The environmental life cycle assessment of selected 1st through 3rd generation biofuels within the context of sustainable development

Livsløpsvurdering av utvalgte første til tredje generasjons biodrivstoff i kontekst til bærekraftig utvikling

Philosophiae Doctor (PhD) Thesis

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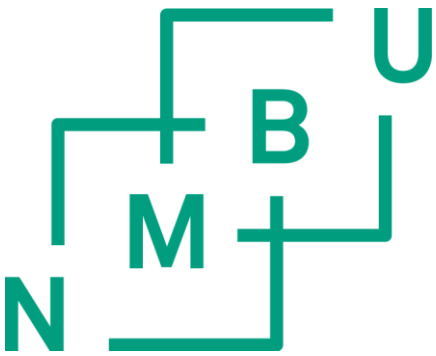

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Summary

Biofuels, at all stages of technological development, exist all around us; primarily the result of society's ambition to mitigate climate change associated with the transport sector. Parallel to this development has been a desire to accurately quantify biofuels environmental impacts through the establishment and development of life cycle assessment methods.

However, a paradigm shift has occurred from one of environmental protection to sustainability (Finkbeiner et al. 2010). This is reflected in the evolution of biofuel relevant policy (EU 2009; EU 2013); the latter introducing a set of sustainability criteria- and evaluation methods. However, these criteria and methods fall short in their ability to adequately address- and assess the concept of sustainability.

The research community proposes the aggregation of several complementary assessment methods to one life cycle sustainability assessment framework (Finkbeiner et al. 2010; Guinee et al. 2010), or the selection a few suitable methods to provide key indicators (Hoekstra 2015). However, no consensus exists, and it appears that not only is the concept of sustainability, but also its assessment, like democracy "universally desired, diversely understood, and extremely difficult to achieve" (Lafferty 2004).

The main hindrance to consensus is a lack of understanding of the structure, or hierarchy, of biofuel solutions and strategies that can ultimately contribute towards the achievement of sustainable development, and the specific limitations of the assessment methods proposed to compare these solutions and strategies. Only through the disaggregation of the concept of sustainable development into easier manageable solutions and sets of strategies, and the identification of the areas of application- or limitations to: environmental- and social (incl. economic) sphere assessment methods can overlap and confusion be reduced, consensus achieved, and advancement towards a sustainable society be achieved.

The four articles contained within this thesis represent on their own a unique contribution to the field of biofuels, and together with this thesis introduction advance the discourse concerning the life cycle assessment of biofuels, with a particular focus on biofuels' role towards the achievement of sustainable development, and life cycle assessment's role in their assessment. A biofuel-sustainability framework, which disaggregates the concepts of sustainable transport and development into manageable sets of strategies and individual solutions is presented, and clearly places individual biofuel pathways and biofuel strategies in context (Article I).

Based on the life cycle assessments performed in Articles II, III and IV it was found that life cycle assessment methodology becomes less significant in comparative assessment at higher levels of the proposed framework. Specifically, this limits life cycle assessment methodologies significance to comparisons at the first four levels in the proposed framework: biofuels (level 5' and 5), alternative fuels (level 4) and improvements in fuels/technology (level 3). Comparisons at higher levels, which deal with broader transport strategies (levels 1 and 2), present such diverse functions, that a collection of meso- or macro level methods are needed to complement life cycle assessment results.

The limitation of life cycle assessment methodology to the comparison of products (and services); and satisfying primarily the first condition of sustainable development: safeguarding long-term ecological sustainability (Brundtland et al. 1987), is discussed.

The scope of Article II, III and IV, covering 1st through 3rd generation biofuels at various stages of technological development: commercial, early commercial and basic and applied R&D respectively, were used to identify a relation between the stage of technological development of individual biofuel pathways and the level of comparative assessments performed with respect to the biofuel-sustainability framework. This relation suggests that the stage of technological development influences both data quality- and quantity, the lack of which can hinder higher-level life cycle assessment comparisons with respect to the proposed biofuel-sustainability framework (Wenzel 1998).

The implications of these findings is that despite the explicit or implicit desire for life cycle assessment results to forecast the sustainability of individual biofuel pathways under development, this is not possible. Instead, one must trace and compare biofuel's contribution via increasing levels of aggregation; via biofuel-, alternative fuel-, technology- and sustainable transport strategies. With increasing levels of aggregation, the function of the elements compared is extended or expanded which requires life cycle assessment to be complemented with additional life cycle approach methods. Ultimately, these solutions and strategies must compare favourably to all four main conditions of sustainable development.

Sammendrag

Biodrivstoff eksisterer rundt oss, ved alle teknologiske utviklingsstadium, noe som i hovedsak er et resultat av samfunnets ambisjoner om å begrense klimaendringene knyttet til transportsektoren. Parallelt med denne utvikling har det vært et ønske å nøyaktig kvantifisere miljøpåvirkninger fra biodrivstoff gjennom etablering og utvikling av livsløpsanalysemetoder.

Imidlertid har det oppstått et paradigmeskifte, fra miljøvern til bærekraft (Finkbeiner et al. 2010). Dette gjenspeiles i utviklingen av politikk relatert til biodrivstoff (EU 2009; EU 2013), der den siste introduserer et sett med bærekraftkriterier- og evalueringsmetoder. Disse kriteriene og metodene kommer imidlertid til kort i sin evne til å vurdere begrepet bærekraft.

Forskningsmiljøet foreslår å samle flere komplementære vurderingsmetoder til et felles rammeverk for livsløpsanalyser med vekt på bærekraft (Finkbeiner et al. 2010; Guinee et al. 2010), eller et utvalg av noen få egnede metoder for å gi nøkkelindikatorer (Hoekstra 2015). Det finnes imidlertid ingen konsensus på området, noe som ser ut til å gjelde både begrepet bærekraft og hvilke vurderingsmetoder som skal benyttes. På samme måte som *demokrati* er *bærekraft* "universelt ønsket, forskjellig forstått, og svært vanskelig å oppnå" (Lafferty 2004).

Det største hinderet for enighet er mangel på forståelse av strukturen, eller hierarkiet, av løsninger og strategier for biodrivstoff, som til slutt kan bidra til oppnåelse av en bærekraftig utvikling, og de spesifikke begrensningene til vurderingsmetodene som foreslås for å sammenligne disse løsningene og strategiene. Bare gjennom oppdeling av begrepet «bærekraftig utvikling» til lettere håndterbare løsninger og sett av strategier, og gjennom identifisering av områder med begrensninger eller –anvendelse; der vurderingsformer for miljø- og sosiale (inkludert økonomiske) sfærer kan overlappe og forvirring reduseres, kan konsensus og fremskritt mot et bærekraftig samfunn oppnås.

De fire artiklene i denne avhandlingen representerer i kraft av seg selv et unikt bidrag til forskningsfeltet for biodrivstoff, og sammen med denne avhandlingsintroduksjonen bidrar de til fremskritt som viktig innlegg vedrørende livsløpsanalyser av biodrivstoff, med et særlig fokus på biodrivstoffets rolle opp mot oppnåelse av bærekraftig utvikling og livsløpsanalysens rolle. Et rammeverk for vurdering av biobrensel-bærekraft, som deler begrepene bærekraftig transport og -utvikling i håndterbare sett med strategier og individuelle løsninger, blir presentert og plasserer tydelig enkelte biodrivstofftraséer og biodrivstoffstrategier i sammenheng (Artikkel I).

Basert på livsløpsanalysene utført i Artikkel II, III og IV ble det funnet at livsløpsanalysemetodikken blir mindre signifikant i sammenlignende vurderinger på høyere nivåer av det foreslåtte rammeverket. Særlig begrenser denne signifikante sammenligninger av livsløpanalysemetoder på de fire første nivåene i det foreslåtte rammeverket: biodrivstoff (nivå 5’ og 5), alternative drivstoff (nivå 4) og forbedringer i drivstoff / teknologi (nivå 3). Sammenligninger på høyere nivåer, som omhandler bredere transportstrategier (nivå 1 og 2), representerer så forskjellige funksjoner, at en samling av metoder på meso- eller makronivå er nødvendig for å fylle ut resultatene fra livsløpsanalysen.

Livsløpsanalysemetodikkens begrensninger for sammenligning av produkter (og tjenester); og tilfredsstillelse av primært den første betingelsen for bærekraftig utvikling: å sikre langsiktig økologisk bærekraft (Brundtland et al. 1987), er drøftet.

Omfanget av artikkel II, III og IV, som dekker første til tredje generasjons biodrivstoff på ulike teknologiske utviklingsstadier: kommersielle, tidlig kommersielle, grunnleggende og anvendt FoU, som henholdsvis ble brukt til å identifisere en sammenheng mellom fasene av teknologisk utvikling av de individuelle biodrivstoff-traséene og nivået på komparative vurderinger utført med hensyn til rammeverket for bærekraftig biobrensel. Dette forholdet tyder på at teknologisk utviklingsfase påvirker både datakvalitet og –kvantitet, hvor mangel av dette kan hindre høyere nivå av livsløpsanalyse-sammenligninger med hensyn til det foreslåtte rammeverket for bærekraftig biobrensel.

Implikasjonene av disse funnene er at til tross for det eksplisitte eller implisitte ønsket om livsløpsanalyseresultater for å forutsi bærekraften i enkelte biodrivstofftraséer under utvikling, er dette ikke mulig. I stedet må man spore og sammenligne biodrivstoffs bidrag via økende nivåer av aggregering; via biodrivstoff-, alternative drivstoff, teknologi- og bærekraftige transportstrategier. Med økende nivåer av aggregering, er funksjonen av elementene sammenlignet forlenget eller utvidet, noe som krever at livsløpsanalyser blir supplert med ekstra livssyklustilnærmingsmetoder. Til syvende og sist må disse løsningene og strategiene komme gunstig ut sammenlignet med de fire viktigste betingelsene for bærekraftig utvikling.

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Numerous people deserve thanks for their contributions to this work. I would like to thank my main supervisor Associate Professor Petter Heyerdahl, whom I have known for ten years, and who has always provided me with guidance and insight regarding my professional work and career as a researcher and teacher. I would also like to thank my co-supervisors. Professor Erling Holden is not only responsible for procuring the financing for my thesis, he is also responsible for developing my understanding of biofuels within the context of sustainable development. His professional life has provided me with an example to strive towards. Senior Researcher Ole Jørgen Hannsen, became co-supervisor midway through my PhD studies, and has been essential in guiding me towards a better understanding of life cycle assessment.

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Ås, March 2016

Geoffrey S. Gilpin

List of Articles

Article I

Holden E, Gilpin G (2013) Biofuels and sustainable transport: A conceptual discussion Sustainability 5:3129-3149

Article II

Gilpin G, Hanssen OJ, Czerwinski J (2014) Biodiesel's and advanced exhaust aftertreatment's combined effect on global warming and air pollution in EU road-freight transport Journal of Cleaner Production 78:84-93

Article III

Gilpin G, Andrae ASG (submitted) Comparative attributional life cycle assessment of European industrial enzyme production for use in second-generation lignocellulosic bioethanol production International Journal of Life Cycle Assessment

Article IV

Gilpin G, Schipper J (planned submission) A cradle-to-gate attributional life cycle assessment of integrated aquaculture in Norway for sustainable food and fuel production Aquaculture Reports

Additional Scientific Contributions

Conference papers with published proceedings:

Gilpin G (2010) The achievement of sustainable mobility requires the co-existence of numerous energy carriers Renewable Energy Research Conference (RERC), Trondheim

Gilpin G, Andersen O (2010) The life-cycle-assessment of combined bio- and fossil fuel blends with exhaust after-treatment in heavy-duty diesel engines European Oil and Gas, Krakow

Oral presentations:

Gilpin G (2009) Resource availability of used cooking oil for first generation biodiesel production in the inner Sogn region of Norway Intelligent Energy for Europe Programme – BioDieNet project meeting, Milan

Gilpin G (2009) Status and potential for used cooking oil biodiesel production in the inner Sogn region of Norway Intelligent Energy for Europe Programme – BioDieNet project summary meeting, Brussels

Gilpin G (2010) Applying life cycle assessment methodology towards the evaluation of biodiesel use in diesel lorries European Economic Area – BioDeg project meeting, Sogndal

Gilpin G (2010) The status of alternative motor fuels in Norway International Energy Agency – Alternative Motor Fuels Task meeting, Ottawa

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Introduction

Background

Transport, all types[1], accounted for 14% of worldwide anthropogenic greenhouse gas (GHG) emissions in 2010 (IPCC 2014). In the European Union (EU) 28 and Norway, transport accounted for 21.9% in 2012 (Eurostat 2015) and 16.5% in 2014 (SSB 2015a) of total GHG emissions respectively. In general, almost 95% of transport related GHG emissions are related to the combustion of petroleum-based fuels, e.g. gasoline and diesel (IPCC 2014). Additionally, for all regions, the share of transport related GHG emissions has seen increases over the past decades, and is expected to increase dramatically in the coming decades, both in Europe and Norway, but also globally (Hamje et al. 2014; IEA 2008).

This knowledge has prompted numerous countries and unions to introduce policy aimed at reducing the level of transport related GHG emissions. In Europe, this process began as early as 1998 with the resolution to promote biofuels as a climate change mitigation strategy (EU 1998); culminating in the most recent directive (EU 2009), which aims for 10% of transport fuel in the EU to come from renewable sources, including biofuels, by 2020. Norway has adopted similar aims, in-line with EU countries through participation in European Economic Area agreements.

The result has been an almost 14 fold increase in European (EU 28) biofuel consumption in the period between 2002 and 2014, from 1.1 million tonnes oil equivalent (Mtoe) to 14.0 Mtoe respectively (EurObserv'ER 2015). In Norway over a similar period, 2006 to 2014, biofuel consumption has increased 25 fold from 5.2 kilo tonnes oil equivalent (ktoe) to 129.0 ktoe respectively (SSB 2015b).

However, with this increase in biofuel production and consumption have arisen numerous and previously unforeseen effects (Article I); most notably: competition with the provision of food (Graham-Rowe 2011), changes to land-use (Laborde 2011), threats to biodiversity (Pedroli et al. 2013) and changes to the earths albedo effect (Cherubini et al. 2012). In fact, while some biofuels offer obvious benefits, others offer little-to-no benefits when compared with conventional fossil fuels (Kendall and Yuan 2013; Larson 2006; Zah et al. 2007).

Awareness of these effects has resulted in a paradigm shift from one of environmental protection to one of achieving sustainability (Finkbeiner et al. 2010), and has led some to question whether-or-not the net effect of biofuel strategies is positive or negative with respect to sustainable development (Tilman et al. 2009).

A clarification should be made concerning the two terms sustainability and sustainable development; some scholars argue that there is a difference between these two terms, for example: that sustainability refers to the environmental dimension of sustainable development, or that sustainability refers to a process whereas sustainable development

[1] Road-, rail-, air- and sea transport

refers to the product (end state). In this thesis, the two concepts entail the same dimensions and the same policy implications, and are used interchangeably (Holden et al. 2013).

Sustainable development was defined in ***Our Common Future*** as "development which meets the needs of the present without compromising the ability of future generations" (Brundtland et al. 1987). From this document, three pillars of sustainability grouped into the two (inseparable) spheres: society (incl. economy) and the environment can be identified. Thus, sustainable development can be interpreted as the achievement of a symbiotic- balanced relationship across these two spheres.

From ***Our Common Future*** (Brundtland et al. 1987), four conditions or characteristics for sustainability can be derived, these are: safeguarding long-term ecological sustainability, satisfying basic human needs, promoting inter-generational equity, and promoting intra-generational equity (Holden 2012). These conditions are absolute, and non-interchangeable (Daly 2007). Additionally, whereas the last three conditions have explicit relevance for both the environmental and social spheres, it can be acknowledged that the first condition is more relevant to the environmental sphere.

Since sustainable development's definition in ***Our Common Future***, and the subsequent derivation of the four aforementioned conditions (Brundtland et al. 1987; Holden 2012), numerous aspects of the environment and society have been identified as central factors towards its achievement; including the transport sector.

With the aforementioned knowledge that the existing transport sector is of concern, several concepts denoted by terms such as sustainable transport and sustainable mobility have been identified. In the literature on transport and sustainable development, these terms are essentially synonymous; though, in this thesis the term 'sustainable transport' is used.

Holden (2012) applied the imperative of sustainable development to the transport sector and derived four conditions or characteristics for sustainable transport. These conditions or characteristics are: a minimum per capita energy consumption for passenger transport, a minimum level of per capita travel distance by motorized transport, a maximum fraction of total population with access to public transport, and a maximum fraction of fuel provided by renewable resources. Similar to the conditions of sustainable development, these conditions are assumed absolute, and non-interchangeable.

Returning to the aforementioned paradigm shift from environmental protection to sustainable transport which is reflected in a change in the formulation of European[2] biofuel policy between 1998 (EU 1998) where environmental protection and climate change mitigation stood in focus, and 2009 (EU 2009) where sustainability is a central theme.

The latter policy has resulted in the introduction of biofuel sustainability criteria, with the three main criteria corresponding to: minimum reductions in GHG emissions, and the protection of land with high- carbon-stocks and biodiversity. Here, the question of biofuel sustainability has been primarily reduced to addressing the first condition of sustainable development which can be derived from Brundtland et al. (1987), i.e. safeguarding long-term ecological sustainability. Furthermore, the methodology proposed for evaluating

[2] Including Norway through European Economic Area agreements

some of these criteria rely heavily on environmental life cycle assessment (LCA) methods and results (Alberici and Hamelinck 2010; Alberici et al. 2010; EU 2013).

These criteria and their proposed methods fall short in their ability to adequately address and assess the concept of sustainable development, which has three additional conditions: satisfying basic human needs, and promoting intra- and intergenerational equity (Brundtland et al. 1987; Holden 2012); the assessment of which lies outside LCA's area of application.

Fortunately, parallel to the promotion of biofuels as a climate change mitigation- and later sustainability strategy, has been the introduction of the theoretical perspective of industrial ecology. Industrial ecology's holistic perspective and set of environmental and social (incl. economy) life cycle approach assessment methods from the product- (micro) to macro levels can offer one solution.

From the field of industrial ecology has emerged the proposal of a life cycle sustainability assessment framework as one means of assessment that can address several of the conditions of sustainable development through the aggregation of complementary environmental- and social life cycle approach methods from the micro- to the macro levels (Finkbeiner et al. 2010; Guinee et al. 2010).

Though despite a consensus concerning the need for a transdisciplinary approach, and standards or guidelines existing for several of the assessment methods (Benoît 2010; ISO 2006), little consensus exists as to which individual methods- and their configuration are most suitable towards addressing and assessing individual biofuels, or their integration in sets of strategies towards the achievement of sustainable development.

Research Questions and Scope of the Thesis

The overarching objective of this thesis is to present an interpretation of biofuel's role towards the achievement of sustainable development, and LCA's role in assessing the sustainability of biofuels through the practical application of LCA to relevant 1st through 3rd generation biofuels. Thereby placing both biofuels and LCA within the broader contexts of sustainability and its assessment.

To achieve this, two research questions are posed:

1. What is the contribution of individual biofuel pathways towards the achievement of sustainable development?
2. What are the limitations of LCA in evaluating the sustainability of biofuels within the context of the biofuel-sustainability framework?

To address these research questions four articles have been written. The rational for the mismatch between the number research questions and articles in this thesis requires an explanation. In order to determine whether a relationship exists between the level of technological development of a specific biofuel pathway, and its level of assessment (research question 2), with respect to the proposed framework, a selection of biofuel pathways at varying levels of technological development was necessary, see Table 1.

Table 1 Summary of the scope of the LCAs performed in Articles II, III and IV

Article	**Biofuel generation**			**Stage of technological development[1]**				**Biofuel-sustainability framework level[2]**			**LCI modelling framework**	**LCIA method**
	1st	2nd	3rd	R&D	Demo.	EC	C					
II	X						X	3			attributional	CML
III		X			x	X			5		attributional	CML
IV			X	X						5'	attributional	CML

Of the four articles written, two (Articles I and II) have been peer-reviewed and published. Of the remaining two; Article III has been re-submitted to a journal for peer-review following revision, and one is considered publishable, but is as-of-yet un-submitted, see Table 2.

Table 2 List of articles written in this thesis

#	**Title**	**Year**	**Journal**
I	Biofuels and Sustainable Transport: A conceptual Discussion	2013	Sustainability
II	Biodiesel's and advanced exhaust aftertreatment's combined effect on global warming and air pollution in EU road-freight transport	2014	Journal of Cleaner Production
III	Comparative attributional life cycle assessment of European cellulase enzyme production for use in second generation lignocellulosic bioethanol production	(20xx)	Under review: The International Journal of Life Cycle Assessment
IV	A cradle-to-gate life cycle assessment of integrated aquaculture in Norway for food and fuel production.	(20xx)	Planned submission to: Aquaculture Reports

In article I, we discuss biofuels as one proposed strategy towards the achievement of sustainable development. We present a hierarchal biofuel-sustainability framework with which to compare biofuels against alternatives, see Figure 7, and affirm that LCA, supplemented by other assessment methods, is suitable to compare individual biofuel pathways against their alternatives. This article develops on the previous work of the co-author Holden (2012).

Tilman et al. (2009) points out though that conclusions concerning biofuel sustainability cannot be drawn about biofuels in general, and that great variation exists depending on the individual biofuel pathway under consideration. This observation is not unexpected when considering the numerous biomass feedstocks available, and biomass-to-biofuel conversion pathways, see Figure 1.

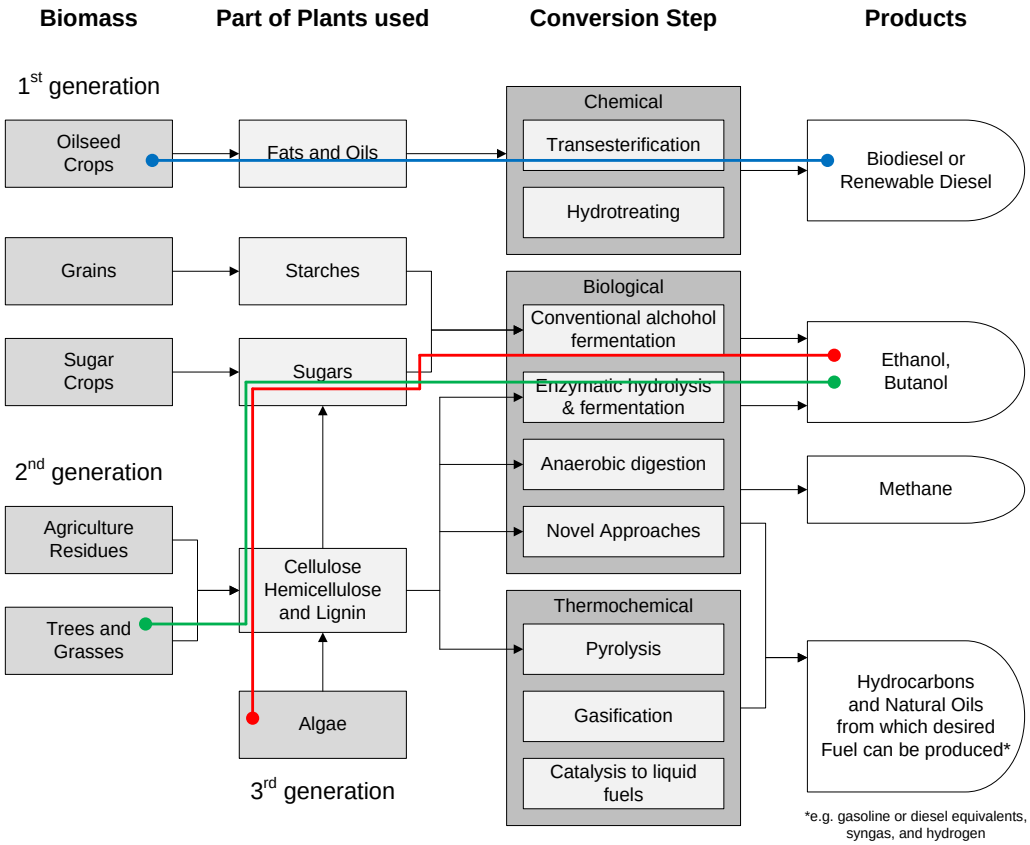

Figure 1 Biofuel conversion pathways (Peña 2008), with blue, green and red lines indicating the biofuel pathways evaluated in Articles II, III and IV respectively

It is common to organize these individual biomass-to-biofuel pathways into either stages of technological development, i.e. advanced- or conventional technology, or generations which differentiate between biofuel feedstock source, i.e. agriculture, silviculture (& waste) and aquaculture, though a strong correlation exists between these two approaches, see Figure 2.

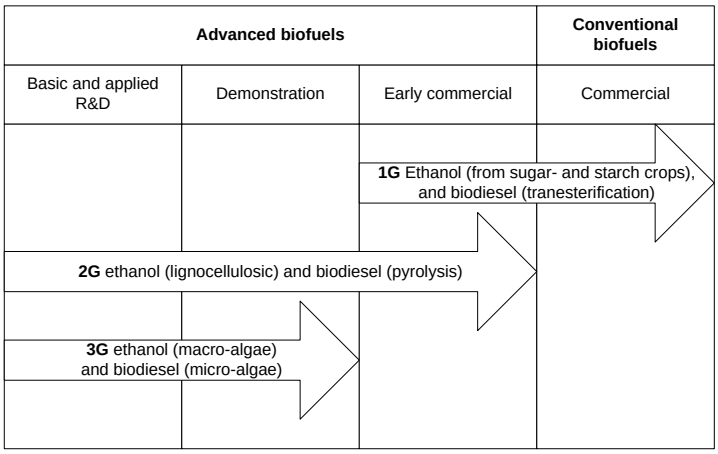

Figure 2 Stages of technological development of selected biofuel pathways illustrating the correlation between technology development (columns) and generations (arrows), adapted from (IEA 2011)

Acknowledging Tilman et al. (2009)'s concern with regards to generalization, the remaining three articles in this thesis (II, III and IV) present LCAs of representative 1st through 3rd generation biofuel pathways in Europe, i.e. 1st generation rape methyl-ester biodiesel, 2nd generation lignocellulosic bioethanol, and 3rd generation macro-algae bioethanol; these are indicated in Figure 1.

In article II, we compare and measure the well-to-wheel global warming potential (GWP) and local air pollution emissions[3] resulting from combining commercial 1st generation rape methyl-ester biodiesel and advanced exhaust aftertreatment in EU road-freight transport. The functional unit (FU) chosen is per tonne kilometre (tkm). Both the introduction of 1st generation biodiesel and advanced exhaust aftertreatment are the result of implementing two EU directives aimed at mitigating transport related- climate change (EU 2009) and local air pollution (EU 2005)(citation) respectively. This article disseminates the results of a European Economic Area project[4].

In article III, we acknowledge the advanced stage of technological development of 2nd generation lignocellulosic bioethanol; however, we question the completeness of existing LCAs of lignocellulosic bioethanol due to the common omission- or lack of consistency in cellulase enzyme production life cycle inventories (LCI). Cellulase enzyme production is a significant process with respect to the environmental impacts of lignocellulosic bioethanol. We determine and compare the environmental impacts of on-site submerged aerobic fermentation production of cellulase enzymes in full-broth based on three carbon sources: glucose, molasses and pre-treated softwood. The FUs chosen are per kilogram (kg) cellulase enzyme and per mega joule (MJ) lignocellulosic bioethanol.

In article IV, based on new cultivation and harvesting data for macro-algae from one integrated aquaculture demonstration site in Norway, we perform a cradle-to-gate LCA of an integrated aquaculture with biorefinery system. This system produces salmon (FU per kg wet-weight) and macro-algae biomass. The latter is further processed in a biorefinery as defined by Cherubini (2010) producing 3rd generation macro-algae bioethanol (FU per

[3] Nitrous oxides, particulate matter, carbon monoxide and non-methane hydrocarbons.
[4] BIODEG – Influence of bio-components content in fuel on emission of diesel engines and engine oil deterioration. Research Project No PL0261.

MJ ethanol) along with one high-value low-volume product (sodium alginate) and one low-value high-volume product (compost) both with FU per kg product.

For consistency, all LCAs apply attributional life cycle modelling (EU 2010; ISO 2006), and CML (non-)baseline life cycle impact assessment (LCIA) methods (PRé 2015). Articles II, III and IV filled at the time of their submission/publication the research gaps for their respective biofuel generations, and individually represent important contributions to this field. Table 1 summarizes the scope of Articles II, III and IV.

The specific goal & scope and research questions of each individual article are presented in the Results of the Thesis section, along with the most important results and conclusions. Complete versions of these articles are presented at the end of this thesis.

Structure of the Work

The remainder of this thesis is divided into five sections: Theoretical Basis, Methodology, Results of the Thesis, Discussion, Implications and Conclusions and finally Recommendations for Further Research. In the Theoretical Basis section a brief introduction to Industrial Ecology is presented. In the Methodology section a description of the key aspects of Life Cycle Assessment methodology is provided. A summary of Results of the Thesis follows with additional information provided in the attached full-articles. The Discussion is divided into two sections, each addressing one of the two research questions outlined in the Research Questions and Scope of the Thesis section. These discussions are summarized in the Implications and Conclusions section that leads to Recommendations for Further Research..

Theoretical Basis

Industrial Ecology

The Institute of Environmental Sciences – Leiden University (CML) provides a spatial representation of the two spheres in which we exist (CML 2003), i.e. society and the environment, see Figure 3. This representation and succeeding explanations can be interpreted as a techno-centric approach to the very complex interactions that occur within and between these two spheres (Giddings et al. 2002); however, this representation does allow one to firmly place LCA within context. As well, Figure 3 provides a spatial backdrop for the discussion of sustainability, and the solutions and strategies towards its achievement.

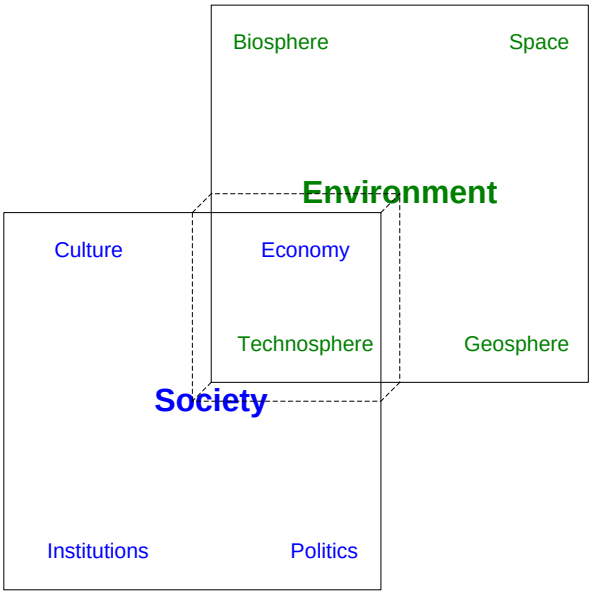

Figure 3 Spatial representation of society and the environment (CML 2003)

The theory of industrial ecosystems was formally proposed by Frosch and Gallopoulos (1989), and elaborated on by Frosch (1992) and Patel (1992) by which time the term ecosystems had been exchanged for ecology.

Based on the foreseen challenges that a growing society and its resulting demand for: resources, their transformation, use and disposal posed to the natural environment; these authors suggested that we rethink the way in which we approach industrial production. At the time (and currently) industrial processes possessed a linearity in which raw materials were extracted from the natural environment, and after transformation and use, were then returned as waste to the natural environment (Figure 4, left). Early industrial strategies for environmental management: dilution, end-of-pipe treatment, recycling and recovery, and cleaner production all to a lesser-or-greater degree follow this linearity.

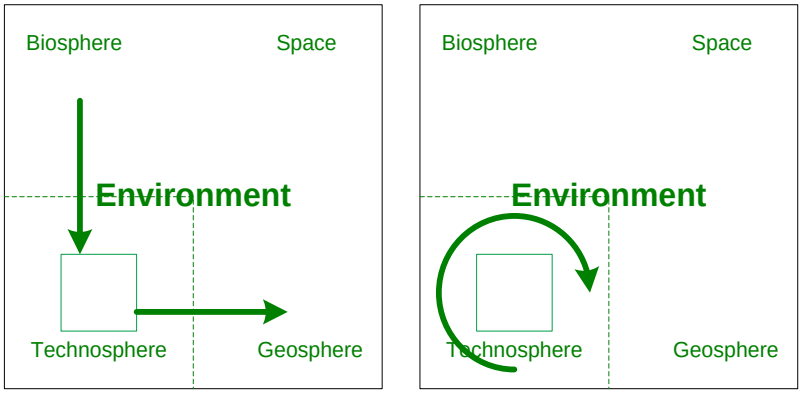

Figure 4 Depiction of product life cycles within the context of the environment, with linear industrial processes (left) and ecological industrial systems (right)

In contrast, Frosch and Gallopoulos (1989) proposed that we approach industrial production from a more holistic perspective, in which we consider the waste of one process as the potential material for other processes. They drew on a parallel between industry and ecosystems present in the natural environment where a symbiosis exists. The adoption of Frosch and Gallopoulos (1989)'s approach requires a departure from linear industrial processes to accepting a web-like nature of interactions between industrial process in the techno-sphere and the natural environment beyond. In fact, all three envisioned that the ideal industrial system would not interact with the natural environment (Figure 4, right) though admitted that this might be unachievable in practice (Frosch 1992; Frosch and Gallopoulos 1989; Patel 1992).

Returning to Figure 3, the interface between the economic and techno-sphere quadrants is where the production of products (techno-sphere) and their consumption (economy) occurs. The remaining three quadrants of the environment represent the natural environment; together, these four quadrants form the physical environment, and accordingly allow themselves to be quantified in physical units, e.g. tkm, kg, MJ etc. In contrast, the economy and remaining three quadrants of society represent the symbolic society, and do not allow themselves to be quantified in physical quantities, but in symbolic ones, e.g. monetary units, standard of living etc. One further distinction can be made between products and services; it can be argued that we do not consume products but the services that these products provide, i.e. services exist in the symbolic society. Correspondingly, products and their physical life cycles exist in the environment, or more specifically the techno-sphere quadrant of the physical environment.

Collecting the concepts of the web-like structure of industrial processes in the techno-sphere, and that products and their life cycles exist in the physical environment and can be quantified in physical units, it is possible to identify how industrial ecology contributed to the study of material and energy flows. These material and energy flows include both interactions within the techno-sphere and extraction of materials from- and emissions to the natural environment. Furthermore, it becomes easier to understand the areas of application of various life cycle approach assessment methods, summarized in Figure 5. These are: environmental input-output assessment (EIOA), and its hybrid-forms with LCA, multi-regional input-output assessment (MRIOA), general- and partial equilibrium models (GEM and PEM respectively), input-output assessment (IOA), life cycle costing (LCC) and social LCA (SLCA).

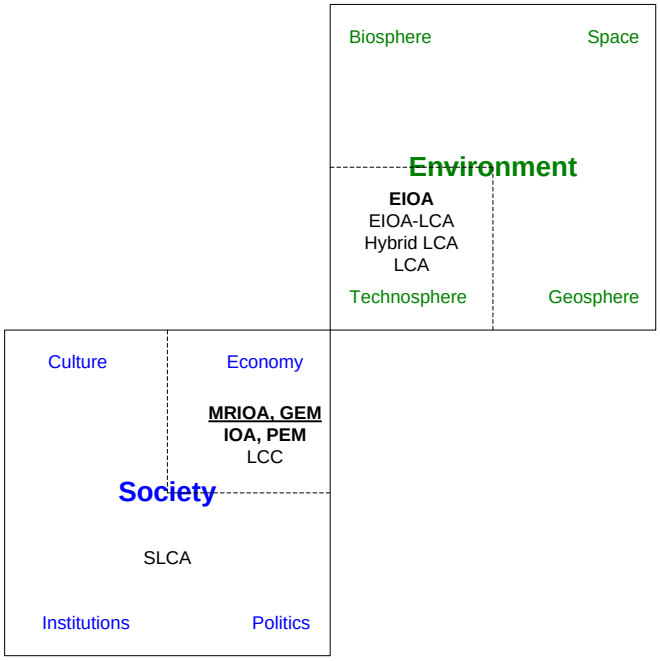

Figure 5 Life cycle approach methods and their areas of application, with macro-level methods (bold and underlined font), meso-level assessment methods (bold font) and micro-level assessment methods (normal font)

Figure 5 is the result of combining Guinee et al. (2010)'s depiction of LCSA with CML (2003)'s spatial representation, and clearly places LCA in its appropriate area of application. Additional life cycle approach methods do exist though are not indicated in Figure 5, but could easily be placed here based on their general descriptions.

Methodology

Life Cycle Assessment

The International Organization for Standardization's (ISO) defines LCA as a method which:

> *"…addresses the environmental aspects and potential* ***environmental impacts*** *throughout a* ***product****'s* ***life cycle*** *from raw material acquisition through production, use, end-of-life treatment, recycling and final disposal."(ISO 2006)*

This definition is consensual with the application discussed under Industrial Ecology, however it should be noted that some definitions include both products and services. While providing the first set of consensually accepted principles and framework the ISO (2006) series of documents remained somewhat vague in their formulation, undoubtedly the result of achieving consensus. In 2010, the publication of the European Commission's ***International Reference Life Cycle Data System (ILCD) Handbook*** (EU 2010) further developed the concepts of this series of standards; providing the detailed (technical) guidance necessary for LCA practitioners to perform and disseminate their studies with consistency. It is with a basis in these two documents that the LCAs performed in Articles II, III and IV of this thesis are performed.

LCA can be divided into four separate stages: goal and scope definition, LCI, LCIA and interpretation. Though separate, these stages are not independent, but form the iterative process of LCA as indicated by the multi-directional arrows in Figure 6.

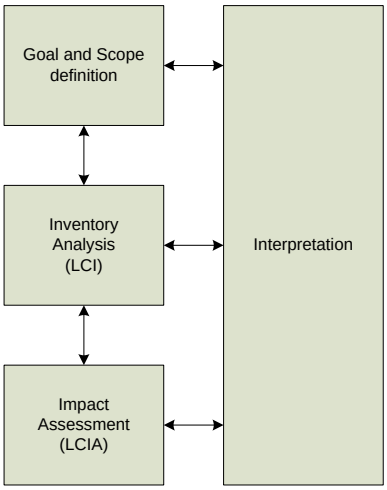

Figure 6 Stages of an LCA

A brief description of these stages is as follows:

Goal and Scope Definition is the first phase of LCA in which sufficient detailed information is provided with respect to: the system under evaluation, LCI and LCIA methodologies applied, data quality, foreseen application/dissemination and limitations of the LCA's results.

Life Cycle Inventory is the "phase of LCA involving the compilation and quantification of inputs and outputs for a product through its life cycle" (ISO 2006).

Life Cycle Impact Assessment is the "phase of LCA aimed at understanding and evaluating the magnitude and significance of the potential environmental impacts for a product system throughout the life cycle of the product" (ISO 2006).

Life Cycle Interpretation is the "phase of LCA in which the findings of… the impact assessment are evaluated in relation to the defined goal and scope in order to reach conclusions and recommendations" (ISO 2006). This phase can be equated with the discussion sections in Articles II, III and IV.

These definitions are consensual with those of the ***ILCD Handbook*** (EU 2010). In addition to these standards and guidelines, a ***Computational Structure of Life Cycle Assessment*** has evolved (Heijungs and Suh 2002).

The computational cornerstone of LCA is given by formula 1 (Heijungs and Suh 2002), and should remind one that the computational structure of almost all: LCA studies, software and databases is based on this simplified- linear treatment of a steady state situation.

$$s = A^{-1} \cdot f \qquad (1)$$

Where:

s	Scaling vector
A	Technology matrix
f	Final demand vector

The technology matrix (A^{-1}) is with some slight differences to an input-output model (Heijungs and Suh 2002) the same inverse matrix developed by Nobel Laureate Wassily Leontief in the 1930s (Leontief 1936). Thus, the A^{-1} matrix provides a solution to the challenge of web-like interconnections and loops existing in industry at the product level, and can quantify the direct and indirect activity of both foreground and background systems.

The f vector represents the LCI of the foreground system, with one of its f^{th} elements equal to one FU and the remaining positions the direct: material, energy and emission flows connected to the production of one FU.

The result is the s vector which presents the total (direct + indirect) activity (flows) necessary to supply the external demand of the FU. The s vector, due to the nature of its determination has the ability to account for a degree of physical transfer effects within the environmental sphere. These previous paragraphs present what entails the LCI stage of LCA.

Multiplying the s vector with the matrix of emission characterisation factors allows one to reduce the scale of results from 1000's of individual emissions to a manageable number of impact categories. Here, a distinction is made with respect to the level of characterization performed along the cause-effect (impact) chain, i.e. midpoint (<100 impact categories) vs. endpoint (<10 impact categories). This process is known as characterisation, and represents

the LCIA phase of LCA. The aim of which is the improvement of clarity and comprehension, however, along with these benefits come sacrifices (Bare et al. 2000). All LCIAs performed in Articles II, III and IV apply primarily the CML (non-)baseline midpoint impact assessment method (PRé 2015)

The reason for presenting the computational structure of LCA is that with an understanding of this, one can also understand the general application and limitations of LCA. From this description, it is clear that LCA is a product level assessment method that accounts for both direct and indirect activity. The determination of the A^{-1} matrix reveals some steady state transfer effects, though only in the environmental sphere. Thus, intra and inter-sphere (environmental and social) consequences are only partially accounted for through decisions made with respect to LCI modelling framework. Additionally, each LCA iteration is performed for one function (FU). Thus, multi-functionality, i.e. the instance of co-products (functions) must be reduced to LCIs providing individual functions.

This highlights three important considerations of LCA that must be addressed. First, numerous industrial processes and the products they produce do not exist, and their interaction with- and introduction to the techno-sphere can have significant consequences. The degree of consequences effected by the processes' or product's introduction can determine the type of LCI modelling framework chosen, and the level of decision support to which the results can be applied. Second, numerous industrial processes produce single products with multiple functions, while others produce multiple products; these products and processes are defined as multi-functional, and how one deals with these is one important aspect of LCA. The third aspect concerns data quality and its effect on LCA results.

Regarding the first consideration, the two LCI modelling frameworks are attributional and consequential. Attributional modelling "depicts the potential environmental impacts that can be attributed to a system over its life cycle… [based on] historical, fact-based, measurable data of known…uncertainty" EU (2010). In contrast, consequential modelling "aims at identifying the consequences that a decision in the foreground system has for other processes and systems of the economy…hence [it is] not reflecting the actual… average supply chain, but a hypothetic generic supply chain… [as] prognostizised along market-mechanisms, and potentially including political interactions and consumer behaviour changes" (EU 2010). Selection of either method is dependant on the decision-context (EU 2010; Tillman 2000; Wenzel 1998), and a LCA should reflect the environmental impacts of the change(s) resulting from those decisions made based on its results (Wenzel 1998). As long as these changes are marginal, attributional LCI modelling is sufficient, however, the broader the reach of change, the greater degree to which the consequences should be understood, i.e. consequential modelling should be used.

For Articles II, III and IV attributional LCI modelling was chosen. Article II presented the LCA of commercial 1st generation rape methyl-ester biodiesel, who's EU production/consumption has begun to level over the previous years (EurObserv'ER 2015). As such, the application of attributional LCI modelling elicits little debate. Concerning Articles III and IV, the EU reported in a recent scenario (Hamje et al. 2014) that 2nd generation lignocellulosic bioethanol will contribute < 1% to total road transport energy demand in 2020, and 3rd generation macro-algae bioethanol receiving no mention. As such

the authors felt the selection of attributional LCI modelling justified along with the addition of sensitivity analysis (Thomassen et al. 2008), and a reminder to policy makers that attributional results can be uncertain when applied at policy level (Brander et al. 2008).

Regarding the second consideration, multi-functionality, or more specifically, which portion of the f vector and resulting environmental impacts are associated with co-products or functions, ISO (2006) suggests a hierarchy of methods:

I. *Subdivision* requires that the process providing the product in focus is isolated from the larger production system, allowing one to formulate a LCI specifically for this product.

II. *System expansion* requires that either additional processes are added to, or processes are subtracted from the system under review in order to make this comparable with other systems.

III. *Allocation* accepts that neither subdivision nor system expansion are possible. In allocation, inputs and outputs are partitioned based on any number of properties, e.g. mass, energy or market price.

The choice between attributional or consequential LCI modelling can dictate which method is selected for dealing with multi-functionality, with subdivision or allocation often selected in attributional modelling, and system expansion in consequential modelling (Thomassen et al. 2008). Due to the complex systems modelled in Articles II, III and IV, subdivision was not an option. Numerous methods and criteria exist for allocation, most common are: main product bears all burden, substitution, energy, economic and mass with, with exergy and newer hybrid methods recently being proposed (Cherubini et al. 2011; Sandin et al. 2015). However, due to the varying functions of the co-products in the articles, results based on: main product bears all burden, energy, mass and exergy would not present representative results. The application of substitution and hybrid methods require that both the products and their substitutes are well known and documented in the literature, which is was not the case for the systems modelled in Articles III and IV. As a result, economic allocation was chosen for foreground processes when necessary.

The third and final consideration is the central role which data quality plays in LCA, and can be divided into four issues: representativeness, completeness, precision and consistency; with representativeness receiving a further division into three further categories: technological, geographical and time related (EU 2010). Furthermore, van den Berg et al. (1999) differentiate between model/method and data, providing two dimensions to the four data quality categories. Practically, representativeness and completeness equate to misused- and missing data, with precision (and accuracy) addressing discrepancies in the actual values chosen and their effect on determined results. Consistency is overarching and mandates both consistency in methods and assumptions, and consistently representative-, complete- and precise data. Insufficient quality can set limitations on a LCA study, and bring into question the validity of the results, their interpretation and application.

Effort was made in each article to achieve representativeness, completeness, precision and consistency with exceptions clearly stated. The most notable exceptions are presented in Table 3.

Table 3 Notable issues regarding representativeness, completeness and consistency in Articles II, III and IV

Article/category	Representativeness	Completeness	Consistency
Article II	Technological -Euro 3 motors were by the time of publication not representative of EU road-freight transport. -European transient cycle (ETC) testing of engines is not fully representative of actual transport	-Limited impact categories (GWP) due to the limited number of emissions measured in ETC testing -Engine wear and deterioration of motor oil were not accounted for.	-Some inconsistencies between primary empirical- and secondary database data.
	Geographical -Some Swiss LCIs used as proxies for Europe.		
	Temporal -Economic allocation values were outdated and not representative.		
Article III	Technological -Unknown representativeness of glucose production LCI.	-There were no notable issues concerning completeness identified in this article.	-No LCI information available for Case B as a result Case A was conditioned and used as a proxy for Case B. -Some inconsistency between the LCIs for carbon sources: glucose, molasses and pre-treated biomass.
	Geographical - Enzyme production LCIs for Europe based USA proxies. -German, Swiss and global LCIs used as proxies for Europe.		
	Temporal - Case C based on LCI data from 1999.		
Article IV	Technological -Biorefinery based on a collection of separate-basic and applied R&D laboratory experiments with unknown commercial representativeness. -Potential mismatch between salmon and macro-algae growth cycles not accounted for	-Due to the heavy reliance on basic and applied R&D derived LCI data for the biorefinery processes, and due to the proprietary nature of the LCI data for macro-algae cultivation and harvesting, it can be inferred that these are incomplete.	-Great variation in LCI consistency between fish- and macro-algae aquaculture & biorefinery due to the various levels of technological development. -Some inconsistency in economic allocation values used for the biorefinery co-products. -Inconsistency in impact categories due to the focus on the accurate determination of GWP and eutrophication potential.
	Geographical -European and North American LCIs used as proxies for Norway.		
	Temporal - Some LCIs for fish aquaculture- feed and infrastructure based on data from 2000 and 2001 respectively.		

Several methods have been proposed to determine the effect: representativeness, completeness, precision and consistency have on results, these are: contribution- and sensitivity analysis, and stochastic data uncertainty calculations (EU 2010). All of these methods were applied during the course of this thesis, see Table 4. In Article II, the use of the pedigree matrix method (Weidema and Wesnæs 1996), a stochastic data uncertainty calculation, was accompanied by a qualitative discussion of uncertainty at both the data and model/method levels. In the final two articles (III and IV) a combination of contribution- and sensitivity analysis accompanied by a thorough discussion of data quality was found to be the best combination of methods to address data quality.

Table 4 Data quality assessment methods applied in Articles II, III and IV

Article/method	**Contribution**	**Sensitivity**	**Stochastic data uncertainty calculation**
Article II	X		X
Article III	X	X	
Article IV	X	X	

Results of the Thesis

In this section, we summarize the research questions and main results/findings of the four articles, listed in Table 2, which constitute the body of this thesis.

Article I

Biofuels and Sustainable Transport: A Conceptual Discussion

Biofuels have been proposed as one strategy towards the achievement of sustainable development. However, the challenge is that there exists no political or scientific agreed definition for sustainable development instead it "is now like democracy': universally desired, diversely understood and extremely difficult to achieve, and won't go away" (Lafferty 2004). This poses a great challenge to anyone wishing to understand how biofuels can contribute towards achieving sustainable development.

The goal of this article was to disaggregate the concept of sustainable development into decreasing levels from the macro- to the micro levels (individual biofuel- strategies and pathways). In doing so, we present a clear path along which individual biofuels' contribution towards achieving SD can be understood, and just as important how we can compare biofuels against alternative solutions and strategies.

We begin by reiterating the conditions of SD from ***Our Common Future*** (level 1): safeguarding long-term ecological sustainability, satisfying basic human needs, and promoting intra- and intergenerational equity (Brundtland et al. 1987; Holden 2012). Second (level 2); we acknowledge that developing sustainable transport systems is crucial towards the achievement of SD. However, this requires re-interpreting the conditions of sustainable development specifically for transport, these are respectively: an upper limit on daily per capita energy consumption, a lower limit on daily per capita travel distance, a minimum of accessibility to transport, and a minimum of renewable transport fuels (Holden 2012). Sustainable transport as a general strategy for achieving sustainable development can be further disaggregated into three specific alternative strategies: improving efficiency, alteration of transport patterns and reduction in transport (level 3). We acknowledge the importance of implementing all three of these sustainable transport strategies, though we continue with a disaggregation of the strategy of improving efficiency. Improving efficiency (level 4) entails both the improvement of existing fuels/technologies, but also the introduction of new- more efficient fuels/technologies including alternative energy carriers (level 5) . Numerous energy carriers can be developed from existing primary energy sources; biofuels are some examples. However, even biofuels demonstrate great variability in both biomass feedstock and conversion technology. This has led to their separation into 1st, 2nd and 3rd generations (level 6), which both differentiates between their biomass feedstocks and resulting conversion technologies (Figure 1).

This conceptual discussion culminates in the development of a hierarchal biofuel-sustainability framework with which to compare biofuel strategies in general, along with individual biofuels against alternative: biofuels, alternative fuels, and technology- and

sustainable transport strategies towards addressing the four conditions of sustainable development, see Figure 7.

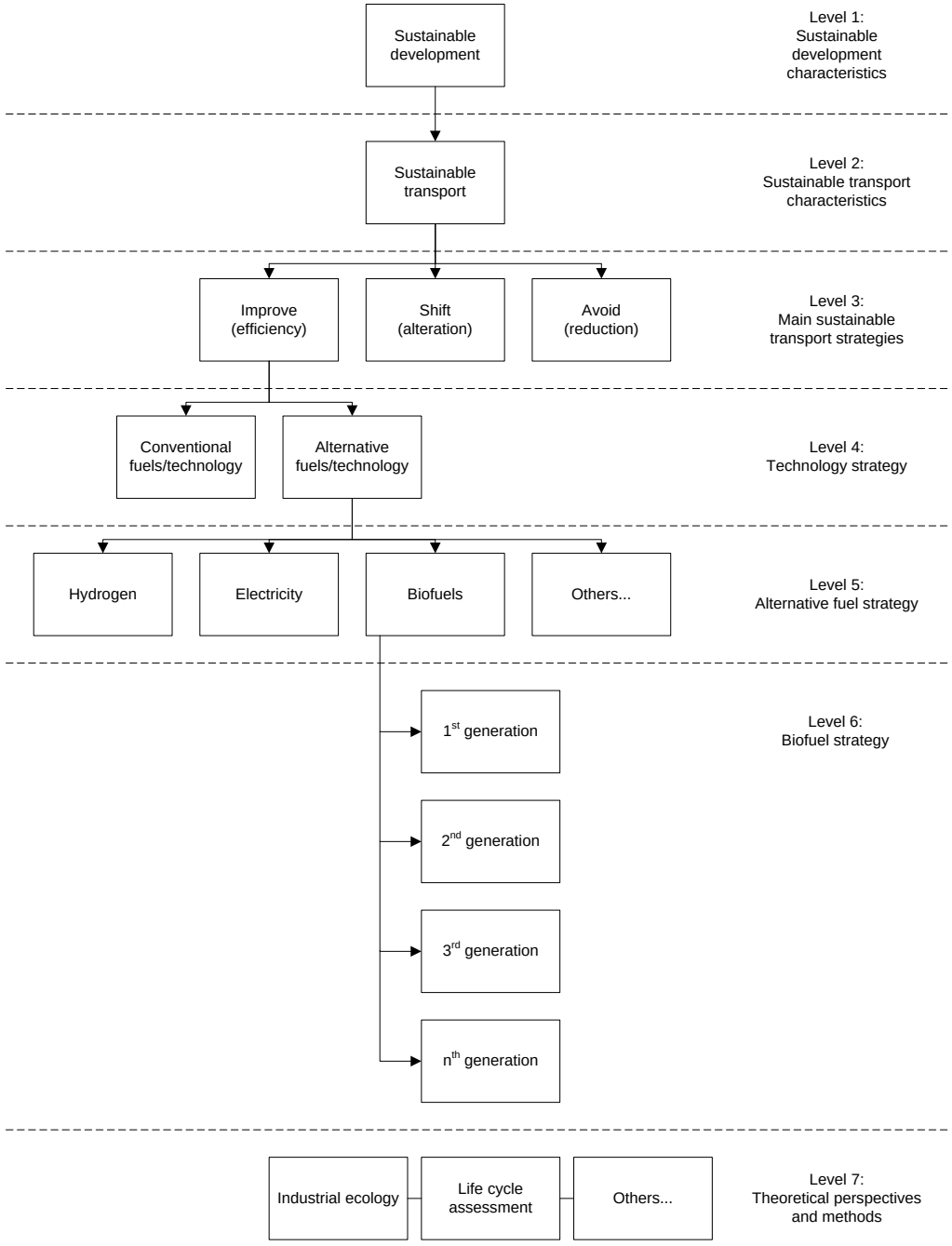

Figure 7 Biofuel-sustainability framework, version 1 (Article I)

In conclusion, we propose that in order to contribute towards the achievement of sustainable development, individual biofuels must compare favourably against other biofuels (level 6). Second, that biofuels in general must compare favourably against other alternative energy carriers (level 5) and other improvements in efficiency of conventional fuels/technologies (level 4). Next, that improving the efficiency of transport systems by

introducing biofuels must compare favourably against potential reductions in- and alterations to the transport system (level 3). Finally, these biofuels must satisfy the four conditions of sustainable transport (level 2) and the four conditions of sustainable development (level 1). Additionally, it must be stressed that the four conditions of sustainable development are subjective preferences (Daly 2007).

Furthermore, in order to facilitate comparison by measuring individual biofuels and biofuel strategies against alternatives at all levels, a reliable theoretical perspective and well-established assessment methods are necessary. We propose that the theoretical perspective of industrial ecology and LCA methodology are suitable when supplemented by other methods.

Article II

Biodiesel's and advanced exhaust aftertreatment's combined effect on global warming and air pollution in EU road-freight transport

As a result of European directives promoting biofuels aimed at mitigating the climate change effects of transport (EU 2009) there has been over a tenfold increase in the consumption of transport biofuel in the European Union between 2002 and 2014, i.e. from 1.1 Mtoe to 14.0 Mtoe respectively (EurObserv'ER 2015). In 2014, 79.7% of all transport biofuel consumed was in the form of biodiesel, particularly 1st generation rape methyl-ester biodiesel either pure or blended with fossil diesel in a 30% (B30) volume concentration. Parallel to this activity, the European Union has introduced directives (EU 2005) aimed at mitigating the local air pollutants: nitrous oxides (NO_X), particulate matter (PM), carbon monoxide (CO) and hydrocarbons (HC) emitted from European Union road-freight transport. Mitigation of local air pollutants has taken the physical form of two advanced exhaust aftertreatment systems: selective catalytic reduction (SCR) aimed at mitigating NO_X, and diesel particulate filters (DPF) aimed at mitigating PM.

LCAs that investigate biodiesel's- mitigating effect on climate change and advanced exhaust aftertreatment's mitigating effect on local air pollution independently are numerous. However, there were no LCAs that investigated the combined effect of these two mitigation measures on GHG emissions and local air pollution collectively (at the time of publication).

We asked the following research questions:

1. How do the different fuels: pure biodiesel (B0), B30 and pure biodiesel and the aftertreatment systems original equipment manufacture (OEM), SCR and DPF affect the life cycle GHG emissions from road-freight transport?
2. How do the different fuels (B0, B30 and B100) and the after treatment systems OEM, SCR and DPF affect the life cycle emissions of NO_X, PM, CO and non-methane hydrocarbons (NMHC) from road-freight transport?
3. What are the trade-off effects regarding fuels' and aftertreatment systems' respective abilities to achieve reductions in both GHG emissions and emissions of NO_X, PM, CO and NMHC?

Towards answering these research questions, the well-to-tank-, tank-to-wheel- and combined well-to-wheel GWP were determined using CML 2 baseline (PRé 2015), along with the well-to-tank-, tank-to-wheel- and well-to-wheel emission of local air pollutants (NO_X, PM, CO and NMHC) for various concentrations of biodiesel and configurations of exhaust aftertreatment, see Table 5. The FU chosen was per tkm. Uncertainty of results was determined using the Monte Carlo method, and based on limitations for calculating the real standard deviation of input values, the pedigree matrix method was chosen to estimate the geometric standard deviation (Weidema and Wesnæs 1996).

Table 5 Aftertreatment systems and fuels considered in this study

Fuel	**Aftertreatment**		
	OEM	SCR	SCR & DPF
B0	x	x	x
B30	x	x	x
B100	x	x	x

Engine operation emissions and -consumables for the tank-to-wheel LCI was based on primary- stationary measurement of an Iveco F1C Euro 3 engine under the European Transient Cycle and step load testing. Remaining tank-to-wheel LCI data was adapted from various sources: urea (Althaus et al. 2007; Nemecek et al. 2007), SCR and DPF (Dones et al. 2007), and vehicle and infrastructure (Spielmann et al. 2007). Well-to-tank LCI data was based on Jungbluth et al. (2007) and Dones et al. (2007) and conditioned to the appropriate fuel concentrations. The well-to-wheel results of this study are presented in Table 6.

Table 6 Well-to-wheel LCIA results for the aftertreatment systems and fuels considered in this study

Fuel	Aftertreatment system	GWP	NO_X	PM	CO	NMHC
		kg CO2 eq./tkm	g/tkm	g/tkm	g/tkm	g/tkm
B0	OEM	0,516	2,933	0,513	1,097	0,686
	SCR	0,518	1,223	0,487	1,125	0,576
	SCR & DPF	0,536	0,966	0,435	0,613	0,547
B30	OEM	0,467	3,298	0,569	1,074	0,597
	SCR	0,481	1,425	0,494	1,115	0,535
	SCR & DPF	0,482	1,095	0,459	0,666	0,491
B100	OEM	0,387	3,635	0,646	1,162	0,438
	SCR	0,396	1,853	0,544	1,209	0,402
	SCR & DPF	0,403	1,702	0,532	0,759	0,382

It was found that the complete substitution of biodiesel (B100) for fossil diesel (B0) decreases the overall GWP. The application of: SCR reduces NO_X, PM and NMHC emissions, and combined with DPF reduces all local air pollutants (NO_X, PM, CO and NMHC). All of these findings verify that independently biofuels and advanced exhaust aftertreatments are effective at mitigating climate change and local air pollution respectively. When biodiesel, SCR and DPF are combined, the trade-offs are increased NO_X, PM and CO with the addition of biodiesel, and an increase in GWP with the addition of advanced exhaust aftertreatment.

In conclusion, despite being independently effective, the combination of these two mitigation measures results in some trade-offs. However, further research is needed to reduce the uncertainty of results, along with developing more complete engine emissions testing regimes.

Article III

Comparative attributional life cycle assessment of European cellulase enzyme production for use in second-generation lignocellulosic bioethanol production

With the implementation of the ***Renewable Energy Directive*** (EU 2009) the European Union has set an ambitious target of 10% renewable energy in the transport sector by 2020, incl. biofuels. As a result, the European Commission projects that despite an overall decrease of 8.5% in demand for road transport by 2020 that demand for biofuels will increase to 21.5 Mtoe of which lignocellulosic bioethanol will contribute 0.7 Mtoe (Hamje et al. 2014).

The most common proposed path for converting lignocellulosic biomass into lignocellulosic bioethanol is via the bio-chemical process of enzymatic hydrolysis and fermentation. Key to this process is the enzymatic hydrolysis of cellulose and hemicellulose into fermentable sugars by cellulase enzymes, of which large amounts are required, i.e. 0.3–2.1 g of cellulase enzymes per MJ lignocellulosic bioethanol (Hong et al. 2013; Humbird et al. 2011; MacLean and Spatari 2009).

The most common proposed method for cellulase enzyme production in LCA literature is submerged aerobic fermentation based on one of three proposed carbon sources; glucose, molasses or pre-treated softwood. This is a resource and energy intensive process, and represents one of the greatest environmental and economic uncertainties connected to lignocellulosic bioethanol production (Foust et al. 2009; Luo et al. 2009a). Despite this, cellulase enzyme production is often excluded from LCAs of lignocellulosic bioethanol (Borrion et al. 2012; Wiloso et al. 2012); who cite the lack of LCI data for enzyme production.

We asked the following research questions:

1. Which cellulase enzyme production path with respect to carbon source provides the lowest environmental impacts?
2. How do the environmental impacts determined in this study compare with those determined in similar studies?
3. If current enzyme production methods are over- or underestimated, or omitted what inferences can be made with respect to the representativeness of existing LCAs of LCB?

Towards answering these research questions the environmental life cycle impacts of on-site submerged aerobic fermentation production of cellulase enzyme in full-broth in Europe based on the three proposed carbon sources: cornstarch glucose (Case A), sugar cane molasses (Case B) and pre-treated softwood (Case C) (numerous authors) were determined and compared. In addition to the CML 1A baseline and non-baseline (land-use LU) impact categories we determined cumulative energy demand (Frischknecht et al. 2007). LCIs were adapted from the literature (Humbird et al. 2011; Wooley et al. 1999), with material consumption calculated from stoichiometric equations based on the elemental composition of cell mass and cellulase enzymes (Atkinson and Mavituna 1991; Humbird et al. 2011; Wooley and Putsche 1996), and nutrient requirements based on volume/mass flows (Schell et al. 1991). We conducted sensitivity analysis' for all major assumptions, including the effect of market changes based on the novel approach of advanced attributional life cycle assessment (Andrae 2015). We consistently and transparently presented all LCIs and results and compared these with similar studies, see Table 7.

Table 7 Summary of LCIA results including a comparison with other relevant studies including their mean, and standard deviation (2σ or 95% confidence), with: eutrophication potential (EP), acidification potential (AP), ozone layer depletion potential (ODP), photochemical oxidation potential (POP).

	GWP100	**EP**	**AP**	**ODP**	**POP**	**LU**	**Cumulative energy demand**
Unit	kg CO_2 eq.	g PO_4 eq.	g SO_2 eq.	mg CFC-11 eq.	g C_2H_4 eq.	m^2a	MJ
Case A	10.6	44.2	49.3	0.4	1.6	0.5	81.2
Case B	9.1	24.8	54.5	0.1	4.9	4.3	62.3
Case C	7.9	8.7	31.6	0.2	2.0	41.4	52.4
Weighted Mean & standard deviation[a]	$(14.7 \pm 13.7)^F$ $(6.3 \pm 4.0)^{NF}$	$(22.2 \pm 18.7)^F$ n/a	$(210.4 \pm 343.1)^F$ n/a	$(1.9 \pm 0{,}76)^F$ n/a	$(11.1 \pm 10.9)^F$ n/a	n/a n/a	$(120.2 \pm 118.9)^F$ n/a
(Olofsson et al. 2015)	5.5^F						69^F
(Agostinho et al. 2014)	21.93^F		7^F				1664^F
(Hong et al. 2013)	10.2^{NF}–16.0^F						
(Dunn et al. 2012)[b]	3.7^F						46^F
(Harding and Harrison 2011)							$(53.5–190.2)^{F,c}$
(Kim et al. 2009)	$(16–25)^F$	$(11.5–18.3)^{F,b}$	$(120–145)^{F,b}$		$(5.8–7.6)^{F,b}$		$(117–207)^{F,b,d}$
(MacLean and Spatari 2009)	2.3^{NF}						$24.8^{NF,e}$
(Harding 2008)	$(-1240$–$-924)^F$	$(22–37)^F$	$(270–510)^F$	$(1.52–2.28)^F$	$(11–20)^F$		$(88.8–190.2)^{F,c}$

[a] Does not include cumulative energy demand of Agostinho et al. (2014) or GWP of Harding (2008)
[b] Determined- from figure, or through calculation
[c] Only (direct) electricity and steam consumption
[d] Only non-renewable energy consumption
[e] Only fossil energy consumption
[F] Formulated
[NF] Non-formulated

It was found that cellulase enzyme production based on pre-treated softwood (Case C) provided the lowest environmental- and energetic impacts, followed by sugar cane molasses (Case B) and finally corn starch glucose (Case A). Some variation exists between the results of this and similar studies, though this studies results correlate well with those of other studies. However, the results are sensitive to process efficiency as estimated by carbon atom selectivity, electricity source, and to some degree market changes.

It was concluded that considering cellulose enzyme requirements; this processes' contribution to total well-to-tank life cycle impacts of lignocellulosic bioethanol production is significant and cannot be overlooked or omitted, as is often the case (Borrion et al. 2012; Wiloso et al. 2012), and that doing so would compromise the representativeness, completeness and consistency of such studies (Luo et al. 2009b).

In addition to the results, discussions and conclusions; this article provided consistent and transparent LCI data which can be applied to LCAs where on-site cellulase enzyme production is present, thus filling the existing data gap (Borrion et al. 2012).

Article IV

A cradle-to-gate attributional life cycle assessment of integrated aquaculture in Norway for sustainable food and fuel production

Two challenges facing society are climate change and feeding an ever-increasing population. With 70% of the world's surface covered by oceans, focus is shifting towards this new frontier for the provision of food and fuel.

Salmon aquaculture is a well-established and growing food production industry (FAO 2014). However, further expansion is hindered by numerous unresolved environmental issues (NMFCA 2011), e.g. marine eutrophication. One proposed solution is the co-cultivation of nutrient- and carbon dioxide sequestering macro-algae in close proximity to salmon aquaculture. This so-called integrated aquaculture system aims to mirror the symbiosis existing in natural ecosystems and in combination with a biorefinery can provide a source for environmentally sustainable: food-, chemical- and energy production. Considering the complexity of these integrated systems and only through an equally integrated LCA can consistency in methodology, data, and results be achieved, and the risk of transfer effects (Article I) and other direct- and indirect environmental consequences (Tilman et al. 2009) be avoided.

We asked the following research questions:

1. What are the environmental impacts associated with integrated aquaculture and biorefinery produced co-products: sodium alginate, ethanol, compost and salmon, and how do these compare with the results of other studies?
2. Based on these results what general observations and conclusions can be drawn concerning integrated aquaculture systems and biorefineries?

Towards answering these research questions we performed a LCA of an integrated- salmon (S. *salar*) and macro-algae (S. *latissima*) aquaculture system producing both fish, and macro-algae biomass for further processing in a biorefinery to multiple co-products in Norway. LCI data for fish aquaculture was adapted from numerous literature sources, and

the feed production LCI was developed from the most recent values for feed composition (FHL 2013). LCI data for macro-algae cultivation and harvesting were based on empirical data collected from a pilot facility in Norway. Nutrient and carbon balances between fish- and macro-algae were based on Wang et al. (2012). The macro-algae biomass is processed in a biorefinery as defined by Cherubini (2010) producing: one high-value low-volume product (sodium alginate 90%), one biofuel (ethanol 99.7%), one low-value high-volume product (compost), with some of the biorefineries energy requirements met by biomass combined heat and power generation. LCI data for the biorefinery represented a novel configuration of processes proposed independently in the literature.

The results indicated that the integration of salmon- and macro-algae aquaculture can be beneficial with respect to nutrient- and carbon dioxide sequestration, i.e. 92.33 g carbon dioxide, 2.45 g nitrogen and 0.31 g phosphor sequestered per one kg wet-weight macro-algae produced. Salmon aquaculture is both resource- and energy intensive particularly those processes related to feed production. The recent trend towards higher concentrations of crop-based feed ingredients negatively effects the environmental profile of salmon aquaculture. The production of the biorefinery co-products carry high environmental impacts due to the chemicals used in the production of sodium alginate and ethanol, of which a portion is economically allocated to compost. However, with this said, this studies results are within the range of those determined in similar studies when considering variations in methodology and LCIs.

Table 8 Absolute LCIA results for the production of all aquaculture and biorefinery products, with cumulative energy demand (CED)

		Sodium alginate	**Ethanol**	**Compost**	**Salmon**
	Unit/FU	1 kg	1 MJ	1 kg	1 kg
GWP	kg CO_2 eq.	102.83	0.31	0.23	3.40
CED	MJ	1556.19	4.67	3.50	14.63
EP	g PO_4 eq.	231.46	0.69	0.52	166.85
AP	g SO_2 eq.	1103.35	3.31	2.48	19.89
ODP	mg CFC 11 eq.	9.893	0.030	0.022	0.372
POP	g C_2H_4 eq.	47.808	0.143	0.107	0.487

In conclusion, integrated aquaculture is beneficial though more research is necessary to determine the actual nutrient- and carbon dioxide sequestration potential (Agnalt et al. 2011), and eventual mismatch in growth cycle between salmon- and macro-algae aquaculture (Broch et al. 2013; van Hal 2012). Additionally, further research is necessary to determine suitable biorefinery configurations, though it can be expected that the extraction of high-value low-volume products can influence the environmental profile of the remaining co-products (Skjermo et al. 2014), which exhibit high sensitivity when economic allocation is applied to products with little-to-no existing market (Burton et al. 2009). Finally, when suitable processes have been investigated and towards the achievement of true integration, the use of macro-algae biomass for fish feed production should be considered.

Discussion

In this section, each research question will be individually reiterated followed by a discussion of how observations from the four articles written contribute towards addressing- and answering each question.

Research Question 1:

What is the contribution of individual biofuel pathways towards the achievement of sustainable development?

Article I developed the understanding of the contribution of individual biofuel pathways towards the achievement of sustainable development; in which we disaggregated the concept of sustainable development into six levels summarized in the biofuel-sustainability framework (version 1) presented in Figure 7.

The framework presented in Figure 7 has evolved somewhat from its original form, and Figure 8 presents a revised version of this framework.

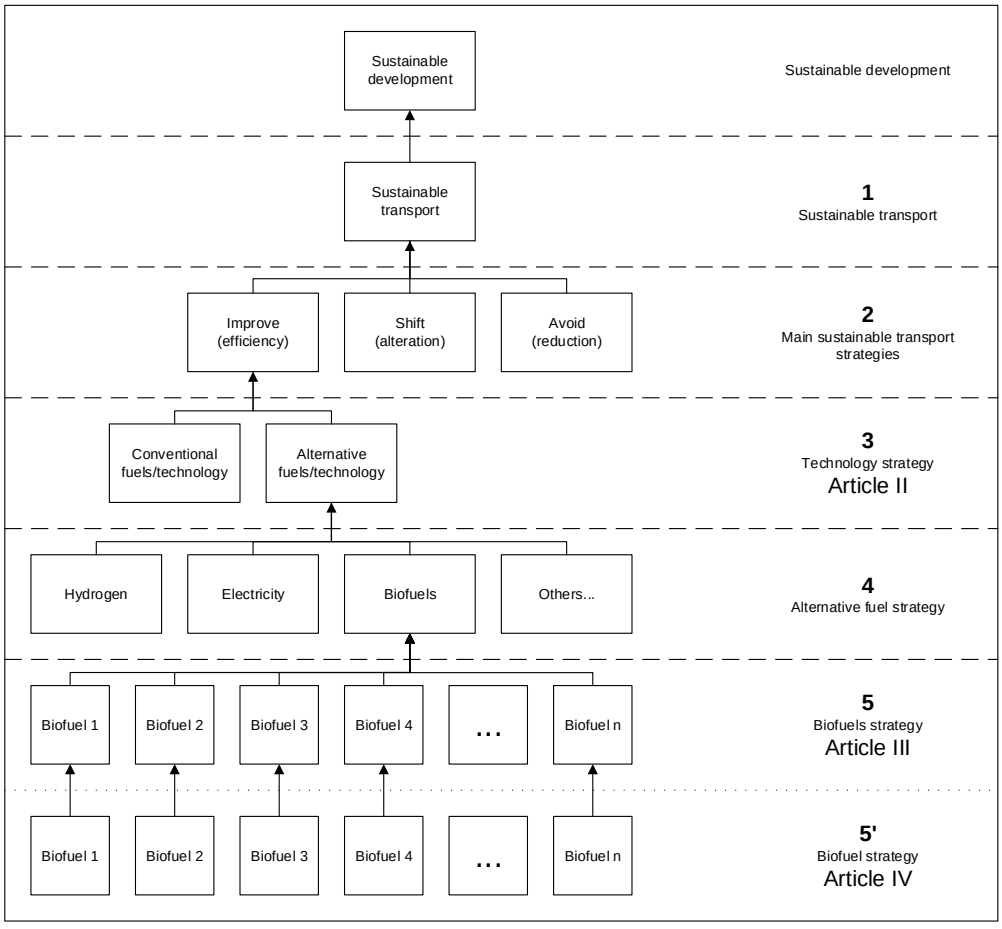

Figure 8 Biofuel-sustainability framework, version 2

The main revisions are first, a new numbering sequence of 5 levels; where the highest level (sustainable development) does not receive a number. As such, numbering represents the levels of separation from addressing the original four conditions of sustainable development (Brundtland et al. 1987; Holden 2012). Sustainable transport (level 1), is then

disaggregated into decreasing levels of aggregation until one eventually arrives at individual biofuel pathways (level 5).

The second change is the addition of a "sixth" level 5', and acknowledges that at an early stage of technological development, comparisons between alternative biofuel pathways are not possible for arguments of maintaining consistency. In other words, at early stages of development, see Figure 2, the lack of data- quality and quantity limits one's ability to perform any meaningful comparison with other: biofuels, alternative fuels or technologies. Instead, at level 5' the only meaningful comparison is between variations within one biofuel pathway. Thus, the outcome of level 5' is the identification of the biofuel pathway's strengths and weaknesses, so-called hot-spot identification, and the application of these findings towards improving the process.

The final revision is the removal of the separation of biofuels into generations (level 6 in Figure 7, and level 5 in the revised Figure 8), in favour of individual biofuels. This acknowledges that regardless of feedstock- or conversion technology, the gains of individual biofuel pathways should compare favourably to other individual biofuel pathways for inclusion in biofuel strategies and advancement to higher levels of comparison. This develops the understanding that in the future it will be necessary to make a shift from a homogeneous- to a heterogeneous alternative fuel strategies (Gilpin 2010).

Article I explains in detail the terms and concepts at each level on the biofuel-sustainability framework. Furthermore, it is important to note that only the direct path linking sustainable development to individual biofuel pathways is depicted, and that other parallel frameworks, e.g. agriculture-sustainability- or tourism-sustainability frameworks exist.

Thus, Article I presents the relationship between individual biofuel pathways and sustainable- transport and development. The biofuel-sustainability framework disaggregates the concept of sustainable development into easier manageable sets of solutions and strategies. Despite the framework's development in a top-down approach, the framework suggests that comparisons between: biofuel pathways, alternative fuels, etc., proceed in a bottom-up sequence. In other words, biofuels, either individually or as part of a strategy, must compare favourably with alternative solutions at each respective- and increasing level on the proposed biofuel-sustainability framework. The comparison, and eventual favouring of one option over another assures that the most suitable combination of solutions or strategies is chosen.

Research Question 2:

What are the limitations of LCA in evaluating the sustainability of biofuels within the context of the biofuel-sustainability framework?

Article I introduced both the theoretical perspective of industrial ecology and LCA methodology. Industrial ecology and LCA were included in Figure 7, however, their relation- or application with respect to the assessment of biofuels or sustainable- transport and development were not specifically identified.

LCA assesses the environmental impacts of a product throughout its life cycle, and compares these either explicitly or implicitly with other product(s) offering the same

function. In the section Industrial Ecology it was explained that LCA's application is limited to the quantification of material- and energy flows in the techno-sphere, and exchange of resources and emissions (impacts) between the techno-sphere and natural environment. Considering LCAs area of application (Figure 5) and the four conditions of sustainable development: safeguarding long-term ecological sustainability, satisfying basic human needs, and promoting intra- and intergenerational equity (Brundtland et al. 1987; Holden 2012), it can be acknowledged that while biofuels have implications for all four conditions (Article I), LCA is most suitable for addressing aspects of the first condition, which is consensual with Finkbeiner et al. (2010), among others.

This is an important re-statement when considering that at the political level, the question of biofuel sustainability has been predominantly reduced to a question of GHG reduction potential- quantified by LCA methodology (Mondou and Skogstad 2012), in addition to effects on biodiversity and land-use. These three criteria all relate to the first environmental condition. Clearly, the paradigm shift from environmental protection to sustainability which has occurred over the past decades at the academic level (Finkbeiner et al. 2010) does not appear in applied policy.

Both Finkbeiner et al. (2010) and Guinee et al. (2010) propose life cycle sustainability assessment as the best means of addressing all four conditions of sustainable development. It should be noted that neither author proposes the expansion of one field of methods to other fields, i.e. micro- or macro level- environmental or social (incl. economy). Instead they propose a collected framework of independent methods rooted in the industrial ecology perspective of holistic life cycle thinking. Which together can provide an indication (indicators) of sustainability. Furthermore, it is only through such life cycle sustainability assessment methods that unintended consequences and transfer effects can potentially be identified within- and between the social- and environmental spheres, at both the product- (micro) and macro levels (Article I).

Some variations in scope of methods exists between Finkbeiner et al. (2010) and Guinee et al. (2010). The former focuses on: environmental-, economic- and social sustainability at the product level and propose: LCA, LCC and SLCA respectively as sufficient assessment methods. These methods are also identified by Guinee et al. (2010) who continue to expand life cycle sustainability assessment from the product- (micro) to the macro levels. Figure 9 presents Guinee et al. (2010)'s life cycle sustainability assessment framework, with Finkbeiner et al. (2010)'s framework represented by the bottom row. It should be noted that methods for assessing the social sphere is wanting; particularly when considering that formal SLCA methods are still at an early stage of development (Benoît 2010).

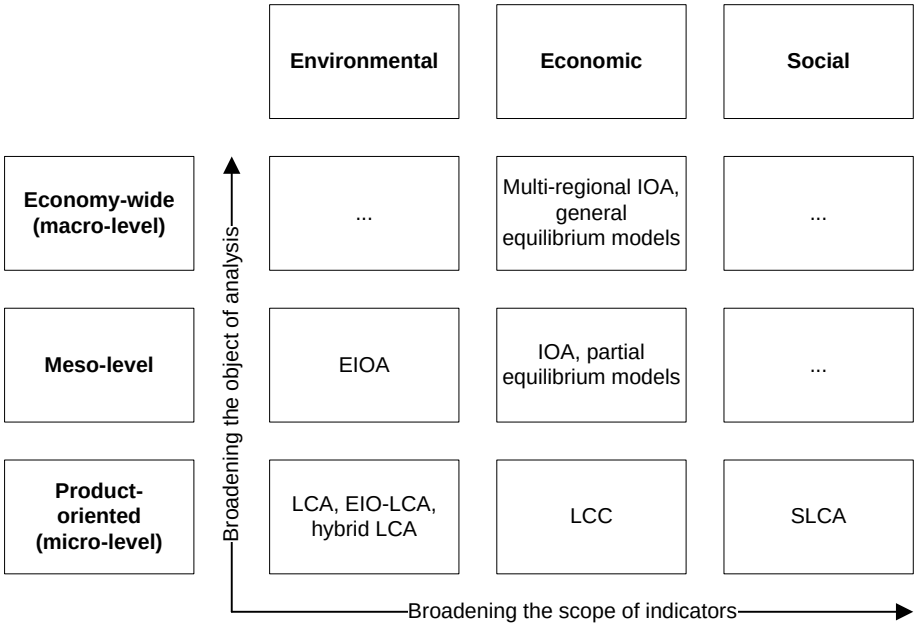

Figure 9 Proposed integrated life cycle sustainability assessment framework (Finkbeiner et al. 2010; Guinee et al. 2010)

Finkbeiner et al. (2010) extended their discussion of life cycle sustainability assessment with the proposal of various methods to characterize individual environmental-, economic and social indicators. This is similar to the LCA process of characterizing individual emissions to midpoint- or endpoint indicators, and aims at improving clarity and comprehension. This is implicitly supported by Hoekstra (2015) who explains that the concept of sustainability is only meaningful at the macro-level, and that the aggregated effect of numerous interconnected activities is what can result in (un)sustainability, i.e. a specific activity is not isolated, but part of a larger context. However, similar to the LCA discussion of midpoints vs. endpoints, characterization of independent values carries with it both benefits and sacrifices (Bare et al. 2000). As an alternative, Guinee et al. (2010) promote the idea of a transdisciplinary integrated framework. Here it is clear that not only is the concept of sustainability, but also the development of its assessment, like democracy "universally desired, diversely understood, and extremely difficult to achieve [consensus]" (Lafferty 2004).

Thus, it is of importance to achieve a consensual agreement of which specific life cycle approach assessment methods are necessary to assess the four conditions of sustainable development as part of a life cycle sustainability assessment framework. Towards this aim, the area of application- and limitations to individual assessment methods must be defined in context- here LCA within the context of the proposed biofuel-sustainability framework.

The preceding discussion presents two alternatives; first, to assume that only macro-level methods are suitable to address the macro-level conditions of sustainable development, this would entail that LCA, which is a product (micro-) level assessment method would become obsolete as individual biofuel pathways are aggregated into fuel-, technology- and transport strategies as suggested by Hoekstra (2015). The second alternative proposes that as one moves from the product level (biofuels) towards addressing the conditions of sustainable development and its assessment, that a broadening of the life cycle

sustainability assessment framework in all dimensions is required, arrows in Figure 9, where LCA remains one element.

However, regardless of whether LCA becomes obsolete or marginalized, equating to the first and second alternatives respectively, the outcome is similar. As one moves from comparative environmental assessments at the product level (biofuel) through the development of: biofuel-, technology and transport strategies towards addressing the conditions of sustainable- transport and development, both alternatives suggest that LCAs of individual biofuel pathways lose their significance at higher levels on the proposed biofuel-sustainability framework. Thus, LCAs significant contribution is restricted to the lower levels in Figure 8, though it remains to determine which levels.

Towards determining this, one must recall two points discussed in the Life Cycle Assessment section. First, the computational structure of LCA allows one function to be assessed at one time, and second, that the product's (or service's) function is limited to the physical environment, as identified by the FU defined in the goal and scope stage of any LCA.

Starting with the second point first. At levels 5 and lower (5') the comparison is between individual biofuel pathways. When comparing individual biofuel pathways MJ is a suitable FU based on the *ceteris paribus* principle, i.e. the vehicle life cycle is the same or similar. LCAs that determine the environmental impacts per MJ are known as fuel-cycle, cradle-to-gate or well-to-tank assessments. However, when comparing alternative fuels (level 4) or technologies (level 3) neither the vehicle's life cycle and/or emissions dominating life cycle phase(s) are the same or similar. As a result, well-to-tank comparisons are no longer feasible; instead, well-to-wheel LCAs are performed, with the FUs: tkm for freight or passenger kilometre (pkm) for passenger transport are used. Thus, advancement from level 5 to 4 and 3 corresponds to an extension or expansion of the FU from MJ to tkm or pkm. This shift in FU also reflects a shift from product to service comparisons.

The real extension, or expansion, in function(s) begins with advancement to higher levels of comparison, e.g. level 2; where individual biofuel pathways might be compared to using a bicycle (shift) or not travelling at all (avoid). Though still representing products or services, the functions they provide vary greatly. For example using a bicycle not only moves a person from A to B, but also improves health, and working from home entails no travelling at all (avoid), and might improve well-being. In order to consistently compare alternatives at these levels, the range of functions must be extended or expanded even further (EU 2010), and will include functions present in the social sphere (incl. economy), which is beyond the area of application of LCA, see Figure 5.

It is now possible to return to the first aspect, i.e. that the computational structure of LCA allows one function to be assessed at one time. Some might suggest applying any one of the three methods for dealing with multi-functionality presented the Life Cycle Assessment section: subdivision, substitution/system expansion or allocation, at these higher levels. However, it is important to reflect on what this would entail, i.e. equating the social- (incl. economy) sphere to the physical environment. This is not the area of application an any single life cycle approach method, but can only be achieved by the collection of several life cycle approach methods, see Figure 5.

Thus, considering these two points, one can determine the range of levels on the biofuel-sustainability framework where LCAs of biofuels represent a significant contribution to the discourse; these are from levels 5' to 3. With this first area of application of- or limitations to LCAs of biofuels with respect to sustainable development identified, other limitations can be sought based on observations from Articles II, III and IV.

In Articles II, III and IV, a selection of representative biofuel pathways were chosen for LCA evaluation; Table 1 summarized their scope. Each of these articles represents a unique contribution to this field. However, a quantitative comparison between these articles is not possible. For this, inter-article consistency in methods, assumptions and data would have to exist. Instead, in the process of addressing each article's respective research question(s) specific variations in LCA methodology, assumptions and data have arisen. As a result, general observations from these three articles forms the discussion of additional limitations to LCA towards evaluating individual biofuel pathways and their eventual sustainability within the context of the biofuel-sustainability framework.

Articles II, III and IV present LCAs of either complete biofuel pathways or aspects of each of the three generations of biofuels, i.e. 1st-, 2nd- and 3rd generation respectively. Article II presented the LCA of 1st generation rape methyl-ester biodiesel blended with fossil diesel in combination with advanced exhaust aftertreatment in EU road-freight transport. Article III presented the LCA of cellulase enzyme production, one significant aspect of 2nd generation lignocellulosic bioethanol production. Article IV presented the LCA of an integrated aquaculture system with biorefinery producing 3rd generation macro-algae bioethanol.

Furthermore, the International Energy Agency defines four stages of technological development of biofuel pathways in ascending order: basic and applied R&D, demonstration, early commercial and commercial (IEA 2011). From Figure 2 one can infer that an inverse relation exists between the numerical generation of a biofuel pathway and the stage of its technological development. This is reflected in Articles II, III and IV, where each of the specific biofuel pathways exists at a different stage of technological development. First-generation rape methyl-ester biodiesel (Article II) is commercially available. Second-generation lignocellulosic bioethanol (Article III) currently spans the frontier between demonstration and early commercial as reflected in the growing number of commercial start-ups. Third-generation macro-algae bioethanol (Article IV) has not advanced past basic and applied R&D, though independent elements of the integrated system are commercially available, e.g. fish aquaculture (FHL 2013), and alginate production (Skjermo et al. 2014).

The LCAs performed in Articles II, III and IV have been placed at their respective levels in Figure 8. Article II compared various configurations of improved conventional fuels/technology (fossil diesel and exhaust aftertreatment) and alternative technology (biofuels), this places the comparison in this article at level 3. Article III, while not explicitly performing a comparison between lignocellulosic bioethanol and other biofuels; the contribution of the improved process of cellulase enzyme production facilitates such a comparison. As a result, the comparison presented in Article III is placed at level 5. Based on the early stage of technological development of the biorefinery process modelled, the

aim of Article IV was to identify areas of major environmental concern as a contribution to the R&D process, placing this article at level 5'.

Each of these articles identified and contributed to the then existing research gap(s) of their respective biofuel generation. Based on these findings, it would seem as if a relation does exist between the stage of technological development, and the level of comparative assessment performed with respect to the proposed biofuel-sustainability framework. However, does this relation exist beyond the scope of this thesis' articles?

Previously, it was acknowledged that the FU chosen can be used to identify the level of comparative assessment on the biofuel-sustainability framework. Additionally, it was identified that as one advances upward in the framework from comparisons between individual biofuel pathways to comparisons between biofuel-, alternative fuel- and technology strategies, and towards facilitating fair and consistent comparisons, an extension or expansion of function(s) will occur. Therefore, it should be possible to use the FU chosen in other LCAs to identify the level of comparative assessments performed with respect to the biofuel-sustainability framework. This, combined with identifying the generation or stage of technological development of the biofuels assessed, should provide an indication if a relation between technological development of individual biofuel pathways and the level of comparative assessment performed exists in the literature.

A literature review of representative 1st through 3rd generation LCAs was undertaken when writing Articles II, III and IV, and I assume that this is sufficient to provide an indication as to whether this relation exists in the literature.

When collecting information for Article IV, a comprehensive overview of relevant literature was possible, reflecting the limited number of LCAs of macro-algae biofuels. Aresta et al. (2005), Aitken et al. (2014) and (Boonstra 2015) all explicitly chose the FU MJ. Whereas Alvarado-Morales et al. (2013) choose the FU per one tonne biomass (dry matter); though provide information which can be used to implicitly determine environmental impacts per MJ. Langlois et al. (2012) chose per pkm as the FU even though they make no comparison with other (bio)fuels. In fact, none of these authors compare the assessed biofuel pathways with other distinctly different (bio)fuel pathways. This clearly places these studies at level 5' on the framework.

In Article III, a similarly comprehensive overview of literature regarding cellulase enzyme production was possible; once again reflecting the limited number of LCAs of enzyme production available. However, towards drawing conclusions about cellulase enzyme's contribution to lignocellulosic bioethanol production two comprehensive literature review studies were cited (Borrion et al. 2012; Wiloso et al. 2012) which encompassed 77 individual literature references encompassing a total of 88 LCAs of lignocellulosic bioethanol. While overlap has not been controlled for, of the 88 LCAs of lignocellulosic bioethanol: 36 were well-to-tank LCAs and 52 were well-to-wheel LCAs, with FU per MJ (or litre) and pkm respectively. The expansion of functional unit from mega joule to kilometre reflects a advancement in the level of assessment from one of evaluating individual biofuel pathways (level 5') to one of comparisons between: bio-, alternative, and conventional fuels. This places the studies reviewed at levels 5, 4 and 3 on the framework.

In Article II, the focus was on tank-to-wheel emissions, and LCI data for well-to-tank 1st generation rape methyl-ester biodiesel was obtained from secondary sources (Jungbluth et al. 2007). No extensive literature review was performed for well-to-wheel LCAs; instead it was assumed that a consensus regarding ranges of inputs/outputs and emissions had been achieved, loosely reflected in this biodiesel's inclusion in LCA software databases in both per MJ and per km (tonne and passenger) forms. Supporting this, both Cherubini et al. (2009) and Cherubini and Strømman (2011), two extensively cited review articles, place the discussion of 1st generation biofuels, incl. rape methyl-ester biodiesel, frequently at the level of well-to-wheel comparisons between biofuels and conventional fuels. This indicates that the level of comparative assessment for 1st generation biofuels in the literature has advanced to levels 4 and 3 on the biofuel-sustainability framework.

The previous three paragraphs indicate that the relation between stage of technological development of individual biofuel pathways and the level of comparative assessment on the framework appears to exist not only in Articles II, III and IV, but also in the literature. However, does an interpretation exist for this relation?

A differentiation between levels[5] of LCA was explained by Wenzel (1998) who discerned between: matrix-, screening- and full LCA. Wenzel (1998) proposed a direct relationship between the level of LCA and necessity for new- empirical data and calculations as opposed to secondary data, and as a result a direct relation between the level of LCA and data- quality and quantity.

Data- quality and quantity is central to performing an LCA (EU 2010). All full LCAs using LCA software and commercial databases, by nature of the applied computational structure (Heijungs and Suh 2002) will rely on a combination of primary- and secondary LCI data to varying degrees. This is achieved either by the collection of primary data or from scientific literature and commercial databases, dependant on data availability and time (Wenzel 1998).

From Articles II, III and IV, it can be observed that as biofuel's technological development advances, secondary data- quality and its availability increases. In Article II, primary data was collected for vehicle operation, and supplemented by conditioned- representative secondary data, allowing for a narrow foreground system. In Article III, new calculations were performed for cellulase enzyme production based on three unique carbon sources. Due to the stage of technological development, i.e. early commercialization, and resulting proprietary nature of higher tier foreground LCI data, it was necessary to expand the foreground system boundaries to achieve consistent data quality. Finally, in Article IV primary data for macro-algae cultivation was acquired from a demonstration integrated aquaculture site in Norway. Due to the early stage of technological development of the proposed integrated aquaculture-biorefinery system, a large foreground was required and relied on conditioning secondary data from basic and applied R&D. The finite availability of either representative- primary or secondary data in Article IV presented a limitation to the level of LCA performed, i.e. a non-comparative LCA.

Thus, it can be interpreted that with advancement in technological development and resulting increase in data- quality and quantity that the ability to perform higher level

[5] Not to be confused with the levels on the proposed biofuel-sustainability framework presented in Figure 8.

LCAs with respect to the biofuel-sustainability framework is possible. In other words, over time, numerous iterations of increasingly higher quality[6] LCAs are made, and the determined environmental impacts of individual biofuel pathways converge to narrower ranges of values. With consensual ranges achieved at the first levels 5 and 5' in Figure 8, the research gap (horizon) advances to higher level(s), i.e. the environmental comparison between biofuels- and other alternative- and conventional fuels, i.e. levels 4 and 3.

[6] Referring to representative, complete, precise and consistent LCAs

Implications and Conclusions

In the thesis, the growing consensual desire to assess the sustainability of products- either individually or combined as solutions and strategies towards the achievement of sustainable development was presented. Additionally, the non-consensual status of recommendations for how the scientific community should approach the life cycle sustainability assessment of products, solutions and strategies was discussed.

It was recommended that towards achieving consensus, the structure, or hierarchy, of possible solutions and strategies that can ultimately contribute towards the achievement of sustainable development and the specific limitations of the assessment methods proposed to compare these solutions and strategies must be disaggregated. Only through the disaggregation of the concept of sustainable development into easier manageable solutions and sets of strategies, and the identification of area(s) of application- or limitations to: environmental- and social (incl. economic) sphere assessment methods can overlap and confusion be reduced, and a movement towards consensus achieved. This thesis proposes one possible disaggregation for the specific case of biofuel's contribution towards the achievement of sustainable development, and the disambiguation of LCAs role towards their assessment.

This was achieve through the research presented in the four articles which constitute the body of this thesis. The specific implications of each individual article's research is appropriately presented in the discussion and conclusions of these articles.

The overarching implication is that despite the existence of standards and guidelines for LCA (EU 2010; ISO 2006) and a wealth of scientific knowledge concerning biofuels, there still exists a misconception concerning the significance of LCA results to the assessment of biofuel sustainability.

This thesis reminds the scientific- and public communities that LCA results alone of biofuels do not constitute a sufficient basis for assessing their sustainability. Instead, this is limited to providing an indication of the environmental impacts of biofuels either individually or as solutions up-to-and-including technology strategies on the biofuel-sustainability framework presented (Figure 8). Above this level, the functions diversify to the extent that several life cycle approach assessment methods are necessary for comparison. This does not imply that LCAs of biofuels are unimportant, only that they are less significant to the discourse at levels of sustainable- transport and development. LCAs of biofuels will though continue to be important tools to discern between proposed individual biofuel pathways, and fuel- and technology strategies.

Furthermore, the eventual presence of a temporal dimension to the spatial biofuel-sustainability framework questions the ability of LCAs of biofuels at early stages of technological development to contribute to the assessment of sustainability in combination with other life cycle approach methods. One could venture so far as to say that only LCAs of biofuels at pre-commercial or higher levels of development are based on suitably high quality data that their results can be applied to the discussion of sustainability.

Finally, this latter implication is not to be confused with- but comes in addition to the debate concerning LCI modelling framework, i.e. attributional vs. consequential. The

definition of consequential LCA (EU 2010) reveals that this LCI modelling framework does not compensate for the lack of data- quality or quantity at early stages technological development. In fact, it is this lack of knowledge concerning the future which constitutes one weakness of consequential LCA methods (Thomassen et al. 2008).

Recommendations for Further Research

Recommendations for further research is divided into two categories: specific recommendations with respect to improvement or additions to the studies presented in articles II, III and IV, and general recommendations with respect to biofuels and LCA in the contexts of sustainable development and life cycle sustainability assessment respectively.

Article II presented the LCA comparison of various bio- & fossil diesel blends and exhaust aftertreatment combinations in EU freight transport. This article, and particularly the data on which it was based, is now outdated. However, the issue that this article addresses, i.e. environmental trade-offs when combining two separate technologies aimed at mitigating two separate environmental impacts is still relevant as both EU directives (EU 2005; EU 2009) are still in use. Furthermore, the vehicle-, aftertreatment- and biofuel technologies along with our understanding of their potential environmental impacts has evolved. A new LCA of relevant combinations of these new technologies would increase to our understanding of their combined benefits and trade-offs. Steps towards fulfilling this recommendation were achieved prior to my departure from the Western Norway Research Institute when I participated on a successful research application to the European Economic Area – Norway Grants investigating the use of lignocellulosic bioethanol in the European transport sector.

Article III presented a LCA comparison of processes for producing cellulase enzymes based on three carbon sources for the production of lignocellulosic bioethanol. A full LCA as defined by Wenzel (1998) was not performed for lignocellulosic bioethanol which included these results, instead a scanning LCA based on secondary sources was performed. Due to the stage of technological development of lignocellulosic bioethanol production (early commercial), and the potentially non-marginal contribution of lignocellulosic bioethanol to transport fuels, a full- consequential LCA of could be highly informative to policy makers and is recommended.

Article IV presented a LCA of integrated aquaculture with biorefinery in Norway; a system which is currently at the technological development stage of basic & applied R&D. As highlighted in the discussion, this limited the level and representativeness of the LCA performed. The co-author and myself have acknowledged that whereas this article is publishable, a more representative article is possible as the stage of technological development advances in the coming year(s). It is the intention of the co-author and myself to focus on the collection of data that will address/solve the concerns we have with the evaluated system. The main concerns are:

- What non-marginal consequences resulting from introducing new macro-algae derived biorefinery products will have on their respective markets; here a consequential LCA could be recommended.
- The central biorefinery process of alginate extraction is based primarily on one source (Langlois et al. 2012). Either supplementing this with additional literature sources or modelling an alternative system based on new empirical data would alleviate this concern.

- The evaluated aquaculture system is integrated but not multi-trophic; for this, products from the biorefinery should be re-introduced (consumed) in the aquaculture system. This can be achieved by including the extraction of macro-algae protein for fish-feed production, a process currently being researched by the co-author.
- Macro-algae-, its composition- and rate of nutrient & CO_2 sequestration are not static; instead, there are seasonal- and growth stage variations, e.g. (Broch et al. 2013). A new LCA incorporating these fluctuations either centrally or as a sensitivity analysis would improve representativeness.
- Finally, including the wider effects on marine biodiversity and marine-use that an integrated- or multi-trophic aquaculture system have would make LCAs of 3rd generation biofuels more comparable to those of 1st- and 2nd generation studies.

A recurring recommendation is the implementation of consequential LCI modelling. This recommendation is made despite little-to-no practical experience with consequential LCI modelling. I have attended numerous Ph.D. courses and participated in numerous national and international research projects involving LCA, however, all have approached the theme of LCA with attributional LCI modelling. This is despite a general acknowledgement of the benefits of consequential LCI modelling under certain circumstances. It is my observation that this is a common situation in the field of LCA, i.e. consequential LCA is repeatedly discussed, but seldom performed. My set of skills would benefit over the coming years from performing consequential LCAs and comparing these with attributional LCA results. Publication of these methodology comparative studies would provide the scientific community and myself with the academic basis for discussing the attributes of these two alternative methods. As an added benefit, the application of consequential LCI modelling would also alleviate the need to apply (economic) allocation to solve for multi-functionality, as performed in Articles II, III and IV. The use of economic allocation between biodiesel and its co-products in Article II, and the biorefinery co-products in Article IV raises particular concern.

In general, this thesis contributes to the discourse concerning the role of biofuels towards the achievement of sustainable development, and LCAs contribution towards their assessment. With this said, this thesis by no means closes these discussions, instead, it opens for more research and discussion. Specifically, it will be important to develop a similar understanding of the area of application of- and limitation to additional life cycle approach methods proposed in life cycle sustainability frameworks, and their contribution towards the assessment of sustainable- transport and development, among other. To achieve this, interdisciplinary research groups encompassing a broad range of life cycle approach methods from the environmental- and social (incl. economy) spheres is necessary. The Sogn og Fjordane University College, in collaboration with national and international researchers is building such an interdisciplinary research group with an increasing collection of projects and publications. Additionally, a broader literature review is necessary to substantiate the relation identified in the

Discussion between the stage of technological development and level comparative assessment. Combined, these points will contribute to the evolution of the biofuel-sustainability framework presented in Figure 8, and its extension to other fields.

Finally, the transport sector, and specifically biofuels will continue to play an important role in our society for at least the near- and mid-term. As argued in this thesis, LCA is an important method with which to evaluate the environmental impacts of individual biofuel pathways. Tilman et al. (2009) points out that conclusions concerning sustainability cannot be drawn about biofuels in general, and that great variation exists between individual biofuels considered. This, combined with the understanding of the relations developed in this thesis clearly indicate that much research work awaits LCA practitioners, particularly in the assessment of yet-to-be commercially developed biofuels, and particularly those involving biorefinery processes.

From a LCA methodology perspective, this will require the refinement of: LCI modelling framework, how one deals with multi-functionality, and the broadening- and development of new- as of yet irrelevant life cycle impact categories and methods for their characterization.

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Article

Biofuels and Sustainable Transport: A Conceptual Discussion

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Abstract: Strategies for sustainably using biofuels must be thoroughly assessed at several levels. First, the use of biofuels must comply with sustainable development's main dimensions. Second, the use of biofuels must comply with sustainable transport's main dimensions. Third, gains from using biofuels strategies must compare favorably to gains from other sustainable transport strategies, such as altering transport patterns and reducing transport volume. Fourth, the gains must compare favorably to gains from improving conventional fossil-fuel-based advanced vehicles. Fifth, the gains must compare favorably to gains from using other alternative fuels. Sixth, the gains from using one generation of biofuels (e.g., first generation) must compare favorably to gains from using others (e.g., second through fourth generations). Performing scientifically sound and fair comparisons demands reliable theoretical perspectives and a well-established methodological basis. Industrial ecology theory and life cycle assessment methodology, respectively, are well-suited for these tasks.

Keywords: ecological sustainability; sustainability assessment and strategies; biofuels; sustainable development; sustainable transport; industrial ecology; life cycle assessment

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Abbreviations

ALCA: attributional LCA
CLCA: consequential LCA
GHG: greenhouse gas
ICE: internal combustion engine
IEA: International Energy Agency
ISO: International Organization for Standardization
LCA: life cycle assessment
LCA-IO: input-output macro-economic model
LCSA: life cycle sustainability analysis
MRIO: multi-regional input-output
OECD: the Organisation for Economic Co-operation and Development
UNEP: United Nations Environmental Programme
RED: Renewable Energy Directive
WIO: waste input-output
WTM: world trade model

1. Introduction

Climate change impacts, together with an increasing demand for energy, volatile oil prices, and energy poverty have led to a search for energy options that will be economically efficient, socially equitable and environmentally sound. One option that has raised significant interest from a wide range of actors is increased use of biofuels. Encouraged by research indicating that biofuels could provide substantial energy while at the same time mitigating climate change, governments have supported production aimed at increasing biofuel use in many countries. Farmers seek additional income, and biofuels may have the potential to promote rural development and access to energy in poorer countries. Industry has invested significantly in production and technology development. The number of scientific publications devoted to biofuels is growing rapidly, as is the number of reviews [1]. Thus, the use of biofuels is seen as an important pathway to achieving sustainable transport.

However, there are major concerns about the negative implications of growing biomass for biofuel production. Currently, biofuels are often made from feedstock crops that also serve as food. Hence, there is a potential risk for competition between food and fuel, which could result in consequences on food prices. Another identified risk is expansion of biofuel feedstock production into ecosystems that support high biodiversity and other services that are crucial to our economies and human life. Moreover, the envisaged positive effects on climate mitigation could turn out to have the opposite effect because of land-use changes associated with expanding agriculture [1].

Thus, two important issues regarding the merits of biofuels must be addressed. Are biofuels really sustainable; that is, do they comply with the main dimensions of sustainable development? Moreover, how do they compete with other strategies and technologies to comply with these dimensions? The aim of this paper is to provide a framework for assessing these issues with a particular focus on sustainable transport.

We argue that an assessment of the merits of biofuels requires discussions at six levels (Figure 1). As a starting point, the main dimensions of sustainable development must be defined. To meaningfully interpret sustainable transport, we must sort out these dimensions. Second, we must translate these dimensions into transport dimensions, giving relevant substance to the sustainable transport concept. Third, we must outline the competing strategies for achieving sustainable transport. The strategies are efficiency (e.g., developing improved fuels and technologies), alteration (e.g., promoting mode shifts) and reduction (e.g., avoiding trips) [2]. Fourth, we must compare technology strategies. At this level, the relative merits of conventional and alternative fuels are compared. Fifth, we must compare the relative merits of various alternative fuels. A large number of alternative fuels (e.g., biofuels, electricity, and hydrogen) have been suggested as appropriate solutions, and their respective qualities must be compared and assessed. Sixth, we must discuss and compare the various generations of biofuels. Biofuels can originate from various sources (e.g., sugar cane, wood, or algae), take different forms (e.g., gas or liquid), and use different technologies (e.g., ICEs or fuel cells). Additionally, we must find appropriate theoretical perspectives (e.g., industrial ecology) and methods (e.g., LCA) to make sustainability assessments at each level.

To assess the comparative merits of a specific solution at a given level (e.g., promoting second-generation biofuels at level 6), two questions must be addressed. (1) How does this solution compare to other solutions *at the same level*? (2) How does this solution compare to solutions *at higher levels*?

A comprehensive assessment at each level can hardly be done in one article. Rather, we discuss some of the main issues at each level and present a few examples to illustrate how assessments can be made. We focus on passenger mobility, but many of the conclusions may eventually be relevant for goods transport as well. Moreover, the arguments are based from a Western (European) point of view. Yet, sustainable transport is a global challenge and therefore throughout the article the challenge of achieving sustainable transport in both developed countries and developing countries is addressed. Thus, our conclusions may turn out to be relevant for a number of developing countries as well.

2. Sustainable Development

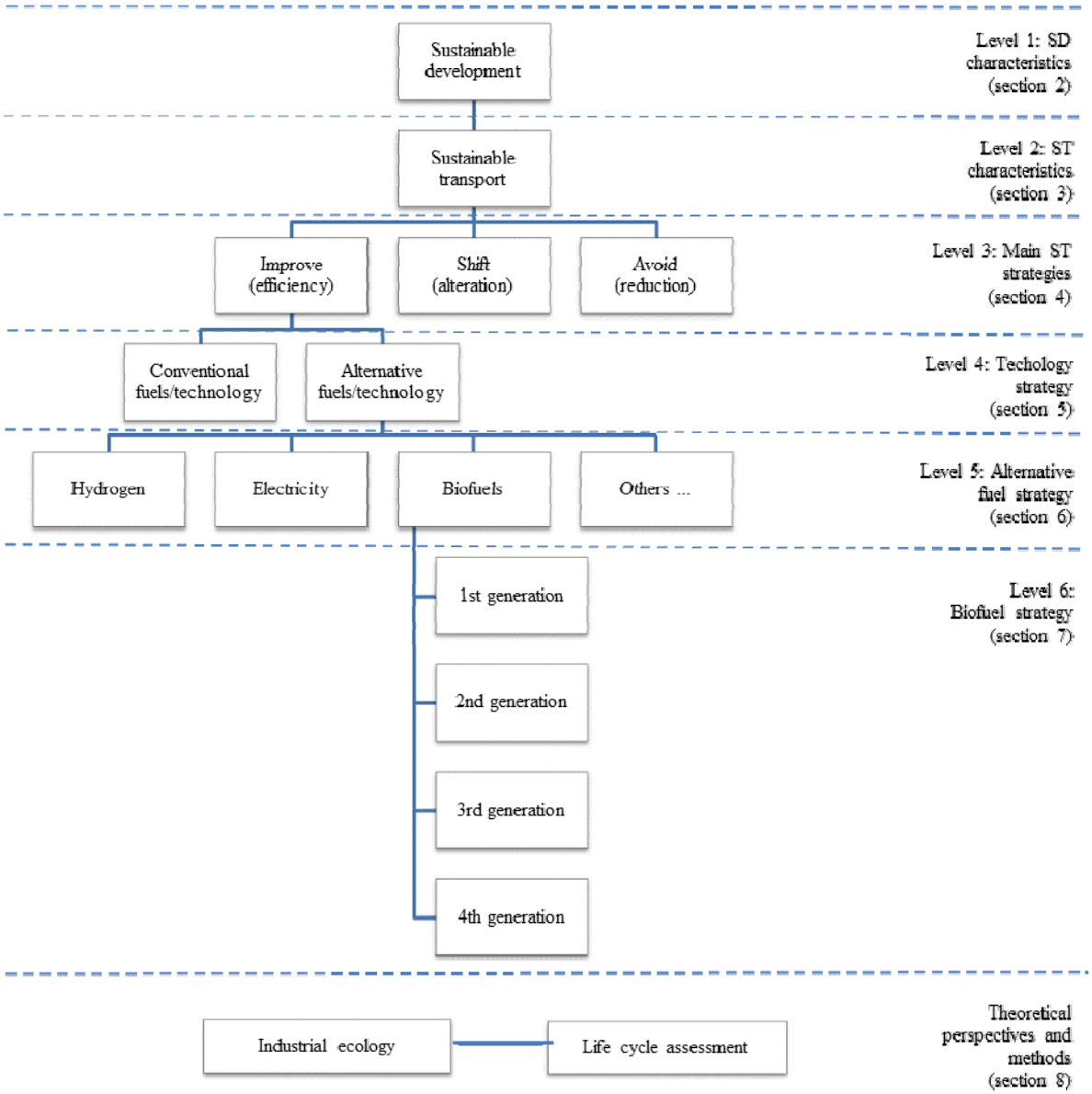
Sustainable development has increasingly been presented as a pathway to all that is good and desirable in society. The list of proposed indicators is long, and it grows longer yearly. Thus, the sustainable development concept has become so comprehensive and complex that it is becoming less helpful in guiding policymaking. Not surprisingly, a number of scholars have argued that the sustainable development concept is about to become useless, if it has not already done so.

Even though there is not yet any political or scientific agreement on a definition of sustainable development, it remains remarkably persistent as an ideal political concept, similar to democracy, justice, and liberty [3]. Indeed, sustainable development "is now like 'democracy': it is universally desired, diversely understood, extremely difficult to achieve, and won't go away" [4].

Unquestionably, sustainable development still is an important concept, which was clearly illustrated at the United Nations Conference on Sustainable Development, held in Rio de Janeiro in June 2012. One of the conference's main outcomes was the agreement by member states to set up sustainable development goals, which could be useful tools in achieving sustainable development. Thus, achieving

sustainable development is still high on the international and national agendas 25 years after the concept was launched with the publication of *Our Common Future*, commonly referred to as the Brundtland Report [5].

Figure 1. Sustainable development (SD), sustainable transport (ST) and biofuels.



Notes: (i) Level 1 to 6 represent a hierarchy of biofuels and sustainable development relations; theoretical perspective and methods represent one way of assessing the relations; (ii) Reference to article sections in brackets on the right-hand side in the figure; (iii) Main ST strategies: Improve (efficiency) = using new, conventional and/or alternative technology; Shift (alteration) = changing the prevailingly transport pattern into one based on public transport systems; Avoid (reduction) = decreasing present transport volume.

However, to become a useful tool, the concept must be clearly defined. Four main dimensions can be derived from the Brundtland Report: (1) safeguarding long-term ecological sustainability, (2) satisfying basic human needs, and promoting (3) intragenerational and (4) intergenerational equity [6]. These dimensions are what Daly refers to as "fundamental objective values, not subjective individual preferences" [7]. Thus, they are not negotiable.

In addition to the main dimensions, Høyer [8] presents a number of *secondary* dimensions, which include preserving nature's intrinsic value, promoting protection of the environment, promoting public participation, and satisfying aspirations for an improved standard of living (or quality of life). These secondary dimensions are subordinate to the main dimensions. Thus, preserving nature's intrinsic value (a secondary dimension) must give way whenever basic human needs (a main dimension) are threatened. Correspondingly, satisfying aspirations for a better life (a secondary dimension) should be subordinate to safeguarding long-term ecological sustainability (a main dimension).

Relevance for Biofuels

The use of biofuels clearly has relevance for all four main dimensions. Biofuels' potential to reduce GHG emissions and thereby safeguard long-term ecological sustainability is one of the reasons why biofuels have been introduced in the first place. Indeed, given the right conditions, biofuels have been shown to have a great potential to reduce GHG emissions.

The "food or fuel" debate clearly shows that biofuels have relevance to satisfying basic human needs [1]. Concern has been growing about negative implications of growing biomass for biofuel production. As stated previously, there are concerns with concern to food and expansion of production into valuable ecosystems.

The relevance to equity is twofold. First, biomass feedstock for biofuels production is readily available in most countries and could promote intragenerational equity. Second, biofuels are continuously renewable and could promote intergenerational equity. The main point, however, is that biofuels' merits must be assessed across all of these dimensions.

This point has been acknowledged for some time though, for example in the 2006–2009 *Bioethanol for Sustainable Transport* project [9]. Yet, the current overall trend is that, as policies surrounding biofuels (and more generally bioenergy) become more holistic, sustainability has become a stronger criterion at the starting point of policy development [10,11]. This has occurred in the EU, the USA and China, but also in many developing countries such as Mozambique and Tanzania. There are now more than 70 registered initiatives worldwide to develop and implement sustainability frameworks and certification systems for bioenergy and biofuels, as well as in agriculture and forestry (more on this in Section 7), but this can also lead to a fragmentation of efforts [10]. The four dimensions presented here should perform as guidelines for all such initiatives.

3. Sustainable Transport

Since launching their 1992 *Green Paper on the Impact of Transport on the Environment*, the EU has had sustainable transport as an overriding goal in its transport policy [12,13]. Since then, the EU has continued pursuing this goal in two *White Paper*s [14,15].

Still, as the EU emphasizes in their 2011 *White Paper*, the transport system is not sustainable: "Looking 40 years ahead, it is clear that transport cannot develop along the same path. If we stick to the business as usual approach, the oil dependence of transport might still be little below 90%, with renewable energy (hereafter, RE) sources only marginally exceeding the 10% target set for 2020. CO_2 emissions from transport would remain one third higher than their 1990 level by 2050. Congestion costs will increase by about 50% by 2050. The accessibility gap between central and peripheral areas

will widen. The social costs of accidents and noise would continue to increase" ([14], p. 4). Thus, finding ways to make transport sustainable remains high on the political agenda.

In fact, in most developed countries, the present transport patterns are unsustainable [16–21]. Moreover, there is still no political or scientific agreement on a definition of sustainable transport or on the required policies to achieve it. Rather, the use of the concept has increasingly reflected socially desirable attributes of local- and project-level problem solving, ignoring the global challenges that the concept was meant to solve [19]. To reflect local or project-specific challenges, a diversity of definitions and interpretations of the concept has been presented in the literature with the risk that the concept has become diluted and will end up as mere rhetoric offering little actually guidance for policymakers and scientists [19].

However, the main dimensions for sustainable transport can be derived from each of the main dimensions for sustainable development, thereby ensuring that the concept of sustainable transport reflects the four main dimensions of sustainability as described in the Brundtland Report. Thus, we argue that the main dimensions of sustainable transport are that it establishes [19]:

- an upper limit on daily per capita energy consumption for passenger transport to safeguard long-term ecological sustainability;
- a lower limit on daily per capita travel distance for motorized transport to satisfy basic transport needs;
- a minimum fraction of the total population that must have accessibility to public transport to promote intragenerational transport equity; and
- a minimum fraction of transport fuel that must be provided from RE sources to promote intergenerational transport equity.

We argue that the four main dimensions (e.g., safeguarding long-term ecological sustainability) and their corresponding indicators (e.g., per capita energy consumption for passenger transport) represent equally important targets where each needs to be fulfilled. This excludes the possibility of trading off an underperformance on one indicator against an over performance on another. Transport policies must reflect this; for example, will an affordable, biofuel-based public transport system reduce per capita energy consumption, satisfy basic transport needs for all population groups and promote RE sources.

Other important dimensions of sustainable transport certainly exist, for example, minimizing noise and reducing congestion. However, focusing on these less important dimensions (that is, "secondary dimensions") rather than on addressing the four main ones will not enhance the core concerns of sustainable transport.

Relevance for Biofuels

The first dimension creates two problems for biofuels. First, some biofuel varieties do not provide a net energy gain—more energy is required to produce the fuels than they provide [1]. Second, many types of biofuels depend on the use of fossil fuels for harvest, production and distribution [10].

The second dimension raises the question of potential and limitations. We need to remember that renewable (in time) should not be confused with limitless (in volume). Estimates of the global long-term bioenergy potential depend critically on assumptions, particularly on the availability of

agricultural land for non-food production. Whereas more optimistic assumptions lead to a theoretical potential of 200–400 EJ/year or even higher, the most pessimistic scenario relies only on the use of organic waste and residues, providing a minimum of 40 EJ/year. More realistic assessments considering environmental constraints arrive at a sustainable potential of 40–85 EJ/year by 2050 [1]. For comparison, predicted global energy demand for transport that year totals 140 EJ/year [22].

Additionally, other production factors are also involved in addition to resources, for example, labor and capital, whose limits may be exceeded sooner than those of resources. Studies have shown that labor and capital are equally important factors concerning the level of production and prices of products and services [23].

The third dimension raises no particular issues in terms of biofuels; numerous studies have shown that biofuels work well in public transport. The fourth dimension generally favors biofuels, provided they are not dependent on fossil fuels for their production.

4. Main Sustainable Transport Strategies

A review of the literature reveals three main sustainable transport strategies: efficiency, alteration and reduction [19,24] (level 3 on Figure 1). These three strategies, under different names, represent established knowledge within the sustainable transport (and sustainable development) literature [25], for example, the IPAT equation [26,27], the ASIF equation [28], the ISA model [29]; the SMART model [19]; social, technical, and infrastructural emission drivers [30]; and the STPM index [31].

The efficiency strategy for achieving sustainable transport suggests that the environmental problems caused by transport can be reduced and that the lack of accessibility for low-mobility groups can be relieved by developing more efficient technology. The concept of "technology" is here used in a broad sense; it includes the use of both "hard technology" (e.g., developing more efficient vehicle technology and fuels) and "soft technology" (e.g., developing more efficient transport logistics). Moreover, technology that is more efficient could be implemented across all parts of the transport system: motorized transport, transport infrastructure and the energy system.

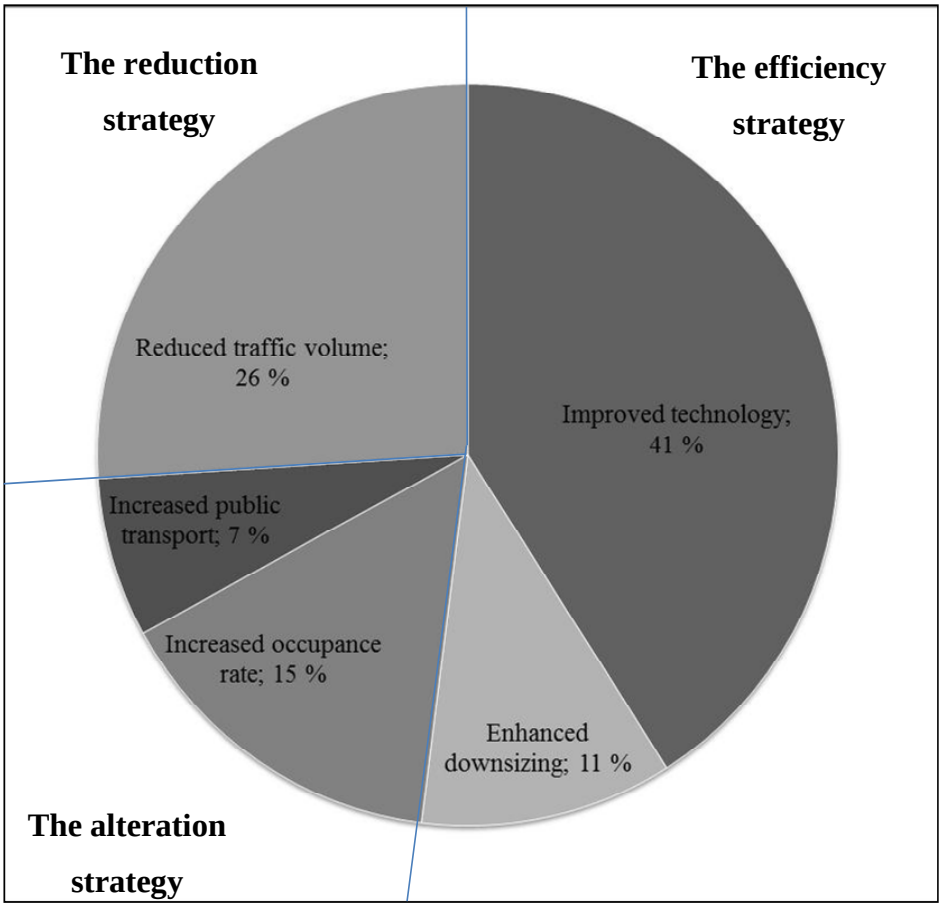
The alteration approach recognizes the urgent need to fundamentally change present transport patterns. Accordingly, the prevailingly transport pattern, dominated by cars and planes, must be changed into one based on collective forms of transport, primarily affordable well-functioning public transport systems [32]. Such systems would lead to increased use of buses, trains and trams, which are all more energy efficient than cars and planes, and therefore reduce the use of cars and planes. Moreover, an affordable well-functioning public transport system would increase accessibility for low-mobility groups.

The reduction approach for achieving sustainable mobility does not question the importance of improved efficiency and increased alteration. Indeed, the latter two approaches would, according to the reduction approach, offer some reductions in energy consumption. However, these reductions are not large enough to meet sustainable transport's energy goal. Moreover, continuous transport growth negates any reductions in energy consumption achieved by implementing new technology and altering transport patterns. Thus, the present transport volume must be decreased—except for those whose basic transport needs are not met—or at least transport growth trends must be changed.

Relevance for Biofuels

Biofuels belong to the efficiency strategy, but it is interesting to know the role they will play as part of the larger picture of looking at all three strategies. A study by the OECD [33] offers some insight on this matter. In their *Environmentally Sustainable Transport* (EST) project, they constructed three sustainable transport scenarios to illustrate the respective importance of the three strategies. The results from what they regarded the most realistic scenario, EST 3, are illustrated in Figure 2.

Figure 2. The relative importance of each sustainable transport strategy in achieving sustainable transport [33]. (The figure shows results from OECD's EST 3 scenario.)



Note: The first two phases of the four-phase EST project established a definition of EST and selected criteria for its attainment (phase 1), and constructed EST scenarios (phase 2). Six criteria were selected: CO2, NOX, VOXs, particulates, noise and land use. Three scenarios were developed for 2030 (that differentiated from the business-as-usual (BAU) scenario): EST1, EST2 and EST3. Each scenario used different assumptions regarding technological progress and transport activity level. 30. Although all three EST scenarios met the EST criteria, the first two scenarios appeared to be too extreme. The EST1 scenario seemed to involve unacceptable economic costs, and the EST2 scenario seemed to involve unacceptable social costs. Accordingly, the research team decided that further work should be confined to assessing how the EST3 scenario might be attained, and to comparing the EST3 and the BAU scenario. It is important to note that the EST3 scenario differs from the BAU scenario in two ways: It implies greater technological progress and less transport activity than the BAU scenario.

An obvious conclusion that can be drawn from the OECD study is that improved technology contributes less than half of what is required to achieve sustainable transport. Thus, the OECD claims that improved technology is a necessary, but insufficient, strategy for achieving sustainable transport.

This claim is supported by Sager *et al.* [30]. In their 2050 light-duty vehicle scenarios, they forecast that meeting GHG emissions targets (an important part of safeguarding long-term ecological sustainability) through technological improvements (*i.e.*, the efficiency strategy) alone would require universal deployment of one or more of the following clusters: electric vehicles running on nearly zero-carbon electricity, cellulosic biofuel-powered vehicles achieving 0.78 L per 100 km, or gasoline-fuelled vehicles achieving in excess of 0.24 L per 100 km. The researchers argue that these performance levels exceed even the most optimistic technology scenarios for the year 2050. Thus, they claim that reducing GHG emissions is also a behavioral issue (*i.e.*, alteration and reduction), not only a technological (efficiency) one.

5. Technology Strategies

The efficiency strategy can further be divided into two main sub-strategies: (1) the use of new, conventional technology and (2) the use of alternative technology [19] (level 4 on Figure 1). The strategy of using new but conventional technology seeks *incremental* improvements in *existing* transport technology, such as advanced ICE with direct injection, energy-efficient hybrid-drive systems, improved catalytic converters, reduced vehicle weight using lightweight materials, advanced motor management, improved aerodynamics, reduced rolling resistance and improved low-sulfur diesel fuel. The alternative strategy seeks to implement *fundamentally new* transport technology, such as introducing new fuels (biofuels and hydrogen) and drive systems (fuel cells).

The current liquid transport-fuel market is dominated by conventional fuels and technology. Taking into consideration the expected future increase in demand from all transport sectors, and the current and foreseen dominance of the ICE, the demand for liquid transport fuels can be expected to persist; therefore, biofuels offer an appealing means of achieving sustainable transport in the near future [34,35].

Relevance for Biofuels

Government policies in various countries have led to a five-fold increase in global biofuel production from 2000 to 2008. Consequently, biofuels, whether pure or blended, accounted for 2% of global road transport-fuel demand in 2008. The production of ethanol and biodiesel increased by 10% and 9%, respectively, in 2009, to 90 billion liters. Biofuels contributed nearly 3% (3 EJ) of global road transport-fuel demand in 2009 [10].

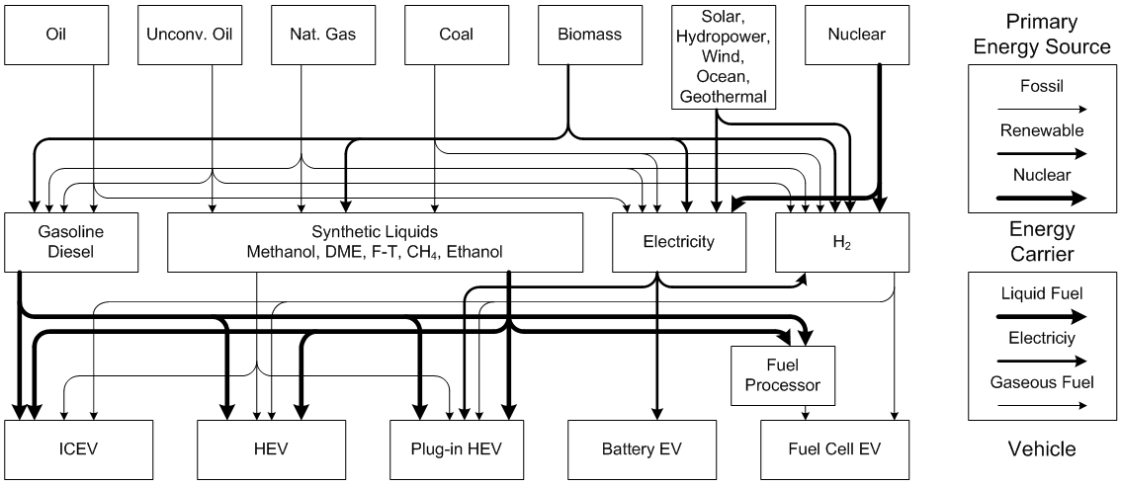
The projected biofuel share needed by 2030 and 2050 have been estimated to be close to 10% (12 EJ per year) and 15% (20 EJ per year), respectively [10,36].

6. Alternative Fuel Strategy

There are a number of possible pathways from the conversion of a primary energy source to an energy carrier (fuel) that can be used to power a vehicle. Possible pathways are shown in Figure 3,

where alternative technologies and fuels include all but those that rely on conventional, oil-based gasoline and diesel. Note that alternative fuels include many varieties based on fossil energy sources, for example, coal-based methanol and electricity (level 5 in Figure 1). Thus, fuels based on RE sources constitute a particular sub-group of the broad category of alternative fuels.

Figure 3. Possible pathways from primary energy sources to vehicular use [10].



Notes: F-T = Fischer-Tropsch process. DME = Dimethyl ether. "Unconventional oil" refers to oil sands, oil shale, and other heavy crudes. ICEV = internal combustion engine vehicles, HEV = hybrid electric vehicles, and EV = electric vehicles.

The IEA (2009) has recently given a good deal of consideration to the impact of alternative fuels and their corresponding compliance with sustainable development. In their Alternative Motor Fuels Platform, the IEA supports extending sustainability criteria beyond environmental indicators and gives the consensual criteria by which any alternative fuel use should be evaluated [37]: (1) it should provide a net energy gain, that is, not use more energy to produce the fuel than it provides; (2) it should provide environmental benefits; (3) it should be economically competitive; and (4) it should be producible in sufficient quantities without impacting the availability or price of the same feedstock when used as food. Preferably, the use of the feedstock for fuel would not compete with its use for food.

Relevance for Biofuels

All these criteria are highly relevant for biofuels. We would argue, however, that demanding that biofuels be "economically competitive" constitutes a necessary means rather than a goal in itself.

7. Biofuel Strategy

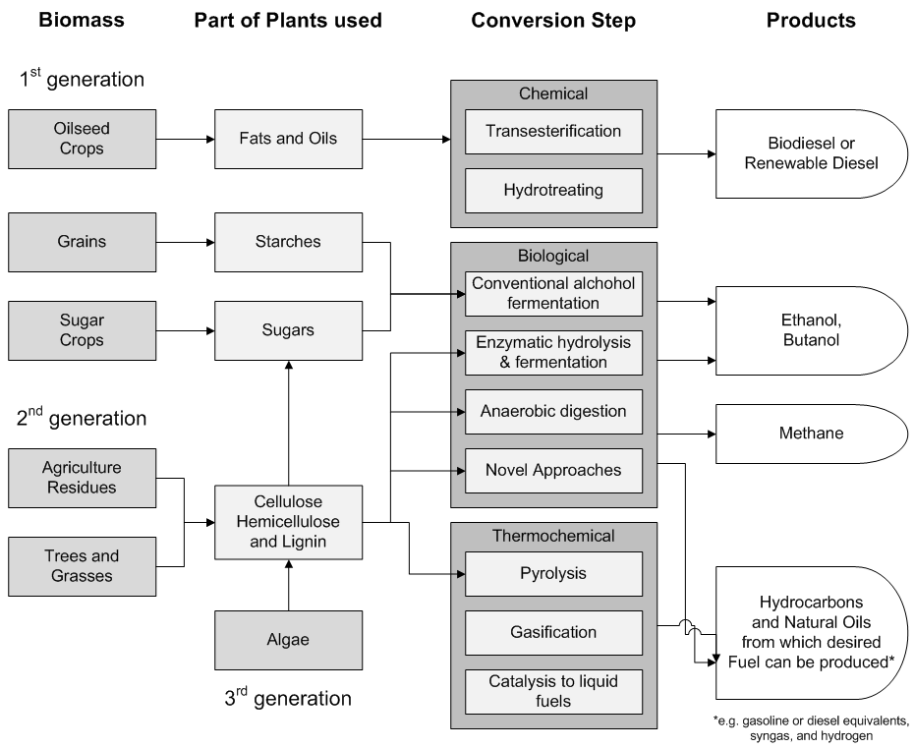
Applying the general precept of sustainable development to the more specific case of biofuels for transport (level 6 in Figure 1) has given rise to a new set of challenges. First, a number of different generations of biofuels must be evaluated. Second, there are a large number of impact assessment frameworks, standards, sustainability criteria, and certifications by which competing generations of biofuels can be evaluated. We discuss both of these challenges briefly here.

7.1. From First- to Fourth-Generation Biofuels

Biofuels for transport are commonly labeled as first-, second- or third-generation biofuels, respectively [1]. Second- and third-generation biofuels are also sometimes labeled "advanced," "next-generation," or "modern" biofuels [10]. This differentiation in generations is not always straightforward, however, because of overlaps in terms of feedstock and processing technologies, as well as uncertainties about environmental impacts.

Many definitions of first to third generations are presented in the literature; UNEP suggests the following definitions [1] (Figure 4).

Figure 4. Current and emerging biofuel pathways [1].



Note: Fourth-generation biofuels are not shown in the figure. 1st generation would also include waste oil (not shown in figure). 2nd generation "Trees and Grasses" include both residues and dedicated crops.

First-generation biofuels are commercially produced using conventional technology. The basic feedstocks are seeds, grains or whole plants from crops such as corn, sugar cane, rapeseed, wheat, sunflower seeds or oil palm. These plants were originally used as food or fodder, and most are still primarily used to feed people. The most common first-generation biofuel is bioethanol, followed by biodiesel, vegetable oil and biogas.

Second-generation biofuels can be produced from a variety of non-food sources. These include waste biomass, the stalks of wheat, corn stover, wood and special energy or biomass crops. Second-generation biofuels use biomass-to-liquid technology through thermochemical conversion (mainly to produce biodiesel) or fermentation (e.g., to produce cellulosic ethanol). Many second-generation biofuels are under development, including biohydrogen, biomethanol, dimethylfuran, bio-dimethyl ether, Fischer-Tropsch diesel, biohydrogen diesel and mixed alcohols.

Third-generation biofuels, also called oilgae, are produced from algae. The algae are feedstock derived from aquatic cultivation for the production of triglycerides (from algal oil) to produce biodiesel. The processing technology is basically the same as that used for biodiesel from second-generation feedstock. Other third-generation biofuels include alcohols such as bio-propanol or bio-butanol, which because of the current lack of production experience, are usually not considered to be relevant as fuels on the market before 2050 [1], although increased investment could accelerate their development. The same feedstock as that used for first-generation ethanol can be used, but its use requires more sophisticated technology.

Although not shown on Figure 4, some have expanded this field to include fourth-generation biofuels [38–40] through the use of genetically modified feedstocks or the use of targeted synthetic microbes to produce synthetic or carbon-negative biofuels. This type of genetic modification has, however, raised serious concerns about the potential environmental impacts of such plants, including gene flow from non-native to native plant relatives [41–43].

Each succeeding generation of biofuel has been developed to reduce the disadvantages and improve on the advantages of the proceeding generation, although as can be expected, each new generation has given rise to new challenges. This complex situation has been noted in the EU relative to the use of second-generation biofuels [44]. Here, the advantages of improved GHG balance, cost competitiveness, fuel quality, land use and food production are stated relative to those of first-generation biofuels in the same characteristics. The same summary states that the challenges confronting second-generation biofuels are primarily related to technology and infrastructure. A European roadmap for biofuels [45] presents a similar mixture of advantages and challenges for first- and second-generation biofuels, and additional studies have presented similar discussions concerning third-generation biofuels [46].

7.2. Sustainability Frameworks, Standards, Criteria and Certification

Governments are stressing the importance of avoiding unacceptable negative effects of bioenergy as they implement regulating instruments. For example, the RED [47] provides mandatory sustainability requirements for liquid transport fuels. Also, in the United States, the Renewable Fuel Standard (included in the 2007 Energy Independence and Security Act [48]) mandates minimum GHG emission reductions from the use of renewable fuels, discourages the use of food and fodder crops as feedstock, permits the use of cultivated land and estimates the effects of land-use change [49] to set thresholds of GHG emission reductions for different categories of fuels [10]. The California Low Carbon Fuel Standard set an absolute carbon intensity reduction standard and periodic evaluation of new information, for example, on indirect land-use impacts. Other examples of this type of regulatory instrument are the UK Renewable Transport Fuel Obligation, the German Biofuel Sustainability Ordinance and the Netherland NT8080 (also known as the Cramer Report).

The development of impact assessment frameworks and sustainability criteria involves significant challenges in relation to methodology, process development and harmonization. As of a 2010 review, nearly 70 ongoing efforts existed to safeguard the sustainability of agriculture and forestry products, including those used as feedstock for the production of bioenergy and biofuels [50]. However, the majority of efforts focus on environmental impacts, which is problematic because of the conflicts that arise between socioeconomic and environmental impacts, especially in developing countries [10,11].

This is going to change though. A review shows that there are (at least) twelve standards and systems currently under consideration worldwide [11]: Rainforest Alliance: Sustainable Agriculture Network; Program for Endorsement of Forest Certification; Social Accountability International; Roundtable on Responsible Soy; Roundtable on Sustainable Palm Oil; The Forest Stewardship Council; Roundtable on Sustainable Biofuels; Better Sugar Initiative; Argentinian Association of Producers for No Tillage; The International Social and Environmental Accreditation and Labeling Alliance; Fair Trade Organization; The Global Bioenergy Energy Partnership; and International Sustainability and Carbon Certification.

The increase of standards that has taken place over the past four years and that continues to advance, shows that certification has the potential to influence local impacts related to the environmental and social effects of direct bioenergy production. Many involved entities conclude that to create an efficient certification system there must be further harmonization, greater availability of reliable data and increased linking of indicators at the micro (e.g., product), meso (e.g., household or municipality) and macro (e.g., economy of states) levels [10].

Thus, sustainability criteria and biomass and biofuels certification have been developed in increasing numbers in recent years as part of voluntary or mandatory systems; interestingly, such criteria do not yet apply to conventional fossil fuels.

8. Theoretical Perspectives and Methodology

Performing scientifically sound and fair comparisons at each of the six levels demands a reliable theoretical perspective and a well-established methodological basis (bottom of Figure 1). We argue that industrial ecology theory and LCA methodology provide such a basis. Industrial ecology acknowledges the complexity and trade-offs involved in comparisons at each level and, moreover, provides input to policies surrounding biofuels in a holistic way. LCA gives quantitative weight to the comparisons. True, LCA can and should be supplemented by other methodologies and environmental management tools in line with industrial ecology, such as social impact assessment, strategic environmental assessment and sustainability assessment. However, this article focuses on LCA.

8.1. Transfer Effects and Industrial Ecology

Particular problems, referred to as “transfer effects,” occur when assessing the sustainability of biofuels (and of alternative fuels generally). Such effects have two forms [51].

Geographic transfer effect: The use of alternative fuels merely transfers energy consumption and emissions geographically (*i.e.*, from the vehicle to the production site and the distribution process); it does not reduce total energy consumption or emissions [1,10,19].

Thematic transfer effect: There are always trade-offs involved in using alternative fuels because their use merely changes environmental impacts thematically rather than reducing the total overall environmental impacts [1,19].

Transfer effects can be positive or negative. Geographical transfer can be justified as shifting emissions from urban to less-populated areas and thereby reducing exposure and the associated impacts on human health. Despite increases in total emissions, some bioethanol blends used in flex-fuel vehicles in Brazil contributed to reductions of as much as 30% in urban emissions because

most emissions originated from farming equipment, fertilizer manufacture and ethanol plants located in rural areas [52]. Thematic transfer can be justified by reducing one particularly important negative impact while at the same time accepting an increase in another less important negative impact.

The occurrence and implications of transfer effects are only evident when one adopts a holistic approach to evaluation, as promoted in the field of industrial ecology. Industrial ecology tries to address the issue of sustainable development within the context of the interrelations between the environment, the economic sphere and the techno sphere. This approach draws on an analogy with natural ecosystems, in which a web of connection exists through which individual organisms live and consume each other and each other's waste [53]. Similarly, industrial ecology can be considered the study of a pattern, or web, of relationships between various industrial activities, their products and the environment with which they interact [54].

One of the important aspects addressed by industrial ecology is that the whole life cycle, including waste and disposal, must be considered in the design process. This cradle-to-grave production philosophy is ideally represented by the unique case in which there is no grave, that is to say, by industrial processes that are environmentally benign during their whole life cycle, including manufacture, use and disposal [54].

8.2. Life Cycle Assessment

LCA provides a well-established and comprehensive methodology to compare RE (including biofuels) technologies with fossil-based and nuclear energy technologies [10]. The methodology has been evolving since the late 1960s [55] and is now supported by international initiatives [56,57] and governed by standards by the ISO [58]. Importantly, though, the ISO never aimed to standardize LCA methods, and there is no single method for conducting LCA [58].

Policies for climate change mitigation are increasingly being informed by the results of GHG-LCAs of alternative energy carriers and delivery pathways. GHG-LCAs are of particular relevance to the biofuel sector because, within the EU, incentives (in the form of obligation certificates) are expected to be related to the life cycle GHG savings relative to conventional fossil fuels [59].

The current use of GHG-LCA for biofuel carbon reporting tends not to distinguish between two different LCA approaches: ALCA and CLCA [10,59–64]. ALCA provides information about the impacts of the processes used to produce (and consume and dispose of) a product, but does not consider indirect effects arising from changes in the output of a product. ALCA generally provides information on the average unit of product and is useful for consumption-based carbon accounting. It informs comparisons between the direct impacts of products and is used to identify opportunities for reducing direct impacts in different parts of the life cycle. CLCA provides information about the consequences of changes in the level of output (and consumption and disposal) of a product, including effects both inside and outside the life cycle of the product. CLCA models the causal relationships originating from the decision to change the output of the product, and therefore seeks to inform policymakers on the broader impacts of policies, which are intended to change levels of production.

Whereas ALCAs are generally based on stoichiometric relationships between inputs and outputs, and the results may be produced with known levels of accuracy and precision, CLCAs are highly dependent upon economic models representing relationships between demand for inputs, price

elasticities, supply and markets effects of co-products. Such models rarely provide known levels of accuracy or precision and should therefore be interpreted with caution [60].

The majority of the available literature on energy technologies is based on ALCAs [10]. A resulting key limitation is that energy system changes that might result from the decision to install additional renewable capacity are excluded.

There is, however, substantial variability in published LCA results. Such variability is due to failure to distinguish between ALCA and CLCA, changing characteristics of the background energy system (e.g., its carbon intensity), technology characteristics (e.g., design, capacity factor, variability, service lifetime and vintage), geographic location, data source type (empirical or theoretical), the potential for double counting when assessing large interconnected energy systems, differences in LCA technique (e.g., process-based LCA or input–output LCA) and key methods and assumptions (e.g., co-product allocation, avoided emissions, study scope, *etc.*) [10]. Moreover, there is a need to take into account more types of externalities (economic and social impacts) and more mechanisms (rebound effects, human behavior, price effects, market dynamics, *etc.*) to meet the shortcomings of existing LCA studies in the field of, for example, biofuels [55].

Inconsistencies when dealing with system boundaries pose a particular problem with LCA; decisions on including or excluding processes in an analysis are typically not made on a scientific basis [65]. Typically speaking; life cycle inventories (one particular stage in LCA) based on process flow diagrams have an appropriate level of detail and aggregation in the preliminary foreground tiers; but because of the exponential nature of the preceding production flows; this level of detail and certainty decreases further upstream. Therefore; the hybridization of LCA with an input–output macro-economic model (LCA-IO) has received increased interest in the field of industrial ecology as a means of clearly solving the inconsistencies described above.

The application of input–output model in the environmental assessment of products and services was first raised by Wassily Leontief [66] and was further developed and supported by Duchin [67], among others. These initial formulations have since evolved, with the aim of integrating challenges resulting from international trade, so-called MRIO models [68], and from the evaluation of waste management scenarios [69].

One of the more recent iterations formulated by Duchin is WTM. This model is suitable for analyzing scenarios about actions that could be taken to achieve the environmental and social objectives associated with sustainable development. Duchin has additionally proposed that the WTM is suitable for evaluating the adoption of biomass-based fuels. With this said, there are some challenges and potential pitfalls associated with this methodology choice, particularly those associated with the highly aggregated nature of IO tables. The proposed LCA-IO promises to combine the detail of the LCA foreground model with the depth of the background IO.

Another extension to or variation of the LCA is WIO, which attempts to take into account the interdependence in the dynamic flow of products and waste [69], and can be implemented to ascertain the environmental impact of various forms of waste and waste treatment on the whole life cycle of a process, an option lacking in previous iterations of IO-type analysis that focus only on products. This methodological development reflects societies' emerging awareness and follows the holistic ideological thread of industrial ecology.

Starting in 2010, we entered the "decade of the LCSA" [55,70–73]. LCSA broadens the scope of current LCA from mainly environmental impacts to cover more dimensions of sustainability. It also broadens the scope from predominantly product-related (micro level) questions to questions related to sector (meso level) or even economy-wide (macro level) levels. In addition, it deepens current LCA to include more than just technological relations, for example, physical relations (including limitations in available resources and land), economic and behavioral relations, and other factors. LCA-IO, MRIO, WTM and WIO all provide important input to the development of LCSA.

We agree that LCSA could be an important tool in assessing the sustainability of biofuels. We also argue that the theory and development of LCSA are currently immature. Further development of LCSA should pay particular attention to two points. First, the "dimensions of sustainability" referred to by Guinée *et al.* [55] must be derived from the main dimensions of sustainable development and sustainable transport as described above. Second, broadening and deepening the scope of analysis may well create a model that is far too complex to use, understand and interpret.

9. Conclusion

Increased use of biofuels is high on the sustainable transport policy agenda. However, rather than take this relation for granted, policies must carefully consider biofuels in terms of their compliance with key characteristics and criteria at six levels. (1) The use of biofuels must comply with the four main dimensions of sustainable development. (2) The use of biofuels must comply with the four main dimensions of sustainable transport. (3) A biofuels strategy must compare favorably with other sustainable transport strategies, such as changing transport patterns and reducing transport volume. (4) Gains from a biofuels strategy must compare favorably to gains from improving fossil-fuel-based advanced ICE vehicles. (5) Benefits from the use of bio-fuelled vehicles must compare favorably to those from the use of other alternative-fuelled vehicles. (6) Benefits from the use of first-generation biofuels must compare favorably to those from the use of next-generation biofuels. In addition, developing robust and scientific sustainability criteria demands a reliable theoretical perspective and a well-established methodological base. Industrial ecology theory and life cycle sustainability assessment methodology, respectively, should prove very useful in that regard.

Achieving sustainable transport will most probably require a full portfolio of strategies. No single strategy such as improving public transport, reducing traffic volumes, or increasing the use of biofuels, plug-in hybrids and long-range-battery electric vehicles will achieve it. Thus, we agree with Sandy Thomas [74], who stated "The triple threats of global warming, energy security and urban air pollution are too great to rely on any one transportation option [strategy or fuels] for the foreseeable future."

Acknowledgments

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Conflict of Interest

The authors declare no conflict of interest.

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Article II

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Biodiesel's and advanced exhaust aftertreatment's combined effect on global warming and air pollution in EU road-freight transport

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ABSTRACT

EU directives promoting the measures of biofuels and advanced exhaust aftertreatment systems aim to mitigate global warming and air pollution, respectively; however, in addition to these claimed benefits, what trade-off effects arise from combining these measures? Based on new-vehicle emissions data for EU road-freight transport combining RME biodiesel, selective catalytic reduction (SCR), and diesel particulate filter (DPF), we determine well-to-wheel (WTW), tank-to-wheel (TTW), and well-to-tank (WTT) greenhouse gas (GHG) emissions as well as the regulated emissions of NO_x, PM, CO, and NMHC.

In comparing results, we draw three conclusions: First, vehicles fuelled by RME biodiesel have reduced WTW GHG emissions and NMHC emissions, but have slight increases in WTW emissions of NO_x, PM, and CO. Second, vehicles fitted with SCR and DPF have reduced WTW emissions of NO_x, PM, CO, and NMHC, but have slight increases in WTW GHG emissions. Third, we conclude that although the measures are independently successful at achieving their specific goals, their combination decreases some of these benefits. Only through improved testing, assessment methods, and transparency can we fully understand the interrelation of mitigation measures, and apply this understanding to policy aimed at reducing the overall environmental and social impacts of transport.

1. Introduction

European Union (EU) statistics report a total of 3831 billion (UK billion) tonne kilometres (tkm) of freight transport in the EU27 for 2010 (ECO, 2012). One tkm is equivalent to 1 tonne of freight transported 1 km, a standard measurement for freight turnover. Road-freight represents 45.8% of all freight transport, including road, rail, inland waterways, pipelines, and sea, and increased approximately 5.3% between 2009 and 2010 (ECO, 2012). The freight transport sector accounted for approximately 5% of EU27 GVA[1] and approximately 5% of EU27 employment in 2009 (ECO, 2012).

Concerning the environment, overall road transport accounted for 19%[2] of EU27 greenhouse gas (GHG) emissions (EEA, 2012). Trends indicate a future increase of GHG emissions from road transport due largely to society's preference for road-freight transport over less GHG-intensive freight transport forms (e.g., rail). Furthermore, anthropogenic GHG emissions are widely known to affect the atmosphere's radiative forcing, and to ultimately contribute to global warming (IPCC, 2011).

In addition to GHG emissions, the road transport sector emits great amounts of mono-nitrogen oxides (NO_x), particulate matter (PM), carbon monoxide (CO), and hydrocarbons (HC). According to the European Environment Agency (EEA) (EEA, 2013), in 2011, road transport accounted for 40% of NO_x emissions, 14% of PM_{10},[3] and

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[1] Refers to gross value added at basic prices.

[2] Road transport represented 94% of domestic transport GHG emissions, which in turn represented 19.7% of EU27 GHG emissions in 2009 (EEA, 2012).

[3] Particulate matter with a diameter greater than 10 μm.

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17% of $PM_{2.5}$[4] emissions, and 26% of CO emissions in the EU27. Mitigating these emissions through improvements in fuel quality, engine technology, and exhaust aftertreatment has been successful, with net reductions of approximately 50% for NO_x, 30% for PM_{10}, 35% for $PM_{2.5}$, and 83% for CO being achieved in the period 1990–2011[5] in the EU27 (EEA, 2013). Reporting statistics for emissions of HC is more complex. EURO emission standards for diesel engines target reductions in total HC emissions, including methane (CH_4) and non-methane hydrocarbons (NMHC), both volatile organic compounds (VOC) (IPCC, 2007). EU emissions reporting practices group CH_4 emissions with GHG emissions (Eurostat, 2012a), and group NMHC, or NMVOC, with long-range transboundary air pollution (EEA, 2013), hence the following division. Road transport accounted for 14% of EU 27 NMVOC emissions in 2011, with reductions of 83% realized 1990–2011 (EEA, 2013). Road transport is a marginal source of EU27 CH_4 emissions, contributing less than 5% of total CH_4 emissions,[6] and experiencing reductions of 68% in the period between 1990 and 2008 (Eurostat, 2012a). Despite these improvements, NO_x, PM, CO, and HC emissions are still considered serious contributors to local air pollution and other environmental problems across Europe, particularly considering the expected future increase in transport (Sessa and Enei, 2009), which will offset additional improvements.

In response to these concerns about the environmental effects associated with current and foreseen transport levels, the EU has independently introduced two directives. The first promotes the use of biofuels and other renewable fuels for transport (EC, 2009) as a mitigation measure for reducing GHG emissions. The second regulates specific pollutants from compression-ignition engines used in vehicles (EC, 2005a) as a medium-term mitigation measure for air pollutants.

In the first directive, the EU set a target of 10% renewable energy in the transport sector by 2020. Subsequently, EU biofuel consumption, particularly biodiesel, has increased over the past decade, and was around 14 million tonnes oil equivalent across the EU27 in 2011 (Eurobserver, 2013). The most common form of biofuel is a blend of rape methyl ester (RME) and fossil diesel.

In the second directive, the EU has introduced successively more stringent limits for the diesel-engine exhaust emissions of NO_x, PM, CO, and HC. The EU's efforts to reduce these emissions seem to be achieving some effect, primarily through the integration of advanced exhaust aftertreatment systems such as selective catalytic reduction (SCR) and diesel particulate filter (DPF).

Studies that investigate independently the environmental effects of RME biodiesel and advanced exhaust aftertreatment are numerous. Less common are studies investigating the specific environmental effects of global warming *or* local air pollution arising from combining the mitigation measures of RME biodiesel and advanced exhaust aftertreatment. Two examples are Sanchez et al. (2012), who investigate these measures' combined effect on GHG emissions, and Soltic et al. (2009), who conversely investigate these measures' effect on only tailpipe emissions of NO_x, PM, CO, and HC. However, there is a need for studies that evaluate GHG and NO_x, PM, CO, and HC emissions resulting from combining REM biodiesel and advanced exhaust aftertreatment, and that shed light on any trade-off effects.

Within this article's context, we define a trade-off effect as a specific ancillary effect occurring when one mitigation measure undermines the targeted environmental benefit of another mitigation measure.

This article evaluates emissions from road-freight transport that uses vehicles fuelled by a variety of diesels and equipped with various exhaust aftertreatment systems. Fuels considered are ultra-low-sulfur diesel (B0), blended bio-fossil diesel (B30), and RME biodiesel (B100). Aftertreatment systems considered are original equipment manufacturer (OEM), SCR, and DPF. Emissions considered are well-to-wheel (WTW), tank-to-wheel (TTW), and well-to-tank (WTT) emissions of GHGs, NO_x, PM, CO, and NMHC.

Based on these results we answer the following research questions:

1. How do the different fuels (B0, B30, and B100) and aftertreatment systems (OEM, SCR, and DPF) affect life-cycle GHG emissions from road-freight transport?
2. How do the different fuels (B0, B30, and B100) and aftertreatment systems (OEM, SCR, and DPF) affect life-cycle emissions of NO_x, PM, CO, and NMHC from road-freight transport?
3. What are the trade-off effects regarding fuels' and aftertreatment systems' respective abilities to achieve reductions in both GHG emissions and emissions of NO_x, PM, CO, and NMHC?

2. Methodology

2.1. Life-cycle assessment

We use life-cycle assessment (LCA) methods (ISO, 2006) to determine the life-cycle global warming potentials (GWPs) of two independent EU Directive measures when implemented simultaneously, and compare life-cycle GWPs with life-cycle emissions of NO_x, PM, CO, and NMHC. LCA's application in developing or reviewing legislative measures is well known (Schenck, 2009; Schleicher, 1996).

2.2. Scope

The system boundaries adopted are TTW, also known as the vehicle cycle, and WTT, also known as the fuel cycle. These two subsystems aggregate to form the WTW life-cycle system boundaries. Fig. 1 depicts their relation.

The functional unit of comparison is tonne kilometres (tkm). GWP is the characterization factor for the environmental impact of climate change, defined here under general terms as the impact of human emissions on the atmosphere's radiative forcing over a 100-year time horizon, denoted as GWP100. In this study, this includes the radiative forcing effects of GHGs: carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) emissions, measured in kilogram carbon dioxide equivalent (kg CO_2 eq.) as described by the International Panel on Climate Change (IPCC, 2007).

The functional unit combined with the characterization factor gives the following: kg CO_2 eq./tkm. The environmental impact assessment for GWP100 is determined by the CML 2 baseline method (Goedkoop et al., 2008), with the use of Simapro v7.3.3 life-cycle assessment software (Goedkoop et al., 2010).

The same functional unit has been adopted for NO_x, PM, CO, and NMHC emissions, giving the following: g NO_x/tkm, g PM/tkm, g CO/tkm, and g NMHC/tkm, respectively. NO_x, PM, CO, and NMHC emissions have been extracted from the life-cycle emissions inventories of the life-cycle impact assessments (see Supplementary material).

[4] Particulate matter with a diameter less than 2.5 μm.

[5] For PM_{10} and $PM_{2.5}$ the reductions given are for 2000–2011.

[6] The 5% refers to all energy-related CH_4 emissions, of which road transport emissions are one element.

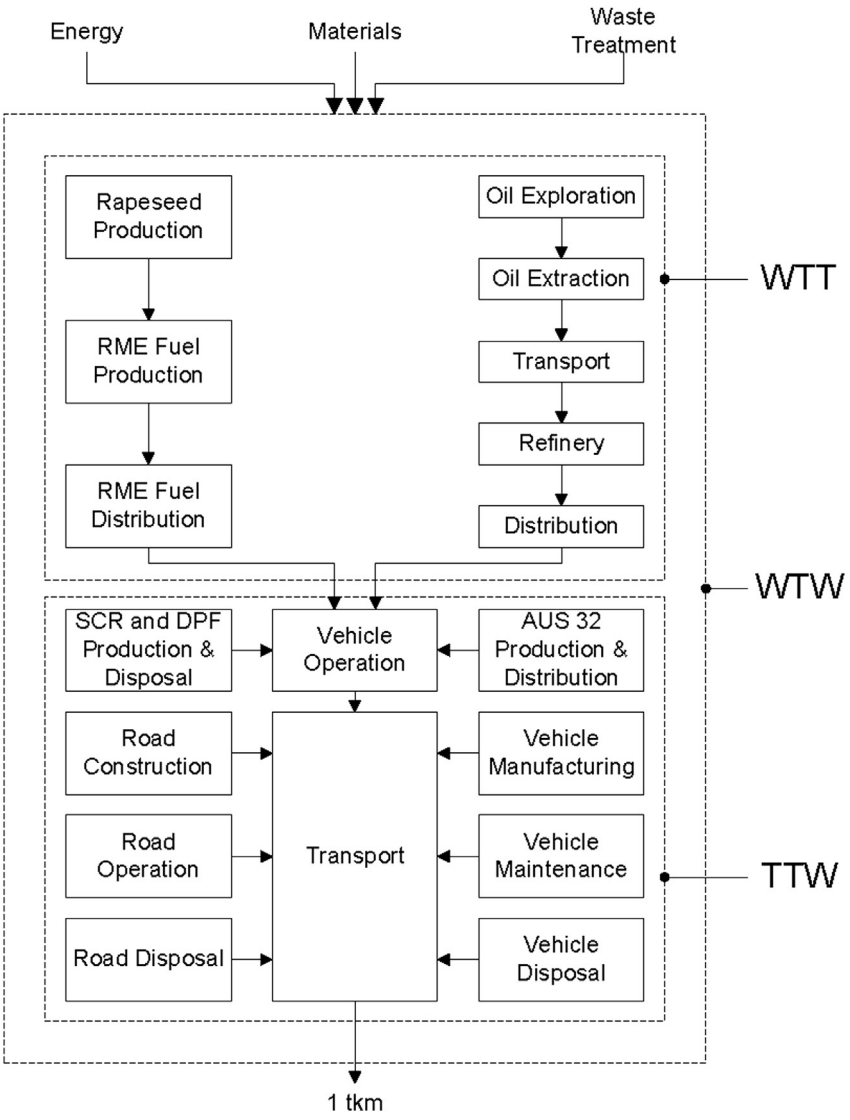


Fig. 1. System boundary for a WTW analysis of transportation, identifying WTT and TTW, adapted from Spielmann et al. (2007) (TTW), Jungbluth et al. (2007) (WTT – RME biodiesel), and Jungbluth (2007) (WTT – Diesel).

2.3. Cases

Stationary measurement and testing have been performed in conjunction with the Laboratories for IC Engines and Exhaust Emission Control of the University of Applied Sciences Biel (AFHB, 2009), on an Iveco F1C EURO 3 diesel engine with various exhaust aftertreatment systems and fuels (see Table 1). EURO 3 emission standards for heavy-duty diesel engines came into force in 2000 (EC, 1999), and were replaced in 2005 by EURO 4 emissions standards (EC, 1999; 2005a). The most recent figures from Eurostat (2012b) indicate that in 2011, vehicles regulated by EURO 3, or older, emissions standards represented 29% of all EU27 road-freight transport, measured in tkm. As such, these results are relevant when considering the retrofitting market.

Table 1
Aftertreatment systems and fuels considered in this study.

Fuel	Aftertreatment system		
	OEM	SCR	SCR & DPF
B0	X	X	X
B30	X	X	X
B100	X	X	X

OEM in this context denotes the vehicle drive train and exhaust aftertreatment system originally manufactured and distributed, without modification or alteration, that is, EURO 3 without exhaust-gas recycling (see Appendix Fig. 1). SCR is an advanced exhaust aftertreatment system that primarily aims to reduce NO_x in the exhaust stream. SCR works by a reducing agent, in this case a homogeneous solution of 32.5% v/v urea solute and water solvent (AUS32) injected into a catalyst-filled canister in the engine's exhaust stream. The NO_x emissions in the engine's exhaust stream are, with the aid of the catalyst and with the addition of AUS32, converted into diatomic nitrogen (N_2) and water (H_2O). The SCR diesel exhaust fluid AUS32 was of the type AdBlue. DPF is an advanced exhaust aftertreatment device that aims to remove diesel PM from the exhaust stream of diesel engines. The biodiesel tested is RME, the most widely used

Table 2
WTT GWP100 (kg CO_2 eq./tkm) results for exhaust aftertreatment configurations for various bio-fossil diesel blends, with SD 95% confidence interval (2σ).

Fuel	Aftertreatment system					
	OEM		SCR		SCR & DPF	
	Value	2σ	Value	2σ	Value	2σ
B0	0.077	0.061	0.076	0.061	0.079	0.063
B30	0.138	0.110	0.143	0.113	0.140	0.112
B100	0.291	0.244	0.296	0.234	0.299	0.254

Table 4
TTW GWP100 (kg CO_2 eq./tkm) results for exhaust aftertreatment configurations for various bio-fossil diesel blends, with SD 95% confidence interval (2σ).

Fuel	Aftertreatment system					
	OEM		SCR		SCR & DPF	
	Value	2σ	Value	2σ	Value	2σ
B0	0.437	0.278	0.442	0.284	0.457	0.288
B30	0.329	0.200	0.338	0.194	0.342	0.198
B100	0.096	0.065	0.100	0.062	0.104	0.062

biodiesel in the EU (Eurobserver, 2013). The Iveco test engine is commonly installed in vans, minibuses, and lightweight truck chassis.

2.4. Life-cycle inventory

Selected life-cycle inventories (LCI) are provided in the Supplementary material.

2.4.1. Well-to-tank

The WTT system includes all phases of the fuel cycle from feedstock production to filling the vehicle's tank. For this study, secondary data from the Ecoinvent v2.2 database were used for both the RME biodiesel and the low-sulfur diesel (LSD) fuel cycles, Jungbluth et al. (2007) and Jungbluth (2007), respectively. For the bio-fossil diesel blend B30, a new LCI was constructed based on a 30% v/v contribution of RME biodiesel, and the remaining 70% v/v was obtained from LCI data for LSD. The authors have adopted economic allocations of 74.3% and 86.9% to rapeseed oil and RME, respectively, consistent with the secondary data source Jungbluth et al. (2007).

2.4.2. Tank-to-wheel

The TTW system is composed of new empirical data and secondary data from the Ecoinvent v2.2 database. As described in Section 2.3, new empirical data have been collected for vehicle operation through stationary European Transient Cycle (ETC) measurement and testing (EC, 1999).

From the ETC tests, concentrations of exhaust emissions were recorded in parts per million (ppm). Concerning the functional unit (tkm), it was necessary to condition these results, first from ppm to grams per kilometre using accepted conversion formulas (EC, 2005a, b), and then to grams per tkm following ratios given by Spielmann et al. (2007).

The ETC testing procedure, in addition to exhaust emissions, provides the vehicle's direct operating inputs (e.g., diesel consumption).

LCI data for AUS32 were based on secondary LCI data for 77.5% v/v deionized water (Althaus et al., 2007), 32.5% v/v urea (Nemecek and Kaegi, 2007); distribution, transport, and blending were modelled on Jungbluth (2007), Speilmann et al. (2007), and Althaus et al. (2007), respectively (see supplementary material).

LCI data for both SCR and DPF advanced exhaust aftertreatment are scaled from secondary LCI data from Heck (2007). Scaling is based on tested SCR and DPF volumes of 7.4 l and 12.5 l, respectively, and on an estimated lifetime of 3500 h of operation at 59 km/h, or approximately 200,000 km.

For the remaining TTW life-cycle stages (vehicle- manufacture, maintenance, and disposal, and road- construction, operation, and disposal), life-cycle inventories were adopted from secondary LCI data provided by Spielmann et al. (2007) with an assumed vehicle lifetime of 540,000 km.

AUS32 consumption and PM emissions were not measured in the ETC testing; these data were obtained from a series of step load testing performed by the AFHB (2009) in conjunction with ETC testing following similar set-up and procedures. These data were conditioned for use in this study.

2.5. Data uncertainty

WTT and TTW data uncertainty has been determined with the Monte Carlo method using 1000 runs. Based on limitations for calculating the real standard deviation (SD) of the primary empirical data statistically (i.e., one set of continuous measurements per engine configuration, see Table 1), the authors have chosen the pedigree matrix method to estimate the geometric SD for both primary empirical data and secondary data from the Ecoinvent v2.2 database.

Table 3
WTT: NO_x, PM, CO, and NMHC emissions (g/tkm) for exhaust aftertreatment configurations for various bio-fossil diesel blends, with SD 95% confidence interval (2σ).

Fuels	Aftertreatment system	WTT emissions							
		NO_x		PM		CO		NMHC	
		Value	2σ	Value	2σ	Value	2σ	Value	2σ
B0	OEM	0.293	0.290	0.046	0.043	0.097	0.112	0.281	0.320
	SCR	0.280	0.254	0.045	0.042	0.095	0.107	0.274	0.309
	SCR & DPF	0.299	0.428	0.047	0.044	0.099	0.117	0.287	0.328
B30	OEM	0.458	0.390	0.074	0.063	0.144	0.158	0.237	0.262
	SCR	0.467	0.378	0.077	0.065	0.147	0.158	0.248	0.265
	SCR & DPF	0.462	0.398	0.075	0.066	0.147	0.174	0.239	0.262
B100	OEM	0.865	0.718	0.147	0.134	0.267	0.309	0.134	0.115
	SCR	0.885	0.698	0.148	0.126	0.266	0.288	0.136	0.109
	SCR & DPF	0.889	0.756	0.150	0.140	0.269	0.313	0.136	0.122

Table 5
TTW: NO_x, PM, CO, and NMHC emissions (g/tkm) for exhaust aftertreatment configurations for various bio-fossil diesel blends, with SD 95% confidence interval (2σ).

Fuels	Aftertreatment system	TTW emissions							
		NO_x		PM		CO		NMHC	
		Value	2σ	Value	2σ	Value	2σ	Value	2σ
B0	OEM	2.640	2.100	0.468	0.374	1.001	1.267	0.405	0.264
	SCR	0.943	0.662	0.442	0.361	1.031	1.277	0.302	0.260
	SCR & DPF	0.667	0.396	0.389	0.276	0.514	0.917	0.259	0.242
B30	OEM	2.840	2.380	0.495	0.422	0.930	1.244	0.360	0.244
	SCR	0.958	0.604	0.417	0.331	0.968	1.300	0.287	0.224
	SCR & DPF	0.633	0.376	0.384	0.277	0.519	0.743	0.252	0.234
B100	OEM	2.770	2.200	0.498	0.420	0.895	1.546	0.304	0.232
	SCR	0.968	0.652	0.396	0.306	0.943	1.562	0.266	0.228
	SCR & DPF	0.813	0.692	0.383	0.280	0.490	0.693	0.246	0.228

3. Results

We provide WTT and TTW values for the emission of GWP100, NO_x, PM, CO, and NMHC. We present WTW results in graphic form only, following WTW = WTT + TTW; the determination of total standard deviation is analogous. For more information, please refer to the Supplementary material.

Table 2 presents WTT results for GWP100.

Table 3 presents the WTT emissions for NOX, PM, CO, and NMHC emissions.

Table 4 presents the TTW results for GWP100.

Table 5 presents the TTW emissions for NO_x, PM, CO, and NMHC emissions.

Fig. 2 presents the WTW results for GWP100.

From Fig. 2, we observe two general trends: first, an average decrease in calculated GWP100 relative to B0 with increasing % v/v of RME biodiesel: 8.8% and 24.4% for B30 and B100, respectively; second, an average increase in calculated GWP100 relative to OEM with the addition of advanced exhaust aftertreatment: 1.9% and 3.0% for SCR, and SCR and DPF, respectively.

Figs. 3–6 present the WTW results for NO_x, PM, CO, and NMHC emissions, respectively.

From Fig. 3 we observe two trends: first, an average decrease in NO_x emissions relative to OEM with the addition of advanced exhaust aftertreatment: −54.7% and −62.8% for SCR, and SCR and DPF, respectively; second, an average increase in NO_x emissions relative to B0 with increasing % v/v of RME biodiesel: 14.1% and 50.5% for B30 and B100, respectively.

Similarly, from Fig. 4 we observe two general trends: first, an average decrease in PM emissions relative to OEM with the addition of advanced exhaust aftertreatment: −11.3% and −15.2% for SCR, and SCR and DPF, respectively; second, an average increase in PM emissions relative to B0 with increasing % v/v of RME biodiesel: 5.9% and 19.9% for B30 and B100, respectively.

From Fig. 5 we see three general trends: first, an average initial increase in CO emissions relative to OEM with the addition of only SCR: 3.5%; second, an average decrease in CO emissions relative to OEM with the addition of combined SCR and DPF: −26.2.3%; third, an average increase in CO emissions relative to B0 with increasing % v/v of RME biodiesel: 1.9% and 12.4% for B30 and B100, respectively.

From Fig. 6 we observe two general trends: first, an average decrease in NMHC emissions relative to OEM with the addition of

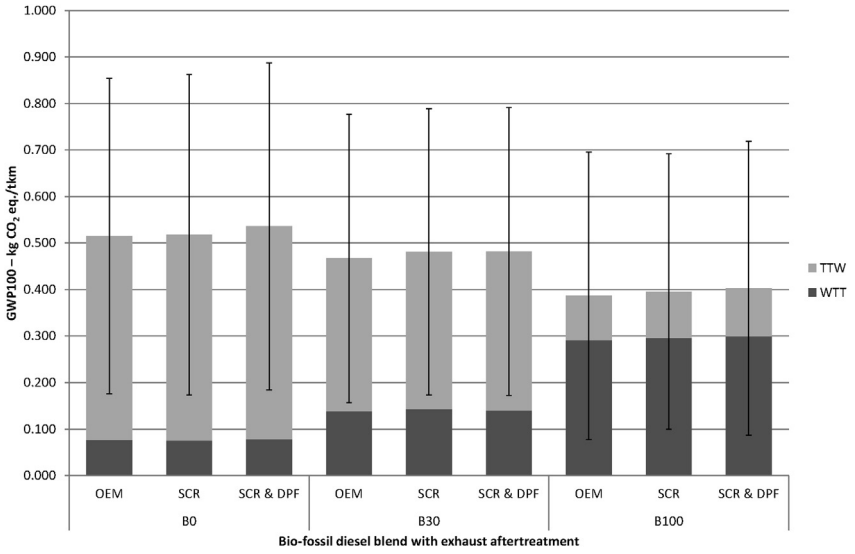


Fig. 2. WTW comparison of bio-fossil diesel blends with exhaust aftertreatment vs. GWP100, with a column division of WTW into TTW and WTT, with SD 95% confidence interval.

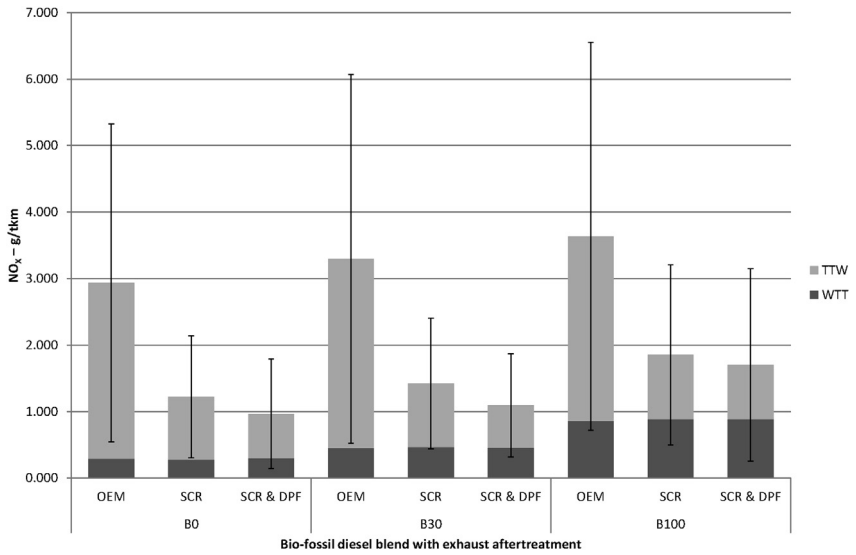


Fig. 3. WTW comparison of bio-fossil diesel blends with exhaust aftertreatment vs. NO_x measured in g/tkm, with a column division of WTW into TTW and WTT, with SD 95% confidence interval.

advanced exhaust aftertreatment: −11.5% and −16.5% for SCR, and SCR and DPF, respectively; second, a similar average decrease in NMHC emissions relative to B0 with increasing % v/v of RME biodiesel: −10.1% and −32.1% for B30 and B100, respectively. Additionally, Fig. 6 includes CH_4 emissions in order to indicate their contribution towards total HC emissions.

4. Discussion

4.1. The effect of RME biodiesel on global warming potential

The WTW results show that complete substitution of RME biodiesel (B100) for fossil diesel (B0) decreases the overall GWP100; this result is consistent with results from other reports, for example, Sanchez et al. (2012) and IEA (2011). This decrease relates to accounting practices of biogenic TTW CO_2 in our determination of GWP100, and to allocation steps applied in the WTT phase; see Section 2.4.

This decrease comes despite a strong increase in WTT GWP100 (see Table 2), an increase which has two contributors; first, an increase in fuel consumption during engine operation (AFHB, 2009) which is due to the difference in upper heating values between fossil diesels and biodiesels (Kousoulidou et al., 2010). The second contributor relates to higher GWP100 emissions associated with biodiesel production when compared to fossil-diesel production (see Jungbluth et al. (2007) and Jungbluth (2007), respectively).

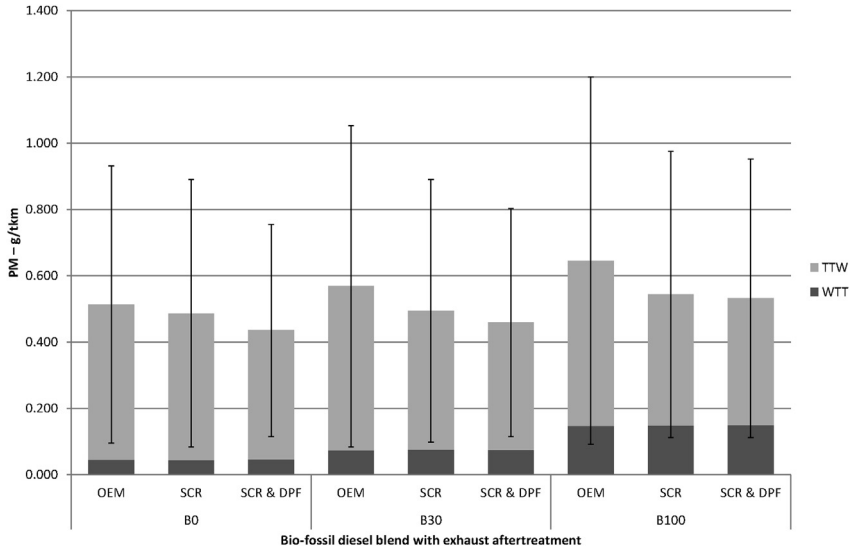


Fig. 4. WTW comparison of bio-fossil diesel blends with exhaust aftertreatment vs. PM measured in g/tkm, with a column division of WTW into TTW and WTT, with SD 95% confidence interval.

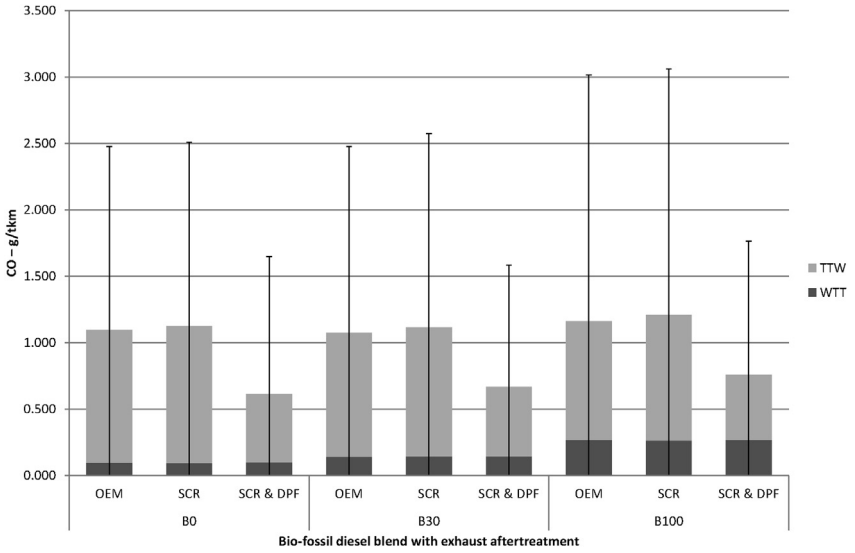


Fig. 5. WTW comparison of bio-fossil diesel blends with exhaust aftertreatment vs. CO measured in g/tkm, with a column division of WTW into TTW and WTT, with SD 95% confidence interval.

4.2. The effect of SCR and DPF aftertreatment systems on the emission of NO_x, PM, CO, and NMHC

The application of SCR exhaust aftertreatment has led to reductions in NO_x, PM, and NMHC emissions. The addition of DPF exhaust aftertreatment has led to reductions in NO_x, PM, CO, and NMHC emissions.

There is one inconsistency in the results: a slight increase in CO with the application of only SCR exhaust aftertreatment. The slight increase in measured CO could be the result of less than optimal combustion parameters (Lewander, 2011), or insufficient oxidation catalyst in the SCR device (Majewski, 2005), as is the case here at both low and high operating temperatures.

Neither these findings nor those of the preceding Section (3) are particularly surprising considering that RME biodiesel was implemented to mitigate GWP100 and that advanced exhaust aftertreatment was implemented to mitigate NO_x, PM, CO, and HC.

4.3. Trade-off effects from the combination of RME biodiesel and SCR and DPF aftertreatment systems

From the WTW results, we can observe that the addition of RME biodiesel increases the overall emission of particularly NO_x, and of PM and CO, while reducing the emission of HC. Increases in TTW NO_x emissions are a common finding in similar studies such as Rounce et al. (2012), Hajbabaei et al. (2012), and Soltic et al. (2009). The precise cause is

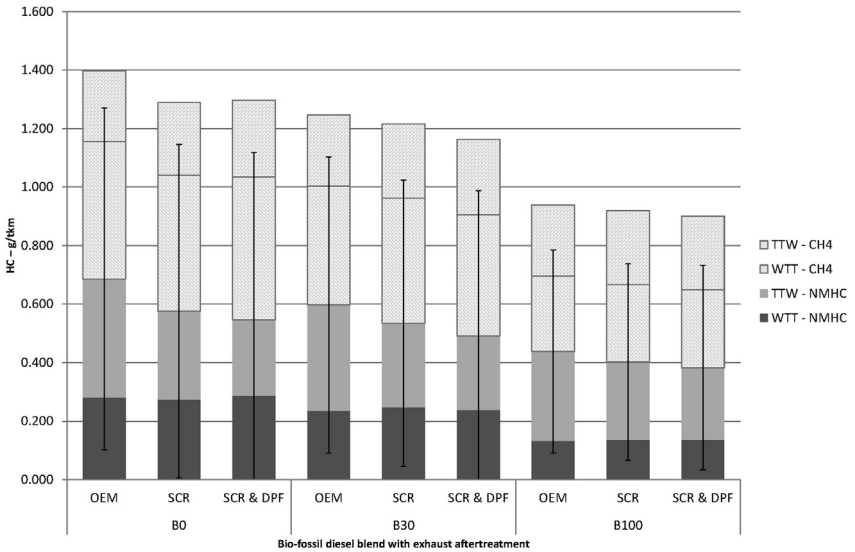


Fig. 6. WTW comparison of bio-fossil diesel blends with exhaust aftertreatment vs. HC measured in g/tkm, with a column division of WTW into TTW and WTT, with SD 95% confidence interval.

unknown, though several hypotheses exist (Hoekman and Robbins, 2012).

Conversely, the addition of advanced exhaust aftertreatment components leads to a slight increase in the GWP100 results. The main cause is increased engine exhaust backpressure with the addition of exhaust aftertreatment, and such backpressure leads to increased fuel consumption (Jaaskelainen, 2007).

Prior to discussing and concluding on these findings, it is important to acknowledge the uncertainties of the results.

4.4. Uncertainty

This discussion is divided into data uncertainties, uncertainties about the correctness of the model and methodology, and uncertainties caused by the incompleteness of the model and the methodology. Furthermore, we will discuss only uncertainties unique to this study, as opposed to general uncertainties common to all LCAs.

The method used for determining data uncertainty is explained in Section 2.5, and presented alongside the results in Section 3; generally, data uncertainty is less for GWP100 than for NO_x, PM, NMHC, and CO. Due to the scarcity of studies with similar scope and methodology, and which provide sufficient transparency, the relevance of a quantitative comparison of data uncertainty is questionable. The basic uncertainty factors provided by Frischknecht and Jungbluth (2007) indicate higher uncertainty of regulated exhaust emissions, since CO_2 emissions can be calculated from fuel input, whereas regulated exhaust emissions are dependent on, and vary, with engine operation characteristics (e.g. Huo et al., 2012). Furthermore, data uncertainty can arise through using the pedigree matrix method to estimate the SD of measured TTW values in lieu of real statistical SD, a method which would provide more realistic SD but perhaps not lessen the uncertainty (see Huo et al., 2012). Finally, the choice of reporting NO_x, PM, CO, and NMHC emissions, as opposed to determining their midpoint impacts, reduces the uncertainty that arises along the cause-effect chain (Bare et al., 2000).

Concerning uncertainties about the correctness of the model and methodology, first it is acknowledged that despite being based on sound decisions and assumptions, the majority of LCI data has been adopted or adapted from secondary databases. Logic tells us that with reductions in operation emissions of GWP100 and NO_x, PM, CO, and HC due to the addition of RME biodiesel and advanced exhaust aftertreatment, respectively, calculating the effects of these measures on their targeted emissions becomes more dependent on secondary data. Conversely, the trade-offs increase operation emissions, reducing the relative contribution of secondary data. Furthermore, recent studies have questioned the representativeness of ETC testing's ability to reflect real-world emissions. One example is Ligterink et al. (2009), who found that real-world vehicle-operation emissions of NO_x were considerably higher than ETC results indicated. The reason given is lower engine efficiency under real-world conditions, explained by Huo et al. (2012) as engine manufacturers' pre-occupation with optimizing for high fuel efficiencies in off-cycle ranges of the European Steady State Cycle. Additionally, we have applied economic allocation (see Section 2.4) to rapeseed oil and RME biodiesel based on the assumption that their co-products of rapeseed meal and glycerine, respectively, have economic value. In Section 4.1 we correlate this decision to reduced GWP100 emissions with the addition of increasing concentrations of RME biodiesel. It is thus important to consider the uncertainty of economic allocation; studying the production of RME biodiesel, Halleux et al. (2008) and Bernesson et al. (2004) have both highlighted that without economic allocation the perceived environmental benefits associated with RME biodiesel are reduced. The final aspect concerning the correctness of the methodology concerns the general choice of performing an attributional (aLCA) as opposed to consequential (cLCA) LCA. Finnveden et al. (2009), citing Curran et al. (2005), define aLCA as focused on describing the environmentally relevant physical flows to and from a life-cycle and its subsystems, and define cLCA by its aim to describe how environmentally relevant flows will change in response to possible decisions. Our choice does not affect the relevance of the results, as we are studying existing policy and measures; however, it does limit the ability of drawing conclusions from these results concerning future policy and measures.

Finally, it is important to consider the uncertainty caused by the incompleteness of the model and methodology. We have chosen to compare GWP100 and NO_x, PM, CO, and NMHC emissions resulting from the implementation and combination of their mitigation measures, RME biodiesel and exhaust aftertreatment, respectively. This limited scope of impact categories allows us to highlight the effects of and trade-offs between two mitigation measures. However, the discourse concerning biofuels contains numerous additional environmental and social impacts, most notably: toxicological (health), direct and indirect land-use, and food security (Andersen, 2013), (Tilman et al., 2009), and (Naylor et al., 2007). Only with this knowledge is it possible to form decisions concerning biofuels and their use, ultimately reducing the generation of further unexpected negative consequences and trade-offs.

This discussion of uncertainties does not diminish the relevance of our findings, but it does limit the conclusions that can be drawn from them.

5. Conclusion

In conclusion, we find that the independent implementation of RME biodiesel and advanced exhaust aftertreatment SCR and DPF in road-freight transport results in the reduction of their targeted emissions GWP100 and the life-cycle emissions of NO_x, PM, CO, and NMHC, respectively. However, when the mitigation measures are combined, trade-offs arise: increased GWP100 with the addition of SCR and DPF exhaust aftertreatment, and increased life-cycle emissions of NO_x, PM, and CO with increasing v/v % of RME biodiesel. As is the nature with trade-offs, the perceived benefits associated with each mitigation measure independently are consequently diminished. Even though these results are determined with uncertainty, they are coherent with the findings of other articles and reports.

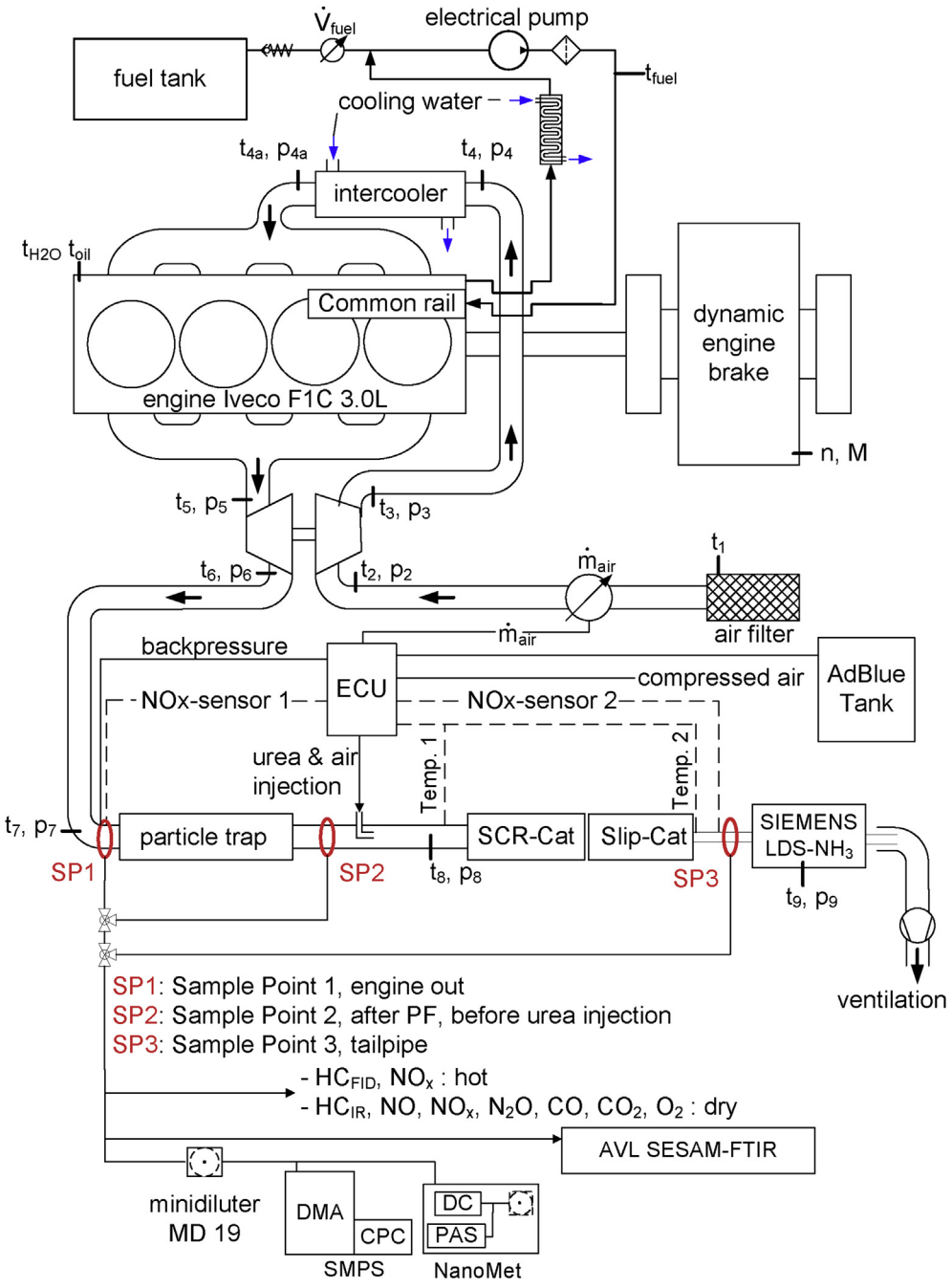
Further research will reduce uncertainty. On-road testing combined with robust life-cycle modelling and assessment methodology, covering a broader range of environmental and social impacts, will provide additional perspectives.

In the future, environmental mitigation policies should account for the environmental and social impacts associated with their proposed measures encompassing the whole life-cycle and be complemented by a sound understanding of uncertainties and trade-offs between foreseen combinations of mitigation measures. Harmonizing these processes would end the preoccupation with reducing isolated emissions, and would redirect focus to reducing the overall environmental and social burdens associated with transport.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jclepro.2014.05.011.

Appendix A. Test engine set-up



Appendix Fig. 1. Test engine set-up (AFHB, 2009).

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Supplementary material for:

Biodiesel's and advanced exhaust aftertreatment's combined effect on global warming and air pollution in EU road-freight transport

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Summary

This supplement contains the life-cycle inventories and their description, and a description of how total NO_X, PM, CO, and HC emissions have been extracted from the life-cycle emission inventories.

Life-cycle inventories

A total of 54 life-cycle inventories (LCI) have been compiled for both transport (functional unit (FU)= tkm), and operation (FU = km), covering: tank-to-wheel (TTW), well-to-tank (WTT), and well-to-wheel (WTW) life-cycle phases, and for all bio-diesel and exhaust aftertreatment configurations. The authors have chosen to provide 1 LCI for transport, as all share the same structure. Additionally, we have included all WTW operation LCIs for all bio-diesel and exhaust aftertreatment configurations.

WTT and TTW data uncertainty has been determined with the Monte Carlo method using 1000 runs. Based on limitations for calculating the real standard deviation (SD) of the primary empirical data statistically (i.e., one set of continuous measurements per engine configuration), the authors have chosen the pedigree matrix method to estimate the geometric SD for both primary empirical data and secondary data from the Ecoinvent v2.2 database. Tables 2-13 provide both inventory values and their units of measure, along

with both the pedigree matrix method determined geometric standard deviation (SD_{g95}), and the reference to the pedigree matrix criteria score, i.e. (U1,U2,U3,U4,U5,U6) which correspond to those scores described in Table 1.

Table 1 Pedigree matrix (criteria scoring) reproduced from Goedkoop et al. (2010).

Score		**1**	**2**	**3**	**4**	**5**
U1	**Reliability**	Verified data based on measurements	Verified data partly based on assumptions OR non-verified data based on measurements	Non-verified data partly based on qualified estimates	Qualified estimate (e.g. by industrial expert); data derived from theoretical information (stoichiometry, enthalpy, etc.)	Non-qualified estimate
		1.00	**1.05**	**1.10**	**1.20**	**1.5**
U2	**Completeness**	Representative data from all sites relevant for the market considered over an adequate period to even out fluctuations	Representative data from >50% of the sites relevant for the market considered over an adequate period to even out normal fluctuations	Representative data from only some sites (<<50%) relevant for the market considered OR >50% of sites but from shorter periods	Representative data from only one site relevant for the market considered OR some sites but from shorter periods	Representativeness unknown or data from a small number of sites AND from shorter periods
		1.00	**1.02**	**1.05**	**1.10**	**1.20**
U3	**Temporal correlation**	Less than 3 years of difference to our reference year (2000)	Less than 6 years of difference to our reference year (2000)	Less than 10 years of difference to our reference year (2000)	Less than 15 years of difference to our reference year (2000)	Age of data unknown or more than 15 years of difference to our reference year (2000)
		1.00	**1.03**	**1.10**	**1.20**	**1.50**
U4	**Geographical correlation**	Data from area under study	Average data from larger area in which the area under study is included	Data from a smaller area than area under study, or from similar area		Data from unknown OR distinctly different area (north America instead of middle east, OECD-Europe instead of Russia)
		1.00	**1.01**	**1.02**		**1.10**
U5	**Further technological correlation**	Data from enterprises, processes and materials under study (i.e. identical technology)		Data on related processes or materials but same technology, OR Data from processes and materials under study but from different technology	Data on related processes or materials but different technology, OR data on laboratory scale processes and same technology	Data on related processes or materials but on laboratory scale of different technology
		1.00		**1.20**	**1.50**	**2.00**
U6	**Sample size**	>100 continuous measurement, balance of purchased products	>20	>10, aggregated figure in environmental report	>=3	unknown
		1.00	**1.02**	**1.05**	**1.10**	**1.20**

The inventory presented in Table 2 has been adopted from Spielmann et al. (2007); vehicle lifetime is assumed to be 540,000 km.

Table 2. WTW LCI for transport, example given for all bio-diesel and exhaust aftertreatment configurations; same values used for all transport LCIs.

Transport, WTW, B0/B30/B100, OEM/SCR/SCR & DPF, lorry 3.5–7.5t, Euro3/tkm/RER	**1**	**tkm**	**SD_{g95}**	**criteria**
Materials/fuels				
Operation, WTW, BO/B30/B100, OEM/SCR/SCR & DPF, lorry 3.5–7.5t, Euro3/RER	1.02E+00	km	2.05	(4,1,1,1,1,na)
Lorry 16t/RER	1.89E-06	p	3.05	(4,1,1,2,1,na)
Maintenance, lorry 16t/CH	1.89E-06	p	3.05	(4,1,1,2,1,na)
Road/CH	2.89E-03	my	3.05	(4,1,1,1,1,na)
Operation, maintenance, road/CH	1.20E-03	my	3.05	(4,1,1,1,1,na)
Waste to Treatment				
Disposal, lorry 16t/CH	1.89E-06	p	3.05	(4,1,1,2,1,na)
Disposal, road/RER	2.89E-03	my	3.05	(4,1,1,1,1,na)

Concerning the inventories presented in Tables 3 to 11:

- All values are based on measurements from AFHB (2009a) unless otherwise stated,
- Values for non-exhaust emissions of PM: Particulates, <2.5 um, Particulates, >10 um, and Particulates, >2.5 um, and <10 um are adopted from Spielmann et al. (2007),
- SCR and DPF values have been scaled from Heck (2007). Scaling is based on tested SCR and DPF volumes of 7.4 l and 12.5 l, respectively, and on an estimated lifetime of 3,500 hours of operation at 59 km/h, or approximately 200,000 km.
- For information on B30 bio-fossil diesel blend, see Table 12.
- For information on urea aqueous solution, see Table 13.

Table 3. WTW LCI for the operation of a lorry 3.5–7.5t running on ultra-low sulphur diesel, without SCR or DPF exhaust aftertreatment.

Operation, WTW, B0, OEM, lorry 3.5–7.5t, Euro3/RER	**1**	**km**	**SD_{g95}**	**criteria**
Materials/fuels				
Diesel, low-sulphur, at regional storage/CH	1.25E-01	kg	1.26	(1,4,3,3,1,5)
Emissions to air				
Carbon dioxide, fossil	3.34E-01	kg	1.26	(1,4,3,3,1,5)
Carbon monoxide, fossil	4.83E-04	kg	5.08	(1,4,3,3,1,5)
Nitrogen oxides	2.30E-03	kg	1.59	(1,4,3,3,1,5)
NMVOC, non-methane volatile organic compounds, unspecified origin	1.56E-04	kg	1.59	(1,4,3,3,1,5)
Methane, fossil	4.96E-06	kg	1.59	(1,4,3,3,1,5)
Dinitrogen monoxide	1.83E-06	kg	1.59	(1,4,3,3,1,5)
Particulates, unspecified	1.13E-04	kg	2.08	(2,4,3,3,1,5)
Particulates, <2.5 um	3.50E-05	kg	3.03	(3,1,3,3,1,2)
Particulates, >10 um	5.66E-05	kg	1.54	(3,3,3,3,1,2)
Particulates, >2.5 um, and <10 um	6.23E-05	kg	2.03	(3,3,3,3,1,2)

Table 4. WTW LCI for the operation of a lorry 3.5–7.5t running on ultra-low sulphur diesel, with SCR exhaust aftertreatment.

Operation, WTW, B0, SCR, lorry 3.5–7.5t, Euro3/RER	**1**	**km**	**SD_{g95}**	**criteria**
Materials/fuels				
Diesel, low-sulphur, at regional storage/CH	1.22E-01	kg	1.26	(1,4,3,3,1,5)
Catalytic converter, SCR, 200 litre/RER	1.80E-07	p	1.26	(1,4,3,3,1,5)
Urea Aqueous Solution, 32.5% at regional storage/RER	3.40E-03	kg	1.27	(2,4,3,3,1,5)
Emissions to air				
Carbon dioxide, fossil	3.36E-01	kg	1.26	(1,4,3,3,1,5)
Carbon monoxide, fossil	5.36E-04	kg	5.08	(1,4,3,3,1,5)
Nitrogen oxides	6.19E-04	kg	1.59	(1,4,3,3,1,5)
NMVOC, non-methane volatile organic compounds, unspecified origin	5.33E-05	kg	1.59	(1,4,3,3,1,5)
Methane, fossil	4.72E-06	kg	1.59	(1,4,3,3,1,5)
Dinitrogen monoxide	2.04E-06	kg	1.59	(1,4,3,3,1,5)
Particulates, unspecified	8.44E-05	kg	2.08	(2,4,3,3,1,5)
Particulates, <2.5 um	3.50E-05	kg	3.03	(3,1,3,3,1,2)
Particulates, >10 um	5.66E-05	kg	1.54	(3,3,3,3,1,2)
Particulates, >2.5 um, and <10 um	6.23E-05	kg	2.03	(3,3,3,3,1,2)

Table 5. WTW LCI for the operation of a lorry 3.5–7.5t running on ultra-low sulphur diesel, with SCR and DPF exhaust aftertreatment.

Operation, WTW, B0, SCR & DPF, lorry 3.5–7.5t, Euro3/RER	**1**	**km**	**SD_{g95}**	**criteria**
Materials/fuels				
Diesel, low-sulphur, at regional storage/CH	1.28E-01	kg	1.26	(1,4,3,3,1,5)
Catalytic converter, three-way, 19.1 litre/RER	3.17E-06	p	1.26	(1,4,3,3,1,5)
Catalytic converter, SCR, 200 litre/RER	1.80E-07	p	1.26	(1,4,3,3,1,5)
Urea Aqueous Solution, 32.5% at regional storage/RER	3.41E-03	kg	1.27	(2,4,3,3,1,5)
Emissions to air				
Carbon dioxide, fossil	3.39E-01	kg	1.26	(1,4,3,3,1,5)
Carbon monoxide, fossil	0.00E+00	kg	5.08	(1,4,3,3,1,5)
Nitrogen oxides	3.28E-04	kg	1.59	(1,4,3,3,1,5)
NMVOC, non-methane volatile organic compounds, unspecified origin	5.57E-06	kg	1.59	(1,4,3,3,1,5)
Methane, fossil	3.93E-06	kg	1.59	(1,4,3,3,1,5)
Dinitrogen monoxide	4.01E-06	kg	1.59	(1,4,3,3,1,5)
Particulates, unspecified	3.08E-06	kg	2.08	(2,4,3,3,1,5)
Particulates, <2.5 um	3.50E-05	kg	3.03	(3,1,3,3,1,2)
Particulates, >10 um	5.66E-05	kg	1.54	(3,3,3,3,1,2)
Particulates, >2.5 um, and <10 um	6.23E-05	kg	2.03	(3,3,3,3,1,2)

Table 6. WTW LCI for the operation of a lorry 3.5–7.5t running on B30, without SCR and DPF exhaust aftertreatment.

Operation, WTW, B30, OEM, lorry 3.5–7.5t, Euro3/RER	**1**	**km**	**SD_{g95}**	**criteria**
Materials/fuels				
B30, at regional storage/RER	1.30E-01	kg	1.26	(1,4,3,3,1,5)
Emissions to air				
Carbon dioxide, biogenic	1.04E-01	kg	1.26	(1,4,3,3,1,5)
Carbon dioxide, fossil	2.29E-01	kg	1.26	(1,4,3,3,1,5)
Carbon monoxide, biogenic	1.28E-04	kg	5.08	(1,4,3,3,1,5)
Carbon monoxide, fossil	2.81E-04	kg	5.08	(1,4,3,3,1,5)
Nitrogen oxides	2.51E-03	kg	1.59	(1,4,3,3,1,5)
NMVOC, non-methane volatile organic compounds, unspecified origin	1.17E-04	kg	1.59	(1,4,3,3,1,5)
Methane, biogenic	1.58E-06	kg	1.59	(1,4,3,3,1,5)
Methane, fossil	3.48E-06	kg	1.59	(1,4,3,3,1,5)
Dinitrogen monoxide	1.84E-06	kg	1.59	(1,4,3,3,1,5)
Particulates, unspecified	1.37E-04	kg	2.08	(2,4,3,3,1,5)
Particulates, <2.5 um	3.50E-05	kg	3.03	(3,1,3,3,1,2)
Particulates, >10 um	5.66E-05	kg	1.54	(3,3,3,3,1,2)
Particulates, >2.5 um, and <10 um	6.23E-05	kg	2.03	(3,3,3,3,1,2)

Table 7. WTW LCI for the operation of a lorry 3.5–7.5t running on B30, with SCR exhaust aftertreatment.

Operation, WTW, B30, SCR, lorry 3.5–7.5t, Euro3/RER	**1**	**km**	**SD_{g95}**	**criteria**
Materials/fuels				
B30, at regional storage/RER	1.32E-01	kg	1.26	(1,4,3,3,1,5)
Catalytic converter, SCR, 200 litre/RER	1.80E-07	p	1.26	(1,4,3,3,1,5)
Urea Aqueous Solution, 32.5% at regional storage/RER	3.40E-03	kg	1.27	(2,4,3,3,1,5)
Emissions to air				
Carbon dioxide, biogenic	1.05E-01	kg	1.26	(1,4,3,3,1,5)
Carbon dioxide, fossil	2.32E-01	kg	1.26	(1,4,3,3,1,5)
Carbon monoxide, biogenic	1.49E-04	kg	5.08	(1,4,3,3,1,5)
Carbon monoxide, fossil	3.28E-04	kg	5.08	(1,4,3,3,1,5)
Nitrogen oxides	6.42E-04	kg	1.59	(1,4,3,3,1,5)
NMVOC, non-methane volatile organic compounds, unspecified origin	4.10E-05	kg	1.59	(1,4,3,3,1,5)
Methane, biogenic	1.46E-06	kg	1.59	(1,4,3,3,1,5)
Methane, fossil	3.21E-06	kg	1.59	(1,4,3,3,1,5)
Dinitrogen monoxide	2.03E-06	kg	1.59	(1,4,3,3,1,5)
Particulates, unspecified	6.04E-05	kg	2.08	(2,4,3,3,1,5)
Particulates, <2.5 um	3.50E-05	kg	3.03	(3,1,3,3,1,2)
Particulates, >10 um	5.66E-05	kg	1.54	(3,3,3,3,1,2)
Particulates, >2.5 um, and <10 um	6.23E-05	kg	2.03	(3,3,3,3,1,2)

Table 8. WTW LCI for the operation of a lorry 3.5–7.5t running on B30, with SCR and DPF exhaust aftertreatment.

Operation, WTW, B30, SCR & DPF, lorry 3.5–7.5t, Euro3/RER	**1**	**km**	**SD_{g95}**	**criteria**
Materials/fuels				
B30, at regional storage/RER	1.29E-01	kg	1.26	(1,4,3,3,1,5)
Catalytic converter, three-way, 19.1 litre/RER	3.17E-06	p	1.26	(1,4,3,3,1,5)
Catalytic converter, SCR, 200 litre/RER	1.80E-07	p	1.26	(1,4,3,3,1,5)
Urea Aqueous Solution, 32.5% at regional storage/RER	3.18E-03	kg	1.27	(2,4,3,3,1,5)
Emissions to air				
Carbon dioxide, biogenic	1.07E-01	kg	1.26	(1,4,3,3,1,5)
Carbon dioxide, fossil	2.35E-01	kg	1.26	(1,4,3,3,1,5)
Carbon monoxide, biogenic	0.00E+00	kg	5.08	(1,4,3,3,1,5)
Carbon monoxide, fossil	0.00E+00	kg	5.08	(1,4,3,3,1,5)
Nitrogen oxides	3.03E-04	kg	1.59	(1,4,3,3,1,5)
NMVOC, non-methane volatile organic compounds, unspecified origin	7.19E-06	kg	1.59	(1,4,3,3,1,5)
Methane, biogenic	1.33E-06	kg	1.59	(1,4,3,3,1,5)
Methane, fossil	2.94E-06	kg	1.59	(1,4,3,3,1,5)
Dinitrogen monoxide	3.76E-06	kg	1.59	(1,4,3,3,1,5)
Particulates, unspecified	2.57E-06	kg	2.08	(2,4,3,3,1,5)
Particulates, <2.5 um	3.50E-05	kg	3.03	(3,1,3,3,1,2)
Particulates, >10 um	5.66E-05	kg	1.54	(3,3,3,3,1,2)
Particulates, >2.5 um, and <10 um	6.23E-05	kg	2.03	(3,3,3,3,1,2)

Table 9. WTW LCI for the operation of a lorry 3.5–7.5t running on B100, without SCR and DPF exhaust aftertreatment.

Operation, WTW, B100, OEM, lorry 3.5–7.5t, Euro3/RER	**1**	**km**	**SD_{g95}**	**criteria**
Materials/fuels				
Rape methyl ester, at regional storage/CH	1.38E-01	kg	1.26	(1,4,3,3,1,5)
Emissions to air				
Carbon dioxide, biogenic	3.38E-01	kg	1.26	(1,4,3,3,1,5)
Carbon monoxide, biogenic	4.03E-04	kg	5.08	(1,4,3,3,1,5)
Nitrogen oxides	2.46E-03	kg	1.59	(1,4,3,3,1,5)
NMVOC, non-methane volatile organic compounds, unspecified origin	6.27E-05	kg	1.59	(1,4,3,3,1,5)
Methane, biogenic	5.66E-06	kg	1.59	(1,4,3,3,1,5)
Dinitrogen monoxide	1.80E-06	kg	1.59	(1,4,3,3,1,5)
Particulates, unspecified	1.47E-04	kg	2.08	(2,4,3,3,1,5)
Particulates, <2.5 um	3.50E-05	kg	3.03	(3,1,3,3,1,2)
Particulates, >10 um	5.66E-05	kg	1.54	(3,3,3,3,1,2)
Particulates, >2.5 um, and <10 um	6.23E-05	kg	2.03	(3,3,3,3,1,2)

Table 10. WTW LCI for the operation of a lorry 3.5–7.5t running on B100, with SCR exhaust aftertreatment.

Operation, WTW, B100, SCR, lorry 3.5–7.5t, Euro3/RER	**1**	**km**	**SD_{g95}**	**criteria**
Materials/fuels				
Rape methyl ester, at regional storage/CH	1.41E-01	kg	1.26	(1,4,3,3,1,5)
Catalytic converter, SCR, 200 litre/RER	1.80E-07	p	1.26	(1,4,3,3,1,5)
Urea Aqueous Solution, 32.5% at regional storage/RER	3.54E-03	kg	1.27	(2,4,3,3,1,5)
Emissions to air				
Carbon dioxide, biogenic	3.44E-01	kg	1.26	(1,4,3,3,1,5)
Carbon monoxide, biogenic	4.47E-04	kg	5.08	(1,4,3,3,1,5)
Nitrogen oxides	6.42E-04	kg	1.59	(1,4,3,3,1,5)
NMVOC, non-methane volatile organic compounds, unspecified origin	1.80E-05	kg	1.59	(1,4,3,3,1,5)
Methane, biogenic	4.35E-06	kg	1.59	(1,4,3,3,1,5)
Dinitrogen monoxide	1.79E-06	kg	1.59	(1,4,3,3,1,5)
Particulates, unspecified	3.66E-05	kg	2.08	(2,4,3,3,1,5)
Particulates, <2.5 um	3.50E-05	kg	3.03	(3,1,3,3,1,2)
Particulates, >10 um	5.66E-05	kg	1.54	(3,3,3,3,1,2)
Particulates, >2.5 um, and <10 um	6.23E-05	kg	2.03	(3,3,3,3,1,2)

Table 11. WTW LCI for the operation of a lorry 3.5–7.5t running on B100, with SCR and DPF exhaust aftertreatment.

Operation, WTW, B100, SCR & DPF, lorry 3.5–7.5t, Euro3/RER	**1**	**km**	**SD_{g95}**	**criteria**
Materials/fuels				
Rape methyl ester, at regional storage/CH	1.42E-01	kg	1.26	(1,4,3,3,1,5)
Catalytic converter, three-way, 19.1 litre/RER	3.17E-06	p	1.26	(1,4,3,3,1,5)
Catalytic converter, SCR, 200 litre/RER	1.80E-07	p	1.26	(1,4,3,3,1,5)
Urea Aqueous Solution, 32.5% at regional storage/RER	3.59E-03	kg	1.27	(2,4,3,3,1,5)
Emissions to air				
Carbon dioxide, biogenic	3.46E-01	kg	1.26	(1,4,3,3,1,5)
Carbon monoxide, biogenic	0.00E+00	kg	5.08	(1,4,3,3,1,5)
Nitrogen oxides	4.76E-04	kg	1.59	(1,4,3,3,1,5)
NMVOC, non-methane volatile organic compounds, unspecified origin	4.99E-06	kg	1.59	(1,4,3,3,1,5)
Methane, biogenic	4.23E-06	kg	1.59	(1,4,3,3,1,5)
Dinitrogen monoxide	3.86E-06	kg	1.59	(1,4,3,3,1,5)
Particulates, unspecified	2.14E-06	kg	2.08	(2,4,3,3,1,5)
Particulates, <2.5 um	3.50E-05	kg	3.03	(3,1,3,3,1,2)
Particulates, >10 um	5.66E-05	kg	1.54	(3,3,3,3,1,2)
Particulates, >2.5 um, and <10 um	6.23E-05	kg	2.03	(3,3,3,3,1,2)

Table 12 presents the LCI for bio-fossil diesel blend B30, and is based on a 30% v/v contribution of RME biodiesel and 70% v/v fossil diesel.

Table 12. LCI for bio-fossil diesel blend B30.

B30, at regional storage/RER	**1**	**kg**	**SD_{g95}**	**criteria**
Materials/fuels				
Diesel, low-sulphur, at regional storage/CH	6.88E-01	kg	1.11	(1,1,3,1,1,na)
Rape methyl ester, at regional storage/CH	3.12E-01	kg	1.11	(1,1,3,1,1,na)

Table 13 presents the LCI for urea aqueous solution including material inputs, production, and distribution.

Table 13. LCI for AUS32.

Urea Aqueous Solution, 32.5% at regional storage/RER	**1**	**kg**	**SD_{g95}**	**criteria**
Materials/fuels				
Urea, as N, at regional storehouse/RER	3.25E-01	kg	1.05	(1,1,1,1,1,na)
Water, deionized, at plant/CH	6.75E-01	kg	1.05	(1,1,1,1,1,na)
Regional distribution, oil products/RER	1.91E-10	p	3.38	(4,5,3,1,4,5)
Transport, lorry >16t, fleet average/RER	1.50E-01	tkm	3.38	(4,5,3,1,4,5)
Chemical plant, organics/RER	4.00E-10	p	3.38	(4,5,3,1,4,5)

Determination of total NO_X, PM, CO, and HC (CH_4 and NMHC) life-cycle emissions

Life-cycle emissions inventories were determined with SimaPro v7.3.3 software (Goedkoop et al., 2010). The LCIA method chosen was CML 2 baseline (Goedkoop et al., 2008). Mean values and standard deviation were determined with Monte Carlo method with 1000 runs.

Table 14. Case life-cycle emissions inventories recorded.

Bio-fossil diesel blend	Exhaust aftertreatment	TTW	WTT
B0	OEM	X	X
	SCR	X	X
	SCR & DPF	X	X
B30	OEM	X	X
	SCR	X	X
	SCR & DPF	X	X
B100	OEM	X	X
	SCR	X	X
	SCR & DPF	X	X

For the determination of total NO_X, PM, CO, and HC (CH_4 and NMHC) emissions, the following conditioning steps were performed with the life-cycle emissions inventories:

1. Only emissions to air were included, next
2. Only emissions measured in kilogram (kg) were included, next
3. European Union emission standards do not specify which HC emissions should be included in the determination of total HC, and it was necessary to perform a cut-off for NMHC emissions. Those emissions included in total NO_X, PM, CO, NMHC, and CH_4 are explained in Table 15.

Table 15. Life-cycle emission inventory emissions included in the determination of total NO_X, PM, CO, and HC for the various bio-fossil diesel blends and exhaust aftertreatment configurations.

Bio-fossil diesel blend	Exhaust aftertreatment	Emission		TTW	WTT
B0, B30, B100	OEM, SCR, SCR & DPF	NO_X		Nitrogen oxides	Nitrogen oxides
		PM		Particulates, unspecified Particulates, >10 µm Particulates, <2.5 µm Particulates, >2.5 µm, and <10 µm	Particulates, >10 µm Particulates, <2.5 µm Particulates, >2.5 µm, and <10 µm
		CO		Carbon monoxide, fossil Carbon monoxide, biogenic	Carbon monoxide, fossil Carbon monoxide, biogenic
		HC	NMHC	NMVOC, non-methane volatile organic compounds, unspecified Hydrocarbons, aliphatic, alkanes, unspecified Pentane Propane Ethane Butane	NMVOC, non-methane volatile organic compounds, unspecified Hydrocarbons, aliphatic, alkanes, unspecified Pentane Propane Ethane Butane
			CH_4	Methane, fossil Methane, biogenic	Methane, fossil Methane, biogenic

To determine total NO_X, PM, CO, and HC (CH_4 and NMHC), the individual mean value of those emissions listed in Table 15 have been summed, e.g.

$$Total\ TTW\ CO = TTW\ carbon\ monoxide, fossil + TTW\ carbon\ monoxide, biogenic$$

For WTW emissions the following calculation was made, e.g.

$$WTW\ CO = Total\ TTW\ CO + Total\ WTT\ CO$$

The same conditioning steps were performed, and individual emissions were included in the determination of the total standard deviation, e.g.

$$Total\ TTW\ \sigma_{CO} = TTW\ \sigma_{Carbon\ monoxide, fossil} + TTW\ \sigma_{Carbon\ moxoxide, biogenic}$$

In order to determine standard deviation with 95% confidence, the total standard deviation was multiplied by two, e.g.

$$Total\ TTW\ 2\sigma_{CO} = 2 \times Total\ TTW\ \sigma_{CO}$$

For WTW standard deviation with 95% confidence the following calculation was made, e.g.:

$$WTW\ 2\sigma_{CO} = Total\ TTW\ 2\sigma_{CO} + Total\ WTT\ 2\sigma_{CO}$$

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Article III

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Comparative attributional life cycle assessment of European cellulase enzyme production for use in second-generation lignocellulosic bioethanol production

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Abstract

Purpose The production of cellulase enzymes (CE) has been identified as one major contributor towards the life cycle environmental and economic impacts of second-generation lignocellulosic bioethanol (LCB) production. Despite this knowledge, the literature lacks consistent and transparent life cycle assessments (LCA) which compare CE production based on the three more commonly proposed carbon sources: cornstarch glucose, sugar cane molasses and pre-treated softwood. Furthermore, numerous LCAs of LCB omit CE production from their system boundaries, with several authors citing the lack of available production data.

Methods In this article we perform a comparative attributional LCA for the on-site production of 1 kg CE in full-broth via submerged aerobic fermentation (SmF) based on the three alternative carbon sources, Cases A, B and C respectively. We determine life cycle inventory (LCI) material consumption based on stoichiometric equations and volume flow, supplemented with information from the literature. All LCIs are provided in a consistent and transparent manner, filling the existing data gaps towards performing representative LCAs of LCB production with on-site CE production. Life cycle impact assessment (LCIA) results are determined with SimaPro 8 software using CML 1A baseline and non-baseline methods along with cumulative energy demand, and are compared to those of similar studies. Sensitivity analysis is performed both for all major assumptions and for market changes with the application of advanced attributional LCA (AALCA).

Results and Conclusion We find that CE production based on pre-treated softwood (Case C) provides the lowest environmental impacts, followed by sugar cane molasses (Case B) and then cornstarch glucose (Case A), with global warming potentials of: 7.9, 9.1 and 10.6 kg CO_2 eq./kg enzyme, respectively. These findings compare well with those of similar studies, though great variation exists in the literature. Through sensitivity analysis, we determine that results are sensitive to assumptions made concerning carbon source origin, applied allocation, market changes, process efficiency and electricity supply. Furthermore, we find that the contribution of CE production towards the overall life cycle impacts of LCB is significant, and that the omission of this sub-process in LCAs of LCB production can compromise their representativeness.

Keywords

Cellulase enzyme, life cycle assessment, glucose, molasses, biomass, lignocellulosic bioethanol

1 Introduction

With the implementation of the Renewable Energy Directive (EC 2009), and its predecessors, the European Union (EU) has set a 10% target for use of renewable energy in the transport sector by 2020. Furthermore, the European Commission reported in a recently released scenario that the total energy demand of all transport in the EU 27+2 was 381 million tonnes oil equivalent (Mtoe) in 2010, of which road transport was 307 Mtoe, with biofuels contributing 13 Mtoe (Hamje et al. 2014). The same report predicts that by 2020, energy demand in road transport will decrease by 8.5% though biofuel's contribution will increase to 21.5 Mtoe, with LCB contributing 0.7 Mtoe, or approximately 1.38 x 10^9 l. The promotion of LCB is partially based on the environmental, social and economic concerns surrounding first-generation biofuels (Tilman et al. 2009) many of which resulted from so-called transfer effects (Holden and Gilpin 2013).

LCB is one of several second-generation biofuels, which are defined as biofuels produced from cellulose, hemicellulose and lignin (Sims et al. 2008), with biomass sources from silviculture, agriculture waste streams and fast-growing high-yield energy crops. The most common proposed method for converting lignocellulosic biomass into bioethanol is via the bio-chemical process of enzymatic hydrolysis and fermentation. Central to this process is the enzymatic hydrolysis of cellulose and hemicellulos into fermentable sugars by CE, see Fig. 1.

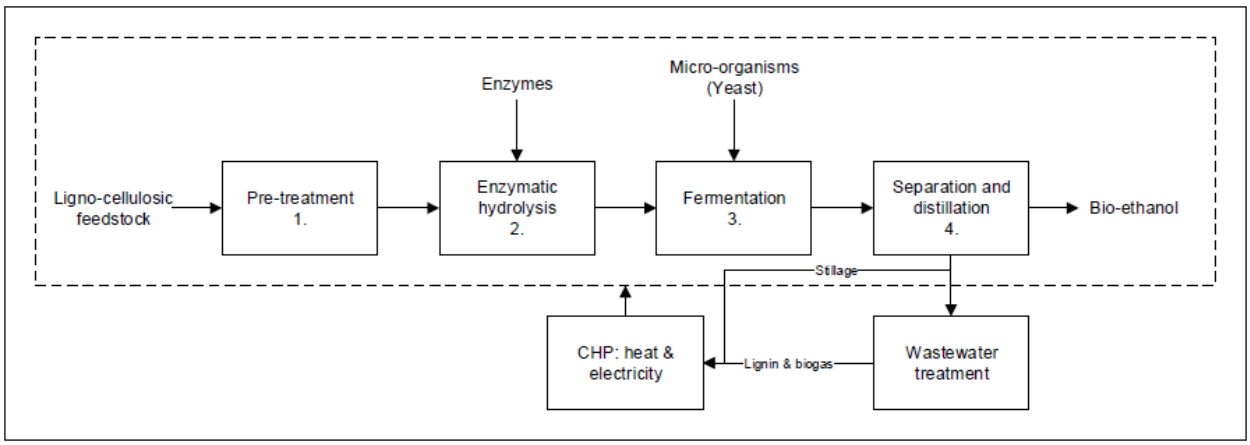

Fig. 1 Simplified process overview of LCB production via enzymatic hydrolysis and fermentation (MacLean and Spatari 2009; Sims et al. 2008)

Large quantities of CE are required for the enzymatic hydrolysis of lignocellulosic biomass, with CE consumption ranges of approximately 0.3–2.1 g of CE per MJ LCB proposed in the literature (Hong et al. 2013; Humbird et al. 2011; MacLean and Spatari 2009), see Supplementary Material.

The most common method proposed in LCA literature to produce CEs results from their secretion by the fungus T. *reesei* cultivated by SmF and fed on a carbon source, see Table 1, though numerous CE secreting organisms and cultivation methods exist. CE production is a resource-intensive process and represents one of the greatest uncertainties and current high costs connected to LCB production (Foust et al. 2009; Luo et al. 2009). Specifically, the high uncertainty and costs are connected to the provision of the carbon source, for which glucose, molasses and pre-treated softwood are alternatives, see Table 1.

Table 1 Comparison of published (environmental studies and economic studies) which highlight the production of enzymes

	Final product[a]	**Type of assessment**	**Carbon source**[b]	**Production**[c]
Enzyme specific assessments				
(Olofsson et al. 2015)	Formulated- and non-formulated cellulase	Environmental & Economic	Pre-treated biomass, molasses	SmF/ on- & off-site/ T. reesei
(Agostinho et al. 2014)	Formulated cellulase	Environmental & Economic	Pre-treated biomass	SmF/ off-site/ C. thermocellum
(Hong et al. 2013)	Formulated- and non-formulated cellulase	Environmental & Economic	Glucose	SmF/ on- & off-site/ T. *reesei*
(Dunn et al. 2012)	Formulated cellulase	Environmental & Economic	Glucose	SmF/ off-site/ T. *reesei* (inferred)
(Harding and Harrison 2011)	Formulated cellulase	Environmental	Pre-treated biomass, corn liquor	SmF & SSC/ off-site/ T. *reesei* & T. *thermocellum*
(Klein-Marcuschamer et al. 2012)	Formulated cellulase	Economic	Pre-treated biomass, glucose	SmF/ off-site (inferred)/ T. *reesei*,
(Barta et al. 2010)	Non-formulated cellulase	Economic	Pre-treated biomass, molasses	SmF/ on-site/ T. reesei
(Kim et al. 2009)	Formulated- aldolase, carbamoylase and hydantoinase	Environmental	Soybean protein, yeast extract	SmF/ off-site/ n/a
(MacLean and Spatari 2009)	Non-formulated cellulase	Environmental	Pre-treated biomass	SmF/ on-site/ T. *reesei*
(Harding 2008)	Formulated cellulase (among other)	Environmental	Pre-treated biomass, corn liquor (among other)	SmF & SSC/ off-site/ T. *reesei* & C. *thermocellum*
(Nielsen et al. 2007)	Formulated- α-amylase, glucoamylase, phytase, protease andamylase	Environmental	Glucose, maltose, cornstarch, sucrose, etc.	SmF/ off-site/ n/a
(Zhuang et al. 2007)	Formulated- and non-formulated cellulase	Economic	Pre-treated biomass	SmF & SSC/ on- & off-site/ C. *thermocellum*
Ethanol assessments with detailed cellulase production				
(Davis et al. 2015)	Hydrocarbons (non-formulated cellulase)	Economic	Glucose	SmF/ on-site/ T. *reesei*
(Davis et al. 2013)	Ethanol (non-formulated cellulase)	Economic	Glucose	SmF/ on-site/ T. *reesei*
(Agostinho and Ortega 2013)	Ethanol (non-formulated cellulase)	Environmental	n/a	n/a/ on-site/ n/a
(Humbird et al. 2011)	Ethanol (non-formulated cellulase)	Economic	Glucose	SmF/ on-site/ T.*reesei*
(Sheehan et al. 2003)	Ethanol (non-formulated cellulase)	Environmental	Pre-treated biomass	SmF/ on-site/ T. *reesei*
(Wooley et al. 1999)	Ethanol (non-formulated cellulase)	Economic	Pre-treated biomass	SmF/ on-site/ T. *reesei*

[a] Formulated refers here to any post-cultivation process applied to prepare the enzymes for storage; which covers the unique/overlapping terms used in the literature: formulation, immobilization, purification, freeze-drying and concentration.
[b] Pre-treated biomass refers to: detoxified pre-treated biomass slurry, cellulose, wood- chips and pulp, and paper pulp.
[c] Sequence is: production process/location/enzyme producing organism. SCC refers to solid-state cultivation, on-site includes co- and adjacent -located.

1.1 Goal Definition

The goal of this LCA study is to compare the environmental impacts associated with European production of CE based on the three alternative carbon sources: cornstarch glucose, sugar cane molasses and pre-treated softwood. In addition, we aim to fill the existing data gaps in existing LCAs of LCB by presenting consistent, detailed and transparent LCI data for CE production. In doing so we answer the following research questions:

1. Which cellulase enzyme production path, with respect to carbon source, provides the lowest environmental impacts?
2. How do the environmental impacts determined in this study compare with those determined in similar studies?
3. If current enzyme production methods are over- or underestimated, or omitted, what inferences can be made with respect to the representativeness of existing LCAs of LCB?

The LCA is performed under the guidance of both the International Organization for Standardization standards 14040 series for LCA (ISO 2006a,b) and the European Commission's "International Reference Life Cycle Data System (ILCD) Handbook" (EC 2010). We apply attributional LCI modelling methods, and prioritize representative-, publically available-, consistent- and transparent data sources over proprietary commercial data. The effect of this choice on results, along with other assumptions described in more detail later, is tested by sensitivity analyses. Classifying this study as Situation A (EC 2010) and applying attributional LCA methodology presents limitations for the interpretation and application of the results, which are not intended to be used for meso- or macro-level decision-making. Instead, the results are intended to provide guidance to research and business development, along with other public and private stakeholders working in the field of CE and LCB development.

1.2 Scope Definition

"Cellulase enzyme" is an umbrella term for a mixture of endo-p-glucanases-, exo-P-glucanases- and β-glucosidase proteins. These three proteins work in sequence, with endo-p-glucanases randomly breaking the cellulose molecular chain into glucose and cello-oligo saccharides, followed by exo-P-glucanases, which then work on the exposed chain ends reducing these to glucose and cellobiose. Finally, the β-glucosidase protein

hydrolyses the cellobiose to glucose. The result is a slurry of fermentable, low-molecular-weight hexose (e.g., glucose) and pentose (e.g., xylose) monosaccharides (Miyamoto 1997).

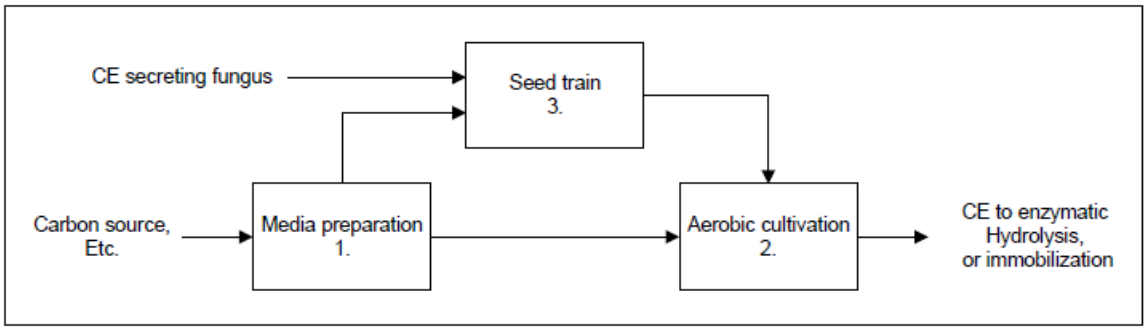

Fig. 2 Process overview of CE production via the SmF method (Davis et al. 2015)

Fig. 2 presents the general system boundary for on-site CE production via the SmF method, from which the specific cases are derived. The product of this process is a full-broth containing CE, which is then utilized in on-site LCB production, specifically the enzymatic hydrolysis of cellulose and hemicellulose to fermentable sugars, step 2 in Fig. 1. The functional unit (FU) of CE production is 1 kg of CE (protein) in full-broth, which is the same FU chosen by all authors of on-site CE-specific assessments in Table 1.

LCA presents the potential environmental impacts of a system or product over its life cycle based on measurable data of existing processes and products with known uncertainty (EC 2010). In this study foreground process LCIs are formulated based on literature review and presented in the succeeding case descriptions. Background processes are derived from commercial databases available in the SimaPro 8 LCA software package (Goedkoop et al. 2013).

Neither CE production nor any of the foreground processes represent multi-functional processes. Of the significant background processes, both corn wet-milling and sugar cane processing are multi-functional processes. Corn wet-milling has the co-products corn steep liquor, corn oil and cornstarch (input to glucose production) among others, and sugar cane processing has the co-product molasses among others. As recommended by the ISO (2006a,b), the methods of subdivision and allocation were considered in this order for dealing with multi-functionality. Using the same logic as Würdinger et al. (2003) (i.e., due to the complexity of corn wet-milling and sugar cane processing, and lack of data with suitable resolution), the preferable solution of subdivision was not feasible. As a result, and with the aim of methodological consistency, allocation has been

applied to all multi-functional processes. Specifically, a preferred causal physical relation does not exist for the co-products of corn wet-milling, and considering that the primary motivation of corn wet-milling operators is profit, background LCI data using economic allocation was selected (Würdinger et al. 2003). Sugar and molasses, the co-products of sugar cane processing, share the causal physical relation of energy; therefore, background LCI data using energetic allocation was chosen (van Zeist et al., 2012). Remaining allocation decisions for *significant* background processes are selected based on the most suitable causal relation, and clearly identified in the LCI tables in this article and in its Supplementary Material.

LCIA was performed using SimaPro 8 LCA software (Goedkoop et al. 2013). CML 1A baseline and CML 1A non-baseline methods (Pré 2015) were used for determining global warming potential 100 year time horizon (GWP) measured in kg CO_2 eq., eutrophication potential (EP) measured in g PO_4 eq., acidification potential (AP) measured in g SO_2 eq., ozone layer depletion (ODP) measured in mg CFC-11 eq., photochemical oxidation potential (POP) measured in g C_2H_4 eq., and land use measured in m^2a. Cumulative energy demand, measured in MJ, was determined using methods developed by Frischknecht et al. (2007).

Sensitivity analysis involves two approaches: first, through traditional testing of sensitivity towards changes in CE yield, source of electricity and various aspects concerning the carbon source. The second approach involves testing the sensitivity of results towards market effects by multiplying Global Change Mix Factors (GCMF) to hexose and pentose flows, a method known as advanced attributional LCA (AALCA) (Andrae 2015). We apply AALCA due to the common criticism of ALCA methodology in its limitation to account for future market effects. Andrae (2015) has shown that AALCA can be performed as a sensitivity check of comparative ALCA results. By applying GCMFs based on real or future market changes, the interdependence of rising and declining global markets and micro-level LCA shifts can be taken into account in comparative micro-level ALCA studies. Therefore, AALCA represents an improvement over ALCA but cannot fully claim, as consequential LCA, to fully reflect consequences of decisions, and as such is not a replacement for consequential LCA.

2 Methods and Case Descriptions

In this LCA study we compare three cases of CE production based on the three more relevant carbon sources: cornstarch glucose in Case A, sugar cane molasses in Case B and pre-treated softwood in Case C. All percentages (%) are given as weight (wt.) percentages unless otherwise stated.

The National Renewable Energy Laboratory (NREL) process descriptions for CE production (Humbird et al. 2011; Wooley et al. 1999) form the general system from which all cases are developed. These designs along with succeeding/preceding LCB assessments produced by NREL, see Table 1, are based on Schell et al. (1991). We have chosen the NREL designs for two reasons: first, NREL provides transparent foreground LCIs of CE production, and second, these designs have been adopted in numerous other studies, among others: (Dunn et al. 2012; MacLean and Spatari 2009; Sheehan et al. 2003; Zhuang et al. 2007). This choice facilitates both the comparison with- and the transfer of results to these and other studies.

The on-site SmF production of CE in full-broth involves three steps:

1. Media preparation – a carbon source, water and other nutrients are mixed in fixed quantities, along with small amounts of slip-stream-produced sophorose, which induces T. *reesei* to produce CE.

2. Seed train – T. *reesei* fungus is stimulated to multiply in optimal conditions and fed by fraction of the media prepared in step 1, producing a T. *reesei* inoculum for step 3.

3. Aerobic cultivation – in this step the T. *reesei* from step 2 is introduced to a fermenter under submerged aerobic conditions (SmF), where it feeds on the carbon source and nutrients prepared in step 1 and secretes CE, see Fig. 2.

Common to all three cases is the assumption that the production of cell mass (T. reesei) and CE, steps 2 and 3 respectively, requires the stoichiometrically balanced dosing of the reactants: carbon source, oxygen (O_2), ammonia (NH_3) and sulfur dioxide (SO_2) with the resulting products: carbon dioxide (CO_2), water (H_2O), CE and cell mass, see Table 2. We assume that the entire reactive carbon source, see Table 3, is consumed during steps 2 and 3. Humbird et al. (2011), citing Atkinson and Mavituna (1991) and proprietary information provided by Novozymes, provide the elemental compositions for CE and cell mass, respectively for Cases A and B, and Wooley et al. (1999), citing Wooley and Putsche (1996), provide the elemental composition for CE and cell mass for Case C, see Table 2.

The efficiency of CE production can be expressed by the molar selectivity of the reactive carbon source's carbon atoms towards the formation of CO_2, CE and cell mass. The higher the carbon selectivity towards CE, the higher the yield of CE per kg of reactive carbon source. Davis et al. (2015, 2013) and Humbird et al. (2011) all apply the

same molar selectivity with glucose as the carbon source, and these authors provide the molar selectivity for Cases A and B, see Table 2. Wooley et al. (1999) assume one molar selectivity for soluble sugars and a different molar selectivity for sugar polymers; these combine to form the molar selectivity presented in Table 2 for Case C. By assessing the elemental compositions of the reactants and products and molar selectivity, we were able to balance the aforementioned stoichiometric equations, see Table 2.

Table 2 Chemical reactions modelled in CE production for Cases A, B and C

	Case A	**Case B**	**Case C**
Carbon source	Cornstarch glucose	Sugar cane molasses	Pre-treated softwood
CE prod. Modelled on:	(Humbird et al. 2011)		(Wooley et al. 1999)
Elemental composition CE	$CH_{1.59}N_{0.24}O_{0.42}S_{0.01}$		$CH_{1.57}N_{0.29}O_{0.31}S_{0.007}$
Elemental composition Cell mass	$CH_{1.645}N_{0.205}O_{0.445}S_{0.005}$		$CH_{1.64}N_{0.23}O_{0.39}S_{0.0035}$
Carbon selectivity	65% CO_2/31% CE/4% cell mass		65% CO_2/29% CE/6% cell mass
Reactants			
$C_6H_{12}O_6$[a]	1.00	1.00	1.00
$C_{12}H_{22}O_{11}$[b]	0.00	0.56	0.00
$C_5H_{10}O_5$[c]	0.00	0.00	0.61
$C_6H_{10}O_5$[d]	0.00	0.00	2.45
O_2	3.86	8.19	15.2
NH_3	0.50	1.05	2.32
SO_2	0.02	0.04	0.05
Products			
CO_2	3.90	8.28	15.4
H_2O	5.07	10.2	18.2
CE	1.86	3.95	6.82
Cell mass	0.24	0.51	1.49

[a]Galactose, glucose, fructose and mannose
[b]Sucrose
[c]Arabinose and xylose
[d]Cellulose, galactan and mannan

Table 3 Composition of reactive carbon source available to cellulase production per 1000 kg of carbon source entering cellulase production

	Cornstarch glucose	**Sugar cane molasses**	**Pre-treated softwood**	**Unit**
Arabinose			3.5	kg
Fructose		146.0		kg
Galactose			6.4	kg
Glucose	850.0	128.0	10.9	kg
Mannose			23.4	kg
Sucrose		292.0		kg
Xylose			17.4	kg
Cellulose			89.0	kg
Galactan			0.2	kg
Mannan			0.6	kg
Total	**850.0**	**566.0**	**151.4**	**kg**

CE production requirements for nutrients and antifoaming agents are based on Schell et al. (1991), see Supplementary Material. LCI data for polysorbate 80 and potassium phosphate were not found in the literature or commercial databases; accordingly, we have determined GWP and cumulative energy demand for polysorbate 80 based on the Swiss Method (Wernet et al. 2008, 2009), and have constructed an LCI for the production of potassium phosphate based on Freilich and Petersen (2005), see Supplementary Material.

Energy requirements (electricity, heating, and cooling) for Cases A and B are adapted from Humbird et al. (2011) and Wooley et al. (1999) for Case C. Similar to Humbird et al. (2011) and Wooley et al. (1999), we assume that electricity and heat are produced by combusting biogas in an on-site combined heat and power (CHP) generation plant. Cooling is produced from the same electricity source and scaled with respect to CE production. Aeration in the form of compressed air is scaled based on the oxygen uptake rate determined stoichiometrically, see Table 2. In doing so we assume that the oxygen transfer rate is proportional to the oxygen uptake rate (Humbird et al. 2011). In addition, we assume the same reactor geometries, equipment and media characteristics as presented in Humbird et al. (2011) for Cases A and B and Wooley et al. (1999) for Case C.

Table 4 presents the final LCIs for the base Cases A, B and C. The specific descriptions of carbon source production are provided subsequent sections.

Table 4 LCIs for Cases A, B and C for the production of 1 kg cellulase enzyme

Product	Case A	Case B	Case C	Unit	Data source[a]
CE	1.0	1.0	1.0	kg	
Input					
Materials/fuels					
Water	19.0	22.9	35.5	kg	Ecoinvent 3
Carbon source	4.7	6.9	28.3	kg	Table 5 (**no allocation**)/ Agri-footprint (**energy**)/ Table 6 (**no allocation**)
Ammonium sulphate	0.037	0.046	0.095	kg	Ecoinvent 3
Potassium phosphate	0.053	0.066	0.135	kg	Supplementary Material (**no allocation**)
Magnesium sulphate	0.008	0.010	0.020	kg	Ecoinvent 3
Calcium chloride	0.011	0.013	0.027	kg	Ecoinvent 3
Polysorbate 80	0.005	0.007	0.014	kg	Own calculation (**no allocation**)
Corn steep liquor	0.269	0.338	0.692	kg	Agri-footprint
Sulfur dioxide	0.028	0.028	0.022	kg	Ecoinvent 3
Ammonia	0.189	0.189	0.254	kg	Ecoinvent 3 (**no allocation**)
Antifoam (corn oil)	0.026	0.033	0.068	kg	Agri-footprint
Energy					
Electricity	6.3	7.3	26.5	kWh	Ecoinvent 3
Heating	2.9	3.6	0.0	MJ	Ecoinvent 3
Cooling	59.8	59.8	43.3	MJ	Ecoinvent 3
Emissions					
Carbon dioxide	3.8	3.8	4.4	kg	

[a] Causal relation used for foreground allocation in bold; when present and significant (≥ 5%) with respect to the contribution analysis of GWP, see either Fig.3 or Supplementary Material

2.1 Cornstarch Glucose

Hobbs (2009) and EIA (2015) provide the LCI data for cornstarch glucose production; this data has been conditioned with respect to glucose output based on Tsiropoulos et al. (2013) and lower dry matter (DM) (85%), see Table 5. In the modelled process, cornstarch containing slurry is treated with hydrochloric acid to lower the pH under high pressure and high temperature for a short residence time, resulting in the longer starch molecules being cleaved into lower-molecular-weight glucose. After conversion to glucose has been achieved, the pH is raised using soda ash (sodium carbonate). The resulting slurry is then centrifuged to remove impurities, then treated with active carbon to remove undesirable flavour and colour. The final step adjusts the pH further and condenses the liquor through evaporation (Hobbs 2009). The composition of reactive carbon sources available for CE production per 1000 kg glucose syrup is presented in Table 3.

Table 5 LCI for the production of 1000 kg glucose 85% DM

	Value	Unit	Data source[a]
Product			
Glucose			
Input			
Materials/fuels			
Activated carbon	15.32	kg	Agri-footprint version 1.0 (**economic**)
Hydrochloric acid	7.66	kg	Ecoinvent 3
Cornstarch	765.77	kg	Ecoinvent 3 (**economic**)
Soda ash	3.06	kg	Ecoinvent 3
Water	6126.13	kg	Ecoinvent 3
Electricity/heat			
Electricity	57.56	kWh	European Life Cycle Database v3.0
Heat	539.969	MJ	Ecoinvent 3
Waste and emissions to treatment			
Waste water	5.92	m^3	Ecoinvent 3

[a] Causal relation used for foreground allocation in bold; when present and significant (≥ 5%) with respect to the contribution analysis of GWP

2.2 Sugar Cane Molasses

Agri-footprint (2014) citing van Zeist et al. (2012) provide the consumption mix and LCI data for sugar cane molasses production and transport to Europe (Netherlands). Furthermore, we have adopted the energetic allocation applied by van Zeist et al. (2012), with 41% of inputs allocated to molasses and the remaining to sugar. We assume that the consumption mix of molasses for the Netherlands is representative of the mix for Europe. Molasses is one product of the multi-product process of refining sugar cane; other products include cane sugar, bagasse and filter cake. Specifically, molasses is the co-product of the multi-stage process of forming and separating crystalized cane sugar (sucrose) from cane juice. The precursor to molasses, cane juice is dried to form various grades of molasses depending on consumption (i.e., edible molasses or feed-grade molasses (blackstrap)) (EPA 1995). The DM content of the raw molasses is assumed to be 73% (van Zeist et al. 2012). DM composition is based on Olbrich (2006), see Supplementary Material, and has a higher ratio of sucrose to glucose and fructose than the ratio used by He et al. (2014). The composition of reactive carbon sources available for CE production per 1000 kg of molasses is presented in Table 3.

2.3 Pre-treated Softwood Biomass

The production of pre-treated softwood, including wood chip handling and pre-treatment processes, has been adapted for softwood chips from Wooley et al. (1999), see Table 6. The softwood chips entering handling and pre-treatment have a DM content of 47.9%, and a DM composition adopted from Ferraro et al. (1999), see Supplementary Material, which is similar to that used by Barta et al. (2010) and Olofsson et al. (2015). In this

process, the softwood chips are initially screened and cleaned of impurities and contaminants. Then a dilute-acid treatment is applied at high temperatures, and during the treatment the hemicellulose polysaccharides are hydrolysed into soluble monosaccharides. This choice of pre-treatment is in consensus with the majority of assessments of LCB (Wiloso et al. 2012). In addition, a portion of the cellulose is converted to glucose, and otherwise exposed for conversion by CE. Lime is then added to raise the pH, and then precipitated as gypsum and removed. The resulting product is a hydrolysate containing cellulose and soluble sugars, among others (Wooley et al. 1999). The hydrolysis reactions for cellulose and hemicellulose are adopted from Davis et al. (2015), see Supplementary Material. The composition of reactive carbon sources available for CE production per 1000 kg of pre-treated softwood is presented in Table 3.

Table 6 LCI for the production of 1000 kg pre-treated softwood biomass

	Value	**Unit**	**Data source**[a]
Product			
Pre-treated softwood biomass	1000	kg	
Input			
Materials/fuels			
Ammonia	3.14	kg	Ecoinvent 3 (**no allocation**)
Lime	1.97	kg	Ecoinvent 3
Softwood chips (DM)	441	kg	Supplementary Material
Steam	177	kg	Supplementary Material
Sulfuric acid	5.15	kg	Ecoinvent 3
Water	759	kg	Ecoinvent 3
Electricity/heat			
Electricity	5.93	kWh	Ecoinvent 3
Waste and emissions to treatment			
Gypsum	6.71	kg	Ecoinvent 3
Waste water	0.42	m^3	Ecoinvent 3

[a] Causal relation used for foreground allocation in bold; when present and significant (≥ 5%) with respect to the contribution analysis of GWP

3 Results

Table 7 summarizes the LCIA results of the present study and compares these with the results of other relevant studies. From Table 7 we observe two things: first, the present results for GWP, EP, AP, POP and cumulative energy demand are within acceptable ranges. All of these impacts (excl. EP for Case A) fall within the standard deviation (2σ or 95% confidence) of the mean of the relevant studies, when considering that non-formulated enzymes can have impacts 36% lower than formulated enzymes (Hong et al. 2013). It should be noted that it was not possible to determine the weighted mean and standard deviation for several impact categories due to limited

sample size. The second observation is that over all impact categories Case C provides the lowest environmental impacts for the production of CE.

Table 7 Summary of life cycle impact assessment results, including a comparison with results from other relevant studies, including the weighted mean and standard deviation (2σ or 95% confidence) of the other studies

	GWP100	EP	AP	ODP	POP	LU	Cumulative energy demand
Unit	kg CO_2 eq.	g PO_4 eq.	g SO_2 eq.	mg CFC-11 eq.	g C_2H_4 eq.	m^2a	MJ
Case A	10.6	44.2	49.3	0.4	1.6	0.5	81.2
Case B	9.1	24.8	54.5	0.1	4.9	4.3	62.3
Case C	7.9	8.7	31.6	0.2	2.0	41.4	52.4
Weighted Mean & standard deviation[a]	(14.7 ± 13.7)[F] (6.3 ± 4.0)[NF]	(22.2 ± 18.7)[F] n/a	(210.4 ± 343.1)[F] n/a	(1.9 ± 0,76)[F] n/a	(11.1 ± 10.9)[F] n/a	n/a n/a	(120.2 ± 118.9)[F] n/a
(Olofsson et al. 2015)	5.5[F]						69[F]
(Agostinho et al. 2014)	21.93[F]		7[F]				1664[F]
(Hong et al. 2013)	10.2[NF]–16.0[F]						
(Dunn et al. 2012)[b]	3.7[F]						46[F]
(Harding and Harrison 2011)							(53.5–190.2)[F,c]
(Kim et al. 2009)	(16–25)[F]	(11.5–18.3)[F,b]	(120–145)[F,b]		(5.8–7.6)[F,b]		(117–207)[F,b,d]
(MacLean and Spatari 2009)	2.3[NF]						24.8[NF,e]
(Harding 2008)	(-1240–-924)[F]	(22–37)[F]	(270–510)[F]	(1.52–2.28)[F]	(11–20)[F]		(88.8–190.2)[F,c]

[a] Does not include cumulative energy demand of Agostinho et al. (2014) or GWP of Harding (2008)
[b] Determined- from figure, or through calculation
[c] Only (direct) electricity and steam consumption
[d] Only non-renewable energy consumption
[e] Only fossil energy consumption
[F] Formulated
[NF] Non-formulated

Figs. 3 through 9 present the LCIA results for the base Cases A, B and C. The results are disaggregated for all sub-processes which contribute significantly towards each impact (≥ 5%). Contribution analysis results are provided in the Supplementary Material.

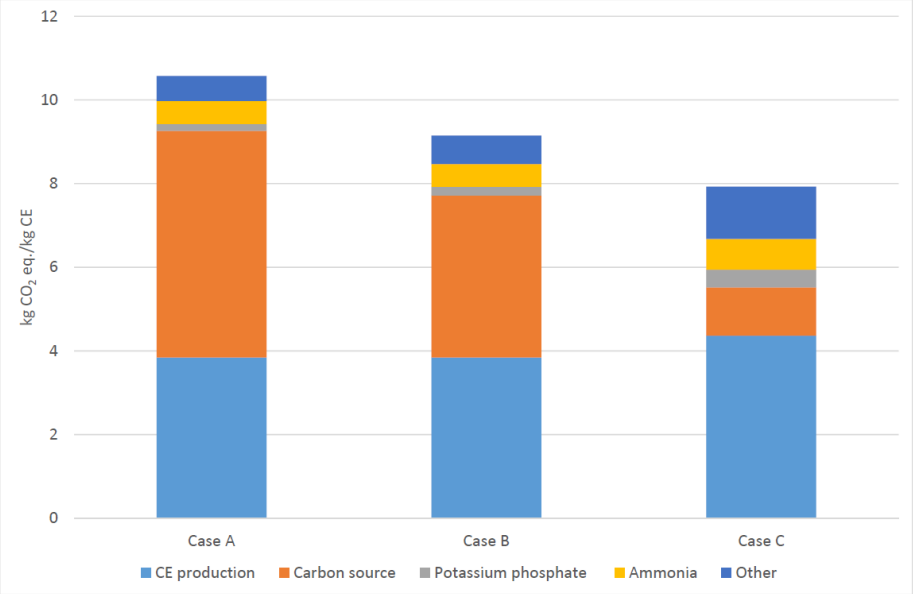

Fig. 3 Global warming potential 100 year time horizon

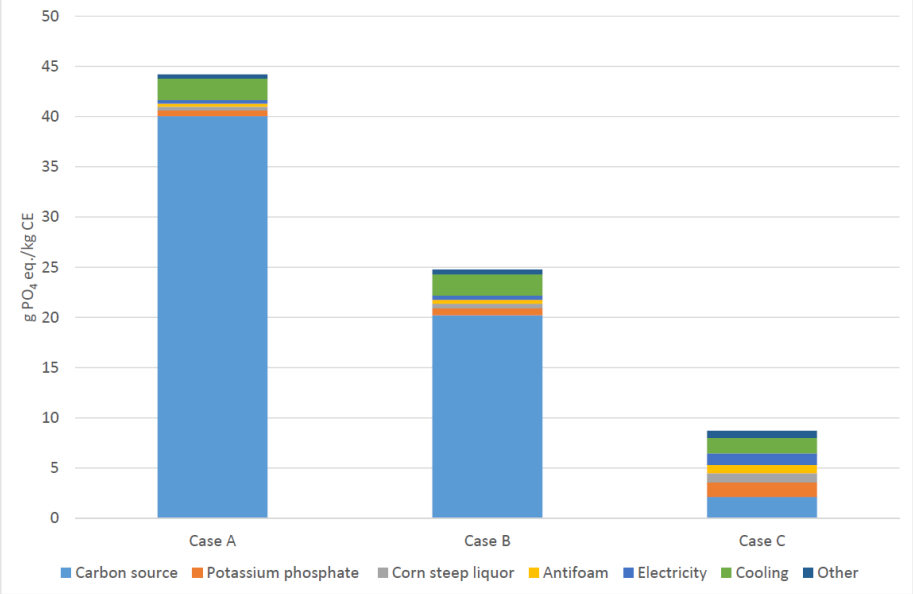

Fig. 4 Eutrophication potential

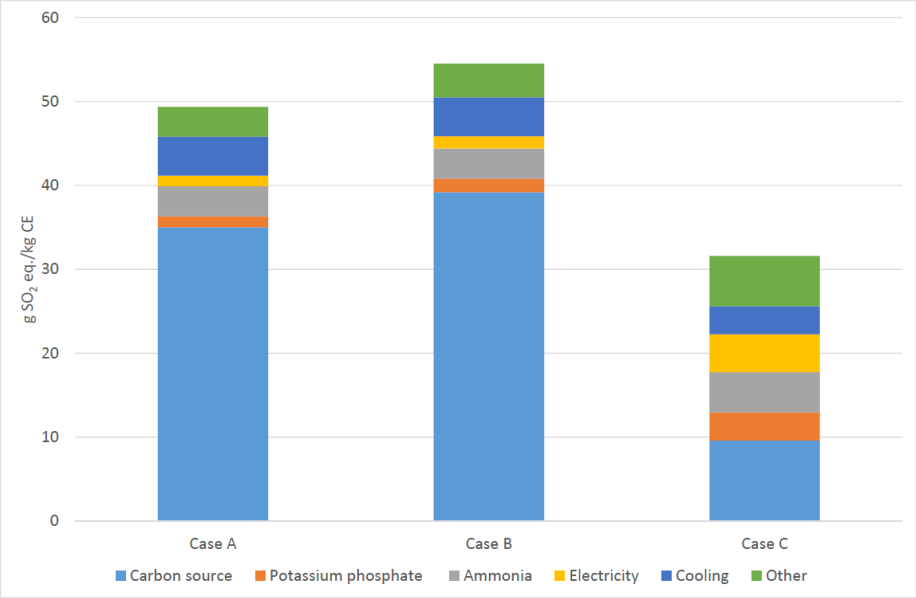

Fig. 5 Acidification potential

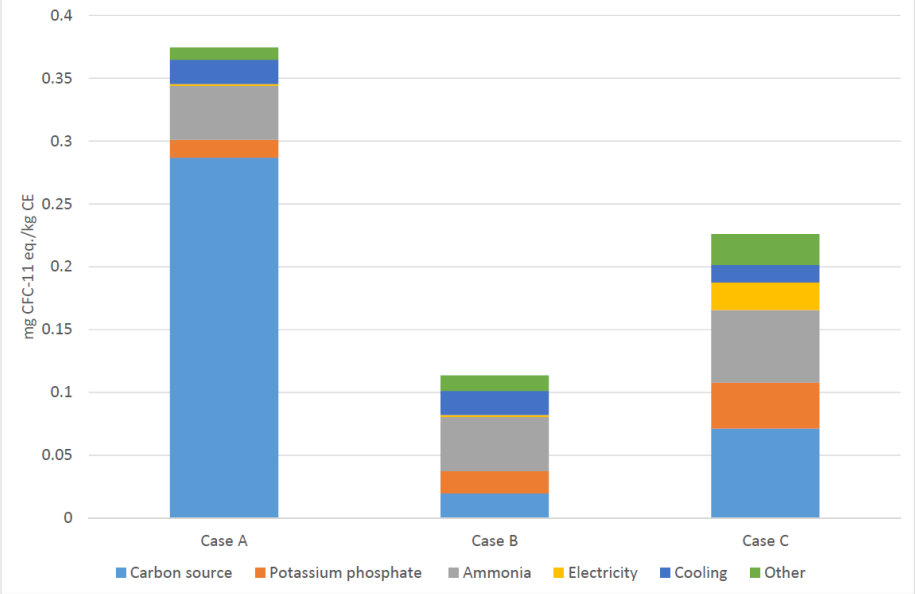

Fig. 6 Ozone layer depletion potential

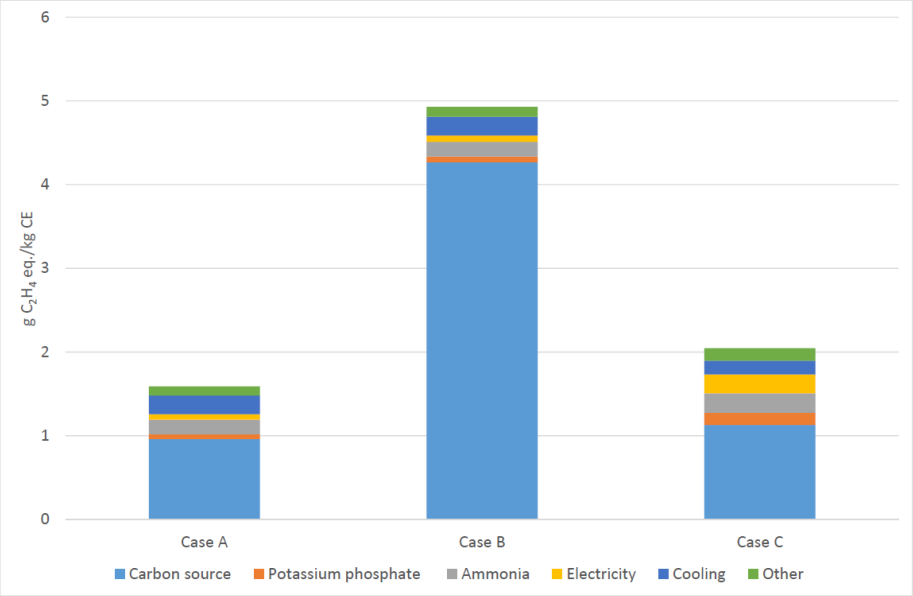

Fig. 7 Photochemical oxidation potential

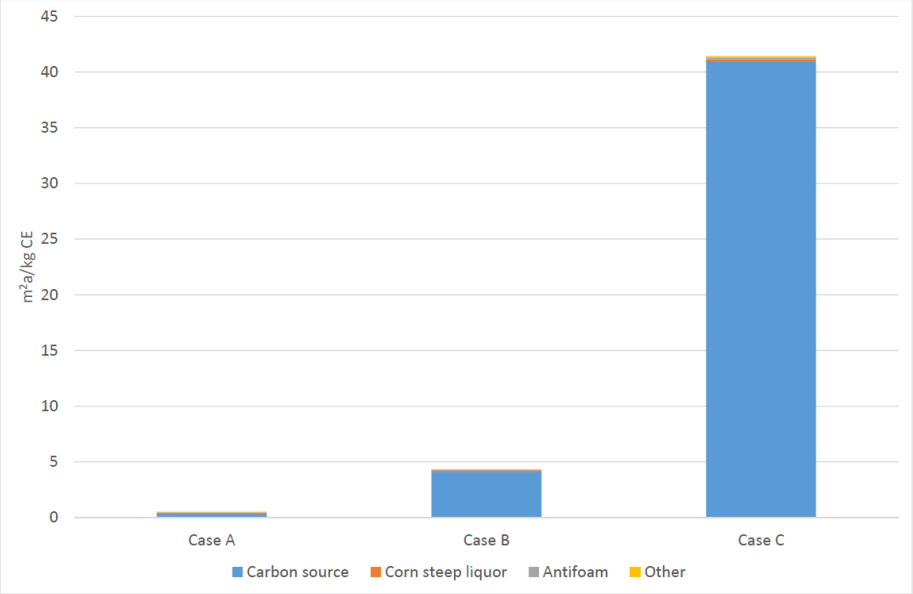

Fig. 8 Land use

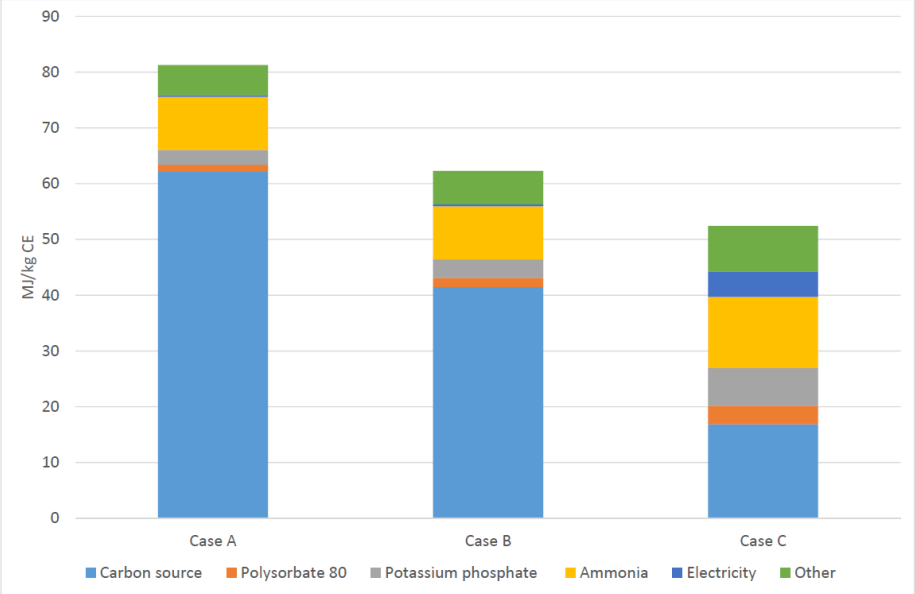

Fig. 9 Cumulative energy demand

Based on the contribution analyses presented in Figs. 3 through 9 and Supplementary Material, it can be observed that the carbon source and electricity are the most significant contributors to all impact categories with the exception of GWP, where CE production-process emissions are as well significant (36%–55%). CE production-process emissions (CO_2) have been determined stoichiometrically based on the assumed molar selectivity of the carbon source, see Table 2; therefore, CE production-process emissions are directly correlated to the assumed molar selectivity and carbon source.

Furthermore, for the impact category land use (Fig. 8) the provision of carbon sources is the largest contributor. Differences in land use impacts are the result of differences in carbon source yield per m^2a. Considering that, corn and sugar cane are annual high-yield crops, and softwood is a slower-growing lower-yielding crop, softwood will occupy the same area for several years to produce the same amount of carbon source, resulting in higher land use values per kilogram carbon source.

A sensitivity analysis is performed for the two significant sub-processes, carbon source and electricity, observing changes in the arbitrarily selected impact categories: GWP and cumulative energy demand. Hereafter we refer to the previously determined results for Cases A, B and C as base cases.

In the base-case scenarios, we have assumed a carbon source molar selectivity of 31% for CE production for Cases A and B (Davis et al. 2015), and 29% for CE for Case C (Wooley et al. 1999). Both Davis et al. (2015) and Wooley et al. (1999) have assumed these selectivities for future n^{th} plant performance based on Meerman et al. (2004) and Hamilton (1998) respectively. Similar to Hong et al. (2013), in the sensitivity analysis, we reduce selectivity for CE production arbitrarily by 5% and 10%; with resulting increases in selectivity for cell-mass production by 5% and 10%, respectively. These changes in molar selectivity for CE production can be interpreted as a reduction from Davis et al.'s (2015) and Wooley et al.'s (1999) n^{th} plant assumptions. The results of this first sensitivity analysis are presented in Fig. 10 and Fig. 11. From these figures, two observations can be made: first, GWP and cumulative energy demand are sensitive to changes in process efficiency, and second, that Case C is least sensitive to changes in carbon source molar selectivity. It should be noted that whereas Humbird et al. (2011) (Case A) and Wooley et al. (1999) (Case C) based their molar selectivity and yields on laboratory results, this study has assumed the same molar selectivity of Case A for Case B based on studies by Olofsson et al. (2015), He et al. (2014) and Barta et al. (2010). From the laboratory results of He et al. (2014) one can determine a molar selectivity of approximately 6% for CE. Therefore, a cautious interpretation of results would choose the 10% reduction for Case B as more appropriate.

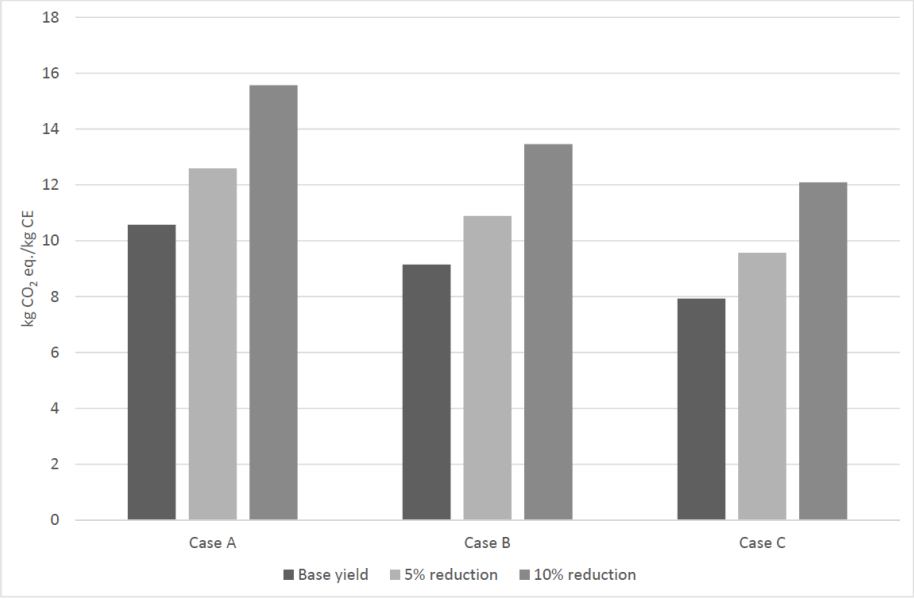

Fig. 10 Sensitivity analysis of GWP results to changes in carbon selectivity (yield)

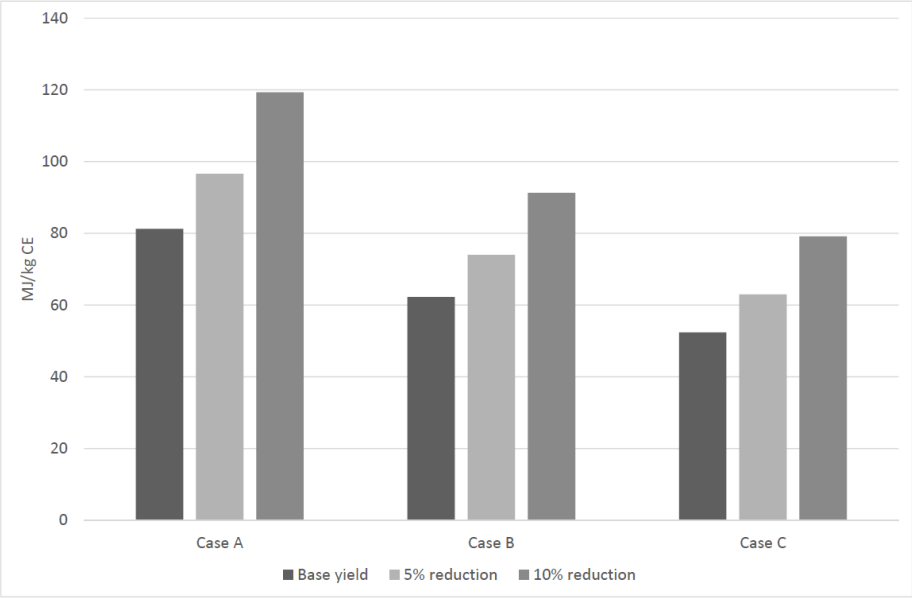

Fig. 11 Sensitivity analysis of cumulative energy demand results to changes in carbon selectivity (yield)

Additional assumptions were made with regard to carbon sources in the base-case LCI models. For glucose, we have constructed a LCI for glucose production. This LCI results in a GWP value of 1.34 kg CO_2 eq./kg glucose DM, which is slightly higher than the 0.95 kg CO_2 eq./kg glucose DM determined by An et al. (2012), and within the range of 0.7 ± 0.2–1.1 ± 0.2 kg CO_2 eq./kg glucose DM determined by Tsiropoulos et al. (2013). Disregarding differences in LCIA, these differences can be attributed to impacts associated with cornstarch production and variations in LCI modelling approaches as explained by both Tsiropoulos et al. (2013) and An et al. (2012). Würdinger et al. (2003) and van Zeist et al. (2012) provide cornstarch LCIs which result in GWP values ranging from 1.41 kg CO_2 eq./kg cornstarch DM (global, economic allocation) to 0.53 kg CO_2 eq./kg cornstarch DM (US, energy allocation) respectively. In this study we have applied a cornstarch LCI resulting in GWP of 1.29 kg CO_2 eq./kg cornstarch DM (German, economic allocation). The application of Würdinger et al.'s (2003) and van Zeist et al.'s (2012) cornstarch LCIs result in GWP values of 1.43 kg CO_2 eq./kg glucose DM and 0.92 kg CO_2 eq./kg glucose DM respectively. Furthermore, Fig. 12 depicts the sensitivity analysis results from substituting into the complete LCI for Case A these two alternative cornstarch LCIs. Fig. 12 indicates that GWP results for Case A are sensitive to changes in cornstarch feedstock, geography of origin, and applied allocation.

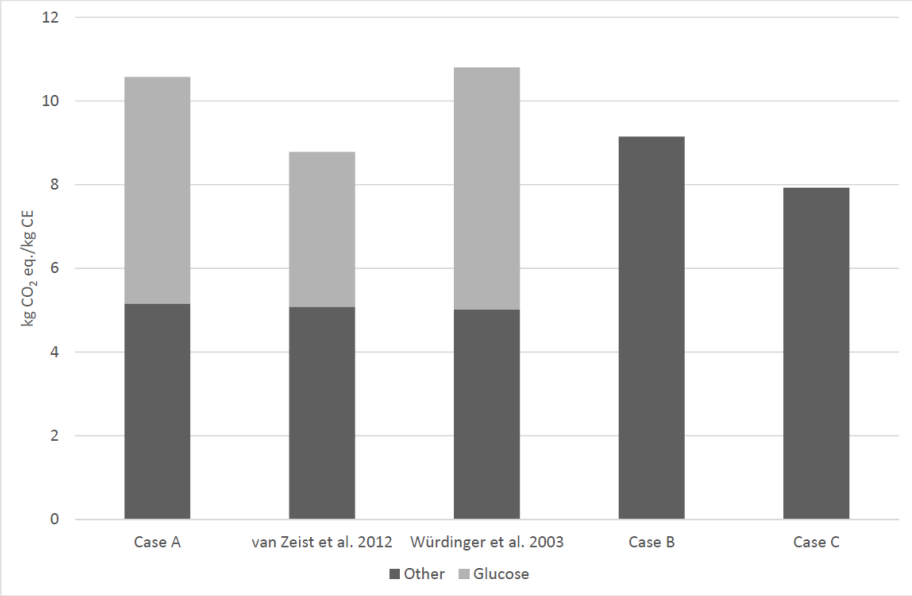

Fig. 12 Sensitivity analysis of total GWP results for Case A under various GWP values for glucose production

For molasses we have based our LCI model on van Zeist et al. (2012) for sugar cane molasses, assuming this LCI is suitably representative. This LCI when applying energetic allocation results in a GWP of 0.76 kg CO_2 eq./kg molasses DM. LCAs of molasses production are scarce due to the dominance of integrated sugar and molasses-ethanol production (Gopal and Kammen 2009), and the resulting lack of sufficiently disaggregated LCIs. Van Zeist et al. (2012) provides values ranging from 0.16 kg CO_2 eq./kg DM for sugar beet molasses (Netherlands, economic allocation) to 1.43 kg CO_2 eq./kg DM for sugar cane molasses (Brazil, mass allocation). This equates to a 79% decrease and a 87% increase in GWP respectively, relative to the base case scenario. As a sensitivity analysis we consider both a 80% decrease and 80% increase in molasses GWP, or 0.15 kg CO_2 eq./kg molasses DM and 1.37 kg CO_2 eq./kg molasses DM respectively. Fig. 13 presents the results of this sensitivity analysis and indicates that GWP results for CE production based on molasses are sensitive to molasses'- feedstock, geography of origin and applied allocation. In comparison, Olofsson et al. (2015) cite a GWP of 0.14 kg CO_2 eq./kg molasses, which upon review is assumed to be for sugar beet molasses. Thus, the 80% decrease column in Fig. 13 is more representative of CE production based on sugar beet molasses, whereas this study's results are representative for CE production based on sugar cane molasses.

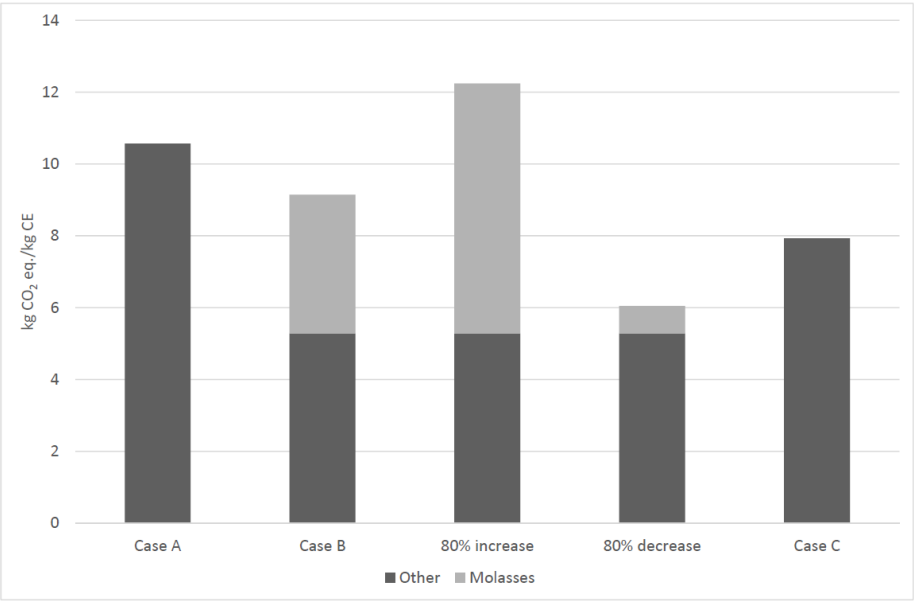

Fig. 13 Sensitivity analysis of total GWP results for Case B under various GWP values for molasses production

For pre-treated softwood, we have assumed softwood cellulose and hemicellulose pre-treatment hydrolysis reactions (conversion) based on Davis et al. (2015), see Supplementary Material. Wooley et al. (1999) provide another set of pre-treatment reactions, see Supplementary Material. As a sensitivity analysis we consider the effect of applying Wooley et al.'s (1999) set of hydrolysis reactions to our pre-treatment process; Fig. 14 presents these results. The reduction in GWP when using Wooley et al.'s (1999) hydrolysis reactions is directly related to their assumption that less cellulose is converted to soluble sugars in pre-treatment, and as a result, more cellulose is available for CE production in succeeding stages. However, the differences in conversion efficiency are only slight, and their effects on GWP results for Case C are less significant.

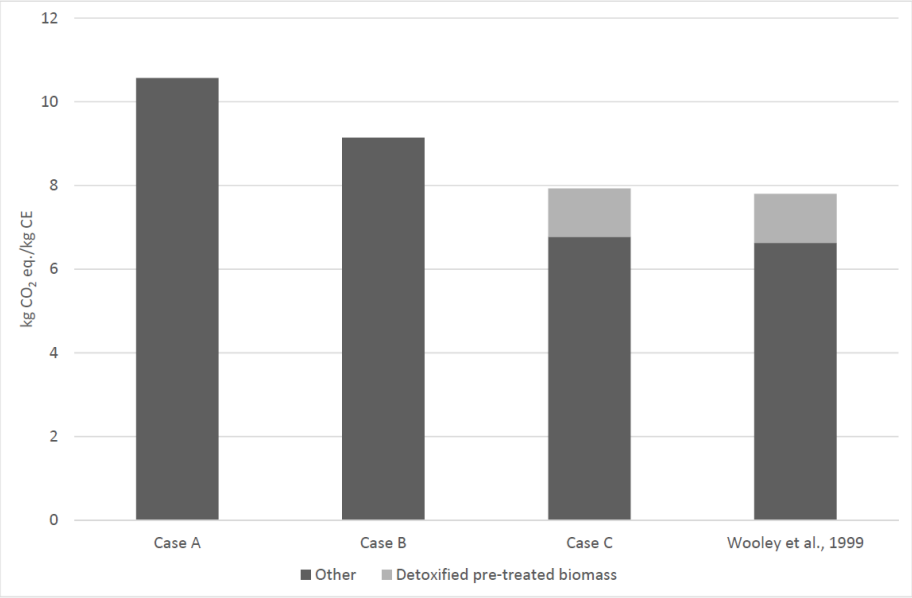

Fig. 14 Sensitivity analysis of total GWP results for Case C under various hydrolysis reaction values

Considering electricity, in the base-case scenarios electricity was modelled on biogas CHP generation (Humbird et al. 2011; Wooley et al. 1999), see Supplementary Material. In the sensitivity analysis we have substituted this method for softwood chip boiler CHP generation and the EU 27 consumption mix, see Fig. 15 and Fig. 16. These figures indicate that both GWP and cumulative energy demand results are sensitive to direct electricity supply. Particularly, the substitution of biogas CHP for woodchip CHP has little effect on GWP and cumulative energy demand results for all cases. However, the substitution of biogas CHP for the EU 27 consumption mix significantly increases impacts for Case C, though less so for Cases A and B. The sensitivity to changes in electricity supply is explained by the variation in direct electricity consumption between Cases A, B and C in increasing order, see Table 4, making Case C more sensitive to changes in direct electricity supply.

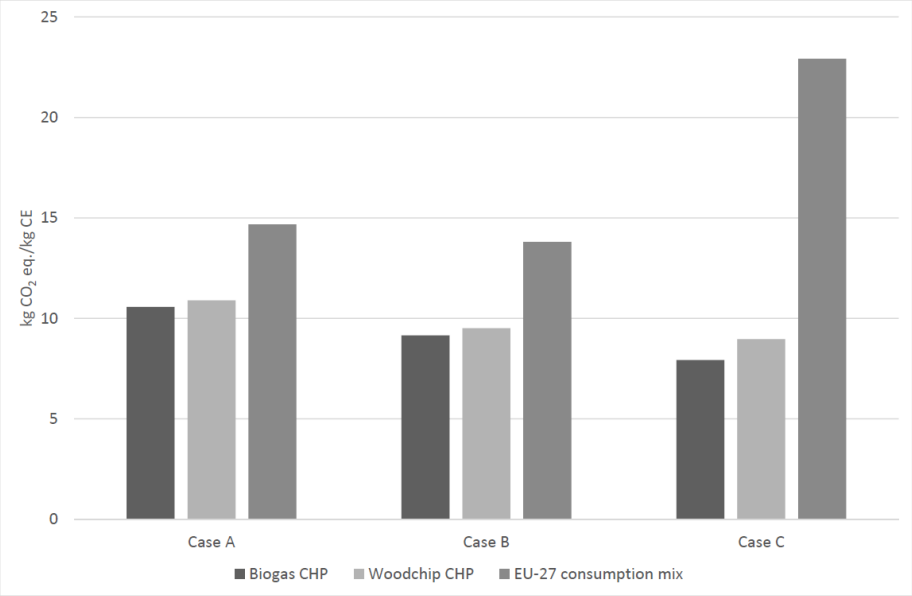

Fig. 15 Sensitivity analysis of total GWP results to changes in electricity generation

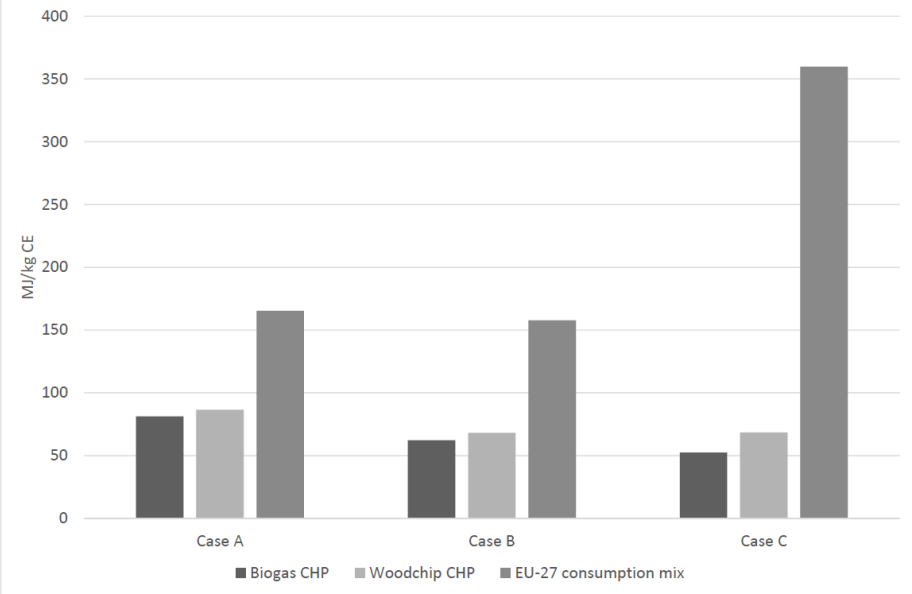

Fig. 16 Sensitivity analysis of total cumulative energy demand results to changes in electricity generation

As a final sensitivity analysis, as an estimate of the effect of market changes on carbon source GWP results, historical GCMF for cornstarch glucose (Case A), sugar cane molasses (Case B) and pre-treated softwood (Case C) are estimated and these GCMF are then multiplied with the reactive carbon source flows, see Table 3, for each Case A, B and C.

The starch and sugar cane markets are large and diverse and in 2013 more than 2 billion metric tonnes were produced (Geohive 2015a). Cornstarch glucose and sugar cane molasses can be derived from starches and sugar canes, respectively, and pre-treated softwood from wood chips.

For cornstarch glucose the GCMF are obtained from the global corn production (Geohive 2015b) and the share of corn sweetener of Unites States corn production (USDA 2015). Globally in 2000 and 2015, 592 million metric tonnes (MMT) (Geohive 2015b) and 972 MMT (WOC 2015) of corn were produced respectively, and the share used to make corn sweeteners were 7.69% and 5.5%, respectively, i.e. 46 MMT and 53 MMT respectively.

For sugar cane molasses the GCMF are obtained from the global sugar cane production (Geohive 2015a) and the yield of molasses per tonne of sugar cane which is approximately three per cent (MM, 2015). In 2000 and 2015, 1,256 MMT and 2,005 MMT of sugar canes were produced respectively (Geohive 2015a,b), i.e. 38 MMT and 60 MMT of molasses cane syrup respectively.

For pre-treated softwood the GCMF are obtained from the annual wood chip production which is used as a starting point for estimating softwood based hydrolysate. The global annual wood chip production increased from around 30 MMT to 60 MMT between 2000 and 2015 respectively (Ekstrom 2011), and the theoretical amount of hydrolysate that can be extracted from woodchips is 1%–12% DM content (Gladyshko 2011). Extraction of hemicelluloses by acid catalyzed hydrolysis is not equal to the actual production of wood hydrolysate for glucose applications as a large share of hydrolysate is used for biofuels and renewable products (Dahlman et al. 2014). Still if an average 6% hydrolysate from wood chips is used and the pre-treated softwood production thereby increased from 1.8 MMT to 3.6 MMT between 2000 and 2015 respectively.

Alternatively the world corn sweetener production (Geohive 2015b; USDA 2015; WOC 2015) and the market share of hydrolysates related to the sweeteners glucose syrup, modified starch, maltodextrin, and cyclodextrin (MM 2015) can be used. In 2000 and 2015, the market shares of hydrolysates are estimated to 7.2% and 11.4% respectively (MM 2015), i.e. 4 MMT and 7 MMT of hydrolysates respectively.

The values used for obtaining- and the GCMF are summarized in Tables 8 and 9, and the results are presented in Fig. 17. The results indicate that Case A, or more specifically GWP associated with glucose production is highly sensitive to market changes. The reasons are the slow market growth of cornstarch glucose leading to a relatively low GCMF, implying a relatively low carbon source allocation in Table 4 in combination with the relatively high share of the

carbon source of the total GWP score (Fig. 3). Pre-treated softwood also obtains a low carbon source allocation due to a low share of the applied GCMF mix. However, the share of the carbon source of the total GWP score for pre-treated softwood is relatively small (Fig. 3), and therefore the effect is less significant as shown in Fig. 17. The sugar cane molasses market grows quicker than those of cornstarch glucose and pre-treated softwood and as such is allocated a relatively high GCMF (Table 9). The share of the carbon source of the total GWP score for sugar cane molasses is relatively high (Fig. 3) leading to a similar GWP score for ALCA and AALCA (Fig. 17).

Table 8 Gross generation of carbon sources for the period 2000–2015

Source of glucose for CE production	**Gross generation in 2000 (Mtonnes)**	**Gross generation in 2015 (Mtonnes)**
Cornstarch glucose (Case A)	46	53
Sugar cane molasses (Case B)	38	60
Pre-treated softwood (Case C)	2	4
Total	85	117

Table 9 Determination of GCMFs for Cases A, B and C

Source of glucose for CE production	**Consequential historical (mass)**	**GCMF**
	Change 2000–2015	Applied mix
Cornstarch glucose (Case A)	8	0.25
Sugar cane molasses (Case B)	22	0.69
Pre-treated softwood (Case C)	2	0.06
Total	32	1.00

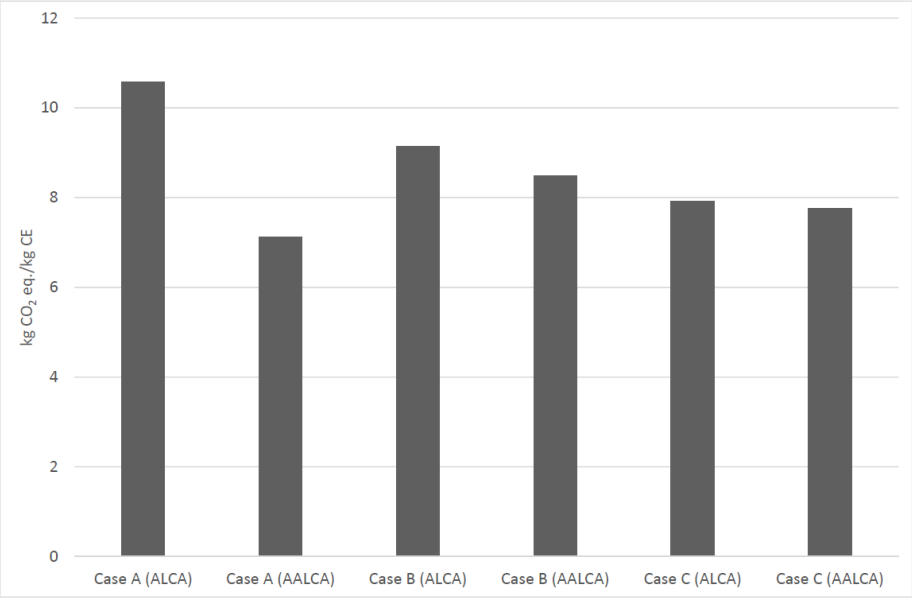

Fig. 17 Sensitivity analysis of total GWP results for Cases A, B and C reflecting market changes

The results of the sensitivity analysis indicate that Case A is most sensitive to assumptions made concerning glucose'- feedstock, geography of origin and applied allocation and market changes. Whereas, Case C is most sensitive to assumptions made concerning molar selectivity and direct electricity supply. Case B indicates the least sensitivity to those assumptions tested in the sensitivity analysis.

4 Discussion

A semi-quantitative comparison of the results of this study with the results of similar non-formulated enzyme assessments, see Table 7, reveals that this study's results are within the determined range (2σ) of Hong et al. (2013) and MacLean and Spatari (2009). Furthermore, when one considers that Hong et al. (2013) found that formulated enzymes can have impacts (GWP) 57% higher than non-formulated enzymes, the results of this study when compared with those for formulated enzymes seem acceptable. However, the results of this study do not share similarities with Agostinho et al.'s (2014) cummulative energy demand- or Harding's (2008) GWP results, see Table 7. Agostinho et al.'s (2014) high value of 1664 MJ/kg CE can be attributed to the use of paper pulp as a carbon source which accounts for 77% of cummulative energy demand. Harding's (2008) low (negative) GWP impacts are the result of the LCI modelling approach he has taken where CO_2 uptake from agricultural inputs during

cultivation exceeds CO_2 releases during cellulase production. Remaining minor differences can be attributed to variations in the background LCIs chosen, and variations in LCI modelling.

Considering the CE consumption values presented in the introduction, and the results of this study, one can estimate the environmental impact associated with CE production per MJ LCB based on this study, see Supplementary Material. The GWP range for all cases is 2 g–22 g CO_2 eq. CE/MJ LCB for low CE consumption Case C to high CE consumption Case A respectively. These results are in the similar to Hong et al. (2013) (12 g CO_2 eq. CE/MJ LCB) and MacLean and Spatari (2009) (3.3 g–3.6 g CO_2 eq. CE/MJ LCB) for non-formulated enzymes, and the higher range of Olofsson et al. (2015) (18 g–30 g CO_2 eq. CE/MJ LCB) and Dunn et al. (2012) (4.6 g CO_2 eq. CE/MJ LCB) for formulated enzymes.

When one compares these values with the total GWP of forest-to-gate LCB production determined by other authors (9.3 g–50.3 g CO_2 eq./MJ LCB: Bright and Strømman (2009), Dunn et al. (2012), and Olofsson et al. (2015), to cite several), and with a recent review of 53 studies by Morales et al. (2015) revealing values ranging from 1.6 g to 123.4 g CO_2 eq./MJ LCB, one can conclude that CE production is significant with respect to the total environmental impacts of LCB production.

Alarmingly, Borrion et al. (2012), who reviewed 53 LCAs of LCB, found that in many instances it is not clear as to whether CE production is within the evaluated system boundary, and Wiloso et al. (2012), who reviewed 22 LCAs of LCB, revealed that only 15 studies incorporated enzyme production in their inventory analyses, several of which have been cited in this article. Morales et al. (2015), Borrion et al. (2012), Wiloso et al. (2012), Singh et al. (2010) and Luo et al. (2009) all point to the existing data gaps concerning CE production and the necessity to include this process in LCAs of LCB in order to represent the true environmental impacts associated with LCB production.

Finally, future research, development and implementation will lead to a better understanding of the best-suited pre-treatments (Kumar et al. 2009), enzyme strains (Seiboth et al. 2011) and location of CE production (Olofsson et al. 2015), in addition to compiling LCIs suitable for assessing additional LCIA categories, for example, water consumption (An et al. 2012).

5 Conclusion

The results of this study show that CE production based on pre-treated softwood as a carbon source provides lower environmental impacts than does CE production based on cornstarch glucose and sugar cane molasses carbon sources. However, to varying degree, results are sensitive to assumptions made in this study.

From the results we conclude that other studies evaluating the environmental impacts of CE production neither over- nor under estimate CE production's environmental impact. Instead, based on sensitivity analysis and review of these sources, we conclude that particular attention should be paid to correct selection of background LCIs, particularly carbon source and electricity, and even nutrient requirements. Additionally, consistent LCI modelling methodology should be applied and transparently described.

Furthermore, we observe that based on estimates for CE's contribution towards the GWP impacts of LCB production, CE production's exclusion from LCB assessments can lead to significant deviations from the true impacts associated with LCB production.

As CE production increases because of the foreseen increase in LCB production, experience will lead to advances in understanding and technology, advances which could prove either favourable or detrimental to assessing the environmental impacts associated with CE production. It is, however, of the utmost importance to find (the most) sustainable processes and avoid competition for finite resources, in line with the philosophy of industrial ecology. This study has filled some of the data gaps associated with the production of LCB in general and has specifically provided a clear and transparent indication of the resource use and environmental impacts of on-site SmF CE in full-broth production. Future research could include defining a pure consequential LCA looking at the consequences of changing cornstarch glucose, sugar cane molasses and pre-treated softwood demands in CE production to substantiate or disregard the findings of AALCA sensitivity analysis.

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Supplementary Material:

Comparative attributional life cycle assessment of European cellulase enzyme production for use in second-generation lignocellulosic bioethanol production

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The supplementary material contains calculations, life cycle inventories, tables and results.

Calculation of CE consumption ranges have been performed (when necessary) by dividing enzyme dosage by ethanol yield, both relative to feedstock mass. When necessary a lower heating value of 26.95 MJ/kg ethanol and density of 0.789 kg/l ethanol are used.

Supplementary Material Table 1 Calculation of CE consumption ranges for non-formulated enzymes

	Note:	**Enzyme dosage**		**Enzyme consumption**	
		Value	**Unit**	**Value**	**Unit**
(Hong et al. 2013)	Low	3.0	kg enzyme/t carbohydrate	0.3 (low)	g enzyme/MJ LCB
	Base	11.5	kg enzyme/t carbohydrate	1.2	g enzyme/MJ LCB
	High	20.0	kg enzyme/t carbohydrate	2.1 (high)	g enzyme/MJ LCB
(Humbird et al. 2011)	Base	20.0	kg enzyme/t cellulose	1.1	g enzyme/MJ LCB
(MacLean and Spatari 2009)	DA-SSCF production	9.2	kg enzyme/t feedstock	1.6	g enzyme/MJ LCB
	AFEX-SSCF production	9,6	kg enzyme/t feedstock	1.5	g enzyme/MJ LCB

Nutrients requirements are dosed based on Schell et al. (1991) and based on weight and mass flow adapted from He et al. (2014), Humbird et al. (2011) and Wooley et al. (1999).

Supplementary Material Table 2 Nutrient requirements for CE production

Nutrient	Amount	Unit
Corn steep liquor	1	% w/w
Corn oil	0.1	% v/v
Ammonium sulphate	1.4	g/l
Potassium phosphate	2.0	g/l
Magnesium sulphate	0.3	g/l
Calsium chloride	0.4	g/l
Polysorbate 80	0.2	g/l

The literature did not provide a suitable LCI for the production of potassium phosphate, as a result we have constructed an LCI for potassium phosphate production based on Freilich and Petersen (2005) presented in Supplementary Material Table 3.

Supplementary Material Table 3 LCI for the production of 1 kg potassium phosphate

Reactants				**Products**		
Phosphoric acid	+	Potassium hydroxide	→	Potassium phosphate	+	Water
Molecular weight						
98.00 g/mol		56.11 g/mol		174.18 g/mol		18.02 g/mol
Stoichiometric formula						
1 H_3PO_4	+	2 KOH	→	1 K_2HPO_4	+	2 H_2O
Mass balance (LCI)						
0.56 kg		1.29 kg	→	1 kg	+	0.41 kg

Supplementary Material Table 4 presents the DM composition of sugar cane derived molasses provided by Olbrich (2006).

Supplementary Material Table 4 DM composition of sugar cane-derived molasses

Component	Value	Unit
Sucrose	40.0	%
Glucose	17.5	%
Fructose	20.0	%
Organic non-sugars constituents	12.5	%
Inorganic constituents	10.0	%
Total	100.0	%

Softwood composition entering pre-treatment are based on Ferraro et al. (1999), and presented in Supplementary Material Table 5.

Supplementary Material Table 5 Softwood composition given as a percent of DM

Component		**Value**	**Unit**
Cellulose	Glucan	39.0	%
Hemicellulose	Glucan	4.3	%
	Xylan	7.4	%
	Galactan	2.8	%
	Arabinan	1.5	%
	Mannan	10.2	%
Lignin		28.6	%
Acetate		0.3	%
Ash		0.9	%
Extractives		5.0	%
Total		**100.0**	**%**

Towards determining the reactive carbon source, we have applied hydrolysis reactions from Davis et al. (2015), and in the sensitivity analysis Wooley et al. (1999), see Supplementary Material Table 6, based on softwood composition provided by Ferraro et al. (1999) see Supplementary Material Table 5.

Supplementary Material Table 6 Pre-treatment hydrolysis reactions and their assumed efficiencies

Reaction	**Reaction Efficiency**	
	Davis et al. 2015	Wooley et al. 1999
Arabinan + Water → Arabinose or $(C_5H_8O_4)n + nH_2O \rightarrow nC_6H_{10}O_5$	0.900	0.750
Glucan* + Water → Glucose or $(C_6H_{10}O_5)n + nH_2O \rightarrow nC_6H_{12}O_6$	0.099	0.065
Galactan + Water → Galactose or $(C_6H_{10}O_5)n + nH_2O \rightarrow nC_6H_{12}O_6$	0.900	0.750
Mannan + Water → Mannose or $(C_6H_{10}O_5)n + nH_2O \rightarrow nC_6H_{12}O_6$	0.900	0.750
Xylan + Water → Xylose or $(C_5H_8O_4)n + n H_2O \rightarrow nC_5H_{10}O_5$	0.900	0.750

Supplementary Material Table 7 LCI for the production of 1 kg DM softwood chips

Materials/fuels	**Value**	**Unit**	**Data source**
Softwood chips	1.00	Kg	Ecoinvent 3
Transport	0.02	tkm	Ecoinvent 3

Supplementary Material Table 8 LCI for the production of 1 kg steam produced from biogas combustion

Materials/fuels	**Value**	**Unit**	**Data source**
Water	1.17	Kg	Ecoinvent 3
Electricity/heat			
Heat	3.07	MJ	Ecoinvent 3

Supplementary Material Table 8 presents all LCIA results for all impact categories and cases including the results of the contribution analysis.

Supplementary Material Table 8 LCIA results for all impact categories including contribution analysis (≥ 5 %)

Impact category (unit)	**Case**	**Contribution**										
		CE production	Carbon source	Potassium phosphate	Ammonia	Corn steep liquor	Antifoam	Electricity	Cooling	Polysorbate 80	Other	Total
GWP (kg CO_2 eq.)	A	3.8	5.4	0.2	0.6						0.6	10.6
	B	3.8	3.9	0.2	0.6						0.7	9.1
	C	4.4	1.2	0.4	0.7						1.3	7.9
EP (g PO_4 eq.)	A		40.1	0.6		0.4	0.3	0.4	2.12		0.4	44.2
	B		20.2	0.7		0.4	0.4	0.4	2.12		0.5	24.8
	C		2.1	1.5		0.9	0.8	1.2	1.53		0.7	8.7
AP (g SO_2 eq.)	A		35.0	1.3	3.6			1.3	4.64		3.5	49.3
	B		39.2	1.6	3.6			1.5	4.64		4.0	54.5
	C		9.6	3.4	4.8			4.5	3.36		6.0	31.6
ODP (mg CFC-11 eq.)	A		0.29	0.01	0.04			0.00	0.02		0.01	0.37
	B		0.02	0.02	0.04			0.00	0.02		0.01	0.11
	C		0.07	0.04	0.06			0.02	0.01		0.02	0.23
POP (g C_2H_4 eq.)	A			0.06	0.17			0.07	0.22		0.11	0.63
	B			0.07	0.17			0.08	0.22		0.12	0.66
	C			0.14	0.23			0.23	0.16		0.15	0.92
LU (m2a)	A		0.3			0.1	0.1				0.1	0.5
	B		4.1			0.1	0.1				0.1	4.4
	C		40.9			0.2	0.2				0.1	41.4
CED (MJ)	A	62.1		2.6	9.5			0.3		1.3	5.4	81.3
	B	41.5		3.3	9.5			0.4		1.6	6.0	62.3
	C	16.9		6.8	12.8			4.5		3.3	8.2	52.4

In Supplementary Material Table 9 we present enzyme GWP contribution results determined per MJ LCB based on enzyme consumption ranges presented in Supplementary Material Table 1, which are based on Hong et al. (2013), Humbird et al. (2003) and MacLean and Spatari (2009)

Supplementary Material Table 9 enzyme GWP contribution per per MJ LCB

	GWP		**Enzyme consumption**		**GWP**	
	Value	**Unit**	**Value**	**Unit**	**Value**	**Unit**
Case A	10.6	kg CO_2 eq./kg CE	0.3	g enzyme/MJ LCB	3.2	g CO_2 eq. CE/MJ LCB
	10.6	kg CO_2 eq./kg CE	2.1	g enzyme/MJ LCB	22.3 (high)	g CO_2 eq. CE/MJ LCB
Case B	9.1	kg CO_2 eq./kg CE	0.3	g enzyme/MJ LCB	2.7	g CO_2 eq. CE/MJ LCB
	9.1	kg CO_2 eq./kg CE	2.1	g enzyme/MJ LCB	19.1	g CO_2 eq. CE/MJ LCB
Case C	7.9	kg CO_2 eq./kg CE	0.3	g enzyme/MJ LCB	2.4 (low)	g CO_2 eq. CE/MJ LCB
	7.9	kg CO_2 eq./kg CE	2.1	g enzyme/MJ LCB	16.6	g CO_2 eq. CE/MJ LCB

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Article IV

Gilpin G, Schipper J (planned submission) A cradle-to-gate attributional life cycle assessment of integrated aquaculture in Norway for sustainable food and fuel production Aquaculture Reports

A cradle-to-gate life cycle assessment of integrated aquaculture in Norway for sustainable food and fuel production.

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Abstract

With 70% of the earths surface covered by oceans, focus has shifted towards this new frontier for the provision of food and fuel. The integration of salmon (S. *Salar*) and macro-algae (S. *latissima*) combined with a biorefinery offer one solution towards the provision of both food and fuel (ethanol), along with other co-products: sodium alginate, compost and energy. Based on new empirical data for the cultivation and harvesting of macro-algae, along with a novel biorefinery configuration, we perform a life-cycle assessment (LCA) of this integrated system.

We determine that the integration of salmon and macro-algae sequesters 0.0923 kg CO_2, 2.450 g nitrogen and 0.310 g phosphorus per kg wet-weight (WW) macro-algae; which leads to net reductions in global warming potential (GWP) and eutrophication potential (EP) of -0.055 kg CO_2 eq. And -1.892 g PO_4 eq. per kg WW macro-algae. Additional reductions are possible by increasing macro-algae yield and extending the grow-out infrastructure life-span. Concerning the biorefinery products the production of one high-value low-yield product (sodium alginate) dominates the life-cycle impact assessment (LCIA) results of all biorefinery products; with chemical consumption contributing to 57% of GWP for biorefinery co-products. We conclude that much research is necessary towards developing biorefinery concepts aimed at reducing material (chemical) consumption, along with investigating alternatives for producing biofuel and fish feed additives from macro-algae.

Highlights

- The integration of salmon and macro-algae aquaculture reduces the overall impacts of GWP and EP.
- Increasing macro-algae yield, and extending grow-out infrastructure life-span can further lower these impacts.

- The production of one high-value low-yield product in a biorefinery influences LCIA results of the complete biorefinery system.

Keywords

Integrated aquaculture, IA, LCA, alginate, ethanol

1. Introduction

Two of the major challenges in modern society are; climate change and providing sustenance to an increasing world population. With approximately 70% of the world's surface area covered by oceans, focus has shifted towards this new frontier for the provision of food and fuel. Aquaculture is the aquatic- marine- or freshwater farming of plants or animals. Aquaculture's most common application is the production of fish, and to a lesser degree seaweed. In 2012 worldwide aquaculture production of fish and aquatic algae (mostly seaweed) was 66.6 million metric tonnes (MMT) and 23.8 MMT respectively, with a value of US$ 137.7 billion (US) and US$ 6.4 billion (US) respectively (FAO, 2014). Additionally, aquaculture production has seen annual growth rates of 8.6% between 1980 and 2012 (FAO, 2014). The predominant uses of seaweed are: food, the production of hydrocolloids (agar, alginate and carrageenan), animal feed, fertilizers and cosmetics (McHugh, 2003).

In 2013, Norwegian aquaculture produced 1.25 million tonnes of fish, generating approximately US$ 4.7 billion (US) (1 USD ≈ 8.5 NOK), and employing 5700 people (SSB, 2015). Although, aquatic plants aquaculture is in its infancy in Norway, approximately 170 000 tonnes of wild seaweed is harvested annually, mainly for the production of alginate, of which Norway is one of the world's leading producers (Valmot, 2012). Despite these promising figures, fish aquaculture has been met with scepticism both in Norway and abroad due to numerous unresolved environmental issues (NMFCA, 2011) and social issues (Grigorakis, 2010).

A similar situation exists for biofuels; where the international adoption of biofuels has developed rapidly over the past decades and stands for approximately 2% of total road transport fuel worldwide, or 100 billion (US) litres (IEA, 2011). Biofuel production and consumption in Norway is low compared to fossil fuel despite national ambitions for 5% inclusion of biofuels in transport fuels (NME, 2010). Biofuels limited success in Norway has several reasons, though most important is a general reluctance to adopt alternative fuels with unresolved

environmental, economic, and social sustainability issues (FAO, 2011). However, careful selection of biomass feedstock and conversion technology can potentially resolve some of these challenges (Tilman et al., 2009).

Common to both aquaculture- and biofuel development several of these sustainability challenges will have to be resolved prior to their wide-spread implementation; stressing the necessity that a symbiosis be found between human and hydrosphere development. A so called integrated aquaculture (IA) system with combined biorefinery is one such idea, and can potentially provide both sustainable food and biofuel.

IA entails the integration of independent aquatic species, occupying different positions along an aquatic food chain into one aquaculture system. This form of aquaculture proposes to mirror the natural ecosystem in which the biomass, and/or the waste of higher trophic level organisms, become inputs for lower trophic level organisms, ideally forming a closed-loop. This symbiotic relationship between industrial processes is central to the concept of industrial ecology (Frosch, 1992). Within the Norwegian context, one form of IA proposed integrates (at least); fish and algae aquaculture, or more specifically; salmon and macro-algae (Agnalt et al., 2011; Olafsen et al., 2012; Skjermo et al., 2014). The benefits of this proposed IA system are most importantly: reduced local eutrophication from fish aquaculture (Marinho-Soriano et al., 2009; Reid et al., 2009; Wang et al., 2012; Broch et al., 2013; Reid et al., 2013), macro-algae driven carbon sequestration (Aresta et al., 2005), and the co-production of food, chemicals and energy through a biorefinery concept (Adams et al., 2009; Burton et al., 2009; DOE, 2010; Goh and Lee, 2010; Holdt and Kraan, 2011; Kraan, 2013; Kumar et al., 2013).

A biorefinery is the practice of "processing of biomass into a spectrum of marketable products and energy" (Cherubini, 2010). Cherubini (2010) adds three conditions which are the production of: at least one high-value chemical/material, one energy product besides heat and electricity (biofuel); as well a biorefinery should aim at running in a sustainable manner, i.e. all the energy requirements of heat and electricity should be supplied internally. However, prior to any large scale implementation of IA systems in Norway, it is important to substantiate the proposed claims of environmental benefits, and determine the feasibility of macro-algae based biorefineries.

Life-cycle assessment (LCA) is one tool for determining the direct and indirect environmental impact of a product's function over its complete life-cycle, (ISO, 2006; EC, 2010). LCA studies of salmon aquaculture are well documented (Pelletier et al., 2009; Winther et al., 2009; Samuel-Fitwi et al., 2012). LCA studies of macro-

algae cultivation and biomass conversion to biofuels and other co-products, have also received some attention in recent years (Aresta et al., 2005; Langlois et al., 2012; Alvarado-Morales et al., 2013; Aitken et al., 2014; Boonstra, 2015).

However to our knowledge no LCA studies have been published which depict the complete IA system with a biorefinery concept according to the definition by Cherubini (2010). Only through an integrated assessment can one assure consistency with respect to methodology, data, results and interpretation, one requirement in LCA (ISO, 2006; EC, 2010).

2. Material and Methods

2.1 Goal Definition

The goal of this study is to assess the cradle-to-gate environmental impacts, and cumulative energy demand, of an integrated open cage Atlantic salmon (S. *salar*)- and macro-algae (S. *latissima*) aquaculture system for the production of food (salmon) and via a biorefinery: one high-value low-volume material (sodium alginate 90 %), one energy product (bioethanol 99.7 %) along with one low-value high-volume product (compost) in Norway, see Fig. 1. The internal by-product biogas is used for co-heat-and-power (CHP) generation supplying some of the internally consumed electricity and heat. Additionally, this article presents results based on new empirical data for macro-algae cultivation and harvesting, and an up-to-date life-cycle inventory (LCI) for salmon aquaculture, along with assessing a new biorefinery configuration based on existing independent and semi-integrated production methods.

In doing so, we aim to answer the following research questions:

1. What are the environmental impacts associated with the IA and biorefinery systems' co-products: sodium alginate, ethanol, compost and salmon, and how do these compare with the results of other studies?

2. Based on these findings, which general observations and conclusions can be drawn concerning IA and biorefinery systems?

Through process contribution and sensitivity analysis we interpret the results from the modelled system. This study contributes to the growing scientific knowledge of IA and biorefinery systems, and the results are intended to provide guidance to stakeholders in the technological development of IA systems for food and fuel production.

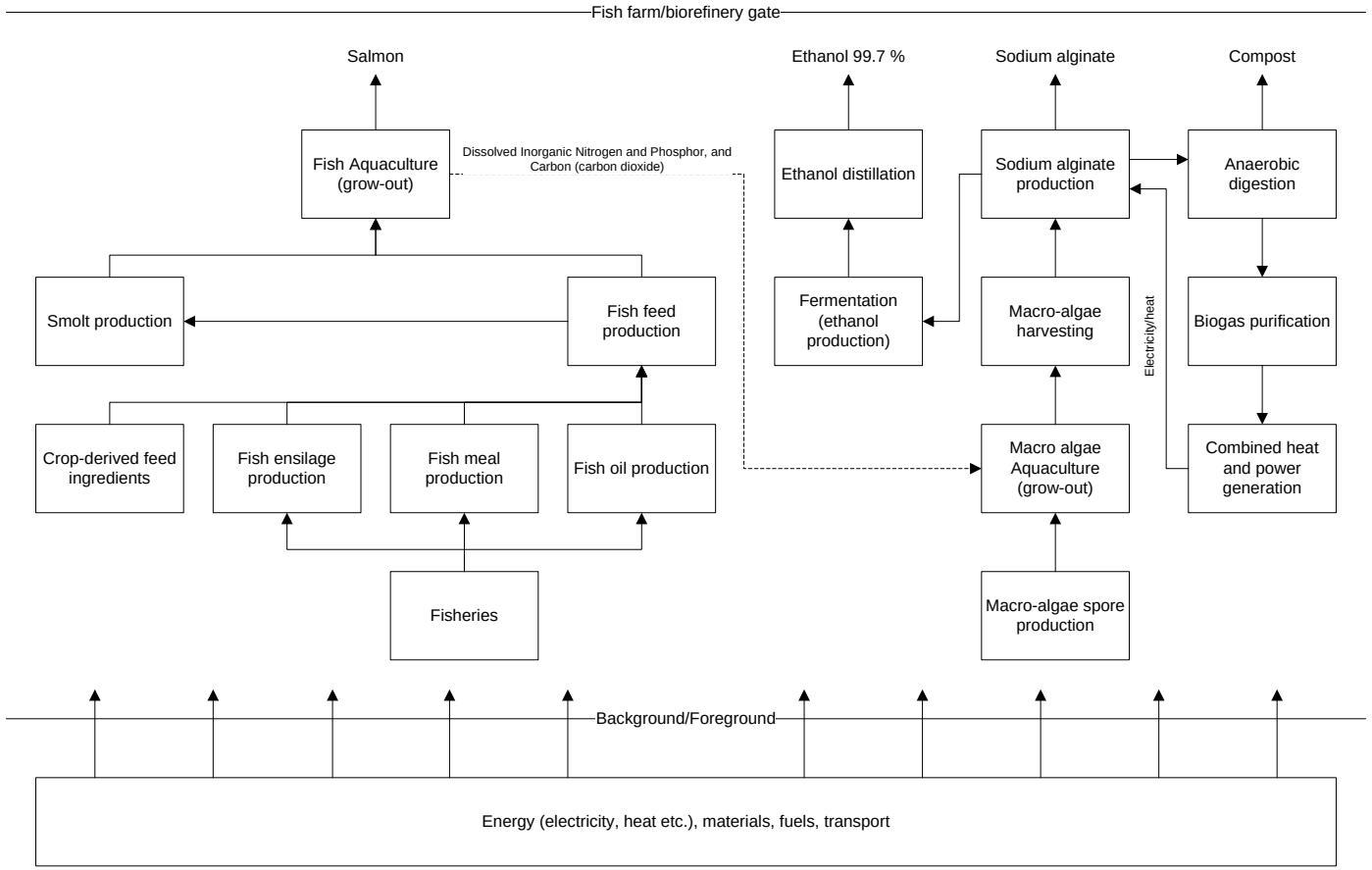

Fig. 1 Integrated salmon and macro-algae aquaculture system with biorefinery indicating system boundaries

5.3 Scope Definition

This is a multi-functional system, with the following functional units (FU): 1 kg WW salmon at fish-farm gate, 1 kg sodium alginate (90 % purity) at biorefinery gate, 1 kg compost at biorefinery gate, and 1 MJ ethanol (99.7 % purity) at biorefinery gate (lower heating value 26.952 MJ/kg (ANL, 2010)). Solving for multifunctionality is performed as specified for biorefineries by Ahlgren et al. (2013) which is based on ISO (2006) and EC (2010). Subdivision is performed between the fish and macro-algae aquaculture systems, though subdivision was not an option for the biorefinery system due to the inseparability of the chemical processes involved. The next preferred method of substitution was considered, however this would require the existence of superseded processes (products), many of which do not exist for macro-algae biorefinery co-products, e.g. alginate. As a result, and under the assumptions that the ratio between biorefinery co-products is inflexible and that no reasonable physical causation exists, we chose to apply economic allocation to the biorefinery co-products based on the values presented in Table A.1.

Macro-algae production is scaled according to the mass of dissolved inorganic nitrogen (DIN) released by salmon aquaculture and available for uptake by macro-algae, since DIN is a limiting factor (Wang et al., 2012). We assume that 30% of DIN is available for uptake by macro-algae (Wang et al., 2012), though other values are present in the literature: (Marinho-Soriano et al., 2009; Broch et al., 2013; Reid et al., 2013) and Wang et al. (2012) citing: (Buschmann et al., 1996; Troell et al., 1997; Troell et al., 2003). All life-cycle impact reductions resulting from the uptake of DIN, dissolved inorganic phosphorus (DIP) and carbon dioxide (CO_2) are assigned to the macro-algae aquaculture system.

LCI modelling is performed using attributional LCA methods. The basis for this decision is as follows: first, the salmon aquaculture system already exists, and we neither advocate nor oppose changes to the salmon aquaculture industry in Norway based on the results of this study. Second, it is not the intention of the authors that the results for the macro-algae system and its biorefinery co-products should be used directly in meso-/macro- level decision making support.

We prioritize secondary LCI data sources providing transparency over the use of confidential manufacturer specific data, with one exception, i.e. the cultivation and harvesting of macro-algae. Foreground LCI data are collected from both private industry and published literature and are conditioned for this study, including both

LCI data for salmon and macro-algae aquaculture grow-out infrastructure. Background LCI data is derived from the commercial databases in the SimaPro 8 software package (Goedkoop et al., 2013) and conditioned to reflect Norwegian conditions, e.g. with respect to energy. Assumptions are based on communication with qualified stakeholders in their respective fields

LCIA is performed with SimaPro 8 (Goedkoop et al., 2013). Environmental impacts are calculated using the CML IA baseline method (Pré, 2015) for the mid-point impact assessment categories: global warming potential (GWP) measured in kg CO_2 eq., eutrophication potential (EP) measured in g PO_4 eq., acidification potential (AP) measured in g SO_2 eq., ozone layer depletion (ODP) measured in mg CFC-11 eq., and photochemical oxidation potential (POP) measured in g C_2H_4 eq.. Cumulative energy demand (CED) measured in MJ is determined using methods developed by Frischknecht et al. (2007). It should be noted that LCI data collection is focused on the accurate determination of GWP and EP, and as a result the remaining environmental impact categories should be interpreted as qualified estimates.

2.3 Case Description

2.3.1 Macro-algae aquaculture

Numerous proposed methods to cultivate macro-algae in general and specifically S. *latissima* exist: (Langlois et al., 2012; Alvarado-Morales et al., 2013; Aitken et al., 2014; Beltran and Guinée, 2014). This LCI of macro-algae aquaculture cultivation is based on commercial data from one semi-industrial producer in Norway, which includes: spore production and macro-algae cultivation and harvesting. The dry-matter (DM) composition of macro-algae used in these inventories is presented in Table A.2.

Due to the proprietary nature of this information the LCI is presented in an aggregated form. In this process, wild harvested S. *latissima* provides the basis for spore production which is carried out in a laboratory under controlled conditions. The growth lines are impregnated with macro-algae spores, and then transported to- and mounted on the floating frames in the fall (November). The macro-algae growth season extends until the late spring (June) at which time they are harvested with the use of a barge, and transported to land.

Table 1 presents the aggregated LCI for macro-algae production and harvesting, and represents ≥ 95% wt. of all inputs. We assume that plastic, steel and concrete will be sent to waste treatment and recycling. We have

assumed an annual repair and maintenance factor of 5% for all infrastructure, and assumed conservative life-spans of: 4, 10 and 8 years for: growth lines, main mooring & buoys, and suspended frames respectively.

Table 1 Aggregated LCI for the cultivation of one kg WW macro-algae harvested and transported to land (biorefinery)

Output	**Value**	**Unit**	**Reference**
Macro-algae	1	kg WW	(Hortimare, 2015)
Input			
Diesel	1.827	ml	(Langlois et al., 2012)
Electricity	0.150	kWh	(Hortimare, 2015)
Plastic	23.640	g	(Hortimare, 2015)
Steel	6.031	g	(Hortimare, 2015)
Concrete	5.934	g	(Hortimare, 2015)
Sequestration			
N	2.447	g	(Reid et al., 2013)
P	0.309	g	(Reid et al., 2013)
CO_2	92.329	g	(Reid et al., 2013)

2.3.2 Salmon Aquaculture

This phase includes the sub-processes: smolt production, fish feed production and salmon aquaculture grow-out and harvesting; including all transport between processes up to fish-farm gate. The LCI for smolt production is adapted from Winther et al. (2009) and Wang et al. (2012), see Table A.3, using an economic feed conversion ratio (FCR) of 1.16 (Wang et al., 2012). Transport of the smolt is included in the LCI for salmon aquaculture. We assume that nitrogen (N), phosphorus (P), carbon and respired CO_2 are emitted to the marine ecosystem.

The LCI for fish feed has been adapted from the composition of fish feed for 2012 provided by the Norwegian Seafood Federation (FHL) (2013), see Table A.4, and supplemented with secondary data from Winther et al. (2009). In this study, we have scaled the known 97.2% of fish feed ingredients to 100%, see Table A.5. In 2012 fish-meal and fish-oil represented over 28% of marine derived feed ingredients (FHL, 2013). In 2012, of the 19 reported fish species: anchovy, blue whiting, capelin, sandeel, sprat, menhaden and trimmings (by-product) constituted approximately 98% and 97% of fish-meal and fish-oil composition respectively (FHL, 2013). We have scaled these 7 species to represent 100% of fish inputs in the production of fish-meal and fish-oil, see Table A.6. A generic LCI for the production of fish-meal and fish-oil has been adapted from Cappell et al. (2007),

Winther et al. (2009), and FHL (2013). The production of fish-meal and fish-oil is a multi-functional process, see Fig. 1, as a result we have applied mass allocation between fish meal (83%) and fish oil (17%) based on Winther et al. (2009) and Pelletier (2007). Tables A.7 and A.8 present the LCIs of fish-meal and fish-oil respectively. Energy inputs to Peruvian and USA fisheries are based on Pelletier (2007), and Winther et al. (2009) for Norwegian fisheries, see Table A.9. Products of Peruvian anchovy and USA menhaden fisheries include transport distances of 10556 km and 7556 km respectively to the west coast of Norway. The LCI for the production of fish ensilage is based on the process described by Tatterson and Windsor (2001), see Table A.10. Crop derived proteins and carbohydrates have been conditioned and scaled based on inventory data provided by Winther et al. (2009) for crop derived meal, with: soy meal, sunflower meal, wheat and wheat gluten meal constituting 50%, 16%, 30% and 4% of total crop derived meal respectively. The complete LCI for fish feed production is presented in Table A.5.

The LCI for salmon aquaculture grow-out is adapted from Pelletier et al. (2009), Winther et al. (2009) and Tyedmers (2000) for energy and materials, and Wang et al. (2012) for nutrient emissions, see Table 2. Both Pelletier et al. (2009) and Winther et al. (2009) are based on empirical data which are representative of the salmon aquaculture industry in Norway. Diesel consumption is for both harvesting and the inspection and maintenance of the fish aquaculture grow-out facility.

Table 2 LCI for the production of one kg WW salmon at fish-farm gate

Output	**Value**	**Unit**	**Reference**
Fish	1	kg	
Input			
Fish feed	1.139	kg	
Smolt	0.017	kg	(Pelletier et al., 2009)
Electricity	72.2	Wh	(Pelletier et al., 2009)
Transport (feed and smolt)	0.307	tkm	(Pelletier et al., 2009)
Heat	0.075	Wh	(Winther et al., 2009)
Diesel	0.015	l	(Winther et al., 2009)
Steel	4.500	g	(Tyedmers, 2000)
Zinc	0.255	g	(Tyedmers, 2000)
Polyethylene (high density)	0.567	g	(Tyedmers, 2000)
Polystyrene	0.241	g	(Tyedmers, 2000)
Nylon	8.653	g	(Tyedmers, 2000)
Polyethylene (low density)	0.345	g	(Tyedmers, 2000)
Emissions to air			
Carbon dioxide	975.8	g	(Wang et al., 2012)
Emissions to water			
Nitrogen	49.1	g	(Wang et al., 2012)
Phosphorus	9.1	g	(Wang et al., 2012)
Carbon (total organic)	123.8	g	(Wang et al., 2012)

2.3.3 Biorefinery

The development of a suitable biorefinery process for macro-algae resulting in the separation and refinement of individual compounds into final products is one of the major research and development challenges facing commercialization (Kraan, 2013; Skjermo et al., 2014). Numerous laboratory scale experiments have been performed targeting individual, or a limited number of compounds and final products, e.g. protein (Barbarino and Lourenço, 2005; Kim et al., 2011; Harnedy and FitzGerald, 2013), mannitol and laminaren (Horn, 2009), and alginates (Phycocolloids) (Langlois et al., 2012), among others. Based on extensive literature review we assume that it is feasible to extract/isolate those compounds leading to the co-production of: sodium alginate, bioethanol and compost. Table 3 presents the LCI for the biorefinery.

Table 3 LCI for a biorefinery producing: sodium alginate, compost and bioethanol from one kg WW macro-algae

Output	**Value**	**Unit**	**Reference**
Sodium alginate	0.042	kg	(Langlois et al., 2012)
Compost	0.490	kg	(Langlois et al., 2012)
Ethanol	0.323	MJ	(Horn, 2009)
By-products			
Electricity	0.049	kWh	(Jungbluth et al., 2007; Langlois et al., 2012)
Heat	0.303	MJ	(Jungbluth et al., 2007; Langlois et al., 2012)
Inputs			
Macro-algae	1	Kg WW	
Water	143.060	kg	(Horn, 2009; Langlois et al., 2012)
Hydrochloric acid	7.944	kg	(Langlois et al., 2012)
Cellulose	0.244	kg	(Langlois et al., 2012)
Sodium carbonate	99.714	g	(Langlois et al., 2012)
Lubricating oil	0.028	g	(Jungbluth et al., 2007; Langlois et al., 2012)
Ammonia	1.040	g	(Horn, 2009)
Sulfuric acid	0.265	g	(Jungbluth et al., 2007)
Sulphate	0.027	g	(Jungbluth et al., 2007)
Phosphate	0.069	g	(Jungbluth et al., 2007)
Electricity	1.145	kWh	(Jungbluth et al., 2007; Langlois et al., 2012)
Process steam	0.595	kWh	(Jungbluth et al., 2007; Langlois et al., 2012)
Heat	0.368	kWh	(Jungbluth et al., 2007; Langlois et al., 2012)
Cooling	0.393	kWh	(Langlois et al., 2012)
Emissions to air			
Carbon dioxide	106.817	g	(Jungbluth et al., 2007)
Methane	0.682	g	(Jungbluth et al., 2007)
Hydrogen sulphide	0.107	mg	(Jungbluth et al., 2007)
Sulphur dioxide	13.770	mg	(Jungbluth et al., 2007)
Mono-nitrogen oxides	8.262	mg	(Jungbluth et al., 2007)
Waste and emissions to treatment			
Waste water	182.318	kg	mass balance calculation

Sodium alginate production forms the corner-stone of this biorefinery system and is based on Langlois et al. (2012). The waste flows, hereafter referred to as side flows from sodium alginate production are determined by mass balance and provide the inputs for the production of ethanol and compost. Both Horn (2009) and Kraan (2013) support the preliminary separation of alginate for reasons of biorefinery efficiency. We assume the macro-

algae DM composition which is presented in Table A.2, based on Holdt and Kraan (2011) and Reid et al. (2013). Additionally, we assume that the total mass of each compound is available for conversion, though we have accounted for conversion efficiency. Sodium alginate production includes the processes: pre-treatment, acid lixiviation, alkaline extraction, rectification and finally conversion to sodium alginate (Langlois et al., 2012). This process is similar to that which is outlined by McHugh (1987) for the acid-method of alginate extraction.

The acid lixiviation process in sodium alginate production is the same extraction method for mannitol and laminaren explained by Horn (2009) in his experiments. Determined by mass balance, we find that the mannitol and laminaren containing side-flows from acid lixiviation has a similar concentration of mannitol and laminaren (26 g/l) as Horn (2009) achieved in his experiments. The production of ethanol from this side-stream proceeds as follows: first it is treated with ammonia solution (25%) to adjust the pH according to Horn (2009). This is followed by fermentation to ethanol (95%) and further distillation (99.7%); both based on Jungbluth et al. (2007). Based on Horn (2009) we assume a 46% wt. conversion efficiency of mannitol and laminaren to ethanol.

The side-stream from alkaline extraction becomes the feedstock for first biogas production via anaerobic digestion and following this the conversion of digester sludge to compost. Biogas production and purification is adapted from Langlois et al. (2012) and Jungbluth et al. (2007) for material and energy consumption, and process emissions. Here we scale emissions based on Nm^3 biogas produced. The purified biogas is then combusted in an on-site CHP plant, providing both heat and electricity consumed in the biorefinery (avoided products). We have assumed a LHV of 18 MJ/Nm^3, based on the methane content of the biogas provided by Langlois et al. (2012). The CHP plant efficiency is 55% for heat and 32% for electricity, the remaining 13% is in the form of waste heat (Jungbluth et al., 2007). After biogas production, some of the water content of the digester sludge is removed and treated; the remainder becoming the final co-product of compost.

3. Results

Fig. 2, 3 and 4 present the normalized LCIA results for salmon and biorefinery co-products: sodium alginate, ethanol and compost, indicating processes contribution (≥ 5%), where direct emissions refer foreground process emissions. The absolute LCIA results for all products are presented in Table 4.

Table 4 Absolute LCIA results for the production all products

		Sodium alginate	Ethanol	Compost	Salmon
	Unit/FU	1 kg	1 MJ	1 kg	1 kg
GWP	kg CO_2 eq.	102.83	0.31	0.23	3.40
CED	MJ	1556.19	4.67	3.50	14.63
EP	g PO_4 eq.	231.46	0.69	0.52	166.85
AP	g SO_2 eq.	1103.35	3.31	2.48	19.89
ODP	mg CFC 11 eq.	9.893	0.030	0.022	0.372
POP	g C_2H_4 eq.	47.808	0.143	0.107	0.487

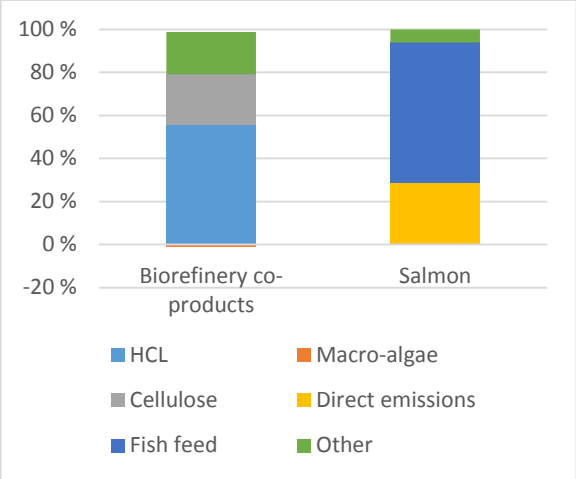

Fig. 2 Normalized LCIA results for GWP indicating sub-process contribution

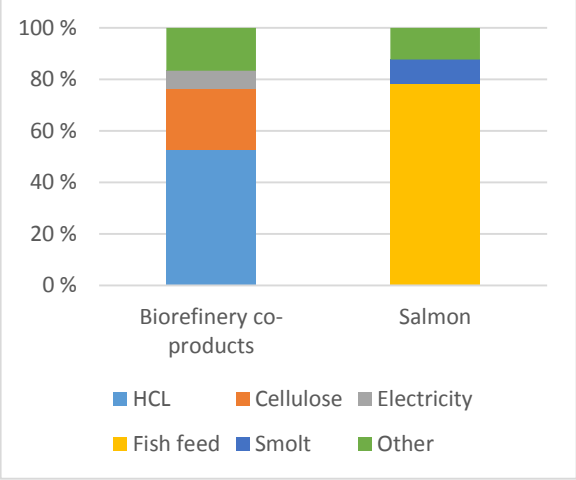

Fig. 3 Normalized LCIA results for CED indicating sub-process contribution

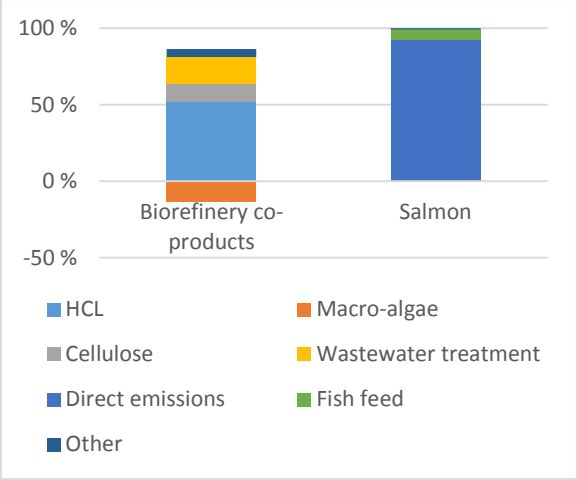

Fig. 4 Normalized LCIA results for EP indicating sub-process contribution

From Fig. 2 through 4, we see that the relative contribution of biorefinery processes is the same for all co-products due to the application of economic allocation. In this study, we have assumed the economic values presented in Table A.1. However, the world market for alginate is currently limited to approximately 30 000 tonnes (Burton et al., 2009; Bixler and Porse, 2011). With a yield of 0.042 kg sodium alginate per kg WW macro-algae, only 0.7 mil. tonnes WW of macro-algae would be necessary to meet current demand. The projected increase in macro-algae production from 0.2 mil. tonnes to 20 mil. tonnes between 2010 and 2050 (Olafsen et al., 2012) will affect market prices for alginate. As a sensitivity analysis, we adjust the market price of alginate by ± 50% and as a result the economic allocation applied between biorefinery co-products. The results of this, and other, sensitivity analysis are presented in Fig. 5 and 6 for sodium alginate and ethanol respectively. Fig. 6 reveals that GWP, CED and EP results for ethanol are highly sensitive to changes in the market price for sodium alginate, and Fig. 5 reveals that GWP, CED and EP results for sodium alginate are less volatile to changes in the market price for sodium alginate Furthermore, one should note that the normalized results for ethanol production are representative for compost production.

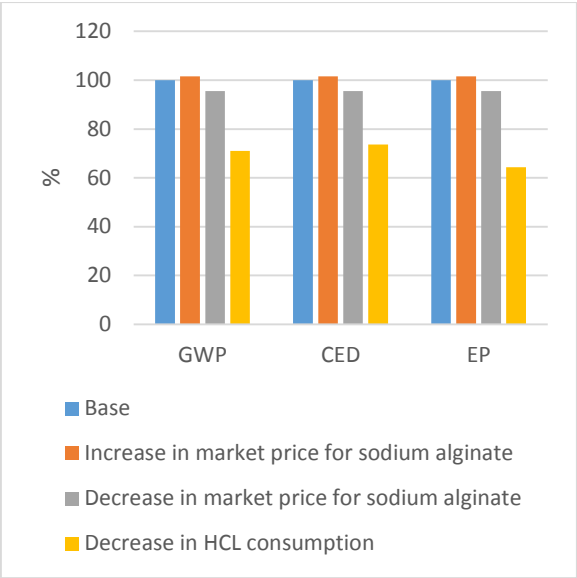

Fig. 5 Normalized LCIA results of the sensitivity analysis for the production of sodium alginate

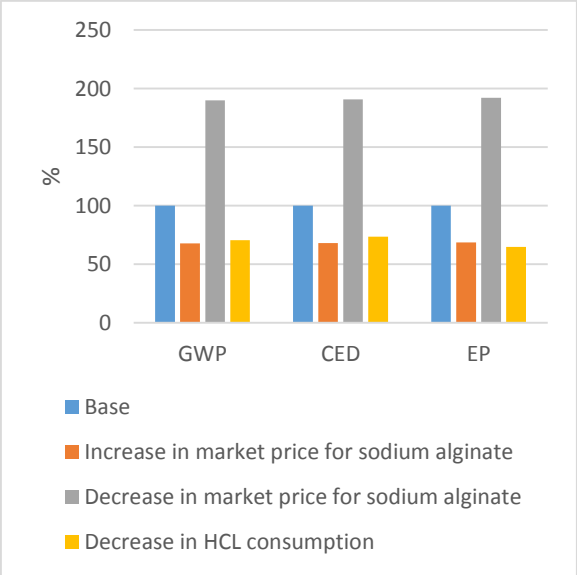

Fig. 6 Normalized LCIA results of the sensitivity analysis for the production of ethanol

Additionally, Fig. 2 through 4 reveal that hydrochloric acid (HCL) consumption dominates the results for biorefinery products. Specifically, HCL contributes to: 57.9%, 52.8% and 52.0% of GWP, CED and EP results respectively. Hernández-Carmona et al. (1998) claim that recycling HCL in the production of sodium alginate is possible with little-to-no effect on the yield. As a sensitivity analysis, we reduce biorefinery consumed HCL by 50%, to simulate the effect of partial recycling of HCL. The results of this sensitivity analysis are as well

presented in Fig. 5 and 6, where we observe that GWP, CED and EP results for both sodium alginate and ethanol are highly sensitive to reductions in HCL consumption.

Furthermore, Fig. 2 and 4 reveal that the use of macro-algae biomass for the production of biorefinery products lowers their GWP and EP results. The integration of macro-algae is an important element in the IA–biorefinery system and responsible for the sequestration of CO_2 and N & P emissions, resulting in lower GWP and EP results respectively. Though some of these benefits are offset by the cultivation and harvesting of macro-algae; Fig. 5 presents the normalized LCIA results for macro-algae cultivation and harvesting indicating process contribution (≥ 5%). The results for all impact categories are presented in Table 5.

Table 5 LCIA results for the cultivation and harvesting of one kg WW macro-algae

	GWP	**CED**	**EP**	**AP**	**ODP**	**POP**
Unit	kg CO_2 eq.	MJ	g PO_4 eq.	g SO_2 eq.	mg CFC-11 eq.	g C_2H_4 eq.
Total (net)	-0.055	0.446	-1.892	0.234	0.010	0.010

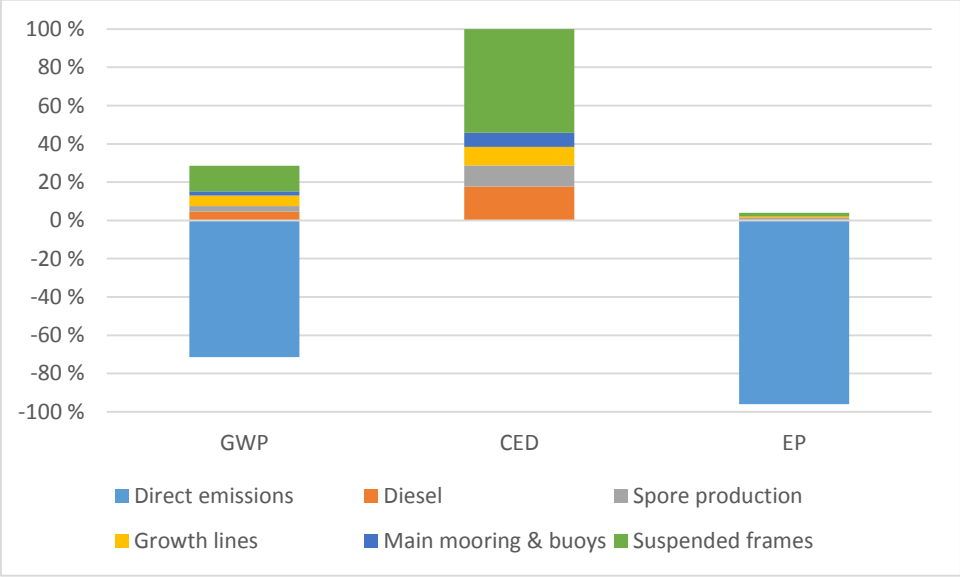

Fig. 5 Normalized LCIA results for macro-algae cultivation and harvesting, indicating sub-process contribution

For these results, we have considered a macro-algae yield of 6 kg per meter growth line (Table A.2). As a sensitivity analysis we consider the effect of either increasing yield to 8 kg/or decreasing yield to 4 kg/m.

Additionally we consider the effect of lengthening/shortening the life-span of the macro-algae cultivation infrastructure by ± 25%. The results of these sensitivity analyses are presented in Fig. 6, from which we observe that the results for cultivation and harvesting of macro-algae are most sensitive to decreases in yield, and shorter life-spans of the cultivation infrastructure.

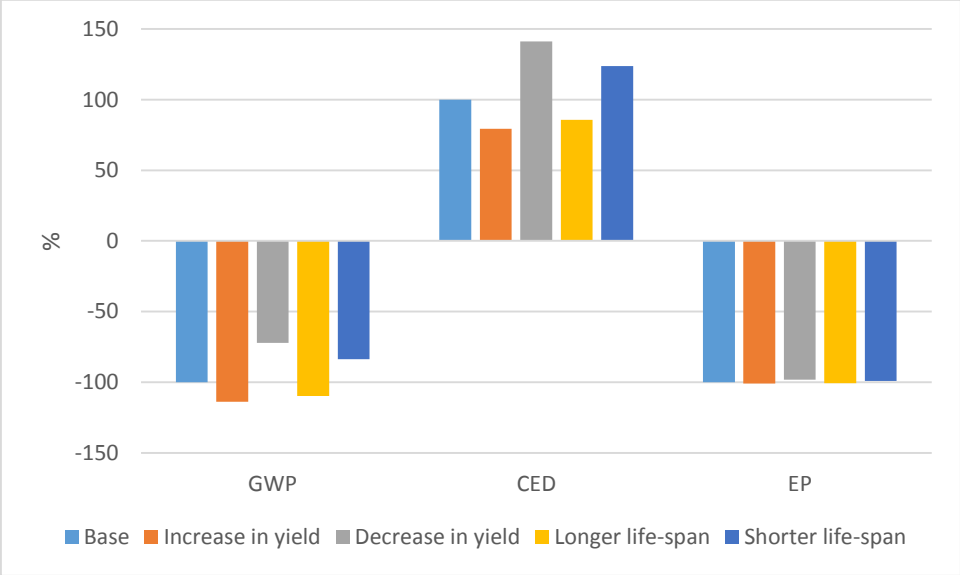

Fig. 6 Normalized LCIA results of the sensitivity analysis for yield and life-span on macro-algae cultivation and harvesting

Finally, Fig. 2, 3 and 4 reveal that direct emissions from fish aquaculture and the provision of fish feed dominate these impact categories for salmon production. Direct emissions cover both N, P and C emissions along with fish respired CO_2, the latter accounting for 0.98 kg CO_2 eq.

4. Discussion

For sodium alginate, we have determined a GWP of 102.83 kg CO_2 eq./kg . Only two other studies have evaluated the environmental impacts of sodium alginate production: Boonstra (2015) who determines a GWP of 207.84 kg CO_2 eq./kg, and Langlois et al. (2012) who present normalized values with no quantities. Our results indicate that HCL and cellulose consumption are the main contributors towards GWP, CED and EP. This is consensual with Boonstra (2015) and Langlois et al. (2012) which is not surprising considering that this studies production of sodium alginate is modelled on the latter. Leceta et al. (2014) performed an LCA of agar (another hydrocolloid), and though not transferable due to their selection of FU, they do identify the consumption of

process chemicals as a significant contributor towards agar production. Whether macro-algae requires such aggressive pre-treatment has been questioned by both Adams et al. (2009) and Hernandez-Carmona et al. (1998). Adams et al. (2009) found that high ethanol yield was possible without the need for lowering the pH with HCL. Hernandez-Carmona et al. (1998), studying various pre-extraction techniques claim that acidic fluids from alginate production can be recirculated with little effect on alginate yield, this claim is supported by McHugh et al. (2001).

For ethanol, we have determined a GWP of 0.31 kg CO_2 eq./MJ and a CED of 4.67 MJ/MJ. Similarly, Aitken et al. (2014) found that for all scenarios producing ethanol there was little-or-no net energy gain. Alvarado-Morales et al. (2013) also found that the net energy balance was negative for the co-production of ethanol and biogas. Both Aitken et al. (2014) and Alvarado-Morales et al. (2013) determined negative GWP values based on their methods for dealing with multifunctionality, i.e. substitution/avoided products.

All similar LCA studies of macro-algae cultivation for biorefinery conversion consider the production of compost, i.e. (Langlois et al., 2012; Alvarado-Morales et al., 2013; Aitken et al., 2014; Boonstra, 2015). Both Aitken et al. (2014) and Alvarado-Morales et al. (2013) deal with co-products through system-expansion and substitution, whereas Langlois et al. (2012) not either consider that co-produced compost will substitute other forms of composts. Boonstra (2015) determines a GWP of 0.08 kg CO_2 eq./kg and a CED of 0.11 MJ/kg compared to the 0.23 kg CO_2 eq./kg and 3.5 MJ/kg compost respectively determined in this study. The differences are explained by variations in production masses and the economic allocation values applied.

In contrast to the other co-products, several LCA studies have been carried out for aquaculture salmon. Table 6 presents a comparison of our study with results from other relevant studies.

Table 6 Comparison of LCIA results of this study with similar studies for 1 kg WW salmon at fish-farm gate

Impact category	**GWP**	**CED**	**EP**
Reference/Unit	kg CO_2 eq.	MJ	g PO_4 eq.
This study	3.4	14.6	167.0
(Buchspies et al., 2011)	2.05		
(Hognes et al., 2011)	2.6	25.3	
(Ayer and Tyedmers, 2009)	2.07	26.9	35.3
(Ellingsen et al., 2009)[a]	2.2 – 2.3		
(Pelletier et al., 2009)	1.79	26.2	41.0

[a] 1 kg fish fillet at fish-farm gate.

For GWP, CED and EP the results of this study varies between -44%, 58% and 120% than the average value of the other studies. It should be noted that Ayer and Tyedmers (2009) have not considered the emission of fish respired CO_2 in their determination of GWP. Additional differences between GWP results can be explained by the higher proportion of crop-derived feed ingredients in this study. CED values from this and the three similar studies is dominated by energy consumption during feed production. The difference between this studies CED results and the others can be partially explained by the lower proportion of fisheries-derived feed in this study, along with the understanding that commercial fisheries are energy intensive (Pelletier et al., 2009). Direct emissions during grow-out contribute most to the EP results in all studies. The difference in total EP values between this study and the two comparative studies is significant. Pelletier et al. (2009) calculated 41.1 g and 5.2 g of direct N and P emissions respectively during grow-out, compared to the 49.1 g and 9.2 g of direct N and P emissions respectively during grow-out in this study. This difference in grow-out emitted N and P is sufficient to explain the variations in total EP results for salmon aquaculture.

Key to this assessment of IA, is the cultivation of macro-algae, and most importantly macro-algae's ability to sequester CO_2 and N & P emissions reducing GWP and EP impacts respectively. The magnitude of emissions sequestration of this and similar studies are presented in Table 7. Differences can be largely attributed to variations in macro-algae composition, see Table A.2.

Table 7 Comparison of emissions sequestration resulting from the production of one kg WW macro-algae

Emission	**CO_2**	**N**	**P**
Reference/Unit	kg	g	g
This study	0.092	2.45	0.31
(Aitken et al., 2014)	0.111	2.80	0.96
(Alvarado-Morales et al., 2013)	0.114	2.10	0.88
(Langlois et al., 2012)	n/a	2.10	0.45

Some of the CO_2 sequestration benefits associated with macro-algae cultivation are reduced as a result of the high consumption of materials, particularly plastics, forming the cultivation structure. The CED for macro-algae cultivation and harvesting of this study is dominated by fuel consumption for repair & maintenance and harvesting.

5. Conclusion

This LCA presents a unique assessment of an IA–biorefinery system for food and fuel production in Norway. We found that integration of salmon and macro-algae aquaculture is environmentally beneficial with respect to CO_2, nitrogen and phosphor sequestration. This is in addition to other positive- and negative aspects of integrated aquaculture, e.g. increased biodiversity (Kraan, 2013) and increased marine area use (Garofalo, 2009). How efficient macro-algae can remove nutrients in practice still needs further investigation (Agnalt et al., 2011) which is reflected in the variation in determinations of uptake of salmon aquaculture emitted DIN by macro-algae, e.g. 10% to 100% (Marinho-Soriano et al., 2009; Agnalt et al., 2011; Broch et al., 2013). This is further compounded by the mismatch between macro-algae and salmon growth periods and resulting seasonal variation in emissions sequestration and composition of macro-algae (van Hal, 2012; Broch et al., 2013).

The use of IA produced macro-algae biomass in a biorefinery has not only the potential to improve the economic performance of the integrated system (Boonstra, 2015), but also the environmental profile of its co-products (Cherubini, 2010; Cherubini and Jungmeier, 2010). However, numerous challenges still confront the implementation of suggested biorefinery concepts (Skjermo et al., 2014). For example, the growth of biorefineries will be limited by the demand for its energy, fuel and chemical products (Cherubini, 2010; Holdt and Kraan, 2011). Currently, very few markets exist for the high-value low-yield products of a macro-algae based biorefinery (Burton et al., 2009) though this is expected to change in the near future (Carlsson and Bowles, 2007) which can affect the environmental profile of its products, suggesting that system-expansion is more suitable than the application of economic allocation as performed in this study. Additionally, further research is necessary to consider if the high-yield extraction of one compound will limit the isolation of others, affecting both the integration of individual processes and the cost-effective cultivation and use of macro-algae and its compounds (Skjermo et al., 2014). In addition, Beltran and Guinee (2014) have indicated that increased integration can lead to increased trade-offs. Most notably, and as identified in this study, the production of high-value low-volume products can affect the environmental profile of its co-products, particularly when applying economic allocation as here, or energetic allocation (Modahl et al., 2015). This is due to the often complex and resource intensive extraction and refinement techniques as is the case for alginate which alternately can be diverted to biofuel production (Horn, 2012). The removal of these high-value low-volume products can improve the environmental performance of biorefineries (Boonstra, 2015), and overall yields of low-value products, like

biofuels, since all products compete for the same carbon atoms. As such, biorefineries focusing on high-yield low-value products (biofuels) should be considered, even though the current trade-off is profitability (Aresta et al., 2005; Beltran and Guinée, 2014), remembering that environmental performance is equally or more important than profit (DOE, 2010).

Finally, only by assessing the complete IA–biorefinery system, is it possible to arrive at the observations arrived at in this study. Reflecting on the high environmental impacts associated with fish feed production, and the relatively high carbohydrate and protein content of S. latissima, feed production is a more sustainable processing route for macro-algae. Further integration between salmon and macro-algae aquaculture are possible, most notably shared infrastructure and transport services.

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Appendix

Table A.1 Values used for economic allocation between biorefinery products

	Economic value		**Quantity (ratio)**		**Total**		**Allocation**	**Reference**
	value	unit	value	unit	value	unit		
Sodium alginate	12.0000	USD/kg	0.4200	kg	5.0400	USD	95.3 %	(Bixler and Porse, 2011)
Bioethanol	0.0360	USD/MJ	3.2332	MJ	0.1163	USD	2.2 %	Personal communication
Compost	0.0271	USD/kg	4.9000	kg	0.1326	USD	2.5 %	(OKR, 2015)

Table A.2 Composition of macro-algae S. latissima used in this study, with: production yield per meter cultivation line and DM percent from producer, DM constituents from Holdt and Kraan (2011), N,P and C content from Reid et al. (2013).

		Value	**Unit**
macro-algae WW harvested per meter		6	kg
DM		10	%
of which	N	2.447	% of DM
	P	0.309	% of DM
	C	25.196	% of DM
Dry matter compounds	Protein	15	% of DM
	Mannitol	12	% of DM
	Phycocolloids	23	% of DM
	other carbohydrates	15	% of DM
	Minerals	10	% of DM
	Laminaren	14	% of DM
	Fatty acids (PUFA)	4	% of DM
	Polyfenols	0.5	% of DM
	Iodine	0.4	% of DM
	Fucoxanthine	0.05	% of DM
	Fucoidan	5	% of DM

Table A.3 LCI for the production of 1000 kg smolt

Output	**Value**	**Unit**	**Reference**
Smolt	1000.0	kg	
Input			
Fish feed	1160.0	kg	(Wang et al., 2012)
Electricity	14129.0	kWh	(Winther et al., 2009)
Emissions to air			
Carbon dioxide	993.1	kg	(Wang et al., 2012)
Emissions to water			
Nitrogen	50.0	kg	(Wang et al., 2012)
Phosphorus	9.3	kg	(Wang et al., 2012)
Total organic carbon	126.0	kg	(Wang et al., 2012)

Table A.4 General composition of Norwegian fish feed for the year 2012 (FHL, 2013)

Marine derived	**Percent**
Fish meal	16.8
Fish oil	11.5
Fish ensilage (from by-products)	3.6
Other marine material	0.1
Crop derived	
Proteins/carbohydrates (meal)	47.0
Rape seed oil	18.2
Total	97.2
Difference	2.8

Table A.5 LCI for the production of 1000 kg fish feed

Output	**Value**	**Unit**	**Reference**
Fish feed	1000	kg	
Input			
Fish meal	168	kg	(FHL, 2013)
Fish oil	115	kg	(FHL, 2013)
Fish ensilage	36	kg	(FHL, 2013)
Rapeseed oil	182	kg	(FHL, 2013)
Soybean meal	235	kg	(Winther et al., 2009; FHL, 2013)
Sunflower meal	75.2	kg	(Winther et al., 2009; FHL, 2013)
Wheat	141	kg	(Winther et al., 2009; FHL, 2013)
Wheat gluten	18.8	kg	(Winther et al., 2009; FHL, 2013)
Water	3000	kg	(Winther et al., 2009)
Diesel	0.22	l	(Winther et al., 2009)
Electricity	0.011	kWh	(Winther et al., 2009)
Heat (light fuel oil)	15	kWh	(Winther et al., 2009)
Heat (natural gas)	51	kWh	(Winther et al., 2009)
Steam	82	kWh	(Winther et al., 2009)
Liquefied petroleum gas	1.3	l	(Winther et al., 2009)
Transport[a]	767.1	tkm	own calculation
Waste and emission to treatment			
Waste water	2971	l	own calculation

[a] Netherlands - Norway 1211 km and Germany - Norway 1087 km

Table A.6 Fish species used in the production of fish meal and fish oil in Norway in 2012, including scaled values used in this study

	Fish meal			**Fish oil**		
Species and assumed origin	FHL (2013)	Conditioned/scaled		FHL (2013)	Conditioned/scaled	
	%	%	kg	%	%	kg
Anchovy (Peru)	33	33.7	336.7	41	42.3	422.7
Blue whiting (Norway)	2	2.0	20.4	< 1	1.0	10.3
Capelin (Norway)	21	21.4	214.3	12	12.4	123.7
Sandeel (Norway)	3	3.1	30.6	2	2.1	20.6
Sprat (Norway)	7	7.1	71.4	12	12.4	123.7
Menhaden (USA)	< 1	1.0	10.2	5	5.2	51.5
Trimming (Norway)	31	31.6	316.3	24	24.7	247.4
Total	< 98	99.9	999.9	< 97	100.1	999.9

Table A.7 LCI for the reduction of fish to 1000 kg fish meal using 87%/17% mass allocation between fish- meal and oil.

Output	**Value**	**Unit**	**Reference**
Fish meal	1000.0	kg	
Input			
Salt water	60.0	m3	(Cappell et al., 2007)
Fresh water	1.5	m3	(Cappell et al., 2007)
Anchovy (Peru)	1302.3	kg	(FHL, 2013)
Menhaden (USA)	38.6	kg	(FHL, 2013)
Pelagic (Norway)[a]	1302.3	kg	(FHL, 2013)
Trimmings	1218.6	kg	(FHL, 2013)
Sodium hydroxide	3.977	kg	(Winther et al., 2009)
Formaldehyde	3.321	kg	(Winther et al., 2009)
Methanol	5.628	kg	(Winther et al., 2009)
Sulfuric acid	1.735	kg	(Winther et al., 2009)
Nitric acid	0.425	kg	(Winther et al., 2009)
Hydrochloric acid	0.317	kg	(Winther et al., 2009)
Heat	5138.3	MJ	(Winther et al., 2009)
Electricity	157.7	kWh	(Winther et al., 2009)
Transport[b]	3632.6	tkm	own calculation
Waste and emission to treatment			
Waste water	64421.4	l	own calculation

[a] Blue whiting, capelin, sandeel and sprat.
[b] Of anchovy and menhaden fish meal to Norway.

Table A.8 LCI for the reduction of fish to 1000 kg fish oil using 87%/17 % mass allocation between fish- meal and oil

Output	**Value**	**Unit**	**Reference**
Fish meal	1000.0	kg	
Input			
Salt water	58.7	m3	(Cappell et al., 2007)
Fresh water	1.5	m3	(Cappell et al., 2007)
Anchovy (Peru)	1597.8	kg	(FHL, 2013)
Menhaden (USA)	196.4	kg	(FHL, 2013)
Pelagic (Norway)[a]	1051.1	kg	(FHL, 2013)
Trimmings	933.3	kg	(FHL, 2013)
Sodium hydroxide	3.889	kg	(Winther et al., 2009)
Formaldehyde	3.244	kg	(Winther et al., 2009)
Methanol	5.511	kg	(Winther et al., 2009)
Sulfuric acid	1.700	kg	(Winther et al., 2009)
Nitric acid	0.416	kg	(Winther et al., 2009)
Hydrochloric acid	0.309	kg	(Winther et al., 2009)
Heat	5022.2	MJ	(Winther et al., 2009)
Electricity	154.2	kWh	(Winther et al., 2009)
Transport[b]	4866.7	tkm	own calculation
Waste and emission to treatment			
Waste water	62971.5	l	own calculation

[a] Blue whiting, capelin, sandeel and sprat.
[b] Of anchovy and menhaden fish meal to Norway.

Table A.9 Fuel inputs for 1000 kg of: Peruvian (anchovy), USA (menhaden) and Norwegian (pelagic) fisheries

Output	**Value**	**Unit**	**Reference**
Fish	1000.0	kg	
Input			
Peru (anchovy)	40.1	l	(Pelletier, 2007)
USA (menhaden)	32.0	l	(Pelletier, 2007)
Norway (pelagic)[a]	93.3	l	(Winther et al., 2009)

Table A.10 LCI for the production of 1000 kg fish ensilage

	Value	**Unit**	**Reference**
Output			
Fish ensilage	1000	kg	
Input			
Fish by-product	966.2	kg	(Tatterson and Windsor, 2001)
Formic acid	33.8	kg	(Tatterson and Windsor, 2001)
Electricity	119.3	kWh	(Humbird et al., 2011)