases
Limitations of available LCA studies with focus on assessing environmental impacts of biochemicals		Variation in assessed life cycle stages: 2 studies assess stages from resource extraction to acid production. 11 include polymerization and 11 assess the whole life cycle	Few studies available	Few studies available	Few studies available	No LCA studies publicly available Nothing about environmental performance is known
Opportunities for applying LCA		Conduct and publish more studies identifying hotspots and burden shifting within the life cycles; more focused research will help increasing environmental sustainability of bio-based substances		Apply- and publish LCA studies on bio-based products and processes		

*Full references of all studies included in our analysis are listed in the Supplementary Information.

(i)Estimated production volume; (ii)Global warming and energy demand impact results are only retrievable from Morales et al.[24]; (iii)One of the studies is not an LCA, but an assessment of selected environmental sustainability metrics[25]; (iv)30 studies follow ISO standard, and 6 studies do not follow ISO standard.

LCA studies have been found for all assessed biochemicals except 1,5-Pentanediamine. Eighty-three percent of the analyzed studies claim to follow ISO standards, requiring LCA studies to consider all relevant life cycle stages and cover a comprehensive set of environmental issues related to the product system being studied[26]. Nevertheless, forty-six percent of these studies only consider one or two impact categories and many assess only a limited number of life cycle stages (see Figure 1 for an example).

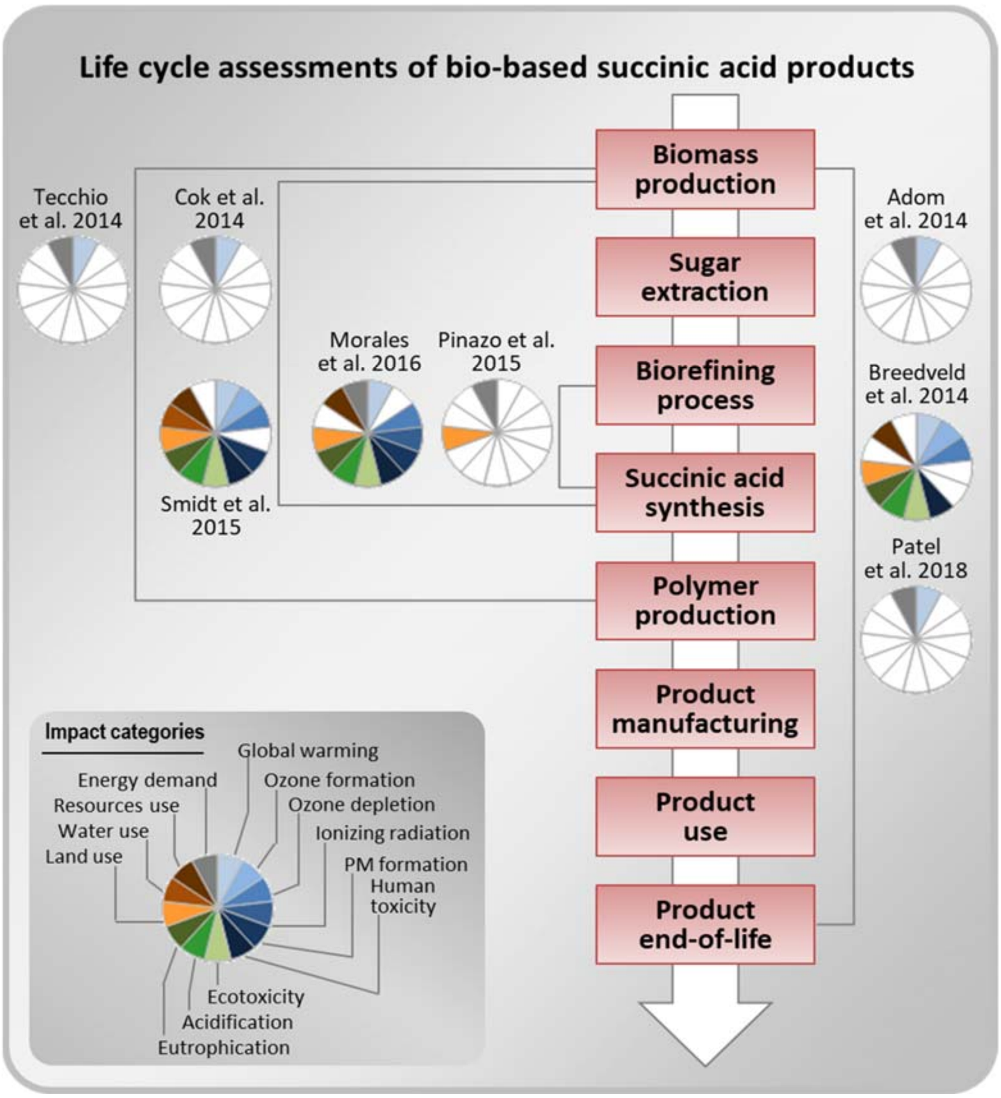


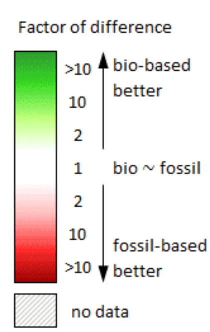
Figure 1 Overview of life cycle stages covered and impact categories considered in seven life cycle assessments of succinic acid production. Full references for all studies listed in this figure are provided in the Supplementary Information.

Three life cycle stages, namely biomass production, polymer production, and end-of-life treatment, drive LCA results for the five biochemicals with available data (see Table 1), either through a combination of involved processes or high impacts for specific processes. For example, when assessed, land-use impacts are in almost all assessed cases more than a factor 10 higher for biochemicals than for petrochemicals[27-29]. Variability in life cycle impacts from biochemical production is predominantly affected by geographical differences in the technology mix of the electricity generation[30,31], while end-of-life impacts vary mainly due to differences in economic development and geographical and cultural waste treatment patterns, yielding a variety of waste disposal options, such as industrial composting, incineration (with or without heat recovery), and landfilling[32]. Impact results variability is further influenced by the choice of allocation approaches in case of multifunctional production systems (system boundary expansion versus economic or energy allocation based approaches)[33]. Both geographical and approach-based variability can be tested in scenarios to assess the sensitivity of LCA results and estimate related uncertainty for each scenario choice, which can help to understand the robustness of results.

Across LCA studies, the single most assessed impact category is global warming from emission of greenhouse gases. Global warming impacts vary widely when comparing production of lactic acid and (poly)lactic acid (PLA) with functionally equivalent fossil-based chemicals and plastics, in e.g. polyethylene terephthalate (PET) and polystyrene (PS) (see Table 2). In a number of studies, PLA shows 5-90% lower global warming impacts than its fossil-based counterparts, with higher CO_2 emissions due to the extraction and processing of fossil resources[34,35]. However, some studies show higher global warming impacts for PLA than for PET[5] and PS[6], mainly associated with CO_2 emissions from electricity generation (due to a fossil-based electricity generation used for the resin production[30]) and from waste management[24,25]. For succinic acid, global warming impacts for bio-based production varies from 22% lower to more than 250% higher than fossil-based production as a function of considering carbon storage during biomass cultivation, different energy mixes during resin production[33], and purification technology[36] chosen in the LCA studies. Going beyond global warming, we observe similar trends and variations with both lower and higher impacts for biochemical options compared to their fossil-based counterparts, as summarized for all considered chemical-impact combinations in Table 2.

Table 2 Environmental impact ratios expressed as factors of difference between bio-based and fossil-based chemicals (color range) and study count (numbers) for different chemical-process-impact combinations (1,5-pentadiamine was not included due to a lack of data). Different colors within a single combination (e.g. global warming (GW) impacts associated with acid production of succinic acid) indicate that multiple scenarios in a single study (i.e. study count *n*=1) or results of multiple studies (i.e. study count *n*>1) show different impact ratios for the same chemical-process-impact combination. This variability is plotted as color range. Full references of all studies included in our analysis are listed in the Supplementary Information.

	Lactic acid			Succinic acid			1,3 Propanediol		1,4 Butanediol	
	Life-cycle stages included in studies									
Impact category*	acid production	polymeri-zation	end-of-life scenario	acid production	polymeri-zation	end-of-life scenario	diol production	end-of-life scenario	diol production	end-of-life scenario
GW	2	11	11	4	1	2	4	2	1	2
OF	1	1	1	1		1	1		1	
OD	1	1	2	1		1			1	
IR	1									
PM	1	1	1	1			1		1	
HT	1	2	1	1		1	1			
ET	1	1	2	1		1	1			
AC	1	4	3			1	2		1	
EU	1	3	3	1		1	1		1	
LU	1		1			1	1	1		
WU	1	3		1					1	
RU	1		1	1		1			1	
ED		9	3	3	1	1	4	2		2



*Impact categories GW: global warming, OF: photochemical ozone formation, OD: stratospheric ozone depletion, IR: ionizing radiation, PM: particulate matter formation, HT: human toxicity, ET: ecotoxicity (terrestrial or aquatic), AC: acidification (terrestrial or aquatic), EU: eutrophication (terrestrial or aquatic), LU: land use, WU: water use, RU: abiotic resources use, ED: (non-renewable) cumulative energy demand.

Burden shifting between life cycle stages is an often-disregarded phenomenon when analyzing the transition from fossil-based to biochemicals. A cradle-to-gate LCA shows, for example, that global warming impacts from a PLA bottle reach only 69-90% of impacts from a PET bottle[6]. When including disposal (cradle-to-grave), the total burden for PLA increases and shifts from ‘harvesting and production’ to ‘use and end-of-life’, due to emissions of the strong greenhouse gas methane from degradation of PLA under anaerobic conditions during landfilling, whereas PET is assumed non-degradable[6]. The advantage of PET over PLA is further increased if the bottle material is recycled, since such systems are currently in function in many places for PET but not for PLA. An additional shift in burden is seen when moving to bio-based lactic acid, where we see strongly reduced global warming impacts for the acid production but strongly increased land use impacts, which may be up to more than 100 times higher due to the use of an agricultural crop-based feedstock (see Table 2).

For succinic acid, LCA studies show that fermentation-related energy consumption, choice of fermentation process, and impacts from end-of-life processes constitute the main environmental performance challenges when moving to biochemicals[33,37,38]. Studies for 1,4-Butanediol and 1,3-Propanediol show more consistent environmental benefits of the bio-based chemicals over the petrochemical. This is mainly linked to petrochemical conversion processes being more energy intensive[39], while including biomass production related impacts, such as land-use and acidification, results in bio-based chemicals either performing worse[28] or results not being very decisive[39].

In summary, results for even the most often included impact category, global warming, vary a lot across bio-based chemicals (see Table 2), rendering generic conclusions impossible without applying a full LCA in all cases. In addition, LCA studies need to consider other potentially important impacts, such as land use and eutrophication, associated with current bio-based production methods to ensure that they identify and address relevant impact tradeoffs and burden shifting along the chemicals’ life cycles.

Improving LCA practice for biochemicals

The large variation in included impacts and life cycle stages across LCA studies reflects current challenges when assessing biochemicals. Each studied system is unique in features and components, rendering it difficult to compare it with functionally equivalent systems or processes. This well-known problem, however, is not unique to biochemicals but applies to many product systems, such as waste treatment systems[40]. For improving LCA practice for biochemicals, we emphasize the key components to be included in each study, such as all life cycle stages, including end-of-life scenarios, and all impact categories. Indeed, it is an ISO requirement that all life cycle stages should be included in an LCA[41] to uncover possible burden shifting along product life cycles, such as environmental benefits or impacts related to certain end-of-life treatments. Below, we detail the needed adaptations of LCA for the biochemicals industry to allow giving a relevant impression of environmental sustainability, including to adhere to existing assessment standards and available practical guidance, and to address the need to estimate currently missing data.

Ensuring coverage of all relevant system components and environmental impacts

The analysis of existing LCA studies on biochemicals revealed that the most relevant impact categories are global warming, land use and water use, eutrophication (fertilizer use) and ecotoxicity (pesticide use) during feedstock production, and energy and water use in biorefineries. The most relevant and variable life cycle stage is feedstock production, where a potentially very important modelling aspect is the impacts from indirect land use changes (ILUC) representing those changes in land use that may result from expansions in cropland induced by an increased demand for crops due to increases in biochemical (or biofuel) production. Biochemical processing has significant potential for sustainability optimization that becomes even more important during upscaling from laboratory to market scale, where the biochemicals industry will still need further innovation for process maturation. Finally, end-of-life treatment is relevant, as biodegradable chemicals are often claimed to be CO_2 emission neutral, but methane emissions from landfilling can offset these benefits.

Because of the special nature of bio-based chemicals originating from biotic resources, all impact categories assessing impacts occurring in the growing phase of the biomass should be included by default in related LCA studies. For end-of-life scenarios, it is especially important to consider those impact categories that address possible toxicity-related effects of waste treatment including ecotoxicity and human toxicity, and to model potential landfill emissions of methane, a strong greenhouse gas. Spatial variability may have an important influence on LCA results, and it should be considered if data and models are available, in particular for locally variable impact categories like freshwater use, eutrophication and ecotoxicity.

When assessing end-of-life scenarios, the most representative setups for relevant product applications should be included, as environmental impacts can vary greatly between disposal methods[30,40]. If end-of-life scenarios are not considered, it is still important to outline applicable scenarios, stating if relevant whether products are compostable, biodegradable under environmentally relevant conditions, or recyclable.

Adherence to existing standards and guidelines

Inconsistent application of well-defined guidelines yields highly variable LCA results even when the same impact categories are considered[42]. To avoid such issues and to strengthen the credibility of LCA results for biochemicals, we strongly suggest that future studies follow the ISO 14040 standards series and the US-EPA LCA principles and practice[43]. Furthermore, for making LCA on bio-based chemicals much more representative, we recommend to follow the specific standard EN 16760:2015[44] for LCA on bio-based products. This standard builds on the ISO standards[14,26] for guidance concerning the general LCA methodology, but gives for example explicit guidance modelling of agriculture, forestry and aquaculture systems, which are recognized to have relevant environmental impacts in bio-based production systems[44].

Overall, a strength of LCA is its broad coverage of impact categories, ensuring that relevant impacts are reflected in the results. It is, however, also a challenge to communicate the array of results. Hence, the choice between alternative products based on LCA results will often require some sort of aggregation of the results across impact categories, based on normalization and weighting of the impact scores or science-based translation into common metrics representing damages to natural ecosystems (e.g. species loss) or human health (lifetime loss)[45]. Comprehensive guidance to address these challenges of interpreting LCA results and using these results as decision support for the biochemicals industry is available for example in the 'LCA: Theory and Practice' textbook[46].

Estimation of missing data

In the absence of real-world data, which is often the case for lab-scale production processes, reference process data, default optimization potentials, and relevant scale-up mechanisms should be considered for a first hot-spot screening. Data then need to be systematically provided for hot-spot processes and related impacts.

We recommend more specifically the following: For modelling feedstocks, focus should be on impacts from emissions of pesticides, nutrients, and use of water and land, which may be estimated based on generic database values adapted from actual practices. For addressing geographic differentiation, chemical emissions and resource uses, inventory-modelling needs to be performed for the specific processes of the life cycle (possibly based on modification of generic inventory database processes and using local grid mix for electricity). In the impact assessment part, spatially differentiated methods are available for all non-global impact categories, which means that impact assessment research is already focused on strengthening the available methods, for example addressing spatial differentiation of life cycle toxicity impacts[47]. For production efficiency, specific data should be available for the studied system and upscaling and learning may be relevant to consider when comparing new and early-stage technologies with conventional alternatives, depending on the scale and maturity of the processes included. For the impact assessment, we can also *a priori* identify the relevant impact categories when we know the specificities of the bio-based chemical life cycle and the conventional chemical that we want to compare. Normally, they are found among climate change (CO_2, N_2O, and CH_4 related to agriculture and energy systems), eutrophication (nutrients from agriculture), ecotoxicity (pesticides from agriculture and from the production of bio-based chemical and conventional alternative), water use (from agriculture if water is critical in the concerned region) and land use (agriculture again).

Toward a sustainable biochemical industry

We identified several environmental sustainability challenges for the biochemicals industry that require additional development efforts. Key focus areas are (a) to systematically include screening LCA at an early stage as part of directing research efforts and identifying key environmental hotspots; (b) to focus on making scale-up data on process performance available to allow for developing LCA for a broader range of products; and (c) to promote biotechnology as significant contributor to solving environmental sustainability problems in areas where most of the impacts are generated. For example in feedstock production (i.e. agriculture) this could be by improving crop yields and reducing use of fertilizer by using plant growth promoting bacteria.

Include LCA to identify hotspots and research priorities

Bio-based chemicals can show lower or higher global warming impacts compared to fossil-based chemicals, and often show higher impacts in other categories, such as land use. LCA is a useful tool to identify hotspots in environmental sustainability profiles of bio-based chemicals[48]. Significant additional research and development efforts are required mainly regarding feedstock production, biorefining and product recycling, for further improving the overall environmental sustainability of bio-based products.

At the early stages of biorefinery development, feasibility studies should include at least screening-level LCA to identify major hotspots in the product system. For assessments where the purpose is to investigate the consequences at societal scale of a change towards first generation bio-based chemicals, LCA should aim to model the consequences at societal scale, and further modeling efforts are required to address indirect land use change impacts. As an example, an increased demand for corn to produce bio-based chemicals in the United States may lead to the expansion of corn production to other regions to meet overall greater demand. This may eventually induce conversion of natural areas into farmed land[49] causing environmental impacts that are potentially large but typically not considered in LCA of individual biochemical products and materials as reported in the present study. Finally, the 'wicked nature of sustainability'[50] calls for considering consumer preferences to a higher degree[51], since traditional methods dealing with optimization problems might not be sufficient and application of multidisciplinary approaches are necessary to boost environmental sustainability of bio-based products.

Make scale-up data on process performance broadly available

When assessing opportunities using lignocellulosic biomass, macro- and micro-algae as next generation feedstock, main challenges are related to data availability and accessibility, as well as targeting environmental sustainability-related impact hotspots in biochemicals production that may differ between feedstock generations. When assessing environmental impacts of biochemicals produced by early-stage technologies, we need to effectively scale up laboratory data to being more representative for commercial scale production, and for better reflecting on the optimization potential of bio-based chemicals, as various production processes are currently still immature. Such efforts may be inspired by comparisons of efficiencies and emissions for laboratory scale processes and commercial full scale processes for other similar biochemicals and materials. It is further possible to define minimum fermentation yield performance and productivity that would be required to become commercially viable, or to soft-link process simulation with LCA, enabling plant-wide design by scaling up lab-scale technologies using scaling factors[52].

Promote biotechnology to solve sustainability problems

In perspective, we observe that socio-economic aspects including population, transportation, and the use of primary energy, water, fertilizers and biotic and abiotic resources grow rapidly over the last decades[53]. These aspects drive increasing impacts on global warming, ocean acidification, eutrophication, stratospheric ozone depletion, and impacts on humans and ecosystems from chemical emissions, and on depletion or degradation of land, water, fossil and other resources. Some of these trends already exceed our earth's capacity for sustaining the current socio-economic development. Hence, just ever being "more environmentally sustainable" is not enough, especially when consumption increases globally[54]. The biochemicals industry should be promoted to explore how innovation can contribute to being environmentally sustainable in absolute terms based on the capacity of sustaining our biophysical earth systems, while meeting the growing needs for viable bulk chemicals in today's and future societies[55]. For LCA practitioners, this means that there is no excuse not to look at all relevant impacts and include all life cycle stages to fully supporting a comprehensive improvement of biochemicals' environmental performance. For biotechnology developers, this means to better integrate LCA as a tool that can quantitatively support a truly sustainable development of biochemicals instead of relying on partially justifiable environmental sustainability claims such as reduction of CO_2 emissions in the chemical production phase alone compared to a petrochemical alternative. We look forward to seeing both fields converging for successfully moving towards a true sustainable future based on biochemicals in line with the global sustainability agenda.

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Author contribution statement:

Ó. Ögmundarson and P. Fantke organized and structured the work, data processing and paper writing. Ó. Ögmundarson gathered, analyzed and visualized the data and wrote the paper. P. Fantke contributed to writing, data analysis and visualization, editing, and provided overall guidance. M. Herrgard, J. Förster and M. Hauschild provided input on paper structure and background information, and edited the manuscript.

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PAPER II

Journal: Energy and environmental science

Working title

Environmental hotspots of different lactic acid production systems

Ólafur Ögmundarson,[a,b] Sumesh Sukumara[b], Alexis Laurent[a] and Peter Fantke[a*]

[a] Quantitative Sustainability Assessment Division, Department of Management Engineering, Technical University of Denmark, Kgs. Lyngby, Denmark

[b] The Novo Nordisk Foundation Center for Biosustainability, Technical University of Denmark, Kgs. Lyngby, Denmark

Corresponding author: phone: +45 45254452, email: pefan@dtu.dk

Abstract

The objective of this study is to assess the environmental hotspots of lactic acid production within three different feedstock generations, with special focus on the effects of energy inputs based on US-American conditions. As an assessment framework, we used Life Cycle Assessment (LCA), a standardized assessment tool, applied within a cradle-to-grave system boundary applying system expansion in an attributional LCA. When assessing and identifying the largest life cycle environmental impacts – so-called hotspots – of three-feedstock processes producing lactic acid from 1st generation corn, 2nd generation corn-stover, and 3rd generation macroalgae, there are certain observable trends, but they are all mostly affected by high energy utility use. In the 1st and 2nd generation feedstock processes, most energy utility inputs happen in the biorefinery stage, while in the 3rd generation feedstock process, energy utilities have the highest effect in the biomass life cycle stage due to drying of biomass, accounting for up to 86% of the related impacts. The 3rd generation feedstock process is in its early stages of development. By assessing the environmental impacts at such an early stage, our results show that by applying the LCA methodology before large investments have happened for scaling up, we have the opportunity to identify environmental hotspots at an early stage of development and use them to make future biochemicals more environmentally sustainable.

Key words

Life cycle assessment, corn, corn-stover, macroalgae, biochemicals, environmental sustainability, uncertainty

Introduction

Our climate is changing and global temperatures are dramatically rising because of anthropogenic greenhouse gas (GHG) emissions. Approximately 78% of total GHG emissions in the world stem from fuel combustion and energy conversion used in industrial processes[1], and within industrial processes, petrochemical production makes up for about 20% of the energy demand[2,3]. This last share might even grow in the nearest future, as the estimated biggest driver behind global oil demand is the increasing global production of petrochemicals, e.g. because of steady rise in demand for consumer goods and services[4].

Overall, moving from fossil-based to bio-based chemicals does not come without challenges[5]. High production costs, establishing matching or better properties compared to conventional counterparts, and questionable superiority in environmental sustainability profiles of biochemicals and derived products are currently the biggest challenges[6–8]. Despite positive examples[9], where e.g. production of lactic acid performs environmentally better than functionally equivalent products, it remains unclear how different feedstocks influence overall environmental performance of bio-based products and what the feedstock-specific impact hotspots are, which might differ also as function of TRL[7].

Utilizing renewable resources for biochemical production has the potential to lower GHG emissions by substituting petrochemicals with bio based chemicals, and, since 2001, subsidies, campaigns, mandates and bans have globally encouraged market entrance of biochemical-based materials[10]. Bioplastics are the main biochemical product group[11] although they currently only contribute with about 1% to total global plastic production. An increase in market share of bioplastics is however foreseen, and in January 2018 the European Commission published a new *European Strategy for Plastics in a Circular Economy*[12] including a “Vision for Europe´s new plastic Economy” until 2030. The focus of this strategy is to tackle the environmental challenges related to conventional plastics along all life cycle stages (LC-stages), i.e. from resources extraction to end-of-life handling, proposing different ways toward more environmentally sustainable plastics[12]. Consequently, worldwide production of bio-based building block chemicals and bio-based polymers is foreseen to increase by about 26% and 10%, respectively, until 2022[5]. Environmental sustainability claims of biochemicals and derived products based on assessing GHG emissions in specific production processes, however, are often not justified[8,7]. Not including all LC-stages can lead to unconsidered large environmental impacts (i.e. so-called hotspots) and burden shifting, for example, when we overlook potentially large impacts or benefits arising from waste management systems[13,14]. Ögmundarson et al.[7] proposed to at least including land use and water use, eutrophication, and ecotoxicity impacts in addition to assessing global warming, to address important life cycle hotspots of biochemical systems for environmental optimization. Current biochemical and related biopolymer production is mostly based on agricultural crops (1st generation feedstock[15]) with a high Technological Readiness Level (TRL)[16] of 8 (first-of-a-kind commercial system) or 9 (full commercial application) (see Figure 1). Biochemicals from non-agricultural crops, e.g. lignocellulosic biomass and wood (2nd generation feedstock[15]), have not yet reached high commercialization levels, due to the cost related to the conversion of 2nd generation feedstocks into fermentable sugars[17] and low technical process immaturity[15] in comparison to cost of utilizing 1st generation feedstocks. Due to technological challenges for utilizing 2nd generation feedstocks for example in pretreatment of the biomass[18,19] and derived economic challenges[20] of biochemical production from 2nd generation biomass[21], completely different feedstock sources, including engineered crops, algae and urban residues like household waste (3rd generation biomass[15]), are receiving increased attention. Based on conducting full LCAs for all three bio-feedstock generations, we emphasize within each generation and its respective TRL the environmental hotspots in order to focus future improvement efforts for the different biochemical production systems.

1st Generation

Feedstock:

- Agricultural crops (with/without pretreatment)
- World production 891.1 million metric tons/annually[1]
- Of which 42% in USA[1], accounting for 42%[2] of US N fertilizer use

TRL 8-9:

- Mature commercial markets
- Well-understood technologies

Pros:

- Easily fermentable sugar rich feedstocks

Cons:

- Competition with edible food
- Impact on food prices

2nd Generation

Feedstock:

- Lignocellulosic biomass (with pretreatment)
- USA production 108.9 million metric tons/annually[3]

TRL 4-5:

- Immature commercial markets
- Partly understood technologies for commodity chemical production

Pros:

- Non-food biomass
- Abundant availability for a low price

Cons:

- Feedstock conversion into easy fermentable sugars

3rd Generation

Feedstock:

- Macroalgae biomass (with pretreatment)
- Industrial use, minimum 8 million tonnes (numbers since 2003)[4]

TRL 1-2:

- Not commercialized
- Poorly understood technologies for commodity chemical production

Pros:

- Not grown on land
- Bioremediation of feedstock in nature

Cons:

- Biomass availability, and for a reasonable price

Figure 1. Overview of bio-feedstock generations considered in the present study. TRL: Technological readiness level. [1)]See reference[22], [2)]See reference[23], [3)]See reference[24], [4)]See reference[25]

To optimize biochemical production in terms of environmental sustainability performance, it is essential that we understand where along the life cycle of such systems environmental hotspots occur for each feedstock, and how any change in inputs and outputs to the production system influences such hotspots. In this effort, life cycle assessment can turn out to be a useful tool. LCA is an ISO-standardized and widely applied methodology for assessing the environmental performance of different product systems and is hence well-suited for quantifying impact hotspots along the life cycle of biochemical production systems using different feedstocks[6].

Existing LCA studies performed on biochemicals show some trends and limitations. When biochemicals and derived products are compared to functionally equivalent fossil-based products, LCA results show that in some impact categories biochemicals perform better whereas in other categories they perform worse than their fossil-based counterparts[7,8]. However, several studies only focus on assessing global warming impacts and do not include other relevant environmental impacts, thus potentially overlooking burden shifting from one environmental impact to another. There is additionally a large variation in the coverage of LC-stages across studies, which too may result in burden-shifting from one LC-stage to another [7]. These trends and limitations suggest that there is a strong need for a more comprehensive overview of the differences in environmental performances between feedstocks, LC-stages and impacts to identify optimization potentials of biochemical production from conventional and emerging feedstocks[7].

To address this need, we aim in the present study to (1) consistently define life cycles of biochemical product systems across bio-feedstock generations focusing on lactic acid as an important building block chemical[26,27]; (2) characterize the environmental performance of lactic acid production systems with a full life cycle assessment; and (3) discuss related environmental hotspots and their potential drivers. That includes showing how hotspot results can be used to inform technology system design, identify optimization potential of future processes, and operationalize decision support. Since the feedstock systems have different TRLs, it has to be stated that their hotspots cannot be directly compared across systems.

Methods

To address the 3 above objectives, our study follows the International Reference Life Cycle Data System (ILCD) Handbook for LCA practice [28] and the EN 16760:2015 standard specific to bio-based products[29].

Scope definition: Lactic acid cradle-to-grave system description

We assess the production system setups along its entire life cycle, i.e. cradle-to-grave. That includes firstly harvesting of renewable biomass, corn, corn-stover and macroalgae. Secondly, the biorefinery stage. Thirdly polymerization of LA to (poly)lactic acid (PLA) as one of the most important LA products[10,27]. Fourthly, end-of-life (EoL) scenarios are also included, despite being sometimes ruled as infeasible[30]. The functional unit, which reflects the systems' function and is the basis of the assessment[31], is defined as "the production and use of 1 kg of lactic acid, with 99.9% purity, for household packaging application in the United States". By household packaging application, we mean food packaging, which follows the waste streams of household waste.

The default geographical location of the feedstock harvesting-, chemical production-, and end-of-life stages is USA (from cradle-to-grave), because this is where most LA and PLA are produced in the world today and therefore it is assumed that future production of LA from 3rd generation is most likely to be developed there as well. Effects of changing the geographical location of the cradle-to-grave assessment of the 3rd generation feedstock process, to China (CN) and Iceland (ICE) are also tested. That is done to see if differences in energy and heat production between the US and China on one hand, and US and ICE on the other, have effect on the study's results. Another assumption made is that the biorefinery is situated by the feedstock source.

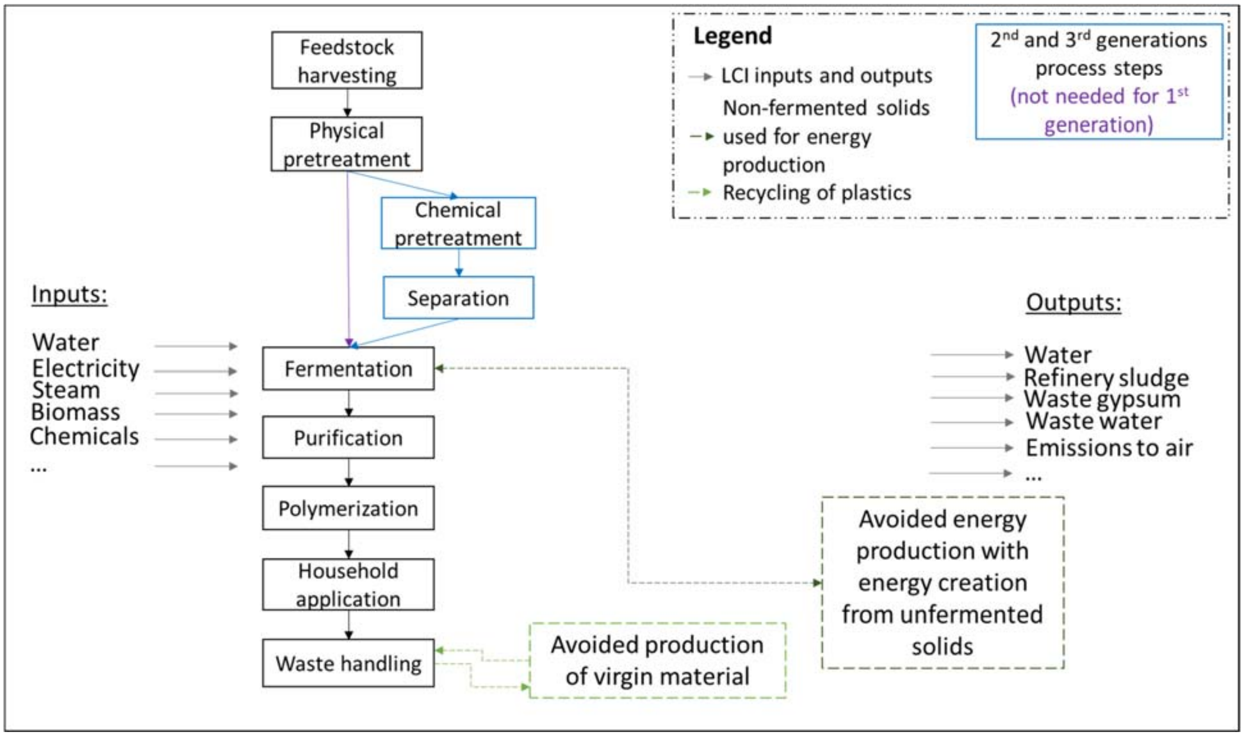

Figure 2 Process flow diagram describing system boundaries of assessment – Utilization of three generations of feedstocks, corn (1st generation), corn-stover (2nd generation) and macroalgae (3rd generation)

Figure 2 shows graphically what is included in the LCA (i.e. system boundaries). For all three assessed feedstock processes, the same LC-stages are assessed. Differences in modeling of the biorefinery stage are due to the different composition of the assessed biomasses. For the 1st generation biomass, pretreatment is not needed, but for both 2nd and 3rd generation biomasses both chemical pretreatment and separation is required before fermentation is possible[32–34]. Inputs, e.g. water, energy utilities and chemicals, and outputs, including waste streams and emissions, are assessed as well as avoided production of both energy (through energy creation from unfermented solids), and avoided production of virgin material (through recycling of plastics from household waste) through system expansion. Among emerging feedstocks, brown macroalgae are getting most attention because of the absence of lignin in the biomass, thus avoiding the need for lignin removal[35], which is costly because of recalcitrance of lignocellulosic cellulose and its toxic effects on microbial properties[36].

The modeled system represents US conditions (referred to as base case) through the whole life cycle for LA production from the three assessed feedstock systems (see Table 1). Because of limitations in current technology, not all carbons can be fermented, like alginate, but they make up a large proportion the carbon content of the biomass. It is therefore interesting so assess what the environmental benefits would be if the alginate was fermentable. That is therefore also assessed, both for the US base case and as well as using ICE and CN country conditions.

System modeling and data collection

The assessed biorefinery scenarios for producing LA from the three different feedstock generations are differentiated by pretreatment practices and purification options downstream for each of the feedstocks. As a pretreatment step, physical pretreatment is common for all the three generations of feedstock.

To secure technological representativeness of the biorefinery LC-stages, that is the conversion of the feedstock to biopolymers, we used Aspen Plus v.8.8 to simulate the processes in a techno-economic assessment (TEA) and create the inventories needed. For production of LA from 1st generation biomass, i.e. corn, the process is modeled based on data availability from process design of commercial production of LA[37]. The same principle applies for the modeling of LA production from 2nd generation biomass, i.e. corn stover[32]. For the 3rd generation biomass, i.e. macroalgae, scientific literature was used when applicable[34], but as there are no existing commercial processes for producing commodity chemicals from macroalgae, academic and industrial experts on microbial design, biomass pretreatment and downstream processing of commodity chemicals were consulted to ensure the chosen technologies' representativeness.

Table 1 – Main data sources for the present LCA study (For more details see SI.1 to SI.4)

	Production of Lactic acid from three different feedstocks					
Life cycle stage	Corn		Corn stover		Macroalgae	
	Foreground system	Background system	Foreground system	Background system	Foreground system	Background system
Biomass production and harvesting	Ecoinvent 3[38]	Ecoinvent 3, including iLUC[38]	Fertilizer value of corn stover[39]	Ecoinvent 3[38]	Harvesting of macroalgae[40]	Ecoinvent 3[38]
Biorefinery	Techno-economic assessment (SI-1a)	Ecoinvent 3[38]	Techno-economic assessment (SI-2)	Ecoinvent 3[38]	Techno-economic assessment (SI-3)	Ecoinvent 3[38]
Polymerization	IHS Markit©* database[41]	Ecoinvent 3[38]	IHS Markit©* database[41]	Ecoinvent 3[38]	IHS Markit©* database[41]	Ecoinvent 3[38]
End-of-life	Country specific handling of household waste[42]	Ecoinvent 3[38]	Country specific handling of household waste[42]	Ecoinvent 3[38]	Country specific handling of household waste[42]	Ecoinvent 3[38]

*IHS Markit©: Market and industrial database

Following the guidance of the ILCD Handbook, an attributional model with use of system expansion and average life cycle inventory (LCI) data was selected[28,38]. By applying Aspen Plus we can populate data that otherwise would not be available, because of lack of readily-available industrial data for production of commodity chemicals[43] as well when assessing future use of emerging biomasses like microalgae Aspen Plus populates the necessary inventory based on given assumptions. The calculated mass flows of the biorefinery from Aspen were used to populate the mass flows for the annual production of 110.000 tons of LA per year. These calculations were used to identify needs of amounts of biomass per feedstock process for harvesting or cultivation.

For the polymerization process, the IHS Markit© database[41] was used to identify mass flows, and for EoL options, the average distribution of waste in 2014 in the US was used[44]. For all background systems, existing processes from Ecoinvent v3 were applied[38] and adapted to country-specific scenarios wherever background information was available (see Table 1 and ESI.1 to ESI.3 for further information).

Life cycle impact assessment

The Life Cycle Impact Assessment (LCIA) was modeled with SimaPro v.8.5. All impact categories included in the ReCiPe 2016 methodology (hierarchist perspective, v.1.02) were assessed at both midpoint and endpoint levels, which differ by that at midpoint, impact indicators are defined somewhere along the cause effect chain, while at endpoint or damage level, impacts are translate to damages to different areas of protection (entire cause-effect chain covered). If increasing environmental sustainability of biochemicals by producing them from alternatives to agricultural feedstocks is the way forward, we need to look beyond individual impact categories and translate the midpoint impacts to damage level.

At midpoint level, impact categories include global warming, stratospheric ozone depletion, ionizing radiation, ozone formation, human toxicity, fine particulate matter exposure, ozone formation, acidification, eutrophication, ecotoxicity, land use, resources depletion, and water consumption. At damage level, impact on three areas of protection are assessed: human health, ecosystem quality and natural resources.[45]

Furthermore, as part of our base case for LA production from corn and because of its particular relevance for crop feedstocks, we included the biophysical indirect land-use change (iLUC) attributional model developed by Schmidt et al.[46]. iLUC contributes to the LCA results through LCI results, where intensification of already farmed land (to meet increased demand for crops) contribute to relevant environmental impacts through increased nitrogen fertilizer use and transformation from secondary forests to croplands contributes to CO2 emissions[46] (see Figure 3).

When assessing iLUC, in this study it is modeled as inputs from technosphere, as potential net primary production (NPP0) kg C / per kg of corn[46].

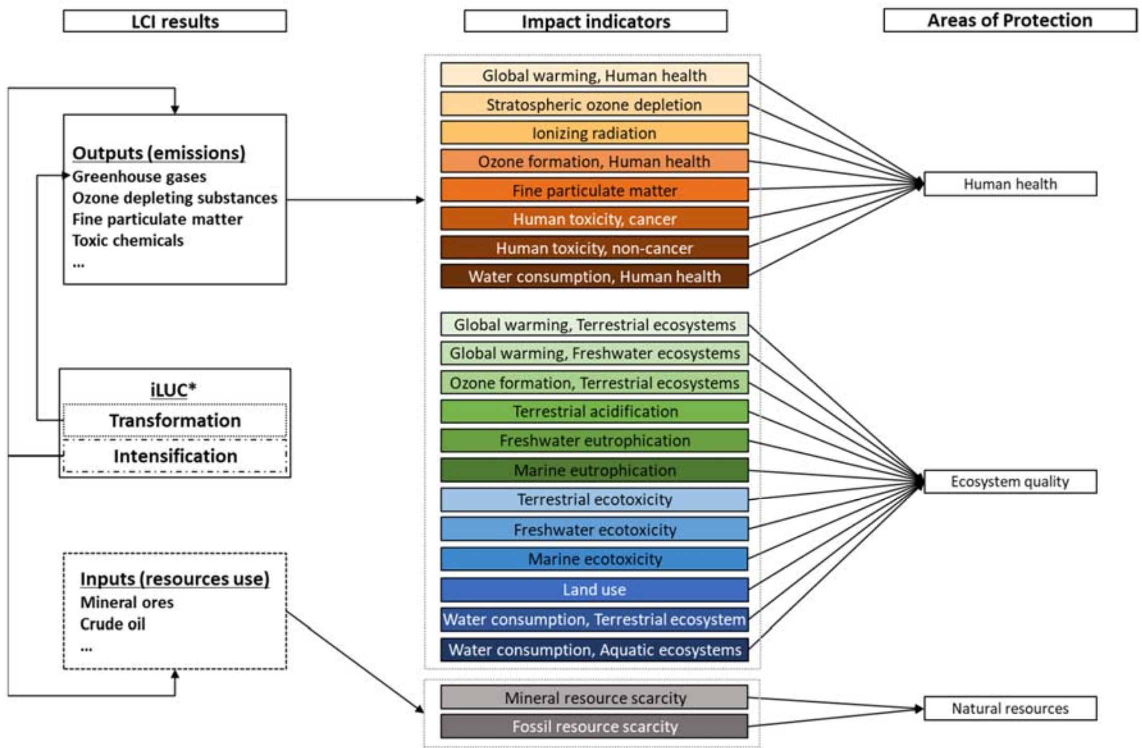

Figure 3 LCI results including iLUC and their contribution to different impact indicators resulting in the three areas of protection assessed (adapted from[47])

When addressing hotspots in our analysis, we define them as most significant impacts (e.g. after weighting). It is the difference between where the largest impacts come from and what the largest impacts are. Based on this definition, we cannot compare changes in hotspots across impact categories because they are expressed in different units. That results in, if we want to compare hotspots across impact categories, we need to translate the LCI results to area of protection. By doing that we can identify environmental hotspots across impact categories.

Analyzed scenarios

Four scenarios were analyzed in the study. When using macroalgae for biorefinery production, only parts of the available carbons are usable for fermentation purposes, namely cellulose, laminaria and mannitol. Alginate, that accounts for about 30% of the carbohydrates composition of the macroalgae [48] cannot be fermented with current technology. To understand what the environmental gains would be if alginate was fermentable, this scenario was modeled. We looked at if geographical location and local energy mix has an effect on the environmental impacts related to energy utilities. We assume harvesting the macroalgae as the way to acquire the macroalgae biomass, and we contrast that with if cultivation of the same biomass would have an effect on our results. Finally, we analyzed if transportation and End-of-Life scenario at a different continent would affect the results.

Table 2 Overview of scenarios analyzed

Scenario	Short description
Effects of fermentation of alginate	To identify the effects fermentation of alginate would have on the environmental impacts of macroalgae
Change in geographical location for LA from 3rd generation feedstocks	To identify if changes in energy mix would affect the LCA results, and secondly in which extent the effect of different energy mixes would affect, the US energy mix of base case scenario was tested against the Chinese and Icelandic energy mixes as scenario analysis.
Harvesting of macroalgae	Scenario of harvesting of macroalgae was tested against macroalgae cultivation results from Seghetta et al.[49] for two impact categories assessed with the same LCI assessment method. (Results presented in ESI.8)
Transportation of polymer and End-of-life scenario in Germany	Transportation is not included in the model, and to test if it would have an impact on the results, transportation of polymer and EoL LC stage in a different geographical area, Germany, where recycling percentages are higher than in USA was tested. (Results presented in ESI.9)

Uncertainty assessment

To address the uncertainty we conducted a Monte Carlo analysis. For all LC-stages of the three feedstock processes (inputs and outputs) we estimated the uncertainty using the Pedigree matrix approach[50] with the exception that the maize grain process was adapted from ecoinvent database and therefore already had assigned geometric standard deviations to flows. The uncertainty data was assessed based on the required data quality criteria (reliability, completeness, temporal correlation, geographical correlation, further technological correlation, and basic uncertainty factor) and uncertainty factors were calculated giving the squared geometric standard deviation for the foreground process. For the background processes already assigned geometric standard deviations in ecoinvent were used. We did

10.000 iterations for the Monte Carlo simulations.

Result and discussion

Dominance of different life cycle stages across feedstock generations

When producing LA from corn, production of biomass and the biorefinery process are the main contributing LC-stage including global warming, stratospheric ozone depletion, freshwater eutrophication, marine eutrophication, human carcinogenic toxicity, land use, and water consumption (see Table 3).

Table 3 Midpoint environmental impact results for lactic acid production from three feedstock generations, including Monte Carlo uncertainty analysis, TRL and hotspots (expressed as % contribution of life cycle stages, BM: Biomass, BR: Biorefinery, PM: Polymerization, and EoL: End-of-life.

		TRL 8-9	Lactic acid from corn Life cycle stages			
Impact categories	Unit	Tot. Res. (2.5th – 97.5th%)	BM	BR	PM	EoL
Global warming	kg CO2 eq	4.2 (1.3 – 4.8)	47.5%	59.1%	1.8%	-8.5%
Stratospheric ozone depletion	kg CFC11 eq	2x10^{-5} (8.4x10^{-06} – 1.6x10^{-05})	95.8%	3.4%	0.1%	0.6%
Ionizing radiation	kBq Co-60 eq	-0.22 (-1.2 – 0.015)	14.9%	-121.6%	3.6%	3.1%
Ozone formation, Human health	kg NOx eq	7.1x10-^{3} (4.4x10^{-3} – 8.9x10^{-3})	37.7%	75.8%	2.4%	-15.8%
Fine particulate matter exposure	kg PM2.5 eq	1.8x10^{-2} (5.8x10^{-3} – 3.6x10^{-2})	18.2%	82.8%	1.0%	-1.9%
Ozone formation, Terrestrial ecosystems	kg NOx eq	7.3x10^{-3} (4.5x10^{-3} – 9.1x10^{-3})	37.6%	77.3%	2.7%	-17.5%
Terrestrial acidification	kg SO2 eq	0.077 (0.037 – 0.13)	20.3%	81.2%	0.4%	-1.8%
Freshwater eutrophication	kg P eq	5.2x10^{-5} (-3.1x10^{-3} – 1.7x10^{-3})	978.6%	-1057.4%	120.0%	58.8%
Marine eutrophication	kg N eq	5.6x10^{-3} (1.8x10^{-3} – 3.5x10^{-3})	84.8%	9.3%	0.1%	5.9%
Terrestrial ecotoxicity	kg 1,4-DCB	12 (7.1 – 19)	20.0%	72.8%	10.8%	-3.6%
Freshwater ecotoxicity	kg 1,4-DCB	0.12 (-0.055 – 0.35)	25.1%	-6.1%	3.6%	77.4%
Marine ecotoxicity	kg 1,4-DCB	0.17 (-0.064 – 0.49)	19.8%	2.9%	4.1%	73.2%
Human carcinogenic toxicity	kg 1,4-DCB	0.048 (-0.19 – 0.24)	73.7%	34.4%	10.0%	-18.1%
Human non-carcinogenic toxicity	kg 1,4-DCB	4.3 (-0.041 – 13)	7.6%	37.7%	4.9%	49.8%
Land use	m2a crop eq	1.4 (0.99 – 1.9)	93.5%	6.5%	0.1%	0.0%
Mineral resource scarcity	kg Cu eq	0.012 (7.8x10^{-03} – 1.8x10^{-02})	33.6%	59.0%	11.8%	-4.4%
Fossil resource scarcity	kg oil eq	0.65 (6.4x10^{-03} – 1.1)	36.6%	137.8%	2.7%	-77.1%
Water consumption	m3	0.43 (-2.0 – 2.8)	75.1%	26.2%	0.6%	-1.9%

		TRL 4-5	Lactic acid from corn stover Life cycle stages			
Impact categories	Unit	Tot. Res. (2.5th – 97.5th%)	BM	BR	PM	EoL
Global warming	kg CO2 eq	7.9 (6.0 – 9.2)	36.7%	66.8%	1.0%	-4.5%
Stratospheric ozone depletion	kg CFC11 eq	3.3x10^{-6} (2.6x10^{-6} – 4.0x10^{-6})	40.7%	54.7%	0.9%	3.7%
Ionizing radiation	kBq Co-60 eq	0.3 (0.038 – 1.3)	0.0%	95.2%	2.6%	2.2%
Ozone formation, Human health	kg NOx eq	1.2x10^{-2} (9.7x10^{-3} – 1.4x10^{-2})	47.7%	60.4%	1.4%	-9.5%
Fine particulate matter exposure	kg PM2.5 eq	0.024 (0.015 – 0.035)	22.0%	78.7%	0.7%	-1.4%
Ozone formation, Terrestrial ecosystems	kg NOx eq	1.2x10^{-2} (9.9x10^{-3} – 1.5x10^{-2})	47.4%	61.5%	1.6%	-10.5%
Terrestrial acidification	kg SO2 eq	0.064 (0.038 – 0.11)	16.2%	85.5%	0.4%	-2.2%
Freshwater eutrophication	kg P eq	3.5x10^{-3} (1.3x10^{-3} – 6.6x10^{-3})	35.3%	62.0%	1.8%	0.9%
Marine eutrophication	kg N eq	1.3x10^{-3} (8.7x10^{-4} – 1.9x10^{-3})	12.1%	61.5%	0.3%	26.1%
Terrestrial ecotoxicity	kg 1,4-DCB	22 (14 – 37)	42.1%	53.9%	6.0%	-2.0%
Freshwater ecotoxicity	kg 1,4-DCB	0.33 (0.16 – 0.69)	25.8%	45.3%	1.3%	27.6%
Marine ecotoxicity	kg 1,4-DCB	0.46 (0.22 – 0.95)	26.3%	45.6%	1.5%	26.6%
Human carcinogenic toxicity	kg 1,4-DCB	0.29 (0.095 – 0.82)	38.3%	63.1%	1.7%	-3.0%
Human non-carcinogenic toxicity	kg 1,4-DCB	9.5 (3.7 – 23)	27.0%	47.9%	2.2%	22.9%
Land use	m2a crop eq	0.17 (0.17 – 0.33)	0.0%	99.8%	0.5%	-0.4%
Mineral resource scarcity	kg Cu eq	0.011 (0.011 – 0.031)	0.0%	91.6%	13.4%	-5.0%
Fossil resource scarcity	kg oil eq	1.8 (1.2 – 2.2)	39.5%	87.8%	1.0%	-28.3%
Water consumption	m3	0.15 (-3.6 – 3.2)	13.5%	90.1%	1.6%	-5.2%

		TRL 2-3	Lactic acid from macroalgae			
			Life cycle stages			
Impact categories	Unit	Tot. Res. (2.5^{th} – 97.5^{th}%)	BM	BR	PM	EoL
Global warming	kg CO2 eq	11 (7.14 – 15.2)	50.9%	51.6%	0.7%	-3.2%
Stratospheric ozone depletion	kg CFC11 eq	$5.8x10^{-6}$ ($3.6x10^{-6}$ – $1.0x10^{-5}$)	51.4%	46.1%	0.5%	2.0%
Ionizing radiation	kBq Co-60 eq	0.27 (-0.13 – 2.7)	45.4%	51.8%	1.5%	1.3%
Ozone formation, Human health	kg NOx eq	0.015 (0.011 – 0.022)	54.5%	51.6%	1.1%	-7.2%
Fine particulate matter exposure	kg PM2.5 eq	0.02 ($8.6x10^{-03}$ – $3.2x10^{-02}$)	50.1%	50.8%	0.9%	-1.7%
Ozone formation, Terrestrial ecosystems	kg NOx eq	0.016 (0.011 – 0.022)	54.9%	51.9%	1.2%	-8.0%
Terrestrial acidification	kg SO2 eq	0.045 (0.032 – 0.061)	51.1%	51.4%	0.6%	-3.1%
Freshwater eutrophication	kg P eq	$3.0x10^{-3}$ ($9.0x10^{-5}$ – $1.0x10^{-2}$)	43.6%	53.8%	1.8%	0.9%
Marine eutrophication	kg N eq	$1.1x10^{-3}$ ($7.3x10^{-4}$ – $2.0x10^{-3}$)	9.2%	62.9%	0.3%	27.6%
Terrestrial ecotoxicity	kg 1,4-DCB	29 (17 – 73)	41.9%	55.4%	4.0%	-1.3%
Freshwater ecotoxicity	kg 1,4-DCB	0.3 (0.078 – 0.74)	28.3%	42.3%	1.3%	28.1%
Marine ecotoxicity	kg 1,4-DCB	0.42 (0.13 – 1)	28.8%	42.9%	1.5%	26.8%
Human carcinogenic toxicity	kg 1,4-DCB	0.26 (0.096 – 1.2)	55.6%	45.5%	1.4%	-2.5%
Human non-carcinogenic toxicity	kg 1,4-DCB	7.1 (2.9 – 22)	24.7%	46.8%	2.5%	26.0%
Land use	m2a crop eq	1.1 (0.74 – 1.4)	86.4%	13.6%	0.1%	-0.1%
Mineral resource scarcity	kg Cu eq	0.022 (0.013 – 0.047)	51.7%	44.5%	6.1%	-2.3%
Fossil resource scarcity	kg oil eq	3.1 (1.9 – 4.6)	61.6%	53.9%	0.6%	-16.1%
Water consumption	m3	0.46 (-2.64 – 3.2)	43.0%	58.2%	0.5%	-1.7%

iLUC contributes the highest to assessed impacts of biomass production, e.g. causing an increase of 35% for marine eutrophication impacts (for calculation of iLUC and contribution to assessed impacts, see ESI.6) demonstrating the importance of assessing iLUC for first generation feedstocks. When looking at the different hotspots of LA from corn in Table 3, the categories global warming, ozone formation, fine particulate matter exposure, and terrestrial acidification, terrestrial ecotoxicity, mineral resource scarcity, and fossil resources scarcity, are driven by high contributions from the biorefinery stage. The single highest contributing process to the biorefinery is the use of triethanolamine in the purification stage of the LA production followed by treatment of refinery sludge. (see LCI for 1st generation process in ESI.1).

When producing LA from corn stover, the biorefinery stage becomes the dominant stage for all impact categories (see Table 3). High energy utility demand is the single most contributing input, as well as use of triethanolamine in the purification of the LA and handling of refinery sludge. Another reason for the high freshwater eutrophication impacts lies in the increased fertilizer use needed to substitute the nutrients that the corn-stover would otherwise have provided to the cornfields if not used as feedstock. (See LCI for 2nd generation process in ESI.2).

For LA production from macroalgae, as can be seen in Table 3, the high utility inputs to biomass harvesting, including drying in the biomass stage, and intensive energy usage in the biorefinery stage, result in environmental hotspots that are almost the same for both LC stages. This is however not the case for the impact categories marine eutrophication, freshwater ecotoxicity, marine ecotoxicity, and human non-carcinogenic toxicity because of the composition of the energy mix used in the LCA model (see SI.3). Process inputs, e.g. use of neutralizing agent, phosphate fertilizer and calcium carbonate in addition to energy utilities in the biorefinery stage cause the highest environmental hotspots for these impact categories. The general trend across feedstock generations is that utilities are the biggest contributing processes (energy inputs), including energy for drying of biomass, steam and electricity use, accounting for up to 86% of the total environmental impacts. Energy utility inputs are also the largest contributor to the other assessed impact categories. This especially is the case for the 3rd generation, which is the most energy intensive across the three assessed feedstock generations, partly because it is also the least technologically optimized generation and also it may be the least energy efficient process if same TRL is considered. This is also reflected in the uncertainty results in Table 3 for all assessed impact categories, showing highest uncertainty for macroalgae scenarios. The high utility demand in the 3rd generation process caused by the high liquid concentration of the macroalgae requiring a drying process before the biomass usage for fermentation (see ESI.3).

Polymerization does not appear as a hotspot in the overall impacts over the cradle-to-grave study in any of the feedstock processes, with one exception. That is for freshwater and marine ecotoxicity, and human non-cancer toxicity impacts in the 1st generation feedstock production process of LA. This is because of proportionally high electricity and chemical inputs, as reported by the IHS Markit© database. The IHS Markit© database is widely used by industry and was selected for that reason. However, it is noteworthy that its reliability has not been checked nor validated; this uncertainty source was factored in when running the Monte Carlo analysis (see ESI.5).

The process in the EoL stage, which are independent of the biomass process, has an overall positive effect on the individual environmental impacts. This is because of the positive impact originating in the proportion of the plastics that are recycled and thereby avoid production of virgin conventional plastics, e.g. 92% reduction of fossil resource scarcity impacts per kilogram LA from corn. Not all impact indicators are however positively affected at the EoL stage, e.g. over 73% increase in freshwater- and marine ecotoxicity. This relates to emission impacts from waste management, municipal waste incineration and sanitary landfilling (see Table 3). If the EoL LC-stage is excluded, neither the these positive and negative impacts would have been caught, demonstrating that it is essential to include the EoL stage when assessing biochemicals.

Tradeoffs between individual impact categories

When looking at individual impact categories, special attention should be given to global warming, land use and water use, eutrophication, ecotoxicity, and indirect land use change for the first generation biomass[7] (see Table 4). When we compare our results with Ingrao et al. (2015), Vink et al. (2015) other studies of similar scope and system boundaries, starting with global warming impacts of LA/PLA production from corn, our results vary from e.g. being within factors of 0.6[51]-2.5[9] for this impact category. For land use, compared to Groot et al. (2010) and Madival et al. (2009), our results are more than one order of magnitude lower[52] as well as for freshwater eutrophication[53] in one study, while Papong et al. (2014) presents very similar results[54]. For marine eutrophication our results are factor 0.3 lower than Landis et al. (2010)[55] present. For water use (Vink et al. 2007)[56] and ecotoxicity (Landis et al. 2010)[55] our results are more than factor 6 higher and respectively more than factor 10 higher. To explain the differences, at least in some extent, in our study we assess iLUC impacts that affect the impact results in two ways, because of intensification of corn production and transformation to arable land because of increased demand for corn products, leading to increasing global warming impacts.[52,53,54] Other factors explaining that our results are higher is the fact that for example Vink et al.[9] assess a specific power plant that uses a highly specific energy mix with a high renewable energy usage[9]. In our study, we use a country specific energy mix that does not take into account local geographical scenarios of energy inputs leading to higher results in our study compared to literature[9,57].

Table 4 Global warming impacts (kg CO2 eq.) of Lactic acid production from 3 generations of biomass, from cradle-to-grave (for calculation of table see ESI.10)

	Unit	Global warming	Global warming without iLUC	Biogenic carbon storage	Net biogenic carbon emissions
Lactic acid from corn	kg CO2 eq	4.19E+00	3.67E+00	2.55E+00	1.64E+00
Lactic acid from corn-stover	kg CO2 eq	7.90E+00	-	2.91E+00	4.99E+00
Lactic acid from macroalgae	kg CO2 eq	1.10E+01	-	4.51E+00	6.52E+00

Previously-published studies assessing LA/PLA production from 2nd generation biomass show lower global warming impacts than we do by more than an order of magnitude[57,58]. This is because we assign fertilizer value per kg corn-stover needed to fulfil the functional unit, as part of the system expansion (i.e. benefits from avoidance of synthetic fertilizer production). In addition, our study assesses the whole product life cycle, and not only the production stage like many previous studies. These discrepancies are thus additional evidence that encompassing a full life cycle is essential in such impact assessment studies.

Macroalgae as a 3rd generation feedstock use for biorefineries is not (yet) a common practice but has large potential because the biomass is accessible almost everywhere along coastal areas. Seghetta et al.[49] studied the life cycle impacts of cultivated macroalgae utilization for energy and feed production. Their results for global warming impacts are higher than ours, because for cultivation practices, their impacts from infrastructure are higher and transportation to and from cultivation areas is more frequent than in our harvesting scenario. For the 3rd generation feedstock, the energy process supporting the drying of the biomass before fermentation is the main driver of climate change impacts.

It is important to note that the level of uncertainty has a big effect on the above comparisons because of differentiating assumptions made for, as example, process design of emerging feedstocks and how heat integration is incorporated[57].

Impacts are highly influenced by if the feedstock is grown, collected or harvested, like terrestrial acidification and marine eutrophication that are highly influenced by fertilizer and or pesticide use, leading to that the environmental hotspots shift between the different feedstock processes.

Optimization potential of using macroalgae as feedstock

This section provides information and discusses different data availability and assumptions made for different TRLs in LCA studies (see Table 5) as well the scenario analysis. Firstly we address optimization of the process itself, and secondly optimization by outsourcing or relocate the production in an appropriate country.

Table 5 List of limitations and assumptions made for assessing the LC impacts for production of LA from three generations of feedstocks

	Corn	Corn stover	Macroalgae
Single product production process	Conceptual biochemical production		
Scaling up	Commercial biorefinery size assumed and scaling up done by applying techno-economic assessment (TEA) for each of the biorefinery LC stage of the feedstock processes		
Level of detail for each of the LC-stages	Feedstock cultivation – High Biorefinery – High Polymerization – Medium EoL - Medium	Feedstock harvesting – High Biorefinery – High Polymerization – Medium EoL - Medium	Feedstock harvesting – Low Biorefinery – High Polymerization – Medium EoL - Medium
Optimization potential	Low	Medium – Process optimization (lignin recovery)	High – Process optimization (optimize for utilities), integrated process development (e.g. alginate fermentation)
Uncertainty (see ESI.5)	Low	Medium	High

Figure 4 presents the Monte Carlo results graphically for the different feedstock processes (for further information see ESI.5 on process of calculating the geometric standard deviation used for uncertainty analysis). Regarding optimization potential of the macroalgae feedstock process, based on the results for human health and ecosystem quality, uncertainty ranges across generations are overlapping in the extent that with the current overall uncertainty it is difficult to identify the most relevant optimization potential in macroalgae scenario. However trends can be observed, in particular the fact that the performances of 3rd generation can be dramatically improved relative to other 2 generations if the drying process is drastically optimized or removed, or by 59-83%. Simulation without removal show the 3rd generation macroalgae to perform better than the other 2 generations, with the exception of natural resources reaching the level of impacts of the 2nd generation.

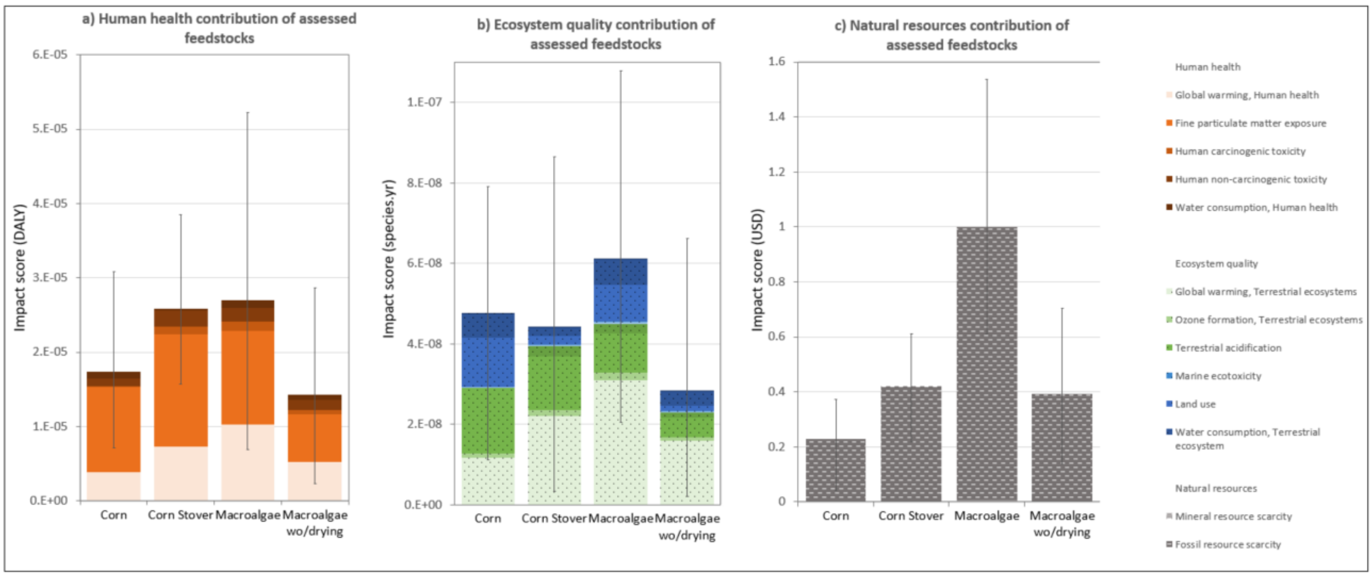

Figure 4 Actual LCA results at damage level including uncertainty ranges for the different generations of feedstock processes. In addition, to assess the optimization potential of not drying the 3rd generation biomass, the figure presents the results and uncertainty range for that scenario. iLUC effects are included in the results.

For scenario analysis we tested different background data inputs. The results are presented in Figure 5a. Fermenting alginate would yield an overall reduction of environmental impacts of 38%, meaning we weighted all impact categories equally. The highest reduction in impacts affecting damages on human health are global warming (15%) and fine particulate matter exposure (19%); both related to the reduction in biomass needed per kg product, resulting in less energy needed for drying of biomass. The lower demand for biomass also drives the reduction in damages on ecosystem quality, where the highest reduction is associated with global warming impacts on terrestrial ecosystems (18%) and land use impacts (9%). For natural resources, fossil resource scarcity is reduced by 36% because of decreased biomass demand.

In addition, increased feedstock yield when fermenting the alginate per FU in China (CN) also sees benefits in the reduction of energy-related impacts. This applies to for example global warming impacts on terrestrial ecosystems and terrestrial acidification (Figure 5b). Despite the energy mix in Iceland (ICE) being more environmentally friendly, harvesting the biomass is driven by energy generation from fossil fuels. By fermenting alginate we can reduce natural resource impacts by more than 50% reduction. For human health and ecosystem quality we see a similar trend (see Figure 5c) as in Figures 6a and c, just below 30% reduction in impacts.

Production location also influences the optimization potential of the macroalgae feedstock process. Changing the location of the LA production from US to CN, we see a drastic increase in damages on human health (108%) and on ecosystem quality (95%). The composition of the energy inputs causes these changes. Damages on natural resources on the other hand are reduced by 30%. The reason for this is that the country-specific ecoinvent processes chosen for modeling the background of the heat-mix production composition in the US cause higher impacts related to fossil resources scarcity than the background processes chosen from ecoinvent to model the CN heat-mix production distribution (see Figure 5d) (See ESI.7).

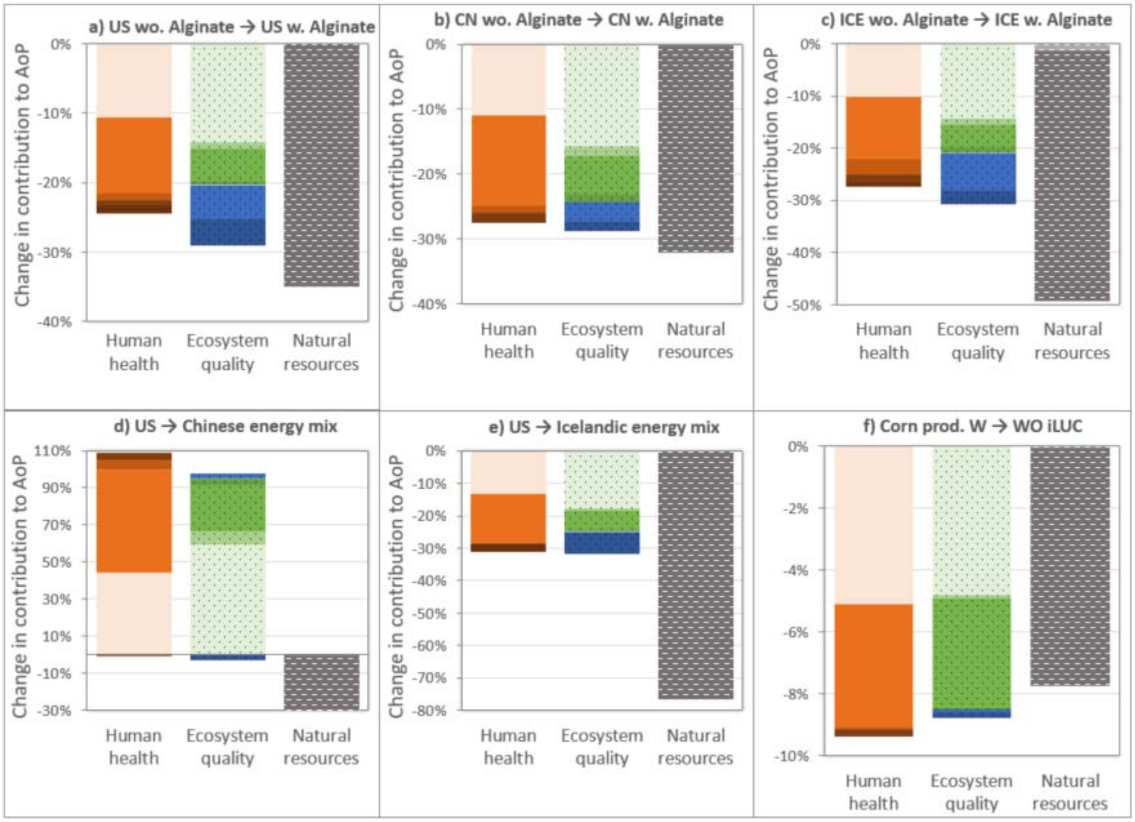

Figure 5 Changes in environmental hotspots at area of protection when assessing the future scenario of fermenting alginate for lactic acid production from macroalgae (5a-5c). Sensitivity scenarios assessing changes in environmental damages when changing geographical location of cradle-to-grave assessments from US to China and Iceland respectively (5d and 5e). Sensitivity scenario showing reduction in impacts when excluding iLUC damages (5f)

Since the energy inputs have the single most dominating impact on the production process of LA from macroalgae, we evaluated if the results change when the considered energy mix has a different composition. When opposing the Chinese and Icelandic mix to that of US, we find that the trend in reduction of damages is only the same for damages to natural resources. Fossil resource scarcity would be reduced by 76,5% in Iceland (ICE) and by 30% in CN. For CN this might come as a surprise, but occur because the lack of a country specific CN heat-mix production processes in the econinvent database and therefore average world processes were selected (for further information about chosen inputs, see ESI.7).

For both human health and ecosystem quality damages there is an opposite trend for CN and ICE when compared to the base case. For Iceland, where 87% of the heat and electricity production stems from renewable resources[59], we see a reduction in damages to human health

(31%) and ecosystem quality (33%), compared to damage increase for human health (130%) and ecosystem quality (99%). USA and China relies to a great extent on fossil resources for their energy mix[60,61]. Geothermal energy conversion emits large quantities of fine particulate matter, which explains the high related reduction presented in Figure 5b for Iceland. Reduction in damages on ecosystem quality differs on the other hand somewhat as compared to the US and CN. This mostly relates to the fact that hydropower energy conversion demands large areas resulting in the land used savings for ICE.

Conclusion and future research needs

In our study, we have demonstrated how LCA can effectively be applied as a process optimization tool at early biochemical design and development stages. Our results show that different biochemical production systems, in our case lactic acid, have different environmental hotspots to focus on for maximizing overall environmental sustainability. As an example, we illustrate that using bacteria in the biorefinery process, which are able to ferment macroalgae without the need to dry the macroalgae first, can significantly reduce global warming and several other impacts, rendering LA production from macroalgae the best-in-class option among different bio-feedstocks.

Applying LCA gives the bio-based industry the opportunity to incorporate environmental sustainability as important decision support aspect. It helps pinpointing relevant environmental hotspots, possibly resulting in the identification of optimization potentials along the entire biochemical life cycle.

Exploiting the potential for maximizing the yield of biofeedstocks would reduce environmental impacts by around 30%. Similar improvements can be reached by producing LA from macroalgae in a country, like Iceland, compared to e.g. the United States, based on differences in the available energy mix.

Finally, by assessing iLUC impacts, we enable considering in any optimization decision the indirect impacts caused by producing LA from 1st generation biomass like corn.

To increase the reliability and accuracy of LCA results for biochemicals, future research should focus on reducing uncertainty of inventory data. This requires access to industrial data and improving assumptions in system models with respect to for example upscaling of early-stage technologies.

To facilitate the assessment of environmental sustainability of biochemicals in practice, we need to systematically combine LCA and TEA for optimal decision support that considers both economic and environmental sustainability aspects. Our study provides a solid starting point for considering the environmental sustainability aspects, and is also applicable to other biochemicals and feedstocks.

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Electronic Supporting Information (ESI) for:

Environmental hotspots of different lactic acid production systems

Ólafur Ögmundarson,[a,b] Sumesh Sukumara[b], Alexis Laurent[a] and Peter Fantke[a*]

[a] Quantitative Sustainability Assessment Division, Department of Management Engineering, Technical University of Denmark, Kgs. Lyngby, Denmark

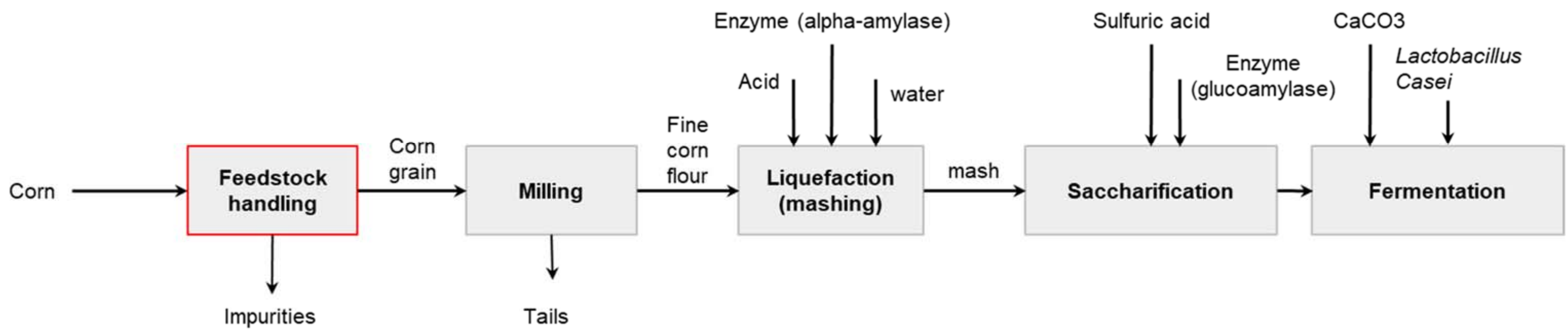
[b] The Novo Nordisk Foundation Center for Biosustainability, Technical University of Denmark, Kgs. Lyngby, Denmark

Corresponding author: phone: +45 45254452, email: pefan@dtu.dk

Contents

SI.1 Lactic acid from corn process[1]

1st Generation LA production: Upstream



Saccharification:
STARCH(CIPSD) + WATER --> GLUCOSE

Fermentation
Frac. conversion GLUCOSE --> 2 LACTI-01
Frac. conversion 2 LACTI-01 + CACO3(CIPSD) --> CA-LAC + WATER + CO2

Inventory processes divided by LC stages

Biomass production[2]

Products					
Maize grain {US}| production | Cut-off, U	1	kg			
Avoided products					
Resources					
Carbon dioxide, in air	in air	1.451188	kg	Lognormal	1.2214
Energy, gross calorific value, in biomass	biotic	15.91	MJ	Lognormal	1.1192
Occupation, annual crop	land	0.62623	m2a	Lognormal	1.4918
Transformation, from annual crop	land	1.0735	m2	Lognormal	1.9993
Transformation, to annual crop	land	1.0735	m2	Lognormal	1.9993
Materials/fuels					
Arable land [NPP0 as C] ecoinvent v3.4 link	1.8	kg	Undefined		iLUC
[sulfonyl]urea-compound {GLO}| market for | Cut-off, U	5.63E-07	kg	Lognormal	1.209	
Acetamide-anillide-compound, unspecified {GLO}| market for | Cut-off, U	1.02E-05	kg	Lognormal	1.209	
Ammonia, liquid {RoW}| market for | Cut-off, U	0.008436	kg	Lognormal	1.209	
Ammonium nitrate, as N {GLO}| market for | Cut-off, U	0.004893	kg	Lognormal	1.209	
Application of plant protection product, by field sprayer {RoW}| market for application of plant protection product, by field sprayer | Cut-off, U	8.25E-05	ha	Lognormal	1.2214	
Atrazine {GLO}| market for | Cut-off, U	9.25E-05	kg	Lognormal	1.209	
Benzoic-compound {GLO}| market for | Cut-off, U	1.73E-06	kg	Lognormal	1.209	
Bipyridylium-compound {GLO}| market for | Cut-off, U	6.14E-07	kg	Lognormal	1.209	
Combine harvesting {GLO}| market for | Cut-off, U	9.59E-06	ha	Lognormal	1.2214	
Cyclic N-compound {GLO}| market for | Cut-off, U	5.78E-07	kg	Lognormal	1.209	
Dinitroaniline-compound {GLO}| market for | Cut-off, U	2.48E-06	kg	Lognormal	1.209	
Diphenylether-compound {GLO}| market for | Cut-off, U	1.93E-07	kg	Lognormal	1.209	
Drying of maize straw and whole-plant {GLO}| market for | Cut-off, U	0.40984	l	Lognormal	1.5051	
Fertilising, by broadcaster {GLO}| market for | Cut-off, U	5.49E-05	ha	Lognormal	1.2214	
Glyphosate {GLO}| market for | Cut-off, U	1.97E-05	kg	Lognormal	1.209	
Irrigation {US}| market for | Cut-off, U	0.244743	m3	Lognormal	1.0936	
Lime {GLO}| market for | Cut-off, U	0.030409	kg	Lognormal	1.209	
Maize seed, for sowing {GLO}| market for | Cut-off, U	0.021471	kg	Lognormal	1.209	
Metolachlor {GLO}| market for | Cut-off, U	4.29E-05	kg	Lognormal	1.209	
Nitrile-compound {GLO}| market for | Cut-off, U	9.39E-07	kg	Lognormal	1.209	
Organophosphorus-compound, unspecified {GLO}| market for | Cut-off, U	9.71E-06	kg	Lognormal	1.209	
Pesticide, unspecified {GLO}| market for | Cut-off, U	6.28E-05	kg	Lognormal	1.209	
Phenoxy-compound {GLO}| market for | Cut-off, U	2.49E-06	kg	Lognormal	1.209	
Phosphate fertiliser, as P2O5 {GLO}| market for | Cut-off, U	0.005848	kg	Lognormal	1.209	
Potassium chloride, as K2O {GLO}| market for | Cut-off, U	0.007193	kg	Lognormal	1.209	
Pyrethroid-compound {GLO}| market for | Cut-off, U	1.23E-06	kg	Lognormal	1.209	
Sowing {GLO}| market for | Cut-off, U	3.8E-05	ha	Lognormal	1.2214	
Tillage, cultivating, chiselling {GLO}| market for | Cut-off, U	0.000126	ha	Lognormal	1.2214	
Tillage, currying, by weeder {GLO}| market for | Cut-off, U	0.0002	ha	Lognormal	1.2214	
Tillage, harrowing, by spring tine harrow {GLO}| market for | Cut-off, U	0.000249	ha	Lognormal	1.2214	
Tillage, ploughing {GLO}| market for | Cut-off, U	8.6E-05	ha	Lognormal	1.2214	
Transport, tractor and trailer, agricultural {GLO}| market for | Cut-off, U	0.002858	tkm	Lognormal	1.8221	
Triazine-compound, unspecified {GLO}| market for | Cut-off, U	2.43E-06	kg	Lognormal	1.209	
Urea, as N {GLO}| market for | Cut-off, U	0.003543	kg	Lognormal	1.209	

Electricity/heat						
Emissions to air						
Ammonia	low. pop.	0.001018	kg		Lognormal	1.3269
Dinitrogen monoxide	low. pop.	0.000398	kg		Lognormal	1.5639
Nitrogen oxides	low. pop.	8.35E-05	kg		Lognormal	1.5639
Water/m3		0.170072	m3		Lognormal	1.4918
Emissions to water						
Nitrate	groundwat	0.011639	kg		Lognormal	1.5639
Phosphate	river	7.51E-06	kg		Undefined	
Phosphorus	river	9.75E-05	kg		Lognormal	1.5639
Water, US	river	0.014934	m3		Lognormal	1.4918
Water, US	groundwat	0.059737	m3		Lognormal	1.4918
Emissions to soil						
2,4-D	agricultura	3.04E-06	kg		Lognormal	1.3269
Acetamide	agricultura	1.01E-06	kg		Lognormal	1.3269
Acetochlor	agricultura	5.94E-05	kg		Lognormal	1.3269
Alachlor	agricultura	4.14E-06	kg		Lognormal	1.3269
Atrazine	agricultura	9.25E-05	kg		Lognormal	1.3269
Bifenthrin	agricultura	2.17E-07	kg		Lognormal	1.3269
Bromoxynil	agricultura	6.5E-07	kg		Lognormal	1.3269
Cadmium	agricultura	-1.9E-08	kg		Undefined	
Chlorpyrifos	agricultura	4.81E-06	kg		Lognormal	1.3269
Chromium	agricultura	4.45E-08	kg		Lognormal	1.5639
Copper	agricultura	-1.5E-06	kg		Undefined	
Cyfluthrin	agricultura	5.05E-08	kg		Lognormal	1.3269
Cypermethrin	agricultura	3.61E-08	kg		Lognormal	1.3269
Dicamba	agricultura	1.73E-06	kg		Lognormal	1.3269
Diflufenzopyr-sodium	agricultura	1.93E-07	kg		Lognormal	1.3269
Dimethenamid	agricultura	5.03E-06	kg		Lognormal	1.3269
Fipronil	agricultura	2.89E-07	kg		Lognormal	1.3269
Flumetsulam	agricultura	3.37E-07	kg		Lognormal	1.3269
Foramsulfuron	agricultura	3.61E-08	kg		Lognormal	1.3269
Glufosinate	agricultura	1.19E-06	kg		Lognormal	1.3269
Glyphosate	agricultura	1.97E-05	kg		Lognormal	1.3269
Imazapyr	agricultura	4.81E-09	kg		Lognormal	1.3269
Imazethapyr	agricultura	1.68E-08	kg		Lognormal	1.3269
Isoxaflutole	agricultura	5.78E-07	kg		Lognormal	1.3269
Lambda-cyhalothrin	agricultura	2.41E-08	kg		Lognormal	1.3269
Lead	agricultura	2.64E-07	kg		Lognormal	1.5639
Mesotrione	agricultura	1.56E-06	kg		Lognormal	1.3269
Metolachlor	agricultura	4.29E-05	kg		Lognormal	1.3269
Nickel	agricultura	-3.3E-07	kg		Undefined	
Nicosulfuron	agricultura	2.65E-07	kg		Lognormal	1.3269
Paraquat	agricultura	6.14E-07	kg		Lognormal	1.3269
Pendimethalin	agricultura	2.48E-06	kg		Lognormal	1.3269
Permethrin	agricultura	1.08E-07	kg		Lognormal	1.3269
Primisulfuron	agricultura	1.2E-07	kg		Lognormal	1.3269
Prosulfuron	agricultura	2.17E-08	kg		Lognormal	1.3269
Rimsulfuron	agricultura	1.2E-07	kg		Lognormal	1.3269
Simazine	agricultura	2.43E-06	kg		Lognormal	1.3269
Tebupirimphos	agricultura	1.01E-06	kg		Lognormal	1.3269
Tefluthrin	agricultura	7.94E-07	kg		Lognormal	1.3269
Terbufos	agricultura	2.7E-06	kg		Lognormal	1.3269
Zinc	agricultura	-5.9E-06	kg		Undefined	
Final waste flows						
Non material emissions						
Input parameters						
m1	0	Undefined				
iLUC	1	Undefined				
Calculated parameters						
m2	1-m1 = 0					

Biorefinery

Biorefinery LC stage_Lactic acid from corn	1	kg				
Avoided products						Explanation
Electricity, low voltage {US}| market group for | Cut-off, U	9.486137	MJ	Lognormal	1.096		Avoided steam production
Resources						
Water, river, US	1.375105	l	Lognormal	1.096		Process water
Water, river, US	54.676	l	Lognormal	1.096		New cooling water substit. evaporated
Materials/fuels						
Soil pH raising agent, as CaCO3 {GLO}| market for | Cut-off, U	1.749801	kg	Lognormal	1.096		CACO3 - Calcium carbonate - Calcite
Sulfuric acid {GLO}| market for | Cut-off, U	0.649614	kg	Lognormal	1.096		H2SO4
Neutralising agent, sodium hydroxide-equivalent {GLO}| soda ash, dense, to generic market for neutralising agent | Cut-off, U	0.017498	kg	Lognormal	1.096		NAOH - Sodium hydroxide
Triethanolamine {GLO}| market for | Cut-off, U	0.43745	kg	Lognormal	1.096		Amin-Amount from IHS - Only avail. in ecoinvent
Chemical factory, organics {GLO}| market for | Cut-off, U	4E-10	p	Lognormal	1.096		Same as in Polylactide, granulate GLO process in Econinvent
Enzymes {GLO}| market for enzymes | Cut-off, U	0.006044	kg	Lognormal	1.096		Enzyme - IHS
Fodder yeast {GLO}| market for | Cut-off, U	0.0008	kg	Lognormal	1.096		IHS - Only yeast found in inventory
Electricity/heat						
Electricity, low voltage {US}| market group for | Cut-off, U	0.212628	MJ	Lognormal	1.096		Electricity
Electricity, low voltage {US}| market group for | Cut-off, U	0.914979	MJ	Lognormal	1.096		Cooling water recirculation
_Heat_US mix production of_Cut-off_U	14.88524	MJ	Lognormal	2.065		Heat/Steam use
Emissions to air						
Carbon dioxide	0.2752	kg	Lognormal	1.096		
Water	54.676	kg	Lognormal	1.096		
Emissions to water						
Water	1.642497	kg	Lognormal	1.096		Water out
Waste to treatment						
Refinery sludge {RoW}| treatment of, hazardous waste incineration | Cut-off, U	0.492	kg	Lognormal	1.096		H2SO4+NAOH+AMIN+YEAST
Waste gypsum {RoW}| treatment of waste gypsum, sanitary landfill | Cut-off, U	1.98	kg	Lognormal	1.096		CASO4 + CACO3

Polymerization

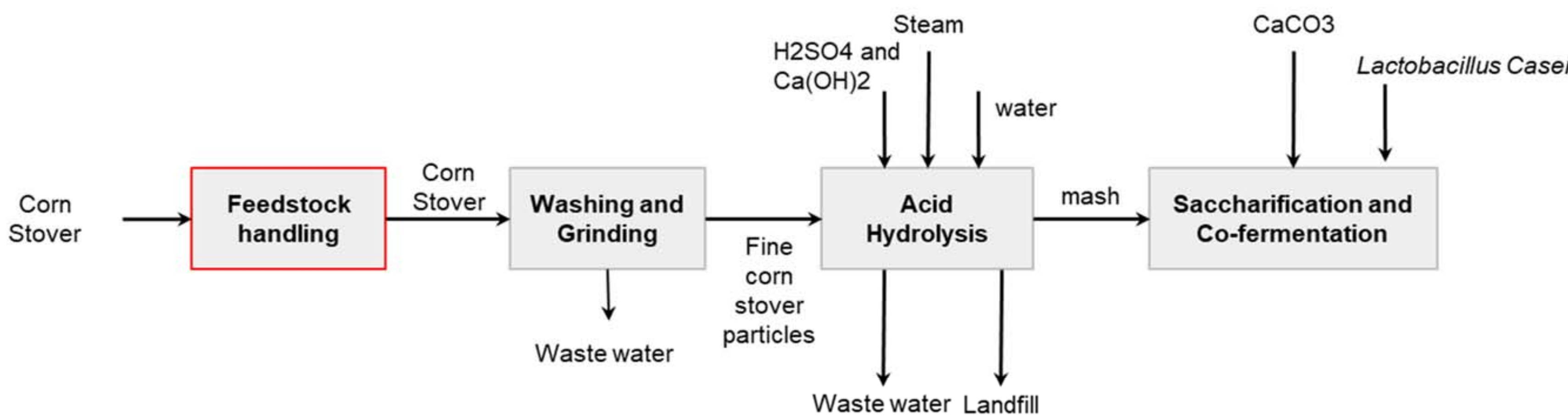
Polymerization	1	kg					
Resources							
Water, cooling, unspecified natural origin/kg		1.887417	kg	Lognormal	1.141		
Materials/fuels							
Tris(2,4-ditert-butylphenyl) phosphite {GLO}| market for tris(2,4-ditert-butylphenyl) phosphite | Cut-off, U	0.001656	kg	Lognormal	1.141			Phosphite stabilizer
Indium tin oxide powder, nanoscale, for sputtering target {GLO}| market for | Cut-off, U	0.000728	kg	Lognormal	1.141			Stannous octoate+Stannous oxide
Electricity/heat							
_Heat_US mix production of_Cut-off_U	0.034145	MJ	Lognormal	1.141			
Electricity, medium voltage {US}| market group for | Cut-off, U	0.129404	MJ	Lognormal	1.141			
Waste to treatment							
Hazardous waste, for incineration {RoW}| market for hazardous waste, for incineration | Cut-off, U	5.39E-05	kg	Lognormal	1.141			Same as for ecoinvent process Polylactide, granulate {GLO}| production | Cut-off, U
Hazardous waste, for incineration {RoW}| market for hazardous waste, for incineration | Cut-off, U	0.002061	kg	Lognormal	1.141			Same as for ecoinvent process Polylactide, granulate {GLO}| production | Cut-off, U
Hazardous waste, for incineration {RoW}| market for hazardous waste, for incineration | Cut-off, U	0.004285	kg	Lognormal	1.141			Same as for ecoinvent process Polylactide, granulate {GLO}| production | Cut-off, U
Waste plastic, mixture {RoW}| market for waste plastic, mixture | Cut-off, U	5.36E-06	kg	Lognormal	1.141			Same as for ecoinvent process Polylactide, granulate {GLO}| production | Cut-off, U
Waste plastic, mixture {RoW}| market for waste plastic, mixture | Cut-off, U	8.23E-05	kg	Lognormal	1.141			Same as for ecoinvent process Polylactide, granulate {GLO}| production | Cut-off, U
Waste plastic, mixture {RoW}| market for waste plastic, mixture | Cut-off, U	0.000912	kg	Lognormal	1.141			Same as for ecoinvent process Polylactide, granulate {GLO}| production | Cut-off, U

End-of-Life

Products				
EoL	1	kg		
Waste to treatment				
_Mixed plastics (waste treatment) {GLO}| recycling of mixed plastics | Cut-off, U	0.35	kg	Lognormal	1.832
Waste plastic, mixture {RoW}| treatment of waste plastic, mixture, municipal incineration | Cut-off, U	0.12	kg	Lognormal	1.832
Waste plastic, mixture {RoW}| treatment of waste plastic, mixture, sanitary landfill | Cut-off, U	0.53	kg	Lognormal	1.832

SI.2 Lactic acid from corn-stover process[3]

2nd Generation LA production: Upstream



Saccharification

CELLULOSE + H2O --> GLUCOLIG
CELLULOSE + H2O --> CELLOBIOSE
CELLULOSE + H2O --> GLUCOSE
CELLOB --> GLUCOSE

Fermentation:

GLUCOSE --> 2 LACTI-01
3 XYLOSE --> 5 LACTI-01
2 LACTI-01 + CACO3(CIPSD) --> CA-LAC + H2O + CO2

Inventory processes divided by LC stages

Biomass – Corn stover fertilizer value

Corn stover fertilizer value	1	kg					
Materials/fuels							
Nitrogen fertiliser, as N {GLO}| nutrient supply from ammonium chloride | Cut-off, U	5.95	g	Lognormal	1.268			Nitrogen (N)
Phosphate fertiliser, as P2O5 {GLO}| market for | Cut-off, U	0.56	g	Lognormal	1.268			Phosphorous (P)
Potassium carbonate {GLO}| market for | Cut-off, U	7.91	g	Lognormal	1.268			Potassium (K)
Calcium nitrate {GLO}| market for | Cut-off, U	2.2	g	Lognormal	1.268			Calcium
Magnesium sulfate {GLO}| market for | Cut-off, U	1.42	g	Lognormal	1.268			Magnesium
Sulfur {GLO}| market for | Cut-off, U	0.47	g	Lognormal	1.268			Solfur
Sodium {GLO}| market for | Cut-off, U	0.15	g	Lognormal	1.268			Sodium
Potassium carbonate {GLO}| market for | Cut-off, U	445	g	Lognormal	1.268			Carbon

Biorefinery

Biorefinery LC stage_Lactic acid from corn stover	1	kg					
Avoided products							
Electricity, low voltage {US}| market group for | Cut-off, U	5.408	MJ	Lognormal	1.527			Avoided steam and heat production
Resources							
Water, river, US	8.654017	l	Lognormal	1.527			Process water
Water, river, US	78.8	l	Lognormal	1.527			New water in per kg. LA
Materials/fuels							
Sulfuric acid {GLO}| market for | Cut-off, U	0.641025	kg	Lognormal	1.527			H2SO4
Triethanolamine {GLO}| market for | Cut-off, U	0.440723	kg	Lognormal	1.527			Amin-Amount from IHS
Neutralising agent, sodium hydroxide-equivalent {GLO}| soda ash, dense, to generic market for neutralising agent | Cut-off, U	0.017652	kg	Lognormal	1.527			NAOH
Ethyl acetate {GLO}| market for | Cut-off, U	0.047743	kg	Lognormal	1.527			Acetate
Lime {GLO}| market for | Cut-off, U	0.041692	kg	Lognormal	1.527			CAH2O2 - Lime
Soil pH raising agent, as CaCO3 {GLO}| lime to generic market for soil pH raising agent | Cut-off, U	0.970855	kg	Lognormal	1.527			CACO3
Chemical factory, organics {GLO}| market for | Cut-off, U	4E-10	p	Lognormal	1.527			Polylactide, granulate GLO process in Ecoinvent
Enzymes {GLO}| market for enzymes | Cut-off, U	0.015602	kg	Lognormal	1.527			Enzymes
Fodder yeast {GLO}| market for | Cut-off, U	0.0008	kg	Lognormal	1.527			IHS
Electricity/heat							
Electricity, low voltage {US}| market group for | Cut-off, U	0.078023	MJ	Lognormal	1.527			Electricity
Electricity, low voltage {US}| market group for | Cut-off, U	8.764045	MJ	Lognormal	1.527			Cooling water recirculation
_Heat_US mix production of_Cut-off_U	9.226606	MJ	Lognormal	1.527			Steam production
Emissions to air							
Carbon dioxide	0.264236	kg	Lognormal	1.527			
Oxygen	0.043378	kg	Lognormal	1.527			
Furfural	0.016313	kg	Lognormal	1.527			
Nitrogen, atmospheric	0.164551	kg	Lognormal	1.527			
Water	78.8	kg	Lognormal	1.527			
Emissions to water							
Water, US	8.865341	l	Lognormal	1.527			Water out
Waste to treatment							
Refinery sludge {RoW}| treatment of, hazardous waste incineration | Cut-off, U	0.7	kg	Lognormal	1.527			H2SO4+ACETIC-ACID+Amin+NAOH+Yeast+Ash
Waste gypsum {RoW}| treatment of waste gypsum, sanitary landfill | Cut-off, U	1.38	kg	Lognormal	1.527			Gypsum

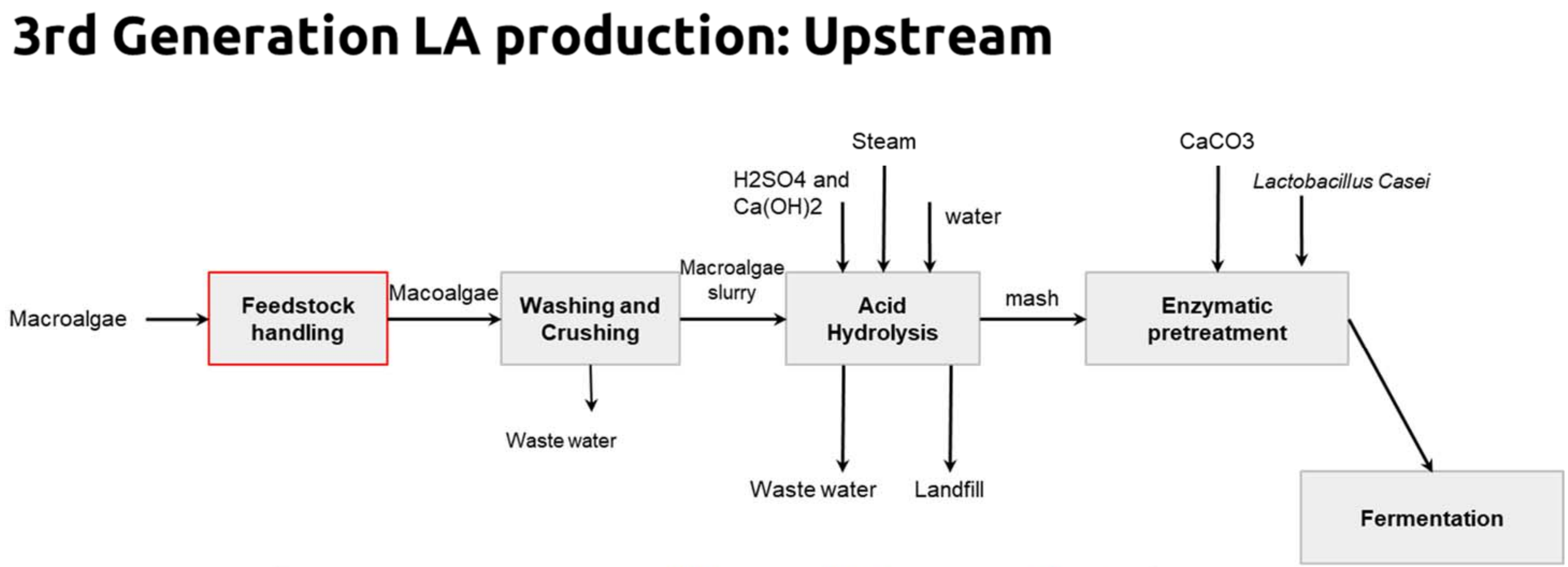
Polymerization

See SI.1

End-of-Life

See SI.1

SI.3 Macroalgae feedstock process[4,5]



	Reactions	ATH + enzymatic conversion	HHW + enzymatic conversion	Heat of reaction (J/kmol)
Laminarin	$(C_6H_{10}O_5)_n + H_2O \longrightarrow C_6H_{12}O_6$	92.5%	75%	-4.8012e+7
Cellulose	$(C_6H_{10}O_5)_n + H_2O \longrightarrow C_6H_{12}O_6$	92.5%	75%	-4.8012e+7
Mannitol	$(C_6H_{14}O_6) \longrightarrow (C_6H_{14}O_6)_{aq}$	95%	95%	-3.0457e+9[a]
Neutralization	$2\,(NH_3) + H_2SO_4 \longrightarrow (NH_4)_2SO_4$	100%	-	-3.53013e+8

	Reaction	Conversion	Component	Heat of reaction (J/kmol)
Oxidation	Mannitol $\longrightarrow$ Fructose + H_2	100%	Mannitol	1.78e+9
LA production	Fructose $\longrightarrow$ 2 Lactic Acid	88.25%	Fructose	-1e+8
Cells growth	Fructose + 0.3704 Proteins + 0.018 DAP $\longrightarrow$ 6 Lactobacillus + 2.4 Water	1.76%	Fructose	-5.07e+9
LA production	Glucose $\longrightarrow$ 2 Lactic Acid	85.02%	Glucose	-1e+8
Cells growth	Glucose + 0.3704 Proteins + 0.018 DAP $\longrightarrow$ 6 Lactobacillus + 2.4 Water	1.79%	Glucose	-5.07e+9
Ca-Lac from LA	2 Lactic acid + Calcium carbonate $\longrightarrow$ Calcium lactate + CO_2 + H_2O	100%	$CaCO_3$	8.48e+7

Inventory processes divided by LC stages

Biomass harvesting and drying

<table>
<tr><td>Algae harvesting</td><td>1</td><td>kg</td><td></td><td></td><td></td><td></td><td></td></tr>
<tr><td></td><td></td><td></td><td></td><td></td><td></td><td></td><td></td></tr>
<tr><td>Materials/fuels</td><td></td><td></td><td></td><td></td><td></td><td></td><td></td></tr>
<tr><td>Petroleum {GLO}| market for | Cut-off, U</td><td>0.00405</td><td>kg</td><td>Lognormal</td><td>1.585</td><td></td><td></td><td>l/kg algae (Asco)</td></tr>
<tr><td>Nylon 6 {GLO}| market for | Cut-off, U</td><td>0.0001</td><td>kg</td><td>Lognormal</td><td>1.585</td><td></td><td></td><td>kg net/kg algae (Asco) each bag used 100 times (10/1000)/100</td></tr>
<tr><td>Reinforcing steel {GLO}| market for | Cut-off, U</td><td>0.00005</td><td>kg</td><td>Lognormal</td><td>1.585</td><td></td><td></td><td>steel/kg algae</td></tr>
<tr><td>Alkyd paint, white, without solvent, in 60% solution state {GLO}| market for | Cut-off, U</td><td>0.00005</td><td>kg</td><td>Lognormal</td><td>1.585</td><td></td><td></td><td></td></tr>
<tr><td></td><td></td><td></td><td></td><td></td><td></td><td></td><td></td></tr>
<tr><td>Emissions to air</td><td></td><td></td><td></td><td></td><td></td><td></td><td></td></tr>
<tr><td>Carbon dioxide, fossil</td><td>0.00629</td><td>kg</td><td>Lognormal</td><td>1.585</td><td></td><td></td><td>Algae cutting algae (petr.usage/kelp petr.usage)*carb em.from trawling</td></tr>
</table>

Biorefinery

Biorefinery LC stage_Lactic acid from macroalgae	1	kg					
Avoided products							
Electricity, low voltage {US}| market group for | Cut-off, U	23.7642	MJ	Lognormal	2.065			Incineration of rest biomass (process unreacted)
Resources							
Water, river, US	44.77226	l	Lognormal	2.065			Washing and process
Water, river, US	64.45075	l	Lognormal	2.065			Cooling water - IHS 0.03% evaporation
Materials/fuels							
Sulfuric acid {GLO}| market for | Cut-off, U	0.450942	kg	Lognormal	2.065			H2SO4
Neutralising agent, sodium hydroxide-equivalent {GLO}| soda ash, dense, to generic market for neutralising agent | Cut-off, U	0.444023	kg	Lognormal	2.065			NAOH - Sodium hydroxide
Enzymes {GLO}| market for enzymes | Cut-off, U	0.013859	kg	Lognormal	2.065			Enzymes
Fodder yeast {GLO}| market for | Cut-off, U	0.0008	kg	Lognormal	2.065			IHS - Only yeast found in inventory
Phosphate fertiliser, as P2O5 {GLO}| market for | Cut-off, U	0.000788	kg	Lognormal	2.065			Diammonium phosphate
Ammonia, liquid {RoW}| market for | Cut-off, U	0.044747	kg	Lognormal	2.065			Ammonia
Calcium carbonate, precipitated {GLO}| market for calcium carbonate, precipitated | Cut-off, U	0.328686	kg	Lognormal	2.065			Limestone
Chemical factory, organics {GLO}| market for | Cut-off, U	4E-10	p	Lognormal	2.065			Preliminary until inventory from Aspen is available
Electricity/heat							
Electricity, low voltage {US}| market group for | Cut-off, U	2.374557	MJ	Lognormal	2.065			Electricity
Electricity, low voltage {US}| market group for | Cut-off, U	24.83835	MJ	Lognormal	2.065			Energy required for water recirculation
_Heat_US mix production of_Cut-off_U	72.0972	MJ	Lognormal	2.065			Steam
Emissions to air							
Hydrogen	0.004317	kg	Lognormal	2.065			H2
Carbon dioxide	0.144528	kg	Lognormal	2.065			CO2
Water	64.45075	kg	Lognormal	2.065			
Emissions to water							
Water	44.77226	kg	Lognormal	2.065			
Waste to treatment							
Refinery sludge {RoW}| treatment of, hazardous waste incineration | Cut-off, U	0.21241	kg	Lognormal	2.065			Fructose-Sulfuric acida-Ammonium sulfate-Diamm. phosp.
Waste gypsum {RoW}| treatment of waste gypsum, inert material landfill | Cut-off, U	0.447089	kg	Lognormal	2.065			Calcium sulfate

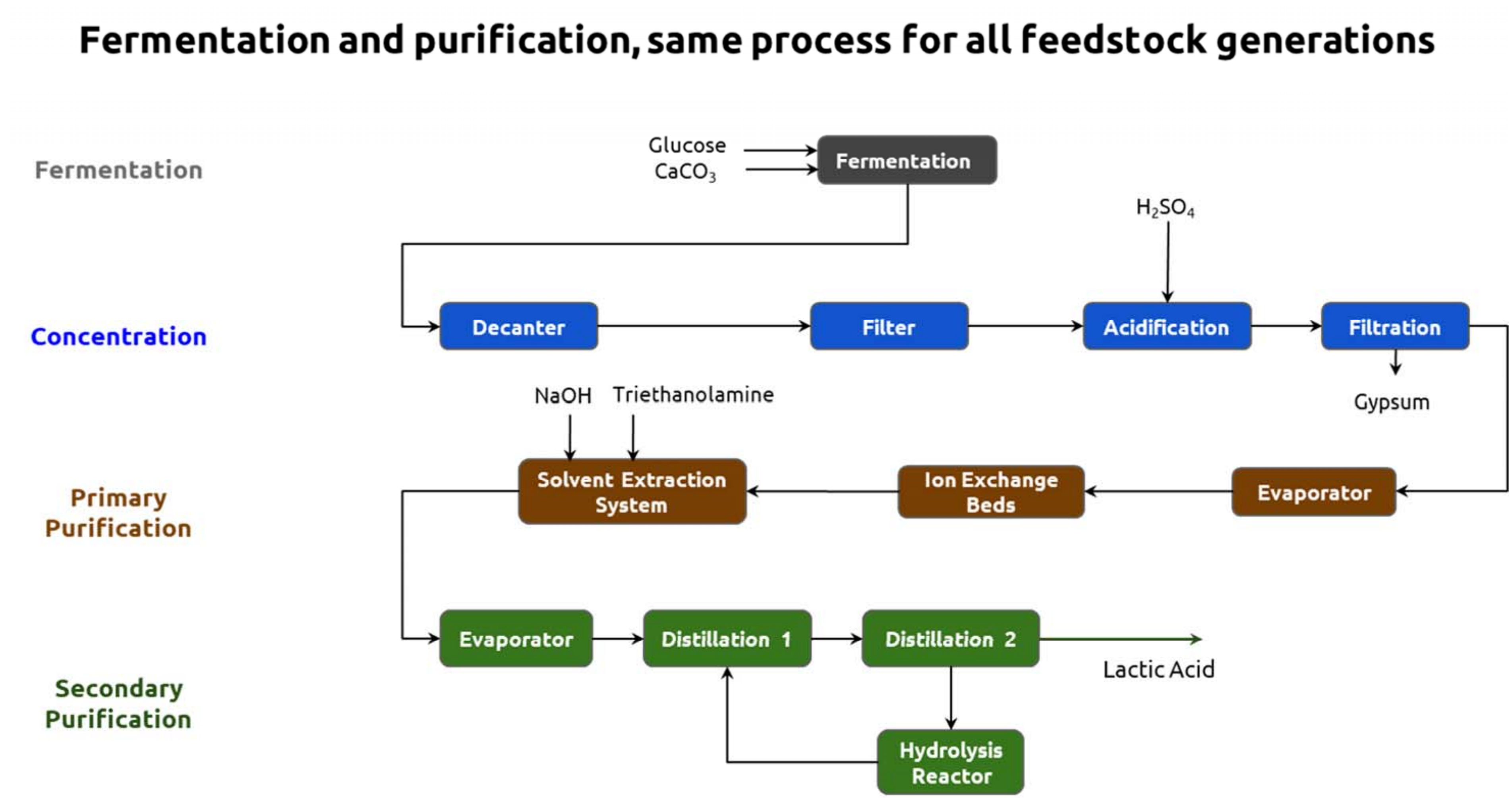
Polymerization

See SI.1

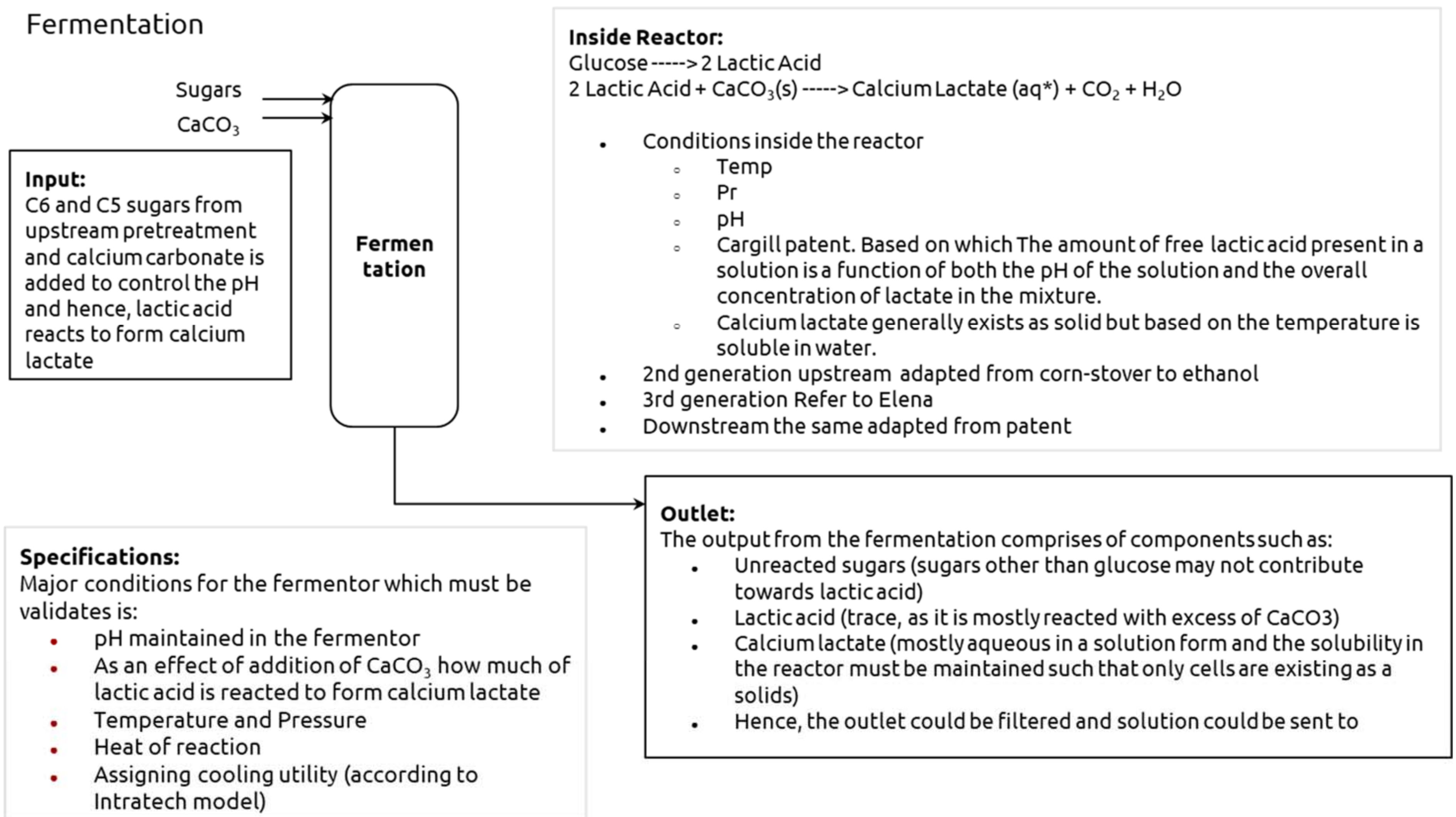
End-of-Life

See SI.1

SI.4 Fermentation and purification, same process for all feedstock generations[1]



Overview of fermentation process[1]



SI.5 Monte Carlo uncertainty analysis

The process of calculating Geometric standard deviation was followed from Owsianiak et al. (2016)[6] and Goedkoop et al. (2016)[7].

Results of calculation of Geometric standard deviation for each assessed LC stage

Feedstock

1st gen	Ecoinvent process already has given Geometric standard deviation from process									
2nd gen	Flows and emissions	U1	U2	U3	U4	U5	Ub	Geometric standard deviation		
		1.1	1.05	1.1	1.02	1.2	1.05	1.268		
3rd gen	Flows and emissions	U1	U2	U3	U4	U5	Ub	Geometric standard deviation		
		1.2	1.1	1.03	1.05	1.5	1.05	1.585		

Biorefinery

Biorefinery										
1st gen	Flows and emissions	U1	U2	U3	U4	U5	Ub	Geometric standard deviation		
		1.05	1.02	1.03	1.001	1.05	1.05	1.096		
2nd gen	Flows and emissions	U1	U2	U3	U4	U5	Ub	Geometric standard deviation		
		1.1	1.05	1.03	1.001	1.5	1.05	1.527		
3rd gen	Flows and emissions	U1	U2	U3	U4	U5	Ub	Geometric standard deviation		
		1.2	1.1	1.03	1.001	2	1.05	2.065		

Polymerization

Polymerization										
Flows and emissions		U1	U2	U3	U4	U5	Ub	Geometric standard deviation		
	US	1.1	1.05	1.03	1.02	1.05	1.05	1.141		

End-of-Life

EoL										
Flows and emissions	U1	U2	U3	U4	U5	Ub	Geometric standard deviation			
US		1.2	1.05	1.5	1.001	1.5	1.05	1.832		

SI.6 iLUC

Calculations are based on Schmidt et al. (2015)[8] - A framework for modelling indirect land use changes in Life Cycle Assessment.

Calculation of amount of Arable land [NPP0 as C] per kg biochemical product from corn

Yield of corn per hectare in the Midwest, USA[9] = 11 t /ha*year

Market for arable land, iLUC user manual[10] = 6.11 t C/ha*year

iLUC of 1 kg corn = Yield/market for arable land = 11 t /ha*year / 6.11 t C/ha*year =1.8 kg C / per kg of corn

Contribution of iLUC to the different impact categories at damage level per kg maize (corn).

Impact category	Unit	Total	Maize grain {US}| production |	Arable land [NPP0 as C] ecoinvent v3.4
Global warming, Human health	DALY	1.00E-06	5.22E-07	4.77E-07
Stratospheric ozone depletion	DALY	5.62E-09	3.11E-09	2.51E-09
Ionizing radiation	DALY	1.48E-10	1.62E-10	-1.36E-11
Ozone formation, Human health	DALY	1.32E-09	1.14E-09	1.84E-10
Fine particulate matter exposure	DALY	1.12E-06	7.45E-07	3.76E-07
Human carcinogenic toxicity	DALY	6.36E-08	5.99E-08	3.71E-09
Human non-carcinogenic toxicity	DALY	4.06E-08	2.32E-08	1.74E-08
Water consumption, Human health	DALY	3.90E-07	3.85E-07	5.93E-09
Global warming, Terrestrial ecosystems	species.yr	3.01E-09	1.57E-09	1.44E-09
Global warming, Freshwater ecosystems	species.yr	8.24E-14	4.30E-14	3.93E-14
Ozone formation, Terrestrial ecosystems	species.yr	1.91E-10	1.65E-10	2.65E-11
Terrestrial acidification	species.yr	1.79E-09	8.70E-10	9.24E-10
Freshwater eutrophication	species.yr	1.85E-10	1.75E-10	1.02E-11
Marine eutrophication	species.yr	4.34E-12	1.55E-12	2.79E-12
Terrestrial ecotoxicity	species.yr	1.49E-11	1.09E-11	3.97E-12
Freshwater ecotoxicity	species.yr	1.12E-11	9.91E-12	1.28E-12
Marine ecotoxicity	species.yr	1.89E-12	1.60E-12	2.96E-13
Land use	species.yr	6.23E-09	6.21E-09	1.25E-11
Water consumption, Terrestrial ecosystem	species.yr	2.37E-09	2.34E-09	3.60E-11
Water consumption, Aquatic ecosystems	species.yr	1.06E-13	1.05E-13	1.61E-15
Mineral resource scarcity	USD2013	5.11E-04	4.54E-04	5.65E-05
Fossil resource scarcity	USD2013	3.66E-02	2.71E-02	9.54E-03

SI7 Heat- and steam mixes, country based

US

USA - sector specific
https://www.eia.gov/totalenergy/data/monthly/pdf/flow/css_2017_energy.pdf

U.S. primary energy consumption by industry	%
Petroleum	38
Natural gas	45
Coal	5
Renewable energy	12

CN

China - sector specific
https://www.iea.org/statistics/?country=CHINA&year=2015&category=Key%20indicators&indicator=WindGen&mode=chart&categoryBrowse=false&dataTable=ELECTRICITYANDHEAT&showDataTable=true

Chinese primary energy consumption by industry	
Production from:	%
- coal	90
- oil	4
- gas	5
- biofuels	0
- waste	1

ICE

The Icelandic heat production is assumed geothermal energy

SI.8 Harvesting of macroalgae feedstock

High demand of biomass for a biorefinery production might require some of the biomass to be cultivated (seaweed farming) and not only harvested (mowing the macroalgae from the sea floor) as is assumed in this study. Therefore, the results of Seghetta et al. (2016)[11] were adapted and used for analyzing the sensitivity of the harvesting data with respect to impacts associated with global warming and marine eutrophication. For these two impact categories, both studies applied the same impact assessment method.

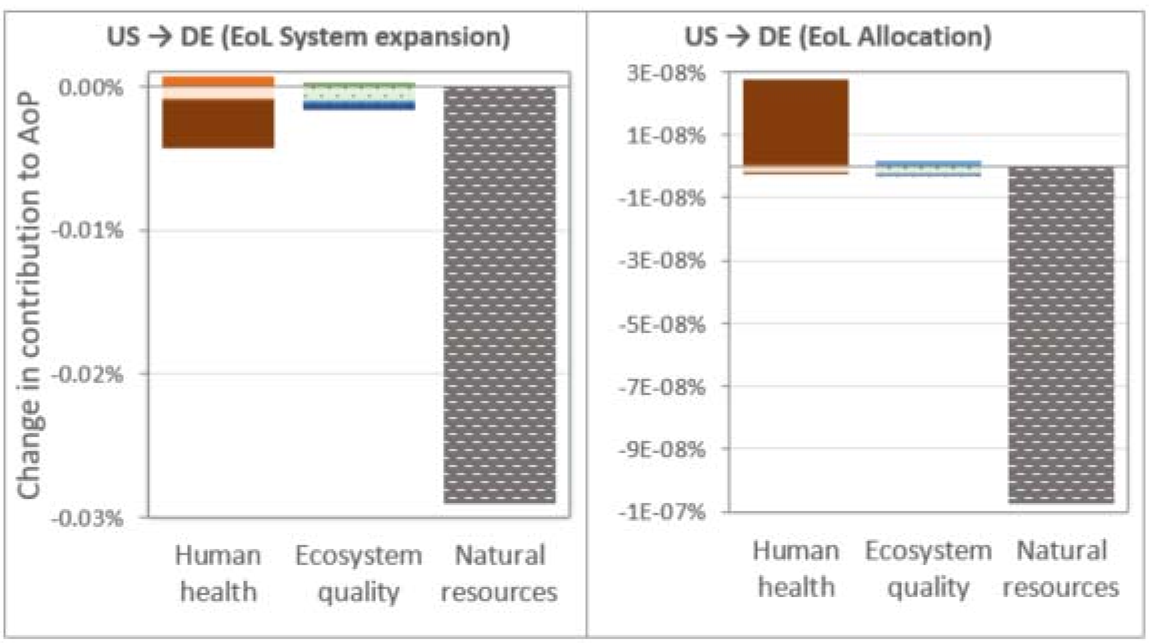
Impact category	Unit/kg macroalgae	Current study	Seghetta et al. 2017 (base case)
Climate change	kg CO2 eq	0.01	0.13
Marine eutrophication	kg N eq	0.001	-0.002

Data for harvesting macroalgae is not publicly available and the data used in this study comes from one industrial contact that produces harvesting equipment. Data on cultivating macroalgae is better documented[11], and they show strong influence on the overall results for macroalgae for global warming (65% of the impact) and therefore having better data for this life cycle stage would have been of high interest. Because if this fact, the available data has high uncertainty expressed in the Monte Carlo analysis.

SI.9 Transportation and End-of-Life in Germany as sensitivity scenario

Sensitivity toward changes in transportation and EoL[12,13] in Germany (DE), with and without system expansion of EoL scenario, compared to the base case. Transportation is not included in the LCA assessment. This was decided based on the results of the sensitivity scenario showing that transportation impacts had a neglectable impact on the study results. Depending on the study, this might though not always be the case[14] and therefore has to be tested. The results of the scenario analysis are following.

EoL with system expansion (base case proxy) showed that reduction in damages on human health was less than 0.01% and reduction of damages on ecosystem quality was less than 0.005%. For damages on natural resources, the reduction was slightly higher with 0.03%, because of avoided production of fossil-based plastics through plastic recycling. For comparison, applying allocation of bioplastic recycling to the recycled material, there are almost no differences with or without the transportation and allocated EoL in Germany.



For assessing transporation the following distances were assessed:

USA	Plant by Norfolk	Hamburg, harbour	Frankfurt Germany
	By the ocean	3784 nautical miles	490 km
		7000 km	
		Freight ship	Truck
		https://sea-distances.org/	googlemaps.com

SI.10 Biogenic Carbon storage and calculations at midpoint

Calculations of Net biogenic carbon emissions according to the CEN 16760:2015[15] standard

Biogenic carbon	Corn	C-to-Grave
Athmospheric Carbon fixation during biomass growth		-2.55E+00
Carbon emitted to air/water and soil during production phase		4.54E+00
Carbon permanently sequestrated in e.g. Co-products or landfilled production wastes		0.00E+00
Biogenic carbon embedded		1.99E+00
Biogenic carbon emissions to air, water and soil and end-of-life		-3.56E-01
Net biogenic carbon emissions		1.64E+00
Biogenic carbon	Corn-stover	C-to-Grave
Athmospheric Carbon fixation during biomass growth		-2.91E+00
Carbon emitted to air/water and soil during production phase		9.31E+00
Carbon permanently sequestrated in e.g. Co-products or landfilled production wastes		0.00E+00
Biogenic carbon embedded		6.41E+00
Biogenic carbon emissions to air, water and soil and end-of-life		-3.56E-01
Net biogenic carbon emissions		6.05E+00
Biogenic carbon	Macroalgae	C-to-Grave
Athmospheric Carbon fixation during biomass growth		-4.51E+00
Carbon emitted to air/water and soil during production phase		1.14E+01
Carbon permanently sequestrated in e.g. Co-products or landfilled production wastes		0.00E+00
Biogenic carbon embedded		6.87E+00
Biogenic carbon emissions to air, water and soil and end-of-life		-3.56E-01
Net biogenic carbon emissions		6.52E+00
Biogenic carbon	Macroalgae (WO drying)	C-to-Grave
Athmospheric Carbon fixation during biomass growth		-2.78E+00
Carbon emitted to air/water and soil during production phase		6.02E+00
Carbon permanently sequestrated in e.g. Co-products or landfilled production wastes		0.00E+00
Biogenic carbon embedded		3.24E+00
Biogenic carbon emissions to air, water and soil and end-of-life		-3.56E-01
Net biogenic carbon emissions		2.88E+00

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PAPER III

Journal: Trends in Biotechnology

Combining economic feasibility and environmental sustainability to optimize performance at early stages - Manuscript draft

Ögmundarson, Ólafur[a,b*], Sukumara, Sumesh[b], Markus Herrgard, Fantke, Peter[a]

[a] Quantitative Sustainability Assessment Division, Department of Management Engineering, Technical University of Denmark, Kgs. Lyngby, Denmark
[b] The Novo Nordisk Foundation Center for Biosustainability, Technical University of Denmark, Kgs. Lyngby, Denmark

[*]Corresponding author: Tel.: +45 21482660, fax: +45 45933435. E-mail: olog@biosustain.dtu.dk

Abstract

In our journey from a fossil-based economy to a bioeconomy to become less dependent on fossil fuels industrial biotechnology plays a key role in the development of alternatives to petrochemicals. There are though still obstacles to overcome, both financially and environmentally. In relation to biochemicals, the obstacles are related to the fact that despite being produced from renewable resources, assessments show that they are not consistently showing a better environmental performance than functionally equivalent petrochemicals. But how can we make them more environmentally sustainable? An operational framework that combines life cycle assessments and techno-economic assessment is the way forward. Combining the results in a decision support framework, from an early stage of optimization in an economic single score will provide insight into developing biochemicals securing that future biochemicals are both economically, and environmentally sustainable.

Global chemical industry thriving for a sustainable alternative

Bio-based production of chemicals has gained significant attention in past decades, due to the promises that it has offered in creating an environmentally benign alternative to dwindling fossil-based products. This period has seen some of the major, large-scale, implementation of bio-based fuel (ethanol, biodiesel)[1] and chemical production (1-3 PDO, 1-4 BDO, ABE fermentation)[2]. The increased potential of scalability, coupled with increased know-how in manipulating cell factories[3] to selective production of molecules, has led to immense R&D efforts around the world, in the quest of finding more bio-based compounds. These technological applications range from production of specific fine (low volume) molecules to the development of commodity (high volume) building block chemicals, which could replace the ones originating from natural products extraction and synthetic fossil-based resources. Moreover, in recent years, several manufacturers have been involved in ventures exploring the possibility of reducing the environmental footprint of their respective products, like the Coca-Cola Company with their PlantBottle™ packaging[4], Lego with their SUSTAINABLE LEGO® BRICKS[5]. The chemical industry is also working on lowering their environmental footprint, Novozymes and Novo Nordisk are examples of chemical manufacturers moving to friendly options in converting their energy use to wind power[6]. Policy makers are also encouraging the shift to more sustainable use, and applications of resources and materials like plastics, for supporting of a more Circular Economy[7]. The drive from manufacturers and policy makers has led to an increased need for replacing the precursors, such as monomers and other chemicals that feed into their products. A recent study showed that two in three stakeholders involved in the commodity chemicals value chain believes that bio-based chemicals could have a significant role to play in the next 5 years[8]. While this ratio could be deciphered as a foreseen consumer demand of renewables by the stakeholders, one must not overlook the challenges that has crippled the development of these technologies for years. Moreover, favorable policies initiatives on international level[9,10], governmental level[11,12], and industrial level[13,14] around the world encourage further green growth.

The potential offered by bio manufacturing has caught ample attention and investment[15,16], yet, there remain a few fundamental concerns which must also be addressed, to bring this vision into action[17]. Presently, several commodities and chemicals are traded around the world, which is based on the regional supply of feedstocks and economic competitiveness of contemporary technologies. Bio-based chemical production has trailed in competing with the fossil-based commodity chemicals, with only a handful of technologies reaching the market[18], thereby replacing only a small fraction of overall global commodities being traded. Moreover, this lack of economic competitiveness has also led bio-based production vulnerable to fluctuations in supply and demands of fossil-based feedstocks. Additionally, the relatively complex supply chain dynamics of bio-based value chains makes these economic obstacles furthermore pronounced (limiting capacity expansions), hindering the implementation[19–21].

Producing biochemicals from renewable resources does not automatically ensure their environmental sustainability. Selection of the bio-based feedstock as carbon source and intensified energy utility for the subsequent conversion and purification of chemicals can cause drastic environmental impacts, thereby, introducing trade-offs, when compared to petrochemical counterparts. Also, claiming that biochemicals are environmentally sustainable, only based on decreased global warming impacts could be misleading, if other vital environmental impacts while assessing biochemicals are neglected, such as water use, land use, acidification and eutrophication[22].

To implement a truly sustainable process for producing commodity chemicals, several of obstacles currently stand in the way, ranging from relatively low economic viability[23–28] to unresolved environmental issues[22,29] as we currently see that moving to biochemicals can increase environmental impacts compared to fossil-based. Especially impacts related to feedstock production and high-energy utility use in pretreatment and the biorefinery process.

Presently, the production of bio-based polymer is largely based on agricultural crops, such as, Lactic Acid by Natureworks, 1-3 Propanediol by DuPont Tate and Lyle BioProducts and 1-4 Butanediol by Novamont[18]. Among these, most of the technologies is essentially based on first generation feedstock, with a relatively high Technological Readiness Level (TRL) of 8 (first-of-a-kind commercial system) or 9 (full commercial application)[30]. Biochemicals from non-agricultural crops, e.g. lignocellulosic biomass, also known as, 2nd generation feedstock[31], have not yet reached high commercialization levels, due to relatively low fermentable sugars content, the cost related to its conversion to those sugars[32] and low technical process immaturity[31] in comparison to cost of utilizing 1st generation feedstocks. Adding to this, immense diversity in feedstock and complexity pertaining to the supply chain[19] further exacerbates the economic viability of 2nd generation feedstock based technologies. Due to these challenges for utilizing 2nd generation feedstocks and derived economic challenges, completely different feedstock sources, including engineered crops, algae and urban residues like household waste (3rd generation biomass[31]), are receiving increased attention. Among emerging feedstocks, brown algae are getting most attention because of the absence of lignin in the biomass, thus avoiding the need for lignin removal (Jung et al. 2013[33], Campbell et al. 2017[3]), which is costly because of recalcitrance of lignocellulosic cellulose and its toxic effects on microbial properties (Daful et al. 2017[34]). As promising feedstocks and associated technologies emerge, it is critical to assess its claim as a viable choice in the long run, as an economical and environmentally friendly alternative.

In a typical industrial biotechnology research and development set-up, several decisions are made during the design phase of a cell factory, which often has huge economic and environmental penalties down the road, while scaling up. For instance, during the strain optimization phase, the strains are selected based on parameters such as yield, titer and productivity. However, there could be scenarios where a presence of specific by-product may lead to higher associated downstream processing cost. Similarly, to achieve a proper upstream conversion or downstream separation of impurities, excess use of chemicals or utilities may also result in increased adverse environmental impacts. Contrarily, one may also imagine cases where extreme environmental measures resulting in a lower economic viability of projects which would also turn out to be undesirable for the stakeholders. Hence, both environmental and economic aspects pertaining to the process must be integrated and looked upon earlier in the technology development process. In order to achieve the proposed integration, various disciplines of engineering sciences and stakeholder’s perspective must be integrated into an iterative framework. This paper presents synergies between these methodologies to speed up the development process using a unique the decision support framework.

TEA and LCA, Context and Fundamentals

The proposed framework encompasses various disciplines and assessment methodologies under one roof to be applied at the early stages of research in the field of industrial biotechnology. Utilizing the underlying concept of Process Systems Engineering (PSE), an executable workflow has been demonstrated. PSE is a branch of process engineering based on the application of holistic methods for the design and optimization of industrial manufacturing processes[35]. This approach has important applications to sustainability, because, by considering manufacturing processes as a system rather than simply as a collection of individual unit operations, insights into overall operation (upstream and downstream), waste generation, and the energy requirement of a process can be achieved. Applying the tools of PSE to the design of sustainable commodity production process is a necessary competence required. Applying the underlying principles of PSE, we propose a decision support framework demonstrating, how to consistently couple environmental and economic indicators to allow for an overall optimization at early development stages of biochemical production.

Techno-Economic Analysis (TEA): TEA is an assessment tool, which gathers upon the information provided, by PSE tools and assigns monetary values to all the materials, energy and other consumables needed to run a production facility. Applying TEA helps users in understanding the long-term economic impact of a process, when in production capacity.

Life Cycle Assessment (LCA): LCA involves cradle-to-grave analyses of production systems and provides comprehensive evaluations of all upstream and downstream energy inputs and multimedia environmental emissions. LCA's can be costly and time-consuming if applied in isolation. However, with our previous research efforts, we have realized that this process could be speeded by applying TEA and LCA simultaneously as the prior can serve as a tool for generating inventories (parameters to proceed with the LCA).

Biomass as a source for chemical production

Converting biomass to biochemicals plays a key role in the bio-based economy making it possible to replace petrochemicals with an environmentally sustainable alternative. Positive examples of environmental benefits when comparing biochemicals or derived products to functionally equivalent petrochemicals, studies show up to a 10 orders of magnitude reduction in climate change impacts, like in the case Lactic acid[36], PLA[37], Succinic acid[38,39], 1,4-Butanediol[40], 1,3-Propanediol[41,42]. Studies assessing the environmental sustainability of 1,4-Butanediol and 1,3-Propanediol show consistent benefits for the environment as demonstrated in Ögmundarson et al.[22] We also see benefits from avoiding toxic air pollution from fossil burning leading to a reduction in non-toxic environmental impacts.

The production of biochemicals has doubled from 2011 to 2018, partly implying towards increased acceptance of bio-based products. Looking forward, in the future, this growth is predicted to sustain[18] with an increased double-digit growth in demand for bio-based materials in certain segments, such as, packaging (42%), automotive (18%), building (13%) and consumer goods (12%) by 2021. Moreover, since 2011, a portfolio of 17 building block chemicals[18] has been identified to be produced via bio-based routes and has led to commercial capacities/ventures. Hence, it would be fair to assume that truly economically viable bio-based technologies could be accepted by the industry and stakeholders, compared to other non-renewable ones, as it provides them a certain marketing edge due to increasing consumer awareness.

Despite the success stories where biochemicals show consistent benefits both economically and environmentally, it is evident that not all biochemicals show a better environmental performance when environmental impacts are assessed that are related to growing of the biomass/feedstock can causing eutrophication[43], acidification[44] or land-use[36]. In the shift to alternative feedstock, it remains unclear how using for example lignocellulosic biomass or macroalgae biomass will contribute to the environmental performance of future biochemicals when the processes have been optimized to the same TRL as biochemical production from corn[22]. To optimize biochemical production in terms of environmental sustainability performance, it is essential that we understand where along the life cycle of such systems so called environmental hotspots occur for each feedstock, and how any change in inputs and outputs to the production system influences such hotspots in a positive or negative way.

Current way of assessing LCA and TEA, separately and combined

Assessing market and economic feasibility of future technologies is becoming an integral part of product development in biotechnology because of the intense cost related to chemical development from idea to product on a market. To assess technical and economic feasibility of products and processes techno-economic assessment are a widely used because they help set the threshold for what is needed to reach technical and economic sustainability. When stating the current landscape of standalone TEA studies for biochemicals process optimization and future development numerous studies can be found. 1st and 2nd generation feedstocks and their feasibility have been covered in great extent and just a few are presented in Table 1 to give an overview of the available literature. When looking at the product categories presented in the literature, most assessments are on technical and economic feasibility of a proposed process configurations and direct comparison of products and processes. NREL has been one of the key contributors to this research field and they have set the standard for high quality assessments.

Environmental sustainability of biochemicals is also getting more and more attention, and is presented in some extent in the literature as demonstrated in Hottle et al.[29] and Ögmundarson et al.[22] and examples from literature are given in Table 1. The limitations of the existing scientific literature as stated in both Hottle et al. and Ögmundarson et al. is that there are surprisingly few studies published assessing especially commodity chemicals, e.g. lactic acid, succinic acid and 1,3-Propanediol. Another conclusion of the studies is that often relevant life-cycle stages are not assess, and lastly the identified studies are limited to assess only global warming impacts from emission of greenhouse gases and other possible impacts from biochemical production are neglected.

When these three points are neglected, we can end up being challenged when defending the actual environmental sustainability of biochemicals. First of all, few studies backing the sustainability claims can spread doubt about how environmentally sustainable they really are, especially when the results are not anonymously stating their environmental superiority when compared to functionally petrochemicals. When not assessing the whole life cycle of product, we can easily overlook burden shifting between life cycle stages resulting in that we miss out on either negative impacts that our product can cause later in an unassessed life cycle stage and also positive impacts that can stem from a high recycling rate of the applications made from our biochemicals. That results in avoided production of virgin material, presumably made from fossil resources. When concentrating LCAs on only assessing global warming impacts and drawing generic conclusions on the actual sustainability is potentially problematic because by not assessing other important life cycle impacts. As example, impacts related to growing of the biomass, such as land use, water use, acidification, and eutrophication, all impacts identified as imperative impacts when assessing biochemical production (Ögmundarson et al.), we potentially overlook environmental trade-offs when transforming our chemical production from fossil- to biochemicals.

TEA or LCA	Chemical	Feedstock	**Assessed environmental impacts**	**Application/ Intention with study and Stage of development**	At an early stage?	Reference
TEA	Biodiesel	Different	-	Process design	-	Zhang et al. (2013)[45]
	Biodiesel and co-production of succinic acid	Glycerine	-	Process design	-	Vlysidis et al. (2011)[46]
	Ethanol	Softwood	-	Process assessment	-	Wingren et al. (2008)[47]
	Drop-in biofuels	Jatropha	-	Process assessment	-	Brown et al. (2012)[48]
	Ethanol, PHB	Sugarcane	-	Process assessment		Moncada et al. (2013)[49]
	Ethanol, lactic acid, methanol	Lignocellulosic residues	-	Process assessment	-	Mandegari et al. (2018)[50]
	Carboxylic acids	Sawdust	-	Process assessment	-	Clauser et al. (2018)[51]
LCA	Succinic acid	Lignocellulosic residues	GHG, CED	Process assessment	-	Patel et al. (2018)[39]
	Succinic acid	Corn	GHG,CED,WU, EU,ET,HT,OD	Process assessment	-	Smidt et al. (2015)[43]

	Lactic acid	Corn	GHG, CED	Process assessment	-	Vink et al. (2015)[37]
	Lactic acid	Lignocellulosic residues	GHG,CED,PM, AC,ET,EU,HT, LU,OD	Process assessment	-	Daful et al. (2016)[36]
	1,3-Propanediol	Corn	GHG,CED,EU, HT,ET,AC	Process assessment	-	Hanes et al. (2015)[41]
	1,4-Butanediol	Corn	GHG,CED	Process assessment	-	Adom et al. (2014)[52]
	1,4-Butanediol	Corn, Lignocellulosic residues	GHG,CED	Process assessment	-	Patel et al. (2018)[39]
Combined TEA and LCA	Methane	Power-to-gas	GHG	Process design		Collet et al. (2017)[53]
	Bioethanol	Rice straw	GHG	Process design		Roy et al. (2012)[54]
	Bioethanol	Lignocellulosic residues	GHG, LUC	Process design		Vaskan et al. (2018)[55]
	Biodiesel	Microalgae	GHG, NER	Process design		Barlow et al. (2016)[56]
	Bioethanol	Lignocellulosic residues	GHG, CED	Process design		Cheali et al. (2015)[57]
	Biodiesel	Microalgae	GHG, EcotA, POP, EUAC, LD50	Process design		Cheali et al. (2015)[58]
	Blendstocks	Lignocellulosic residues	GHG	Product enhancement	Early stage	Dunn et al. (2018)[59]
	Biodiesel	Microalgae	GHG	Process optimization	Early stage	Dutta et al. (2016)[60]
	3-HPA, 1,3-PDO, SA	Bio-feedstock	GHG	Process design	Early stage	Gunukula et al. (2017)[61]
	Butanol, ethanol	Corn-stover	GHG	Process design	Early stage	Hernandez et al. (2018)[62]
	Biodiesel, Glycerol	Macroalgae	GHG	Process design		Kern et al. (2017)[63]

	Butanol	Lignocellulosic residues	HGH	Process design	Early stage	Levasseur et al. (2017)[64]
	Phthalic anhydride	Lignocellulosic residues	GHG, water depletion, fossil depletion	Process design		Lin et al. (2015)[65]
	Phenol Formaldehyde resins	Lignocellulosic residues	GHG, NRR, NER	Process design	Early stage	Mansoornejad et al. (2017)[66]
	Higher alcohols	Ethanol	GHG	Process design		Patel, A.D. et al. (2015)[67]
	1,3-Butandiene	Bioethanol and naphtha	GHG, CED	Process design		Patel, A.D. et al. (2012)[68]
	Energy and biofuels	Lignocellulosic residues	GHG,AC,EU,OD,NRR	Product selection		Rajendran et al. (2017)[69]
	Biodiesel	Microalgae	GHG	Process design	Early stage	Rickman et al. (2013)[70]
	Biogas	Manure	GHG	Process design		Shah et al. (2016)[71]
	Cellulosic isobutanol, cellulosic ethanol, n-butanol	Lignocellulosic residues	GHG,EROI,CAPs,CED	Product comparison		Tao et al. (2014)[72]
	Succinic acid and biofuels	Lignocellulosic residues	GHG,HTPI,HTPE,ATP,TTP,OD,PCOP,AC	Process design		Cheali et al. (2015)[58]
	Biogas and biofuels	Lignocellulosic residues	GHG,CED	Process design	Early stage	Patel, A.D. (2013)[73]

Table 1 Overview of studies applying LCA and TEA separately, and combined.

When presented in the scientific literature, the application of LCA and TEA combined, the methodologies, as an example, are used for optimizing processes (e.g. Dutta 2016) and to assess different process designs (e.g. Martinez Hernandez 2018;Levasseur 2017). Despite literature demonstrating how to use LCA and TEA combined at an early stage of design and process development, the idea of using them as decision support tools for target compound selection, at a stage earlier than process design and development, has not been explored. To perform a combined assessment of LCA and TEA possible we need to translate the individual results from both methods to a combined monetary score.

Early Stage Assessment Framework - Combined application of TEA and LCA, and monetarization of environmental impacts

Combining the TEA results and natural resources results from the LCA is easy as they are both assessed using a monetary score. DALYs and species.yr results are on the other hand by default not expressed as monetary values. The next step in the framework therefore requires monetarizing DALYs and species.yr, which allows combining the results into a single economic score, or cost per functional unit (see Figure 1).

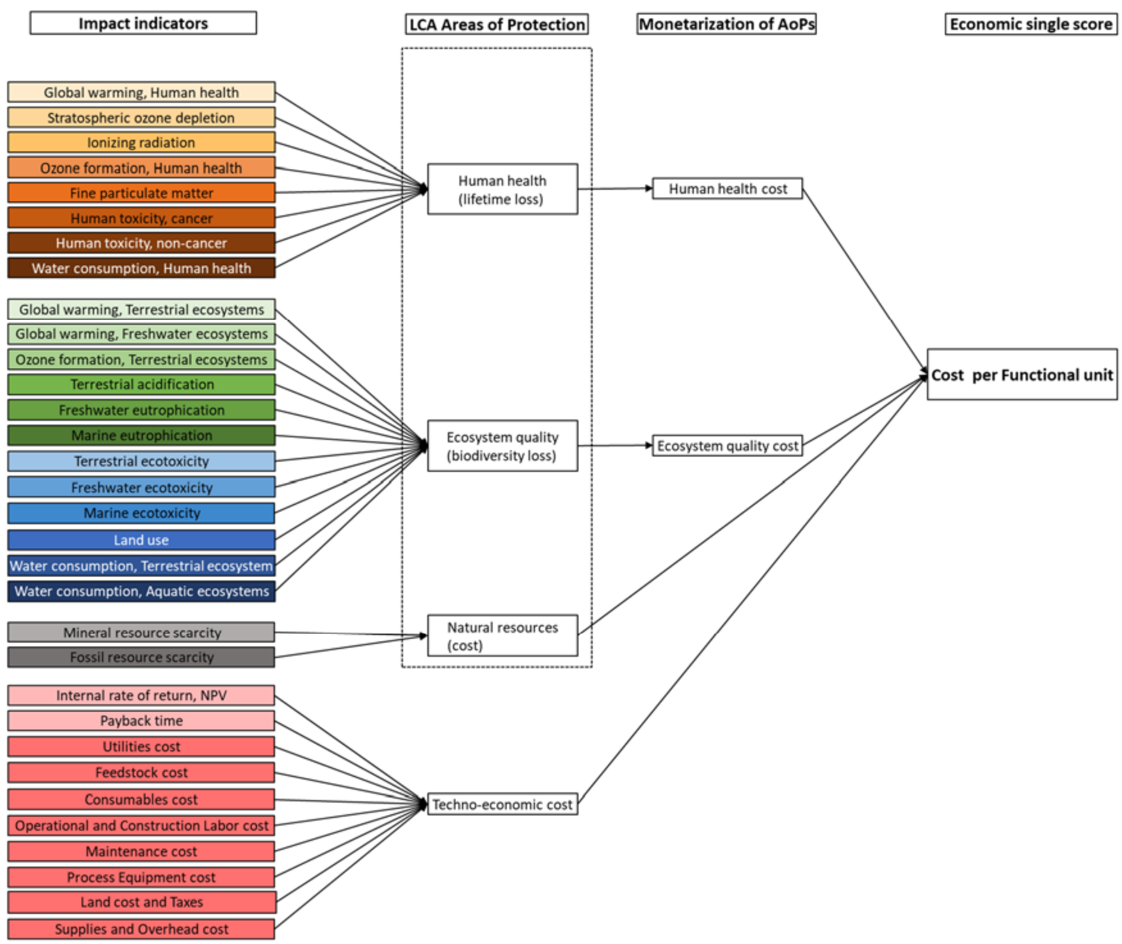

Figure 1 From impact indicators in LCA, to LCA areas of protection (AoP) translated to monetarization of AoPs combined with TEA for an to economic single score.

The economic single score then reflects the real cost of producing products and should be used for identifying environmental and monetary hotspots and tradeoffs between these two pillars of sustainability and is demonstrated in Table 2 and Figure 2.

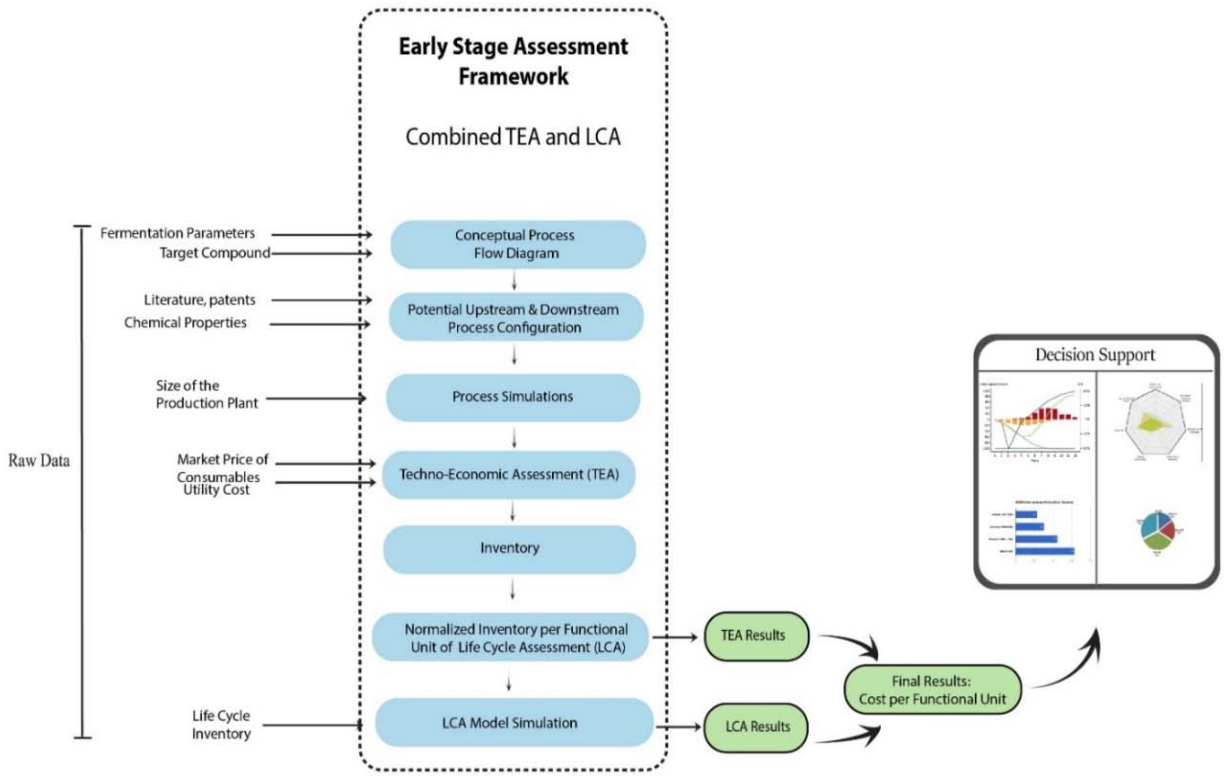

Figure 2 Early stage assessment framework for applying LCA and TEA as a decision support tool in biotechnology.

The publication "Evaluating the monetary values of greenhouse gases emissions in Life Cycle Impact Assessment" by Dong et al. (2018)[74] gives a good overview of the monetizing values that have been presented in the scientific literature, and based on that I then calculated the average monetary values for DALYs, which is 100.000$. For species.yr I used the value provided by Weidema (2009)[75], which is 65.000$.

Monetarization of environmental impacts is not new[75–77], but to my knowledge, it has never been adapted and presented before via an economic single score to combine LCA and TEA results. By doing this, for the first time, I exploit the potential of both LCA and TEA to identify tradeoffs between the results of both methodologies, which makes it possible to incorporate them as a decision support tool in biotechnology.

Framework results, in a total economic single score

The results presented in this section are based on the LCA results presented in Chapter **Error! Reference source not found.**. The functional unit of the study was "the production and use of 1 kg of lactic acid, with 99.9% purity, for household packaging application in the United States". For the LCA study the system boundaries were from cradle-to-grave, but the TEA assessed the system from cradle-to-gate (polymerization included). This is because it was not possible to get monetary values for the waste scenario for the TEA as this is highly dependent on country/region specific waste handling processes and costs are highly dependent on regulations and societal norms[78].

Table 2 Monetarization of areas of protection and economic single score per functional unit, for 1 kg of PLA, in $. Monetarization values for every DALY is 100.000$ based on average value from Dong et al.[74] and species.yr is 65.000$ based on Weidema[75]. TEA results for the 1st and 2nd generation feedstock processes were done by Sumesh Sukumara[79] and Elena Tomás Grasa did the 3rd generation feedstock process simulation.[80,79]

1st generation LCA results			**TEA results**		**Total economic single score**	
DALY	1.74	$				
Species.year	0.003	$				
USD	0.23	$				
Total	**1.97**	$ Cost per functional unit	**3**	$ Cost per functional unit	**4.97**	$ Cost per functional unit
2nd generation LCA results			**TEA results**			
DALY	2.59	$				
Species.year	0.003	$				
USD	0.42	$				
Total	**3.02**	$ Cost per functional unit	**2.14**	$ Cost per functional unit	**5.16**	$ Cost per functional unit
3rd generation LCA results			**TEA results**			
DALY	2.70	$				
Species.year	0.004	$				
USD	1.00	$				
Total	**3.70**	$ Cost per functional unit	**4.5**	$ Cost per functional unit	**8.20**	$ Cost per functional unit
3rd generation LCA results without drying			**TEA results**			
DALY	1.42	$				
Species.year	0.002	$				
USD	0.003	$				
Total	**1.43**	$ Cost per functional unit	**4.21**	$ Cost per functional unit	**5.64**	$ Cost per functional unit

When looking at the TEA results for LA from corn (1st generation) and comparing them to the LA from corn stover (2nd generation), the monetary cost of the 2nd generation is only higher by 2%. In spite of lower overall yield, the feedstock cost for the 2nd generation process is one fourth compared to that of the 1st generation. This calculation highly relies on the monetary value assigned to a unit of corn stover[81] which will increase significantly with the size of the plant due to the supply chain dynamics and low fraction of fermentable sugars present in 2nd generation feedstock as demonstrated in Sukumara et al 2014.[19] Also, despite the 2nd generation process is a factor of 3.6 more energy demanding compared to the 1st generation, it does not show in the TEA results, except by 0.03$ per kg LA. This is despite the currently lower level of optimization of the 2nd generation process and lower TRL, and despite the physical composition of the 2nd generation biomass (more fiber rich) requiring a more intense separation process demanding higher chemical concentrations and more intensive energy use.

As demonstrated in Table 2, the economic single score of the LCA results are 25% higher for the 2nd generation process than for the 1st generation process. This is due to that the energy use in the 2nd generation feedstock process is 3.6 times higher, compared to the 1st generation feedstock processes. The increased energy consumption is e.g. because of separation of the fiber rich biomass that in this study is done by steam explosion, which is not needed in the pretreatment of the 1st generation biomass. The higher energy use is mostly visible for the DALY and USD results because the modeled energy is 88%[82] from fossil resources which environmental impacts contribute mostly to human health (DALYs) and environmental resource use (USD).

When analyzing the results in further detail, we can see that for both the 1st and 2nd generation processes, environmental impacts are the highest expressed in DALYs. For the 1st generation the highest impacts come from indirect land use change (iLUC). This is related to increased global warming impacts due to increased demand for arable land[83]. The highest environmental impacts for the 2nd generation process are also global warming impacts, but from a different source, namely high energy demand of the biorefinery stage. This is due to intensive energy use in the pretreatment stage.

In the future, 3rd generation biomass could become a viable biomass, and with the framework presented here, we can assess the potential of producing LA from the biomass and analyze which environmental hotspots could be optimized to make the process more compatible, compared to the 1st generation feedstock process.

First, by looking at the TEA results for the 3rd generation process, the feedstock cost accounts for almost 50% of the total cost. Energy utilities stand for about 20% of the total cost with drying, and the process without drying has under 10% lower total cost than the process with drying. This is despite the fact that not drying the biomass results in a cut down on steam use of more than 100 MJ per kg product. The reason for the low effect on the total cost per functional unit is that steam bears a low price, and despite high amounts used the steam therefore has a minimal impact.

On the other hand, when analyzing the reduction in steam use in the LCA results, $ cost per functional unit, the benefits of reducing steam usage and thereby lowering the environmental impacts of 1 kg LA from macroalgae by more than 40% becomes evident. With the economic single score based on both LCA and TEA results for the macroalgae processes, we identify tradeoffs between drying and not drying the biomass that only assessing the TEA would not have revealed stating he benefits of incorporating environmental and economic aspects within one framework. This framework makes it possible to identify tradeoffs that only assessing either the economic or environmental aspects would not identify, which could lead to unnecessary environmental impacts that could have been avoided from the earliest stages of development of future biochemical production processes.

Concluding remarks

The execution of proposed framework demonstrates the value that could be unleashed by coupling the TEA and LCA under a roof and applying in the early stages of research and development. With growing consumer demand for truly sustainable technologies, industrial biotechnology could immensely benefit from early assessment that could provide insights in developing economically and environmentally viable processes. With this framework a combined single score can be calculated that could be used as a yardstick by research organizations and companies to rank projects based on its long-term impacts. Furthermore, this decision support tool could be used iteratively by the research units to optimize the entire production scheme in the inception of a viable idea both from an economic and environmental perspective.

The economic and environmental hotspots determined by the integrated assessment provides insights on the impacts that could be mitigated by adopting certain strategies early in the development process. Energy utility use is a hotspot appearing both in the LCA and TEA assessment, as demonstrated in Table 2, and for the 3rd generation biomass, drying the biomass has the biggest impact (accounting for more than 50% of the energy utilities). 3rd generation LCA results show a value of 3.7$ cost per functional unit but when drying is not included in framework, LCA cost per functional unit is reduced to 1.43$, a reduction of 61%. Respectively, the TEA cost per functional unit is reduced by 7%. This shows us that in the TEA model, drying (heat and steam) does not carry a high cost. But environmentally, when comparing the reduction in cost between drying the biomass and not drying, high reduction in cost is achieved. This especially applies in countries relying of fossil fuels as source to produce energy utilities from[78]. If we want to produce environmentally, and economically benign biochemicals, our operational framework helps achieve that.

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PAPER IV

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Toward a more sustainable biochemical industry

Early stage assessments and methodological overlaps between life cycle- and techno-economic assessments of biochemicals

Ólafur Ögmundarson[1&2], Sumesh Sukumara[1], Peter Fantke[2]

[1]DTU Biosustain, The Novo Nordisk Foundation Center for Biosustainability, Kgs. Lyngby, Denmark
[2]DTU Management Engineering, Quantitative Sustainability Assessment Division, Kgs. Lyngby, Denmark
E-mail contact: olog@biosustain.dtu.dk

1. Introduction

Existing Life cycle assessment (LCA) studies of biochemicals reveal that there are challenges that need to be overcome in order to reach an overall high sustainability performance. While in some cases biochemicals have lower global warming impacts compared to fossil-based chemicals, other impacts may become higher, like eutrophication, which is directly related to fertilizer use in the feedstock production of biochemicals. One of the major sources of environmental impacts of biochemicals is the growing of biomass, which in most cases today is corn [1]. This has led to investment in assessing opportunities of using side streams, like leftover agricultural lignocellulosic biomass, or non-cultivated biomass like algae, as carbon source [2]. Lignocellulosic biomass is an interesting feedstock because it introduces the potential for utilizing the entire biomass and not only the corn, and the economic feasibility of using lignocellulosic biomass has been assessed to some extent with techno-economic assessments (TEA) [3]. Presently, most of the processes are conceptually designed for economically viable operation, utilizing all the three major fractions of biomass such as, cellulose, hemicellulose and lignin, for biochemical and utility production at the biorefinery facility. Macro-algae is one such potential source given that they grow without being farmed, and that benefits are put in relation to algae filtering N and P from the ocean and use these compounds for growing, while simultaneously being an important sink for CO_2 [4].

The objective of this study is to identify trade-offs between assessed environmental impacts and possible burden shifting between macro-algae compared to more conventional feedstock´s like maize and lignocellulose.

While it is imperative that any change in process configuration reflects in TEA and LCA, there are very few studies which couples these two assessment demonstrating the trade-offs for improved decision support. Hence, the focus of this contribution is to explore methodological overlap between the two assessments strategies and develop a framework, supported by a proof-of-concept

2. Methodology

Assessing the opportunities of using macro-algae compared to maize and corn-stover, as carbon source, relies mainly on data availability and derivation of data representing a scaled up biorefinery. When working with less evolved feedstock, such as macro-algae, we need methods for scaling up of laboratory data to assess the actual sustainability and optimization potential of bio-based chemicals before large investments in new processes take place.

The approach used to bridge the data gap for the biorefinery process is applying techno-economic assessments (TEA) on the three different feedstocks, macro-algae, corn and corn-stove.

Applying TEA provides an insight if it is technologically and economically feasible to utilize alternative feedstock sources for biorefineries, which is today the main driver if money should be invested in developing new production pathways of chemicals from biorefineries. If the TEA then gives a feasible outcome, performing an LCA will complement that information by addressing the possible environmental impacts of the new chemical production process which otherwise are not a part of a TEA and would therefore not be considered as being relevant for related decisions. TEA and LCA are described further in more detail in the following paragraphs.

TEA accounts for all costs incurred due to the mass and energy flows, operational expenditures and capital investment required for scaled up production of biochemicals. In order to compare the economic impact of three generations of biobased feedstock, a proof-of-concept is performed for targeted production of lactic acid. The tools used for this appraisal is a widely used process simulator, Aspen Plus® and complementary modules from AspenTech® [5].

The LCA is a cradle-to-gate and a cradle-to-grave assessment with the focus on the environmental hot-spots associated with the production of bio-based lactic acid, and derived (poly)lactic acid (PLA) plastic bottles from macro-algae, corn and corn-stover. The functional unit of the study is *one single use PLA-plastic bottle to contain 500 milliliters of water*.

The scope of the study ranges from the extraction of the raw material for all the three generations of feedstock. Followed by the processes of their resin production, through bottle formation, followed by their use stage and end-of-life disposal. The assessment includes the following impact categories of global warming, ozone formation, ozone depletion, ionizing radiation, particulate matter formation, human toxicity, ecotoxicity, acidification, eutrophication, land use, indirect land-use change, water use, resource use and energy demand. The LCA is a consequential LCA applying system expansion.

3. Results and discussions

When contrasting current results from the TEA and LCA cradle-to-gate study, some interesting trends were observed. The TEA show that it´s biggest hot-spots are identified as feedstock cost which is a function of growing, transportation of biomass and if drying is taking place at the refinery site or closer to the harvesting sites of the feedstock. Whereas, the LCA shows the biggest environmental hot-spots occur in relations to growing of biomass, if it requires external application of nutrients and intensity of chemical pretreatment.

4. Conclusions

Today decisions on if chemicals are further developed companies mostly rely of results from TEAs. Our results show that the methodological overlap between TEA and LCA are of that magnitude that justifies the appraisal of this integrated methodology.

Introducing LCA as a decision support tool would integrate sustainability requirements in development of technology and solutions. All technologies and products have a life cycle, and by analyzing their impacts, we put numbers on sustainability and benchmark the solutions.

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