Climate Impact and Energy Balance of Emerging Biorefinery Systems

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Abstract

Use of fossil fuels is the main contributor to anthropogenic emissions of greenhouse gases (GHG). Biorefineries, which are facilities that produce a set of valuable products from biomass, have been suggested as alternatives to fossil refineries, for the production of fuels, chemicals and materials. Emerging biorefineries are introducing new technologies, which can lead to increased use of biomass not previously utilised for industrial processes, such as harvesting residues from agriculture and forestry. Biomass is a renewable resource, but production and processing of biomass are associated with environmental impacts.

This thesis examined the climate impact and energy balance of emerging biorefinery systems, paying particular attention to the use of residues as feedstock. Three biorefinery systems were assessed and compared, all producing transportation fuels in combination with different co-products. These systems were: (1) co-production of ethanol, biogas, electricity and heat from straw in a lignocellulosic biorefinery; (2) co-production of ethanol, protein feed and briquettes from faba bean in a green crop biorefinery; and (3) co-production of biodiesel, biogas and electricity from straw in a lignocellulosic biorefinery. The analytical method used was life cycle assessment (LCA). Methodological issues when using LCA for assessing the climate impact of biorefinery systems were also discussed.

Ethanol and biodiesel produced from straw and forest residues in emerging biorefinery systems were found to have a lower climate impact and better energy balance than fossil fuels. Moreover, the biorefinery system producing ethanol and co-products from straw had a lower climate impact and more beneficial energy balance than that producing biodiesel and co-products. However, when using residues from agriculture and forestry or when harvesting the whole crop as biorefinery feedstock, specific consideration of effects on soil organic carbon is needed. The study on faba bean showed that using a biorefinery feedstock that is currently used for other purposes, such as feed, can cause indirect effects that affect the overall climate performance of the system. To improve the potential value of LCA studies on biorefinery systems, selection of functional unit, allocation method and treatment of biogenic carbon fluxes over time need further attention.

Keywords: Bioenergy, lignocellulosic biorefinery, green biorefinery, LCA methodology, GHG accounting, biofuel policy

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Klimatpåverkan och energibalanser av nya bioraffinaderisystem

Abstrakt

Användning av fossila bränslen är den främsta källan till antropogena utsläpp av växthusgaser. Bioraffinaderier är anläggningar som per definition producerar ett flertal värdefulla produkter från biomassa. För produktion a bränslen, kemikalier och material har bioraffinaderier föreslagits som viktiga framtida alternativ till fossila raffinaderier. Introduktionen av nya bioraffinaderier leder till att ny teknik används samt en ökad användning av biomassa som traditionellt inte har används för industriella processer i stor utsträckning, såsom skörderester från jordbruk och skogsbruk. Biomassa är en förnyelsebar resurs, dock är produktion och bearbetning av biomassa förknippad med miljöpåverkan.

Denna avhandling syftar till att bidra till en bättre förståelse av klimatpåverkan och energibalansen av nya bioraffinaderier, med ett särskilt fokus på användningen av jord- respektive skogsbruksrester som biomassa. Tre bioraffinaderisystem utvärderades, alla producerade transportbränslen i kombination med olika samprodukter. Systemen var: (1) samproduktion av etanol, biogas, el och värme från halm i ett lignocellulosa-bioraffinaderi; (2) samproduktion av etanol, proteinfoder och briketter från åkerbönor i ett grönt-bioraffinaderi. (3) samproduktion av biodiesel, biogas och el med halm som råmaterial i ett lignocellulosa-bioraffinaderi. Metoden som användes var livscykelanalys (LCA), och användningen av LCA för att studera bioraffinaderisystem diskuterades också.

Resultaten visar att etanol och biodiesel som produceras från halm och skogsrester i nya bioraffinaderier har lägre klimatpåverkan och mer fördelaktiga energibalanser än fossila bränslen. Bioraffinaderiet som producerar etanol med samprodukter från halm visade bättre klimatpåverkan och energibalanser än systemet som producerar biodiesel med samprodukter. När rester från jordbruk och skogsbruk används eller när man skördar hela grödan som bioraffinaderimaterial, krävs särskild försiktighet för att begränsa den negativa effekter på markkol. Studien på åkerböna visade att användning en gröda som bioraffinaderiråvara som för närvarande används för andra ändamål, kan ha indirekta effekter som är viktiga för klimatpåverkan från systemet. För att förbättra det potentiella värdet av LCA studier på bioraffinaderier bör hänsyn tas till val av funktionell enhet, allokeringsmetoder och hantering av biogena kolflöden över tid.

Nyckelord: Bioenergi, lignocellulosabioraffinaderier, gröna bioraffinaderier, LCA metodik, växthusgasbalanser, biodrivmedel, policy

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Dedication

To Douglas and Heidi

Contents

List of publications

This thesis is based on the work contained in the following papers, referred to by Roman numerals in the text:

I Ahlgren, S.*, Björklund, A., Ekman, A., Karlsson, H., Berlin, J., Börjesson, P., Ekvall, T., Finnveden, G., Janssen, M. & Strid, I. (2015). Review of methodological choices in LCA of biorefinery systems – key issues and recommendations. *Biofuels, Bioproducts and Biorefining* 9(5), 606-619.

II Karlsson, H.*, Börjesson, P., Hansson, P.-A. & Ahlgren, S. (2014). Ethanol production in biorefineries using lignocellulosic feedstock – GHG performance, energy balance and implications of life cycle calculation methodology. *Journal of Cleaner Production* 83, 420-427.

III Karlsson, H.*, Ahlgren, S., Strid, I. & Hansson, P.-A. (2015). Faba beans for biorefinery feedstock or feed? Greenhouse gas and energy balances of different applications. *Agricultural Systems* 141, 138-148.

IV Karlsson, H.*, Ahlgren, S,. Sandgren, M., Passoth, V., Wallberg, O. & Hansson, P-A. (2016). A systems analysis of biodiesel from wheat straw using oleaginous yeast: Process design, mass and energy balances. *Biotechnology for Biofuels* 9, 229.

V Karlsson, H.*, Ahlgren, S., Sandgren, M., Passoth, V., Wallberg, O. & Hansson, P-A. (2017). Greenhouse gas performance of biodiesel production from straw: Soil carbon changes and time-dependent climate impact. *Biotechnology for Biofuels* 10, 217.

Papers I-V are reproduced with the permission of the publishers.

*Corresponding author.

The contribution of Hanna Karlsson to the papers included in this thesis was as follows:

I Contributed to writing the paper, the discussions on recommendations given in the paper and the underlying literature study concerning mainly goal and scope definition, functional unit and allocation.

II Planned the study with the co-authors. Performed data collection and impact assessment and wrote the paper with input from the co-authors.

III Planned the study with the co-authors. Performed data collection and impact assessment and wrote the paper with input from the co-authors.

IV Planned the study together with the co-authors. Performed data collection, process simulation and assessments of energy balance, and wrote the paper with input from the co-authors.

V Planned the study together with the co-authors. Performed data collection and impact assessment and wrote the paper with input from the co-authors.

Abbreviations

ALCA	Attributional life cycle assessment
CH_4	Methane
CHP	Combined heat and power generation
CLCA	Consequential life cycle assessment
CO_2	Carbon dioxide
CO_2eq	Carbon dioxide equivalents
dLUC	Direct land use change
DM	Dry matter
EE	Energy efficiency ratio
EJ	Exajoule
FAME	Fatty acid methyl ester
FFRP	Fossil fuel replacement potential
FT-fuels	Fischer-Tropsch fuels
FU	Functional unit
GHG	Greenhouse gases
GWP	Global warming potential
Ha	Hectare
HVO	Hydrotreated vegetable oil
iLUC	Indirect land use change
ISO	International Standardisation Organisation
LCA	Life cycle assessment
LHV	Lower heating value
MJ	Megajoule
N_2O	Nitrous oxide
NER	Net energy ratio
PFAD	Palm fatty acid distillate
RED	Renewable energy directive
SOC	Soil organic carbon
TWh	Terawatt hour

1 Introduction

Climate change is one of the greatest environmental challenges of our time. Global actions are needed to keep the average temperature increase below 2 °C relative to pre-industrial levels and one such action is to substantially lower emissions from energy systems (IPCC, 2014). Fossil resources are the main contributor to climate change, with combustion of these resources creating around two-thirds of anthropogenic emissions of greenhouse gases (GHG) (IPCC, 2014).

Fossil resources are used in the energy sector and for producing multiple products, including fuels, chemicals and materials, *e.g.* plastics. The transport sector is the major user of fossil resources, with this sector alone using almost two-thirds of yearly crude oil production globally (IEA, 2015). Around 85% of the energy used in the Swedish transport sector is based on fossil resources, making it the sector with the highest fossil fuel dependency (SEA, 2016). The dependency on fossil resources is problematic, not only because of the environmental consequences, but also since fossil resources are finite. Therefore, good renewable alternatives with a lower environmental impact are needed.

Biomass can replace fossil resources in the production of fuels, chemicals and materials (Keegan *et al.*, 2013). In so-called biorefineries, biomass can be processed into many of the products that are currently produced from fossil resources. Biorefineries can therefore play a central role in creating a fossil-independent society. When biomass is used for feed and food, or for paper and pulp, in combination with new uses for fuels, chemicals and materials, demand for biomass will increase. Although biomass is considered a renewable resource, it is also a limited resource that requires efficient utilisation. The biorefinery concept, involving efficient processing of biomass, is gaining increasing interest for efficient utilisation of biomass (IEA, 2009; Kamm *et al.*, 2007).

Biomass is often considered carbon-neutral, since the carbon dioxide (CO_2) emitted during combustion was previously absorbed from the atmosphere by the plant. However, production and processing of biomass is associated with environmental impacts, including climate impacts and land use change. Land use change can lead to changes in biogenic carbon stocks, resulting in climate impacts. All of these aspects need to be evaluated in an environmental perspective. Globally, agricultural production and land use change are important sources of global anthropogenic GHG emissions (IPCC, 2014), indicating that biomass production can be associated with substantial climate impacts.

Conventional food and feed crops, such as wheat and maize, are the main feedstocks used for biofuel production today (REN21, 2017). The environmental gain of these so-called first-generation biofuels has been widely discussed in research and in the public debate (Brander *et al*., 2017). The main criticisms are the competing use for food and feed crops, the land use required for producing biofuels and the link to indirect land use change. As a consequence, lignocellulosic biomass has been suggested as an alternative feedstock to biofuel production. This type of biomass can be a residue from agriculture or forestry and therefore does not require additional land use.

Biofuels are used with the intention of decreasing the climate impact and fossil fuel dependency of the heavily fossil fuel-dependent transport sector. With the increased demand for lignocellulosic biomass to produce *e.g.* transportation fuels, there is a need for increased understanding of the climate effect of increased biomass harvesting, meaning whole crop harvesting or harvesting of straw and forest residues, on the overall climate impact of biofuels and biorefinery systems. In addition, new technologies and, to some extent, new products are being introduced with the emerging biorefining concepts. Increased insights into the environmental impacts from new biorefinery systems and use of residues can help decision and policy making towards more efficient use of available resources and can assist in meeting future climate and environmental targets.

Life cycle assessment (LCA) is a commonly used tool to assess the potential environmental impact of products and services. This approach can also be used to assess the environmental impact of biorefinery systems and their products and is currently used in policy for assessing the climate impact of biofuels. However, applying LCA to biological production systems is associated with several methodological challenges, including handling of land use change and changes in biogenic carbon stocks over time, and definitions of system boundaries. Moreover, in LCA studies on biorefineries that produce multiple products, definition of functional unit and allocation may be especially difficult. Hence, to improve LCA studies of biorefineries in both research and policy applications, method development and evaluation of LCA for use in assessing biorefinery systems are important.

2 Aim and structure

2.1 Aim

The general aim of this thesis was to provide a better understanding of the climate impact and energy balance of different emerging biorefinery systems producing transportation fuels in combination with different co-products. The main focus was on residues from agriculture and forestry as feedstock. Specific objectives were:

- To evaluate, in terms of climate impact and energy balance, three different biorefinery systems: (1) co-production of ethanol, biogas, electricity and heat from straw and forest residues in a lignocellulosic biorefinery; (2) co-production of ethanol, protein feed and briquettes from faba bean in a green crop biorefinery; and (3) co-production of biodiesel, biogas and electricity from straw in a lignocellulosic biorefinery.
- To analyse the effects of different methodological choices in LCA studies for biorefinery systems, by elaborating on how to handle co-products, selection of functional unit and biogenic carbon changes in LCA, and by discussing how LCA results can be evaluated and compared for different biorefinery systems.

2.2 Structure of the thesis

The structure of this thesis is illustrated in Figure 1. Based on the objectives (described above), two main themes were established, namely assessing the *greenhouse gas performance and energy balance of biorefinery systems and their products* and contributing to the discussion about *LCA methodology for biorefineries*. However, during the course of the work a third theme, *impacts due to increased biomass harvesting*[1] *on LCA results*, was introduced, since this was found to have a large effect on the climate impact. The three themes are represented by the light grey boxes in Figure 1. All papers included in this thesis cover at least one of these themes.

Paper I presents a review of LCA methodology for biorefinery systems and includes recommendations on critical methodical issues. Paper II assesses the GHG performance and energy balance of ethanol and biogas produced from lignocellulosic feedstock, using two different LCA calculation methodologies. Paper III describes a consequential LCA (CLCA) on the use of faba beans as biorefinery feedstock and on the impact of changed use of current faba bean production.

Papers IV and V are presented together in this thesis, since these publications refer to the same biorefinery system. Paper IV describes the process design, mass and energy balance of a newly developed biorefinery system that uses straw as feedstock for producing biodiesel by using oleaginous yeast. The greenhouse gas performance of the biodiesel and co-products is assessed in Paper V.

The third theme, *impacts due to increased biomass harvesting on LCA results*, is covered to different extents in Papers II, III and V. In Paper II, soil organic carbon and nitrogen removal are included, using literature values. Paper III assesses the impact of whole crop harvesting of faba bean by modelling the impact on soil organic carbon, including effects on nitrogen leaching, nitrous oxide emissions and nitrogen fertiliser demand. Paper V includes nitrogen removal and soil organic carbon changes due to straw harvesting. The soil organic carbon impact is modelled and a method to assess the time-dependent temperature impact from increased biomass harvesting on global mean surface temperature change over time is tested. Paper V also investigates to what extent soil organic carbon decrease can be mitigated by returning part of the lignin residues from the biorefinery to the field.

[1] *Increased biomass harvesting* covers: crop residue harvesting, forest residue harvesting and whole crop harvesting.

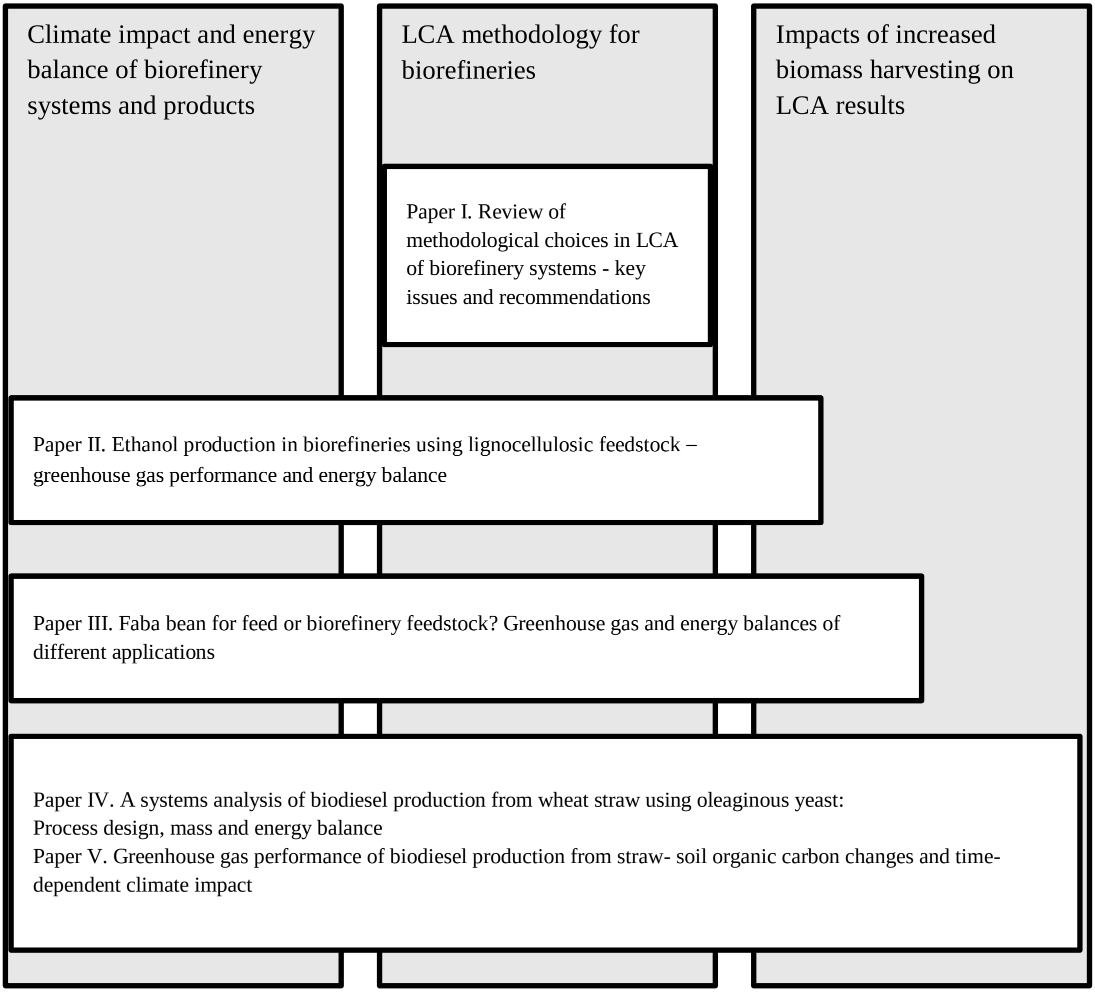

Figure 1. Illustration of the structure of the studies performed in this thesis work.

2.2.1 Definitions

Biorefinery systems

Several definitions of the term 'biorefinery' have been proposed, with the common feature that biorefineries are facilities for producing a spectrum of products from biomass. The definition by the International Energy Agency (Bioenergy Task 42 on Biorefineries), that biorefineries are "…the sustainable processing of biomass into a spectrum of marketable products and energy" (IEA, 2009, p. 7), is commonly cited.

The term *biorefinery system* as used in this thesis refers to the larger biorefinery system, including the foreground system with biomass production, harvesting and transport and the biorefinery plant, but also the background system supplying inputs of energy and materials. Furthermore, potential effects of replacing equivalent products are included in the LCA studies (Papers II, III and V) (Figure 2). This definition of biorefinery system is also used in Paper I. The term *emerging biorefinery systems* is used to describe newly or not yet commercialised biorefinery concepts. Further definitions of biorefinery systems are provided in section 3.3 of this thesis.

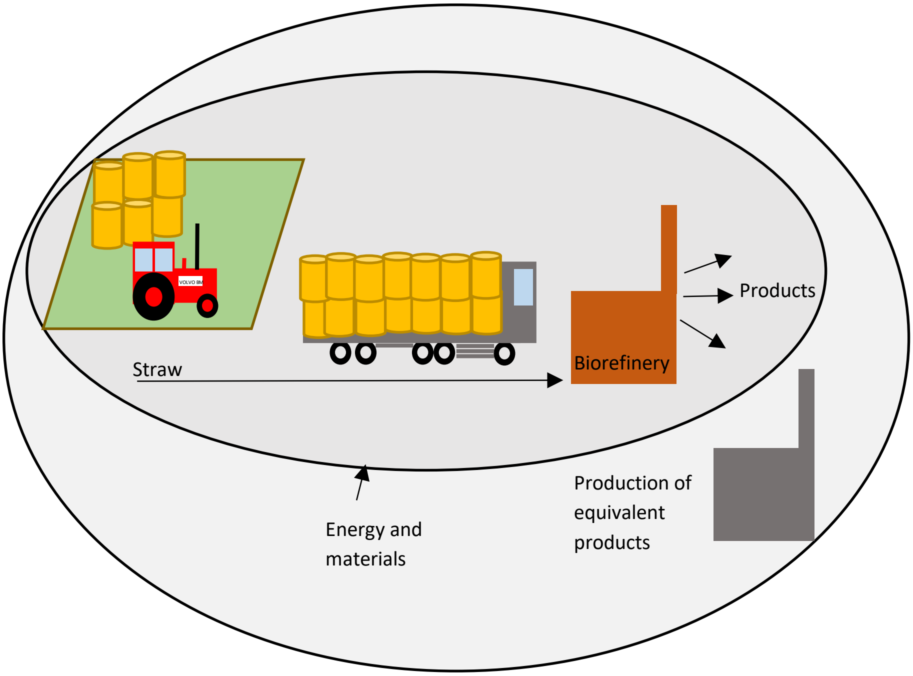

Figure 2. Illustration of the larger biorefinery system, including biomass production, harvesting and transport and effects on the surrounding system by substitution of equivalent products.

Straw and forest residues

The term *residue* is often used to describe biomass from agriculture and forestry that is not the main product, including straw and forest residues (tops and branches). Therefore this term is also used in this thesis. However, the term residue can indicate that the side-stream product is more or less a waste or that it lacks economic value. Therefore it is important to emphasise that crop residues are a valuable resource that has an economic value in many cases, and is also valuable in terms of carbon and nutrient sources for the production system from which it originates.

Biofuels

The term biofuels is used in this thesis to describe liquid and gaseous fuels produced from biomass, primarily intended for the transport sector. There are several types of biofuels produced from a variety of different forms of biomass. In this thesis, the term *first-generation biofuels* is used to describe fuels that are produced from primarily starch, sugar and oilseed crops. *Second-generation biofuels* are defined as fuels produced from lignocellulosic materials. These definitions are in line with those presented in Saladini *et al.* (2016).

Biodiesel

Diesel-like fuels produced from biomass are given different names in the literature, depending on process and feedstock. In this thesis, the term biodiesel is used for all diesel-like fuels produced from biomass. When needed, different biodiesel fuels are divided into: fatty acid methyl esters (FAME), rapeseed methyl esters (RME), fatty acid ethyl esters (REE), hydrotreated vegetable oils (HVO), dimethyl ether (DME) and Fischer-Tropsch diesel (FT-diesel).

Biogenic carbon

In this thesis, biogenic carbon is defined as carbon bound in biomass and in soil organic matter.

3 Background

3.1 Biomass

3.1.1 Biomass for bioenergy today

Globally, biomass is the largest renewable energy source, accounting for 10% of the energy supply (WEC, 2016). Fuelwood is the single largest biomass resource, comprising 68% of the energy supply from biomass, while liquid biofuel makes up a relatively small proportion of the total energy supply from biomass, *e.g*. ethanol (4%), biodiesel (2%), biogas (2%) and HVO (0.3%) (figures for 2013) (WEC, 2016).

In Sweden, biomass provides approximately 30% of the total energy supply (in 2014), and the majority of biomass is used directly in industry or for combined heat and power production (SEA, 2016).

3.1.2 Biomass potential

Biomass potential, especially for the bioenergy sector, has been estimated in many previous studies (see *e.g*. Creutzig *et al.*, 2015; BEE, 2011; Berndes *et al.*, 2003). Future biomass potential depends on several factors. Hoogwijk *et al.* (2003) listed six factors affecting biomass availability for bioenergy: future food demand depending on population growth and diet, future crop production systems, productivity in forestry and energy cropping, use of biomass for material production, amount of degraded land that can be used for bioenergy cropping and competing uses of land, such as using surplus land for reforestation.

Börjesson *et al.* (2013a) reviewed a number of studies on the potential for increased biomass production and harvesting for biofuel production in Sweden. The largest potential was found for stump harvesting and forest residues, while straw contributed approximately 4 TWh (0.014 EJ) out of an estimated total potential of 56-69 TWh (0.20-0.25 EJ) per year in a short-term perspective. In a longer-term perspective (30-50 years), the potential was estimated to be 80-98 TWh (0.29-0.35 EJ) per year or higher (177-195 TWh (0.64-0.70 EJ)), where the higher potential assumed increased stem wood production and fertilisation of forest land (Börjesson *et al.*, 2013a).

Estimates of future biomass potential for bioenergy in Europe also vary greatly, with one review reporting a variation from approximately 2.8 to 24 EJ by 2020 (BEE, 2011). Another review on global biomass potential found estimates in the literature ranging from less than 50 EJ per year to more than 1000 EJ per year, and concluded that there is good agreement that up to 100 EJ per year is the sustainable technical potential (Creutzig *et al.*, 2015). For comparison, total primary energy use globally was 400 EJ in 2015 (IEA, 2017).

Competition for use of biomass by other emerging sectors is sometimes not considered in biomass availability studies (Keegan *et al.*, 2013; Berndes *et al.*, 2003). Novel uses of biomass, in combination with conventional uses for food, feed, building material, pulp and paper *etc*., will increase the pressure on available biomass resources (Keegan *et al.*, 2013). Efficient use of available biomass resources is therefore crucial in meeting the future demands for biomass.

3.1.3 Liquid transportation fuels from biomass

Globally, the transport sector is dependent on fossil fuels to 90% and road transport is even more dependent, with fossil fuels representing 95% of total energy use (2012) (IEA, 2015). In 2015, biofuel use for road transport accounted for 2.6% of final energy use. Ethanol is the most important liquid biofuel globally, with 72% (in energy terms) of total production, while FAME accounted for 23% and HVO 4% of total biofuel production in 2016. Although second-generation biofuel production is now starting to be commercialised, the vast majority of biofuels are produced from starch, sugar and oilseed crops (REN21, 2017).

In Sweden, the share of biofuels is higher than the global average, *e.g.* in 2016 biofuel use in the Swedish transport sector was 18.8%, HVO is the most common biofuel, followed by biodiesel (FAME) and ethanol (SEA, 2017a). Use of HVO has increased greatly in Sweden during recent years, *e.g.* it increased 15-fold from 2015 to 2016 (SEA, 2017a). The HVO sold in Sweden is mainly produced from residues from the food industry (38%), while around 23% of the raw material is palm fatty acid distillate (PFAD), a residue from palm oil production. The three most common biofuels in Sweden (HVO, FAME and ethanol) are either imported as fuels or produced from imported raw material to a great extent (96% of raw material for HVO, 98% for FAME and 84% for ethanol was imported in 2016). Sweden has substantial domestic ethanol production and the reason for the low rate of domestically produced ethanol in the fuel mix is exports to Germany (SEA, 2017a).

To increase the use of residues and wastes in the production of liquid biofuels, these type of fuels are double counted towards the 10% goal in the Renewable Energy Directive (RED) of the European Union (EC, 2009). The effect of this regulation can now be seen in the Swedish biofuel mix. In 2011, 19% of the raw materials used for liquid biofuel production could be classified as residues or waste according to the RED, while the rate had increased to 66% in 2016 (SEA, 2017a).

3.2 Biorefinery systems

The term biorefinery system is defined in section 2.2.1.

3.2.1 Biorefinery classification

Considering all available biomass types, processing technologies and end-products, there are many ways in which a biorefinery can be structured. Biorefineries have been categorised differently (see *e.g.* Cherubini *et al.*, 2009; Kamm & Kamm, 2004). For example, Cherubini *et al.* (2009) suggest a nomenclature for biorefineries based on the platforms (intermediate products), feedstock and products. This nomenclature is suitable for describing individual biorefineries. To discuss and describe biorefinery concepts, the four types of biorefinery concepts suggested by Kamm and Kamm (2007) are applicable:

- Lignocellulosic biorefineries, using lignocellulosic feedstock such as straw, wood and grass
- Whole crop biorefineries, using *e.g.* whole crop cereals and maize
- Green biorefineries, using fresh or conserved non-dried biomass such as grass, clover, immature cereals and alfalfa
- A biorefinery two-platform concept, which includes a sugar platform and a syngas platform.

The concepts described above are not comprehensive and additional biorefinery concepts have been suggested, including conventional biorefineries (based on sugar and starch feedstocks), marine biorefineries, liquid-phase-catalytic processing biorefineries and forest-based biorefineries (Cherubini *et al.*, 2009).

Sections 3.2.2 and 3.2.3 describe in detail the two different biorefinery concepts covered in this thesis: *lignocellulosic biorefineries* and *green biorefineries*.

3.2.2 Lignocellulosic biorefineries

In a lignocellulosic biorefinery, lignocellulosic materials are used as feedstock to produce *e.g.* ethanol, chemicals or bioplastics. Lignocellulosic materials include *e.g.* straw, paper waste and forest residues, which are relatively inexpensive feedstocks (Kamm & Kamm, 2007). The lignocellulosic biorefinery is considered to be one of the most promising biorefinery concepts, for two main reasons: the relatively cheap substrate and the fact that the products which can be produced in lignocellulosic biorefineries are already established on the current market.

For lignocellulosic materials there are two main process routes, thermochemical and biochemical. The thermochemical process involves gasification or pyrolysis. The biochemical process route is outlined below, since it was studied in Papers II, IV and V.

Lignocellulosic materials consists of three main components: cellulose, hemicellulose and lignin. When processing lignocellulosic materials, it is important to gain access to the carbohydrates in the lignocellulose, in particular glucose, from which a wide variety of products can be produced (Kamm & Kamm, 2004) (Figure 3). Cellulose can be hydrolysed to glucose using enzymes or strong acids such as sulphuric acid. Hemicellulose can also be hydrolysed using enzymes (hemicellulases) or acids to yield a mix of pentoses and hexoses (xylose, arabinose, galactose, glucose and/or mannose) (Zheng *et al.*, 2009).

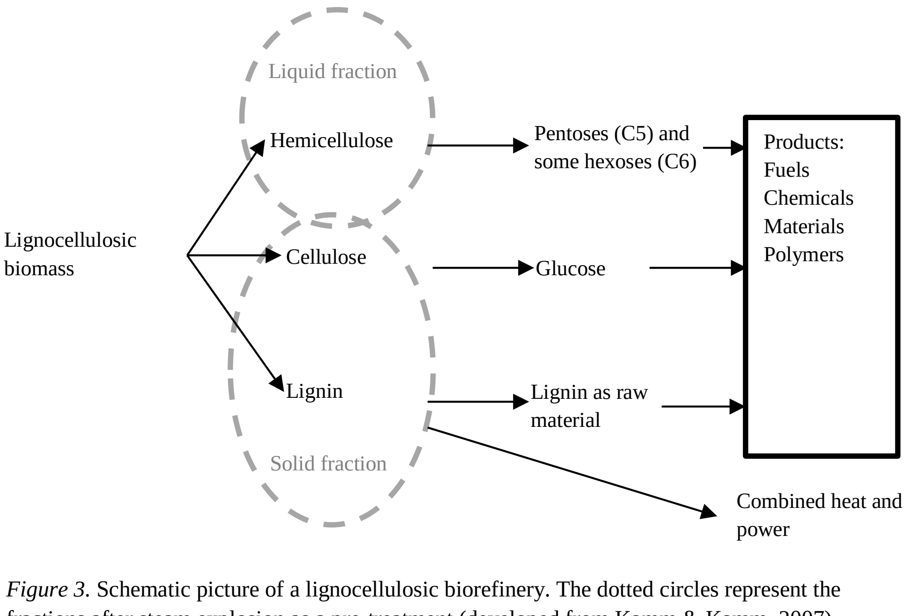

Figure 3. Schematic picture of a lignocellulosic biorefinery. The dotted circles represent the fractions after steam explosion as a pre-treatment (developed from Kamm & Kamm, 2007).

Before the hydrolysation step, the lignocellulosic biomass is pre-treated. The primary aim of the pre-treatment is to disrupt the structure of the biomass, so that it can be hydrolysed, *e.g.* by enzymes (Galbe & Zacchi, 2012). The monomeric sugars released can then be used for *e.g.* ethanol or lipid production using yeast. Lignin is a by-product from this process. Lignin is one of the three major polymers in biomass, and in woody biomass the lignin content may be as high as 30%. Today, most of the lignin generated industrially is from the pulp and paper industry. With the introduction of lignocellulosic biorefineries, substantial amounts of lignin would be generated and this lignin would have different chemical properties than that from the pulp and paper industry (Pye, 2010). Lignin can be used for the production of high-value products such as aromatic chemicals and fuels, which could improve the viability of lignocellulosic biorefineries (Azadi *et al.*, 2013).

3.2.3 Green biorefineries

In a green biorefinery, fresh or ensiled biomass is used to produce a variety of high-value products (Kromus *et al.*, 2010). Green crops (*e.g.* perennial grasses, immature cereals, legumes, forage leys *etc*.) are rich in carbohydrates, proteins, lipids and lignin. The yield may be as high as 20 metric tonnes (ton) of dry matter (DM) per hectare (ha) and year, and the protein harvest can be up to 4 ton/ha (Kromus *et al.*, 2010). Consequently, in a green biorefinery there is great potential to produce large amounts of protein and organic material that can be further processed into high-value products.

By processing green crops, the protein content, which is conventionally only accessible to ruminants, can be converted to a form that is accessible to humans and monogastric animals.

In the first step of processing, the green biomass is separated into a fibre-rich press cake and a nutrient-rich green juice (Kamm & Kamm, 2007) (Figure 4). The green juice contains proteins, among other compounds. Using different technologies, the protein can be separated out from the green juice. This can be done *e.g.* by heat, acid treatment, anaerobic digestion and centrifugation (Carlsson, 1997). Apart from protein products, other target products from the green juice include lactic acid and ethanol. The press cake can be further processed to *e.g.* feed pellets or syngas, or used for biogas production (Kamm & Kamm, 2007).

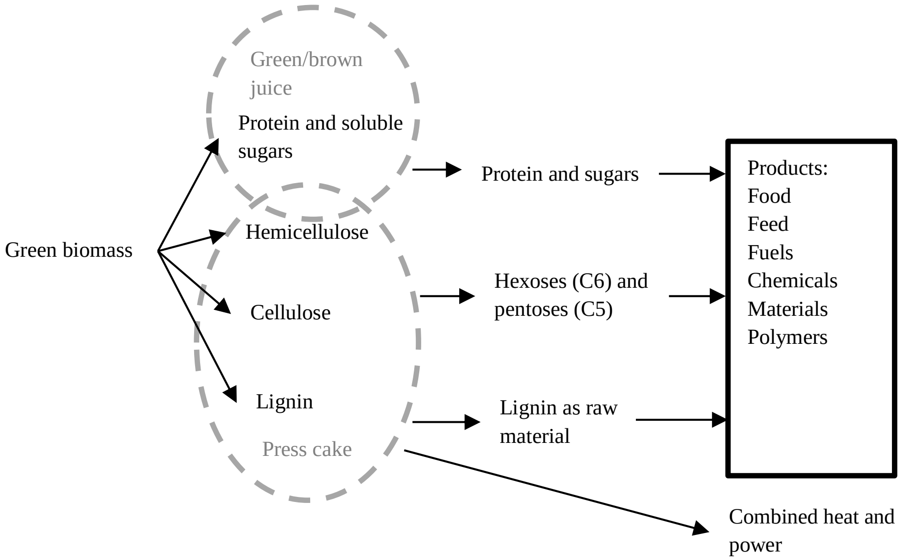

Figure 4. Schematic picture of a green biorefinery. The dotted circles represent the fractions after pressing the biomass (developed from Kamm & Kamm, 2007).

3.3 Life cycle assessment (LCA)

Life cycle assessment is a method for quantifying the potential environmental impact of a product or a service from cradle to grave, *i.e.* from resource extraction to waste management via manufacturing, transport, use and maintenance of the product or service. Among several environmental assessment tools available, LCA is unique for its focus on products and services while considering the whole life cycle (Finnveden *et al.*, 2009).

The LCA method is standardised in International Standardisation Organisation standards ISO 14040 and ISO 14044 (ISO, 2006b; ISO,

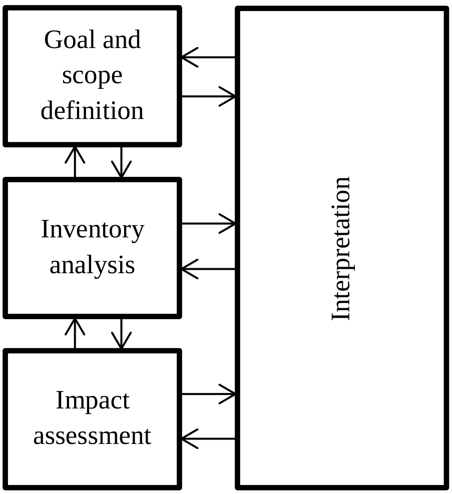

Figure 5. Schematic figure of the life cycle assessment (LCA) methodology.

2006a). Several steps are involved in LCA, including goal and scope definition, inventory analysis, impact assessment and interpretation (Figure 5). In the goal and scope definition step, the aim of the study is stated, together with specifications for the modelling. The inventory analysis is where data on resource use and emissions are collected. In the impact assessment step, individual emissions and resource use are grouped into different environmental impact categories by applying impact assessment methods. The purpose is to describe potential environmental impacts for different environmental impact categories. Interpretation of the results is carried out with regard to the initial aim of the study, the data and the impact assessment method used.

At least two different modelling approaches in LCA can be distinguished, namely accounting (or attributional) LCA (ALCA) and consequential LCA (CLCA). ALCA describes all immediate physical flows to and from a life cycle (Ekvall & Weidema, 2004) and one common use of ALCA is in product declarations and hotspot analysis (Weidema, 2003). CLCA "aims at describing how the environmentally relevant physical flows to and from the technical system will change in response to changes in the life cycle" (Ekvall & Weidema, 2004, p. 161). CLCA is suitable for assessing the impact of changes, for example from the current situation to potential future situations.

3.3.1 LCA of biorefineries

Life cycle assessment is commonly used to assess the environmental performance of bioenergy and biorefinery systems. It is well known that the results depend not only on the production system itself, but also on methodological choices (see *e.g*. Borrion *et al.*, 2012; Börjesson & Tufvesson, 2011; Whitaker *et al.*, 2010; Gnansounou *et al.*, 2009). Paper I identifies six key issues for LCA studies on biorefinery systems: (1) goal definition, (2) choice of functional unit (FU), (3) allocation issues with biorefinery outputs, (4) allocation issues with the production of biomass feedstock, (5) land use and (6) biogenic carbon and timing of emissions. Some of these key issues are more general and applicable to basically all LCA studies, such as goal definition, choice of functional unit and allocation issues (*i.e*. partitioning of the environmental impact between co-products), while others relate more specifically to the use of biomass, in particular land use issues, including indirect land use changes, biogenic carbon changes and timing of emissions.

The term biorefinery *per se* implies that more than one product is generated in the same production plant. When analysing biorefineries using LCA, two of the general key issues are therefore particularly relevant due to the multi-functionality of the system. First, the choice of functional unit becomes very important. The functional unit is the function of the system under study and serves as a basis for the calculations. For biorefineries, it can be difficult to identify one main product or function (Paper I). Second, an allocation or multi-functionality problem arises when several products or services share or partly share a production system. The fact that biorefineries do not produce a main product, but rather a set of valuable co-products that can have different functions and physical attributes, can complicate the handling of multi-functionality problems (Paper I). The biomass used in emerging biorefineries is often a residue or in some cases can be categorised as a waste. For this reason, allocation is also important for the biomass used in a biorefinery (Paper I).

General principles for handling allocation problems in LCA are specified in the ISO standard (2006b):

- If possible, allocation of the environmental impact between co-products should be avoided. This can be done by increasing the level of detail in the modelling (identifying product-specific flows) or by system expansion.
- If allocation cannot be avoided, the multi-functionality problem can be handled by first partitioning the inputs and outputs based on physical relationships between the products. If this cannot be done, partitioning should be based on other characteristics such as economic value, mass or energy.

3.3.2 LCA methodology in biofuel policies

Life cycle assessment is used in biofuel policies, for example the European Union's Renewable Energy Directive (RED) (EC, 2009), the UK Renewable Transport Fuel Obligation, the US Environmental Protection Agencies Renewable Fuel Standard and the California Air Resources Board's Low Carbon Fuel Standard (McManus *et al.*, 2015). In policies, LCA is used mainly for GHG accounting, *i.e.* to assess whether a biofuel gives climate impact savings in relation to fossil fuels. The use of LCA for this purpose accentuates several important issues such as land use, market effects and time aspects in relation to *e.g.* carbon storage and technological development (McManus & Taylor, 2015). These are examples of aspects that the original LCA was not designed to deal with and LCA use in biofuel policy is therefore associated with several challenges (McManus *et al.*, 2015). McManus and Taylor (2015) argue that biofuel policies are driving many of the changes seen in current LCA methodology development, particularly the increased use of consequential LCA studies, including indirect land use change (iLUC) and prospective studies looking at future scenarios. This is exemplified by the much debated issue of land use and indirect land use change focusing on biofuel production, despite land use being a much wider issue that is related to land management and to production of food and particularly animal feed.

The present thesis focuses on the RED (Papers II and V), and therefore it is described in brief below. In the RED, LCA methodology is applied for GHG accounting of biofuels. The RED includes a mandatory target for biofuel use in the transport sector of 10% of total energy consumption in the EU by 2020 and GHG reduction requirements from a fossil fuel reference including the current requirement of 50% and forthcoming 60% reduction requirements in 2018 for installations that started operation after January 2017 (EC, 2009). The RED is currently under revision and a proposal has been presented (EC, 2017b). Some of the changes to the original directive include: the inclusion of GHG reduction targets for solid biomass used for heat and electricity, removal of the 10% target for the transport sector, a cap on biofuels produced from food and feed crops, continued promotion of advanced biofuels and the introduction of a new fossil reference (EC, 2017b). In the proposal for a new RED, the reduction targets are set to 50% for installations in operation before October 2015, 60% for installations starting operation from October 2015 and 70% for installations starting operation after January 2021 (EC, 2017b).

The method used to calculate the climate impact of fuels is based on the LCA methodology, with standardised procedures for system boundaries, functional unit and allocation. The GHG performance is included in the sustainability criteria for liquid and gaseous biofuels listed in the RED and these criteria must be met in order for the biofuel to count towards the target (10% target). Due to this, the calculation method in the RED is potentially highly influential for the European biofuel market.

Although CLCA has been argued to be the most suitable LCA approach for policy applications (Brander, 2017; McManus & Taylor, 2015; Plevin *et al.*, 2014), the LCA method in RED is largely based on an ALCA approach. However, the RED is moving towards a consequential approach, for example iLUC factors have been introduced into the directive (EC, 2015).

3.3.3 Assessing biogenic carbon stock changes in LCA

The amount of soil organic carbon (SOC) depends on carbon inputs and decomposition rate, and the balance between inputs and decomposition is altered when a larger proportion of the crop is harvested. Losses of SOC increase GHG emissions from the system, but also affect the long-term productivity (Cowie *et al.*, 2006). There are three mechanisms by which higher biomass removal influences SOC (Cowie *et al.*, 2006):

- Lower biomass input results in a decrease in SOC
- Biomass contains nutrients and therefore lower biomass input decreases nutrient availability, leading to lower productivity and resulting in lower biomass residue input from roots *etc*.
- Lower SOC in itself decreases soil productivity.

Biogenic carbon is commonly considered to be climate-neutral in LCA, as it is assumed that the carbon from CO_2 sequestered during growth of the biomass equals the carbon released when the biomass is combusted. However, this assumption has been questioned for being too simplistic, since there is a time lag between CO_2 sequestration and CO_2 release (Brandão *et al.*, 2013). This becomes especially relevant for biomass systems with long rotation times. The terrestrial environment, *e.g*. vegetation and soil, is one of the major reservoirs of carbon, containing around 2-3 times the amount of carbon in the atmosphere (of which two-thirds is in the soil) (Houghton, 2003). Therefore it is perhaps not surprising that LCA studies which include changes in carbon stocks in soils and living biomass often find that these changes have a great influence on the climate impact (see *e.g*. Hammar *et al.*, 2014; Whittaker *et al.*, 2014; Cherubini & Jungmeier, 2010).

However, including changes in carbon stocks in soils and living biomass in LCA studies is challenging, for two main reasons. First, LCA normally accounts for point emissions that occur within the same year, so gradual emissions and uptake that can occur over decades, as in the case of SOC changes or a growing forest, are difficult to assess using common LCA methodology. Although several methods have been proposed to improve assessment of the climate impact of changes in biogenic carbon pools, there is no consensus on how to handle this in LCA (Brandão *et al.*, 2013). Second, SOC changes are long-term processes occurring over many years and there is a lack of data on SOC changes over time. For this reason, SOC changes are often modelled. There is no generally accepted method for estimating SOC changes in LCA and different methods are currently used, including emission factors, simple models, dynamic crop-climate-soil models and measurements (Goglio *et al.*, 2015). Soil organic carbon has been found to be one of the most important sources of uncertainty in biofuel LCAs (Whitaker *et al.*, 2010).

Apart from climate effects, decreases in SOC have an impact on soil quality and, in the long run, the productivity of the soil, and therefore wide-scale harvesting of crop residues has been questioned (Lal, 2004). The effect on soil quality was not examined in this thesis, but it is of critical importance for the long-term sustainability of agroecosystems.

3.4 Climate impact and energy performance of bioenergy and biorefinery systems

3.4.1 Biofuels on the market today

There are numerous LCA studies on first-generation biofuels produced from oilseed, sugar and starch crops. Many of these studies show a favourable climate impact and energy balance for first-generation biofuels compared with fossil fuels, especially when effects of direct land use changes (dLUC) and indirect land use changes (iLUC) are excluded from the assessment (see *e.g.* Edwards *et al.*, 2014; Khatiwada *et al.*, 2012; Souza *et al.*, 2012; Wang *et al.*, 2012; Börjesson *et al.*, 2010; Dias De Oliveira *et al.*, 2005). When dLUC and iLUC are included, studies sometimes show a less favourable or even negative climate impact for first-generation biofuels compared with fossil fuels (Malça *et al.*, 2014; Dunn *et al.*, 2013; Hertel *et al.*, 2010; Searchinger *et al.*, 2008). Land use change effects, combined with concerns about competition between food and fuels, are the main criticism of first-generation biofuels. This, and the relatively high cost of food and feed crops, has led to a search for new raw materials for biofuel production.

3.4.2 Lignocellulosic biofuels

A review by Morales *et al.* (2015) found that second-generation ethanol (produced from lignocellulose) has a more favourable climate impact and energy balance than fossil fuels and first-generation ethanol. Reviewing 53 studies mainly on biochemical conversion of lignocellulose to ethanol, Borrion *et al.* (2012) found climate impact reductions for lignocellulosic ethanol of 46-90% compared with fossil fuel, with energy savings ranging from 56% to nearly 100%. Alongside biochemical conversion, the other main process route to produce liquid biofuel from lignocellulose is thermochemical conversion, involving the Fischer-Tropsch process. The climate impact of Fischer-Tropsch diesel is estimated to be 61-115% lower than that of fossil diesel (Sunde *et al.*, 2011).

Lignocellulosic biomass can be either a dedicated energy crop or a residue such as straw and forest residues. Dedicated energy crops are associated with land use and can thereby be associated with direct and indirect land use changes. Residues, on the other hand, are not associated with dedicated land use and thereby not linked to possible indirect land use change. Consequently, all lignocellulosic biomass (both dedicated energy crops and residues) can result in direct land use change, meaning that it can cause changes in biogenic carbon stocks and in SOC. In the case of harvest residues, a larger proportion of the target crop in agriculture and tree in forestry is harvested (Papers II, III and V), which affects the carbon balance as it decreases the amount of carbon added to the soil. When these effects are included in assessments of lignocellulosic biofuels, they have a large influence on the overall climate impact (Liska *et al.*, 2014; Whittaker *et al.*, 2014) and the effects of biorefinery systems (Cherubini & Ulgiati, 2010).

3.4.3 Biorefinery systems

Assessing the climate impact of entire biorefinery systems, and not allocating impacts between the different co-products, is sometimes done in order to *e.g.* perform hotspot analysis (González-García *et al.*, 2011) or identify the best use of side-streams in biorefineries (Gilani & Stuart, 2015). When studying the whole biorefinery (without allocating), the portfolio of products can be compared with a reference system with conventionally produced products (Cherubini & Jungmeier, 2010).

4 Methods

4.1 System descriptions

The three different biorefinery processes studied in this thesis are described below. Two of the biorefineries used lignocellulosic biomass and can therefore be classified as lignocellulosic biorefineries. Whole crop faba bean was used as feedstock in Paper III. The faba bean was not dried but ensiled before being processed, so this concept may be classified as a green biorefinery. All biofuel production systems included were biochemical processes, meaning that microorganisms were used to convert the substrate to valuable products (in this case sugars to fuels).

The following sections describe the methods used in Papers II-V in more general terms. For detailed descriptions, please see the respective papers.

4.1.1 Lignocellulosic biorefineries

Ethanol, biogas and electricity from straw and forest residues

The biorefinery system studied in Paper II is illustrated in Figure 6. Straw and forest residues were used as feedstock, and were analysed in two different scenarios. The feedstock was first impregnated with diluted acid and pre-treated using steam explosion, followed by simultaneous saccharification and fermentation where hemicellulose and cellulose were converted to sugars using enzymes and ethanol was produced using yeast and then distillation to separate out the ethanol. The liquid remaining after distillation and filtration was anaerobically digested to produce biogas. In the scenario using forest residues, the liquid fraction from the pre-treatment containing the majority of the pentose sugars was assumed to be fed directly to the anaerobic digester, as represented by the dotted line in Figure 6. In the scenario using straw, all sugars, hexoses and pentoses, were assumed to be fermented into ethanol, although in practice this would require a genetically modified yeast strain that can utilise both hexoses and pentose sugars for ethanol production. The solid fraction containing most of the lignin was combusted in a combined heat and power (CHP) plant to supply heat and electricity for the refinery. Both scenarios generated excess heat that could be used *e.g*. in district heating and excess electricity that could be sold.

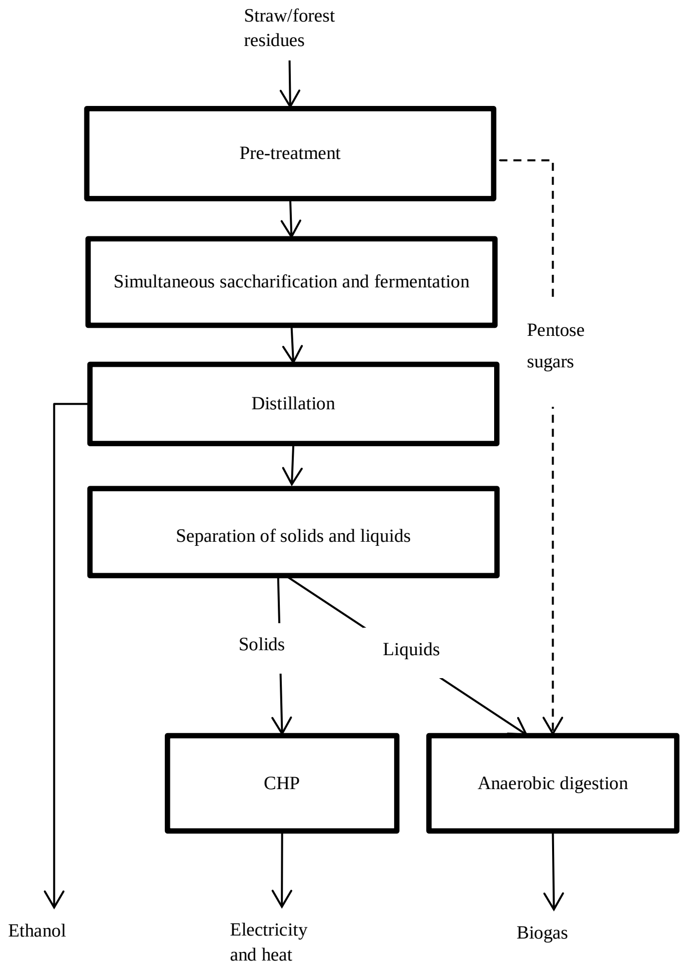

Figure 6. The biorefinery system considered in Paper II (modified from Börjesson *et al*., 2013b). The dotted arrow represents feeding of pentose sugars from the pre-treatment directly to the anaerobic digestion (forest residue scenario) as opposed to all sugars going to the hydrolysis and fermentation (straw scenario).

Biodiesel, biogas and electricity from straw

The biorefinery system studied in Papers IV and V is illustrated in Figure 7. Paper IV studied plant design and the energy balance of biodiesel produced from straw, while Paper V assessed the climate impact of the same system. The straw was first pre-treated using steam explosion of dilute acid-impregnated straw, followed by enzymatic hydrolysation (saccharification). The solid fraction containing most of the lignin was then separated out and combusted in a CHP plant, to produce electricity and heat required in the process. The sugars were used to produce lipids, using oleaginous yeast grown in a bioreactor for lipid accumulation (Figure 7). Under nitrogen limitation, many oleaginous yeast can naturally utilise both pentoses and hexoses to accumulate lipids, and therefore all sugars were assumed to be used for lipid production. The lipids were extracted using hexane and the lipids were transesterified to produce FAME

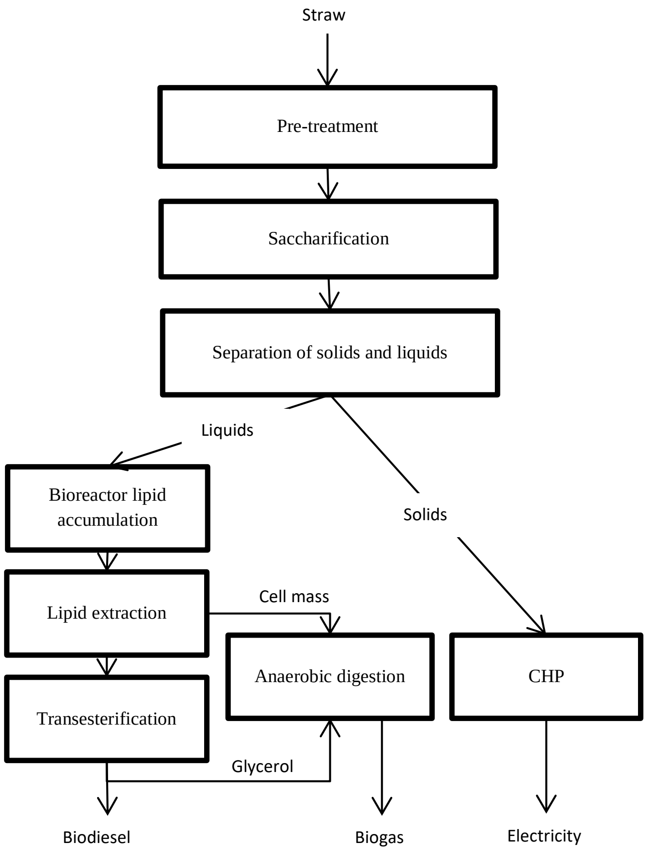

Figure 7. The biorefinery system considered in Papers IV and V.

(biodiesel). The cell mass remaining after lipid extraction was anaerobically digested, together with the glycerol generated in the transesterification process, to produce biogas. The solid fraction remaining after liquid/solid separation contains most of the lignin and was assumed to be combusted in a CHP plant. This generated all process electricity and heat needed in the refinery, and excess electricity that could be sold.

4.1.2 Green biorefineries

Paper III analysed the climate impact, land use and energy balance of a green biorefinery system using ensiled faba bean as feedstock. The biorefinery concept is illustrated in Figure 8. In the plant, the beans were first separated from the rest of the plant mechanically. The starch and protein in the beans were then separated and the starch was used for ethanol production and the protein for animal feed. The remaining biomass was rolled to extract a juice that is high in protein, which was used as animal feed. The press cake remaining after rolling was used for production of solid biofuels in the form of briquettes.

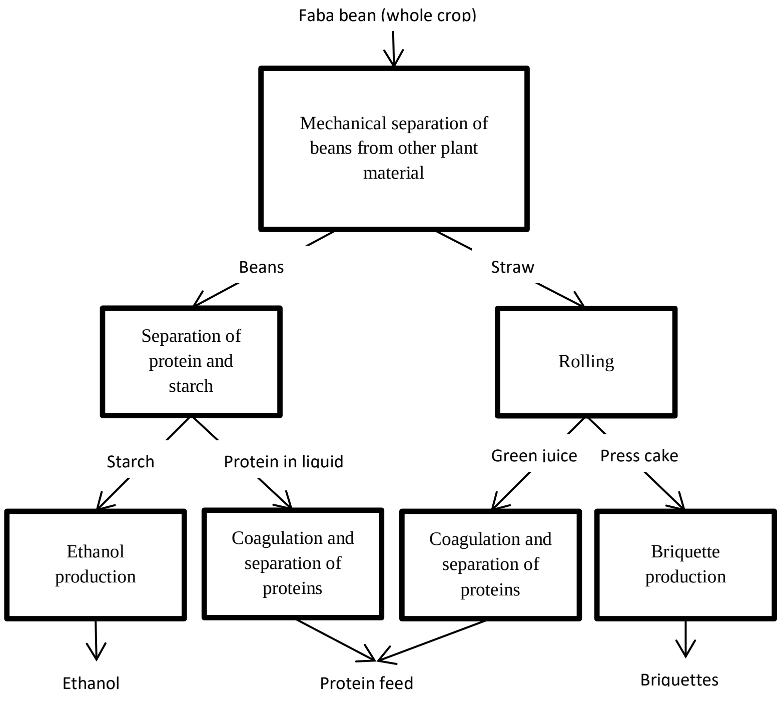

Figure 8. The green crop biorefinery considered in Paper III (Figure from Paper III).

4.1.3 Biorefinery design and performance

In Paper II, the plant design, input requirements and production of energy carriers were based on two earlier studies: Ekman *et al.* (2012) for the straw scenario and Barta *et al.* (2010) for the forest residues scenario. In Paper III, the whole biorefinery system, including the plant, was modelled in Microsoft Excel 2013. The biorefinery plant in Papers IV and V was modelled in Aspen Plus™ (see Paper IV) and the greater surrounding system was modelled in Microsoft Excel 2013 (Papers IV and V). Energy Analyser™ was used to model the heat exchange system in the biorefinery (Papers IV and V).

4.2 LCA methodology

Different LCA methods were used in the different papers. A summary of method choices is presented in Table 1.

Table 1. *Life cycle assessment (LCA) method applied in Papers II, III and V. RED = Renewable Energy Directive, ISO = International Standardisation Organisation, CLCA= consequential LCA, ALCA = attributional LCA*

	Paper II	**Paper III**	**Paper V**
Type of LCA	RED & ISO[a]	CLCA	ALCA & RED
Functional unit	1 MJ ethanol	1 ha faba bean cultivation	1 kg of straw and 1 MJ biodiesel
Handling of multifunctionality	Energy allocation & system expansion	System expansion	Energy allocation[c] & GHG substitution potential
Choice of data	Average	Marginal	Average
System boundaries	Cradle to gate[b]	Cradle to gate	Cradle to gate[b]

[a]This method uses the preferred method to handle multifunctionality in LCA according to ISO (ISO, 2006b), system expansion and includes upstream impacts from residue harvesting.
[b]In the RED method, residues from agriculture and forestry are considered to be 'free' up to the point of harvest.
[c]Used for allocating impact between the energy carriers produced when the function unit 1 MJ biodiesel was used.

In all papers (II-V), the LCA model was built in Excel 2013.

4.2.1 Goal and scope of the LCA studies

Methodological choices in LCA are guided by the objective of the study. The objective in Paper II was to estimate the climate impact and energy balance of ethanol produced from two different feedstocks and to analyse the impact of using two different calculation methods. One of the calculation methods was based on the RED (described in section 4.2.2.) (EC, 2009) with Method I (ISO), which uses system expansion to avoid allocation in accordance with ISO (ISO, 2006b) and includes upstream impacts from harvesting residues. The intended audience and application were companies looking for guidance on how the RED calculation method is applied and policy makers reviewing the impact of methodological choices in climate impact assessments.

The objective of Paper III was to assess the climate impact and change in arable land use and fossil energy use of changing from the current use of faba bean as a protein feedstuff to two types of whole faba bean plant utilisation: biorefinery processing and roughage feed. The specific objective was to determine the most environmentally beneficial use of available faba bean production of the three scenarios assessed and with regard to the three impact categories assessed. Since the aim was to analyse impacts of a change in use of the same biomass, CLCA was applied. In CLCA, allocation is generally avoided by system expansion and data from the technologies affected (marginal data) should be included, rather than average data. The intended audience was researchers interested in CLCA and its applications, but also biorefinery owners, faba bean producers and policy makers.

The objective of Paper V was to assess the climate impact and energy balance of biodiesel produced from straw using oleaginous yeast. The study focused on whether the return of parts of the lignin fraction could mitigate the effect of SOC change due to straw harvesting, which was analysed in different scenarios. ALCA was judged as being the most suitable method for the goal of the study. In addition, to my knowledge Paper V is the first LCA study on biodiesel produced from straw using oleaginous yeast, which is why it was interesting to identify hotspots in the system. For this purpose, ALCA is most suitable (Paper I). The intended audience was policy makers within biofuels and biorefineries and researchers within LCA and biorefinery process and design.

4.2.2 Renewable energy directive methodology

The calculation method from the RED (EC, 2009) was used in Paper II and Paper V. To facilitate comparison between different fuels, the RED has standardised procedures for setting the functional unit (1 MJ biofuel), system boundaries and energy allocation (based on the lower heating value (LHV) of the products). The LCA calculation method assumes that crop and forest residues are free from impact up to harvesting. For the systems studied in Papers II and V, climate impact was also calculated using the RED method, but including upstream impact from residue harvesting in the form of SOC changes and compensation for nitrogen removed with the residues. This method was called RED+SOC in this thesis.

4.2.3 Climate impact

Global warming potential

The commonly used climate impact indicator Global Warming Potential (GWP) was used in Papers II, III and V. It describes the cumulative radiative forcing of a pulse emission of a greenhouse gas relative to the cumulative radiative forcing of CO_2 for a specific time period, (most commonly 100 years (GWP_{100}) (Fuglestvedt *et al.*, 2003).

Time-dependent climate model

A time-dependent climate model described by Ericsson *et al.* (2013) was used in Paper V. The model accounts for the timing of emissions (or uptake) of the three major greenhouse gases (CO_2, nitrous oxide (N_2O) and methane (CH_4)) and estimates climate impact as temperature response over time. Yearly emissions were estimated over 100 years (Paper V). For straw used for liquid fuel production, uptake and release of CO_2 in living biomass were assumed to occur during the same year, and therefore these changes were not accounted for. However, changes in SOC due to straw harvesting occur over several years and were estimated by modelling (see section 4.3.1 of this thesis).

4.2.4 Energy balance indicators and GHG substitution potential

There are several energy performance indicators available (Djomo *et al.*, 2011). In Papers IV and V, three different energy balance indicators were used to show the energy balance of the system in different perspectives (Figure 9). These were: 1) Energy efficiency ratio (EE), calculated as energy carriers produced (LHV) divided by the energy in the feedstock (LHV), which shows the proportion of the energy in the feedstock that is converted to the final product; 2) net energy ratio (NER), calculated as total primary energy input divided over the energy carriers produced, which shows the amount of fossil energy used in production of the biofuel; and 3) fossil fuel replacement potential (FFRP), calculated by subtracting the primary energy in the products that could potentially be replaced by the biofuel(s) from the total use of primary fossil energy in the whole production chain for 1 kg of dry matter (DM) feedstock input into the biorefinery. A positive value of FFRP indicates that use of fossil energy in the biorefinery system exceeds the bioenergy produced, while a negative value indicates the proportion of fossil fuels that could be replaced. In Paper II the energy balance was calculated, using the NER indicator (Method II RED).

The three different energy balance indicators were calculated as follows (see also Figure 9, taken from Paper IV):

$$EE = (E_{prod1} + E_{prod2} + E_{prod3}) / E_{biomass}$$
$$NER = E_{inputs} / (E_{prod1} + E_{prod2} + E_{prod3})$$
$$FFRP = E_{input} - (E_{repl1} + E_{repl2} + E_{repl3})$$

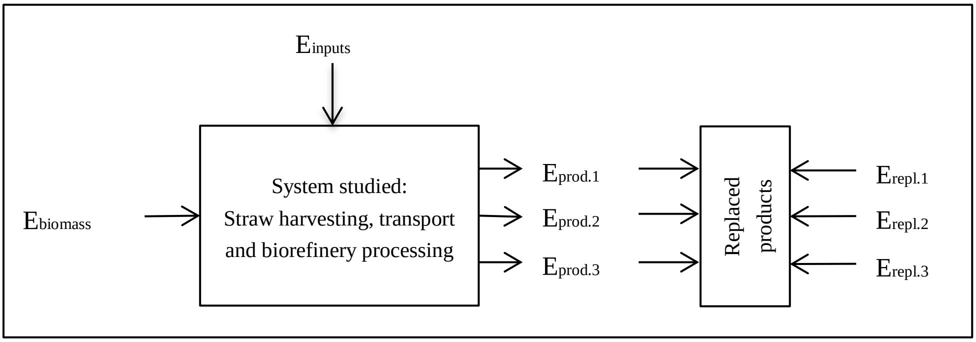

Figure 9. Variables for energy balance indicators used in Paper V. $E_{biomass}$ and $E_{prod.1-3}$ are given in lower heating value (LHV) and $E_{repl.1-3}$ are given in primary fossil energy.

Paper V used an indicator called substitution potential. For clarity, it is referred to here as GHG substitution potential. It was calculated by subtracting the impact of the reference system from that of the system studied (Figure 10). The reference system in this case represents a collection of products equivalent to the products produced in the biorefinery. A negative value for the GHG substitution potential indicates that the system under study has a lower climate impact than the reference system.

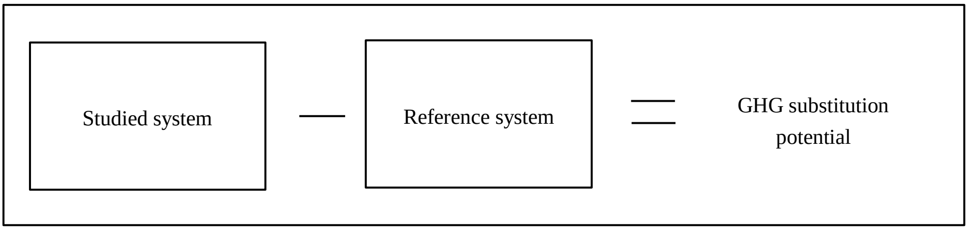

Figure 10. Method used to calculate the greenhosue gas (GHG) substitution potential of emissions from the reference system, with equal amounts of conventional comparable products deducted from the emissions of the studied system.

4.3 Methods for assessing the effects of increased biomass harvesting

4.3.1 Soil organic carbon changes

Soil organic carbon changes resulting from increased biomass harvesting, *i.e.* using a larger proportion of the crop or tree, were accounted for in Papers II, III and V. In Paper II emission factors from the literature were used, while in Papers III and V the Introductory Carbon Model (ICBM) (Andrén & Kätterer, 1997) was used to model SOC losses due to increased biomass harvesting. The ICBM is a two-compartment model that considers two soil carbon pools, one young and one old. The model can be adapted to handle different types of biomass with different humidification rates. Paper III used three types of biomass, namely aboveground and belowground crop biomass from faba bean and manure. Paper V also included three different types of biomass, aboveground and belowground biomass of wheat and the lignin residues from the biorefinery process.

Apart from estimating the effect on soil carbon contribution from different biomass fractions, another advantage with using a soil carbon model is that it allows site-specific factors such as climate to be accounted for. Furthermore, it gives yearly changes in the carbon pool so that climate impact effects over time can be estimated (Paper V). This was used in the time-dynamic climate modelling described in section 4.2.2 of this thesis. In the GWP calculations in Paper III and Paper V, results from the ICBM were used to calculate emission factors, *i.e.* dividing the total loss of carbon over a selected number of years to get an estimate of yearly emissions over that period.

4.3.2 Nutrient losses

In comparison with the reference land use with no residue harvesting, more carbon but also more nutrients are removed from the field or forest when residues are harvested. In Paper II and V this was handled by compensating for all the nitrogen removed in the biomass.

In Paper III, where CLCA that included the whole agricultural system of faba bean production and other crops in the crop rotation was performed, the following effects on the nitrogen cycle were included: 1) Effects on direct (and indirect) N_2O losses; 2) effects on nitrogen leaching; and the nitrogen fertiliser effect on the succeeding crop.

4.4 Scenarios

In order to help the reader to follow the results and discussion presented in Chapter 5, the different scenarios used in Papers II-V are briefly summarised in Table 2.

Table 2. *Description of the scenarios used in Papers II-V.*

	Scenario	Description
Paper II	Straw	Ethanol production in a lignocellulosic biorefinery co-producing ethanol, biogas, electricity and heat using straw as feedstock
	Forest residues	Ethanol production in a lignocellulosic biorefinery co-producing ethanol, biogas, electricity and heat using forest residues as feedstock
Paper III	Base case	Use of faba beans (only the beans) as cattle feed (scenario 1)
	Biorefinery	Whole crop harvesting and use of the whole faba bean plant in a green biorefinery (scenario 2)
	Roughage	Whole crop harvesting and use of the whole faba bean plant as roughage for cattle (scenario 3)
Paper IV	Base case	Biodiesel, biogas and electricity produced from straw
	DRY 10/5%	Drying the yeast before lipid extraction, with 10% or 5% lipid loss
	LIPID 60/40%	Assumed lipid content of the yeast after lipid accumulation phase of 60 and 40% (base case assumed 50%)
	SUGAR+/-10%	Sugar concentration in the hydrolysate varied by +/- 10%
	TIME+/-1	Residence time for lipid accumulation varied by +/- 1 day in relation to base case
Paper V	Base case	Biodiesel, biogas and electricity produced from straw as described in Paper IV
	No excess el	Combusting only the lignin needed to satisfy plant demand for electricity. The remaining lignin was returned to soil
	Biogas for internal H&P	Biogas combusted to meet heat and electricity demand in the plant
	External el prod.	Combusting only the lignin needed to satisfy plant demand for heat. The remaining lignin was returned to the soil and electricity was produced from natural gas

5 Results and discussion

In this section, the three themes of the thesis (Figure 1) are dealt with in three separate sections. Section 5.1 presents and discusses the climate impact and energy balance of the biorefinery systems and products studied, section 5.2 discusses the impact of methodological choices in LCA, with examples from the studies included in the thesis, and section 5.3 describes the impacts due to increased biomass harvesting on LCA results and how changes in biogenic carbon stocks can be included in LCA. All three sections are interlinked. There might therefore be some overlap between the sections.

5.1 Climate impact and energy balance of bioenergy systems and products

5.1.1 Conversion efficiencies

Production of energy carriers (Papers II, III, IV and V) and protein feed (Paper III) from 1 kg dry weight feedstock for all biorefinery systems studied is shown in Table 3. Due to the different fuel efficiencies, comparison between different fuels is best done using vehicle km driven as the basis (Gnansounou *et al.*, 2009). Ethanol and biogas from forest residues (Paper II) generated the most passenger-vehicle km (6.41 km), followed by ethanol and biogas production from straw (6.02 km) (Paper II) and biodiesel and biogas production (5.50 km) (Papers IV and V) (Table 3). Electricity was not included as a transportation fuel in these comparisons. Processing of 1 kg faba bean (whole crop) (Paper III) generated some ethanol with a driving distance of 1.85 km. Comparing the values in Paper III with those in Papers II, IV and V is difficult, since ethanol was produced from faba beans in Paper III but lignocellulosic material was used for fuel production in the other studies. In Paper III, other valuable products including protein feed and briquettes were produced, but these products are not included in the comparison when vehicle km is used as the basis of comparison. Table 3 also presents climate impact and fossil energy use per km (for Papers II, IV and V). The climate impact presented in Table 3 was calculated using two methods, one following the RED method and adding SOC changes and nitrogen replacement (RED+SOC), and one that strictly followed the RED methodology (RED). The climate impact per km was not calculated for Paper III, for the methodological reasons listed above.

The results in Papers IV and V on production of energy carriers are largely the same, but vary some due to changes in the CHP model. Values from the later study (Paper V) are presented in Table 3.

Table 3. *Production of energy carriers and other products per kg dry matter (DM) of feedstock for the base case scenarios in Papers II, III and V*

Feedstock	**kg DM/kg wet**	**Products in:**	**L**	**kg**	**MJ**	**Km[1]**	**Total km**	**Fossil energy (MJ/km)**	**CO_2eq/km RED+SOC/ RED**	**CO_2eq/MJ RED+SOC/ RED**
Straw (Paper II)	1 kg DM/1.22 kg	Ethanol	0.37	0.29	7.88	5.71	6.02[2]	0.18[3]	37.5/10.3[4]	24.7/6.78
		Biogas	0.00	0.01	0.46	0.32				
		Electricity			0.82					
Forest residues (Paper II)	1 kg DM/1.85 kg	Ethanol	0.25	0.19	5.22	3.78	6.41[2]	0.25[3]	69.4/18.6[4]	45.4/9.6
		Biogas	0.00	0.07	3.80	2.62				
		Electricity			0.77					
Straw (Paper V)	1 kg DM/1.22 kg	FAME	0.12	0.11	4.02	3.40	5.50[2]	0.44[3]	51.2/21.7[4]	38.5/16.3
		Biogas	0.00	0.05	3.05	2.10				
		Electricity			0.24					
Faba bean (Paper III)	1 kg DM/2.27 kg	Ethanol	0.12	0.10	2.56	1.85	1.85			
		Protein feed		0.27						
		Briquettes		0.41	7.07					

[1]Vehicle km (assuming 138.6 MJ/100km for E85, 137.9 MJ/100km for E100 (estimated based on values for E85 and petrol), 142.4 MJ/100 km for petrol, 145.1 MJ/100km biogas (estimated from values for compressed natural gas) and 118.5 MJ/100km for fatty acid methyl ester (FAME) (Huss *et al.*, 2013))

[2]Total vehicle km for ethanol/biodiesel and biogas.

[3]Renewable energy directive (RED) calculations including nitrogen (N) compensation. The fossil reference (diesel) is 170 MJ fossil energy/km, assuming 118.5 MJ/km (Huss *et al.*, 2013) and 1.19 MJ fossil per MJ diesel (Edwards *et al.*, 2011).

[4]RED+SOC are calculations with soil organic carbon (SOC) changes and N compensation. While RED follows the calculation in the Directive (EC, 2009). The fossil reference (diesel) is 99.3 g CO_2-eq/km assuming 118.5 MJ/km (Huss *et al.*, 2013) and 83.8gCO_2eq/MJ (EC, 2009).

5.1.2 Lignocellulosic biorefineries

Ethanol, biogas and electricity from straw and forest residues

Ethanol production from straw and forest residues was assessed in Paper II. The co-products biogas, electricity and heat were handled using either energy allocation (Method II; RED) or system expansion (Method I; ISO).

The study was published in 2014 (Paper II), but since then there has been considerable development regarding enzyme production. The climate impact of enzymes has decreased from around 8 kg CO_2eq/kg enzyme product to around 1 kg CO_2eq/enzyme product and, although the dose of the most recently developed products is higher, the overall contribution to fossil energy use from enzyme production has decreased by around 70%. The lower carbon footprint of the newly developed enzyme products is explained by a reduction in processing steps when producing the less concentrated product and by a considerable increase in the amount of renewable energy used in the production plant (Jesper Kløverpris, personal communication 2016). In this thesis, the results from Paper II were recalculated with the new enzyme product and dose[2].

In Paper II, literature values were used to estimate the impact on SOC of harvesting straw (75 g C/kg straw) and forest residues (90 g C/kg forest residues). However, in Paper V SOC changes due to straw harvesting were modelled using the ICBM and yearly emissions were calculated as the average SOC loss over 100 years (36 g C/kg straw). To facilitate comparison between the ethanol (Paper II) and biodiesel (Paper IV and V) produced from straw, the results for straw-based ethanol were recalculated using average SOC losses over 100 years from Paper V. In the forest residues scenario, SOC losses over 120 years were kept as in Paper II. Furthermore, nitrogen compensation due to straw removal was calculated with the assumption that 17% of the straw came from oilseeds, which have a higher nitrogen content than cereal straw (Paper II). In this thesis, nitrogen compensation was accounted for in the same way as in Paper V, where only wheat straw was used.

The new results are presented in Figure 11. Calculated per MJ ethanol, climate impact was estimated to be 87-96% lower than the fossil fuel reference from the RED (83.8 CO_2eq/MJ). Net energy ratio was estimated to be -0.83-0.14 MJ primary energy use per MJ ethanol. These were considerable changes to the results in Paper II, particularly for straw-based ethanol, due to the changed value used for SOC changes (as discussed in section 5.3 of this thesis), and the climate impact decreased in total by 75-35%. The contribution of SOC changes in the straw scenario decreased by around 50%.

[2]The new product and dose was used in Paper V.

The impacts from enzymes decreased by 79%, which explained some of the decreased climate impact for Method I (ISO), and all of the decrease when using Method II (RED). For the forest residues scenario, changing the enzyme product resulted in enzymes no longer being the most important contributor to the total climate impact of all process inputs, which was instead nitrogen used in the biorefinery process. For straw-based ethanol, enzymes were still the most important contributor of all process inputs to both climate impact and primary energy use. Changing the enzyme dose altered the conclusion in Paper II that ethanol from forest residues generally has a lower climate impact than ethanol from straw. Using the RED method, ethanol from forest residues showed a slightly higher impact than ethanol from straw. The reason for this was that the enzyme dose in the forest residues scenario was slightly lower, and was therefore not affected to the same extent when the enzyme product was changed. However, the difference between the climate impact for straw-based and forest residue-based ethanol was small for the RED method.

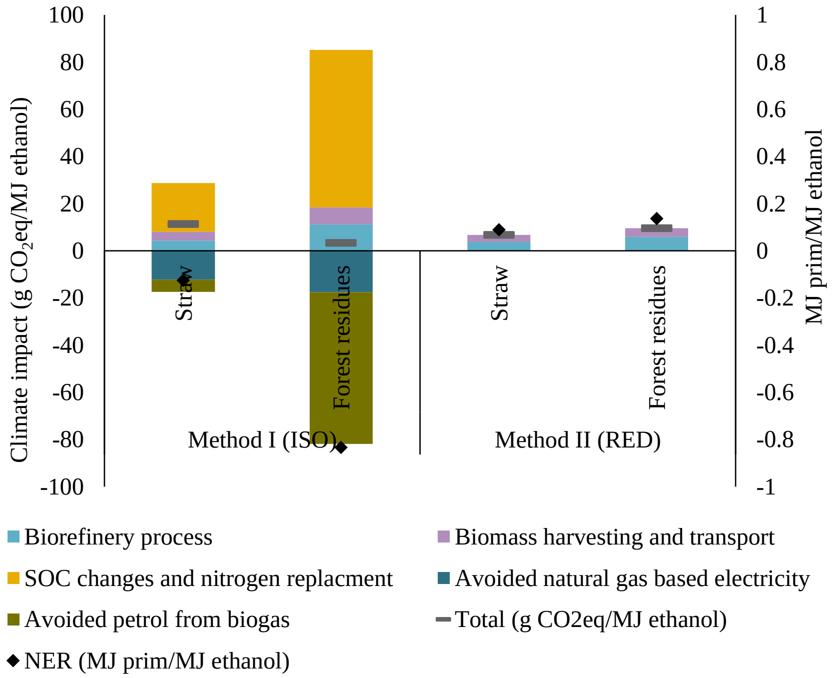

Figure 11. New results for climate impact (CO2eq) and primary energy use (MJ prim) for straw-based and forest residue-based ethanol, using calculation Method I (ISO) and Method II (RED).

Enzyme production is known to be energy-intensive and previous studies have shown that enzymes can have a large impact in the life cycle of lignocellulosic ethanol (MacLean & Spatari, 2009; Slade *et al.*, 2009). Despite this, many previous studies on lignocellulosic ethanol have not included production of inputs such as chemicals and enzymes (Borrion *et al.*, 2012; MacLean & Spatari, 2009). The potential high influence of enzyme production on overall climate and energy balance has been increasingly recognised. A recent study assessed the effect of on-site production of enzymes on overall climate impact and found that this decreased the climate impact of ethanol (Olofsson *et al.*, 2017). However, the climate impact of the off-site enzyme product considered in that study had a higher impact than the newly developed enzyme product. Therefore, the gain from on-site production of enzymes may be smaller if the new enzyme product is already used.

Biodiesel, biogas and electricity from straw

The climate impact for biodiesel produced from straw was estimated to be 54% (RED+SOC) and 81% (RED) lower than the fossil fuel reference (83.8 CO_2eq/MJ from the RED) and NER was estimated to be 0.33 MJ primary energy use per MJ biodiesel (base case) (Paper V).

Mass and energy balance in a systems perspective of biodiesel production from lignocellulose using oleaginous yeast is poorly described in the literature. Therefore, in Paper IV a thorough analysis of potential process design and mass and energy balances covering the whole system was performed, including biomass harvesting and biorefinery processes. It was found that the lipid accumulation step, which is the stage where the oleaginous yeast utilises the sugars from the biomass and accumulates lipids, is energy demanding, since it requires aeration and agitation. Approximately 66% of the total electricity use in the plant was used for this step (Paper IV). By burning the lignin residues, process heat and electricity requirements could be satisfied, with some excess electricity that could be sold (Paper IV). Biogas was produced from the residual yeast biomass, which greatly increased the energy yield. In total, 41% of the energy in the biomass was converted to energy products.

In Paper V, the time-dependent climate impact was calculated for four scenarios. Figure 12 shows the time-dependent climate impact for the base case with reference scenario (comprising equivalent fossil products). The impact from the biorefinery system including SOC is represented by the solid black line, while the reference with equivalent amounts of fossil products is represented by the dashed line. The potential avoided warming from using the biorefinery products instead of fossil products (the reference) is represented by the black dotted line. During the first seven years, the climate impact from the biorefinery system was higher than that of the reference due to the SOC losses. After seven years, the climate impact of the biorefinery system fell below that of the reference and potential avoided warming could be achieved by replacing fossil products with the products from the biorefinery (Figure 12).

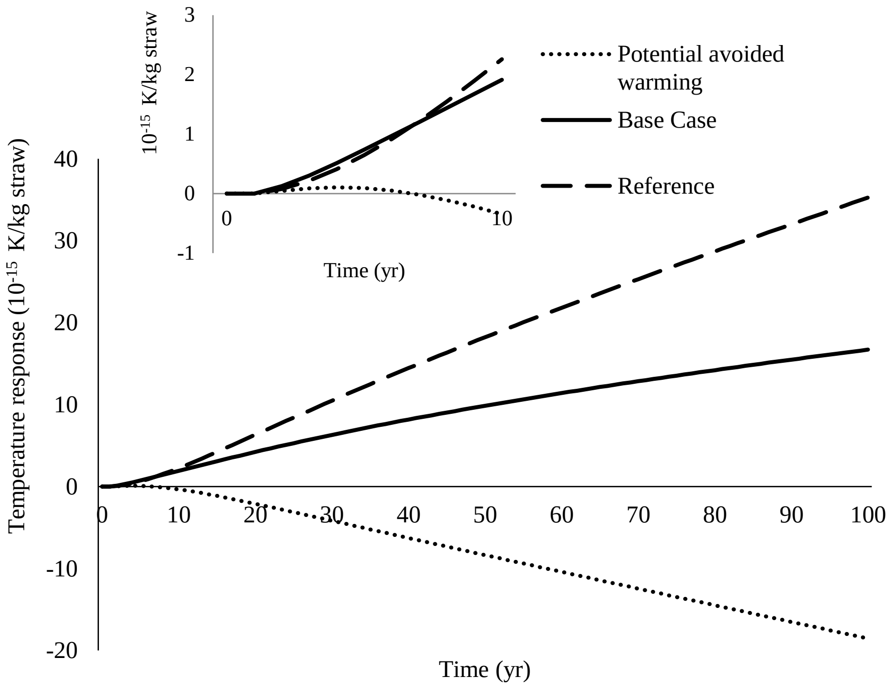

Figure 12. Temperature response of the base case in Paper V (solid black line), with a reference system producing equivalent fossil products (diesel for biodiesel and biogas and natural gas electricity for electricity) (dashed line) and potential avoided warming from using the biorefinery products instead of the reference products (dotted line). Reference system: 1MJ biodiesel was assumed to equal 1 MJ fossil diesel, 1 MJ biogas to equal 0.82 MJ fossil diesel and 1 MJ electricity to equal 1 MJ natural gas electricity.

Comparing ethanol and biodiesel from straw

Figure 13 compares the energy yield of the straw scenario in Paper II with the base case in Papers IV and V. Biodiesel from straw showed lower energy yield per kg straw processed in the biorefinery (Figure 13).

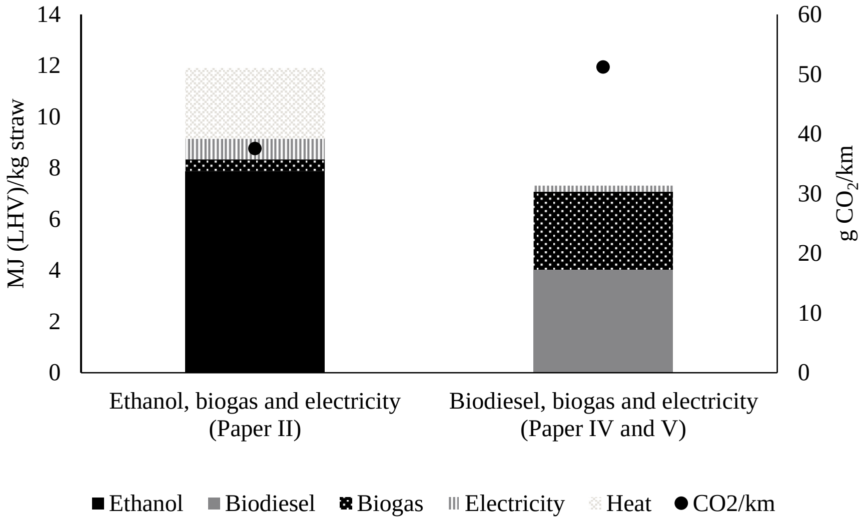

Figure 13. Results for the straw scenario in Paper II and for the base case in Papers IV and V. Energy yield (left axis) in MJ (LHV)/kg straw and CO_2/km (right axis) driven in a passenger vehicle (climate impact calculated according to RED+SOC, see Table 3). The fossil fuel reference (diesel) is 99.3 g CO_2/km (Table 3).

Direct comparison of ethanol and diesel based on energy content (MJ) is not completely accurate, since fuel efficiency differs between these two fuels. As discussed earlier (section 5.1.1), climate impact per vehicle km is more suitable for comparing different fuels and when this was done the difference between the two fuels was lower (Figure 13). However, climate impact per km driven by a passenger vehicle gave a 62-90% reduction for straw-based ethanol and 48-81% reduction for straw-based biodiesel compared with fossil fuels (Table 3). The lower reduction was achieved when SOC and nitrogen replacement were included (method RED+SOC). When comparing the climate performance of these two production systems, it is important to highlight that there has been considerable research on optimisation of the process for ethanol production from lignocellulose, while production of biodiesel from straw using oleaginous yeast is not described and optimised to the same extent in the literature. Consequently, there might be considerable potential for improvement in biodiesel production from straw using oleaginous yeast.

5.1.3 Green biorefinery

Ethanol, protein feed and solid biofuels from faba beans

Paper III analysed the impact of changing from the current use of faba beans as animal feed to use of the whole crop in a green biorefinery for the production of ethanol, protein feed and solid biofuels. To do this, CLCA was used.

The difference in climate impact from the base case with faba beans for feed compared with whole crop faba beans used in a green biorefinery is shown in Figure 14. Substitution of petrol and solid fuel resulted in avoided CO_2 emissions, but the increased need for grain when the faba beans were no longer used for feed, in combination with the higher impact from the cropping stage (mainly from soil carbon changes due to increased biomass harvesting), resulted in an net increase in GWP for the biorefinery compared with the base case. Arable land use and energy use, on the other hand, decreased by 20 and 100% respectively, for the biorefinery scenario compared with the base case (Paper III).

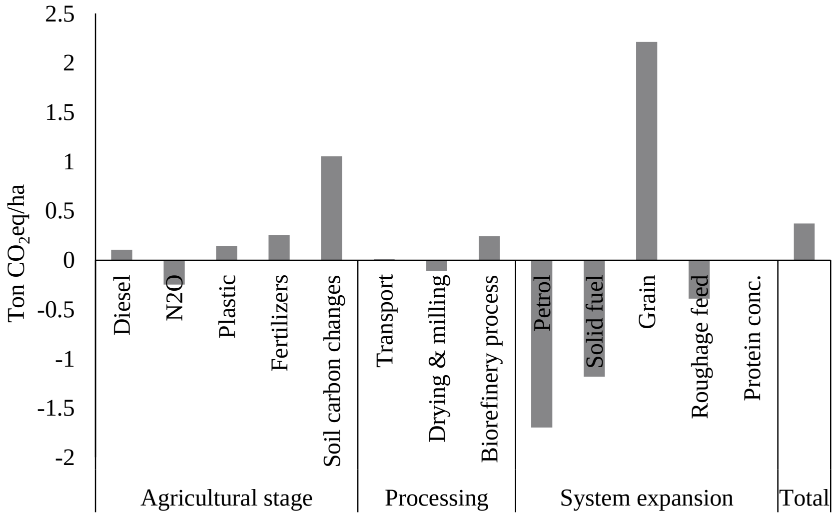

Figure 14. Disaggregated climate impact for the biorefinery system in comparison with the base case. Positive bars show increased impact compared with the base case, while negative bars show decreased impact.

This type of analysis (CLCA of a change in use of biomass) highlights that changing the use of a crop that is currently used as feed can have effects on animal feed demand, which can be important for the overall climate impact and energy balance of the system. The feed rations and the degree to which the new protein feed products produced in the biorefinery replace concentrate feed and other products are important for this assessment. It is important to highlight that this type of analysis assumes constant feed demand, but changes in *e.g.* dietary habits could change the demand for milk and meat and thereby change the demand for animal feed.

Choice of marginal technologies is challenging and can have large impact on the results. Sensitivity analyses were performed where the avoided technology was changed (Paper III). The most influential assumption concerned the product that the briquettes replaced, wood chips, assuming replacement of oil instead would notably decrease the climate impact of the biorefinery system, with overall lower climate impact than the base case (Paper III).

The functional unit of the study was one hectare of land. This functional unit is suitable for comparing best use of land or biomass (Paper I). However, the use of this FU makes it difficult to compare the results with those of other studies that have output functional units, as discussed in section 5.2.1.

5.1.4 Comparing fuels from residues and dedicated energy crops

The results in this thesis calculated using the RED methodology (Papers II and V) can be compared against the default values in the RED, since similar methodology was used. The values calculated using the RED methodology are shown in Table 3. For straw-based and forest residue-based ethanol, the climate impact (6.8 and 9.6 g CO_2eq/MJ ethanol, respectively) was substantially lower than the climate impact for sugar cane, wheat and sugar beet ethanol in the RED, and in line with the default values for second-generation biofuels in the RED (EC, 2009). The climate impact of biodiesel from straw (16.3 g CO_2eq/MJ biodiesel) was also in line with the default values for second-generation ethanol in the RED, but higher than the climate impact reported for diesel-like fuels produced from lignocellulosic biomass, which ranges from 4.2 g CO_2eq/MJ for FT-diesel and DME produced from forest residues to 6.7 g CO_2eq/MJ for DME from wood (EC, 2009).

When the system boundaries were widened from those of the RED to include changes in SOC due to harvesting of residues and compensation for nitrogen removed, the climate impact of the biofuels assessed in this thesis increased (see RED+SOC in Table 3). Ethanol from straw then had a climate impact of 25 g CO_2eq/MJ, ethanol from forest residues a climate impact of 45 g CO_2eq/MJ and biodiesel from straw a climate impact of 39 g CO_2eq/MJ. In comparison with the default values in the RED (EC, 2009), straw-based ethanol was then in line with sugar cane ethanol, while sugar beet and wheat ethanol had a higher impact. Ethanol produced from forest residues had a higher impact than sugar cane and sugar beet ethanol, but a lower impact than wheat ethanol. The climate impact of biodiesel from straw, calculated according to RED+SOC, was lower than the default climate impact for rapeseed biodiesel, soybean biodiesel, palm oil biodiesel and rapeseed HVO, but higher than *e.g.* the default values of climate impact for palm oil HVO and sunflower HVO in the RED (iLUC factors are not included) (EC, 2009).

Management changes such as increased straw harvesting is a direct land use change (dLUC). Other types of dLUC include conversion of forest land into agricultural land and changes from one crop to another, such as from grassland to annual crops (Figure 15). Therefore climate impacts that include dLUC effects (SOC changes and nitrogen compensation, *i.e.* RED+SOC) due to residue harvesting are perhaps best compared with those reported in studies on first-generation biofuels that assess dLUC.

Börjesson *et al.* (2010) used two different land use references to assess the impact of direct land use change, grain cultivation and unfertilised grassland. For rapeseed biodiesel, they found that the climate impact ranged from around 30-60 g CO_2eq/MJ biodiesel when unfertilised grassland was used as a reference to 0-30 g CO_2eq/MJ biodiesel with grain as a reference (the variation within the results when using the same land use reference can be explained by different allocation methods) (Börjesson *et al.*, 2010). For ethanol, the values ranged from 20-50 g CO_2eq/MJ when unfertilised grassland was used as a reference to 5-30 g CO_2eq/MJ when grain was used (Börjesson *et al.*, 2010). Again, the results varied depending on allocation method and feedstock. These results show that dLUC can also be important for the climate impact of first-generation biofuels (Börjesson *et al.*, 2010).

The results in this thesis suggest that the climate impact of second-generation biofuels is lower than that of first-generation biofuels when dLUC effects due to residue removal are not included. When these effects are included the picture becomes more complicated and second-generation biofuels sometimes have lower and sometimes higher climate impacts than first-generation biofuels.

When comparing the results, it is important to emphasise that dLUC effects are not always included in studies on first-generation biofuels. Furthermore, using residues for feedstock has a clear advantage over first-generation biofuels, since the use of residues does not demand extra land and is therefore not associated with iLUC (Figure 15). Overall, however, iLUC can be very important for the climate performance of biofuels (Dunn *et al.*, 2013; Hertel *et al.*, 2010; Searchinger *et al.*, 2008).

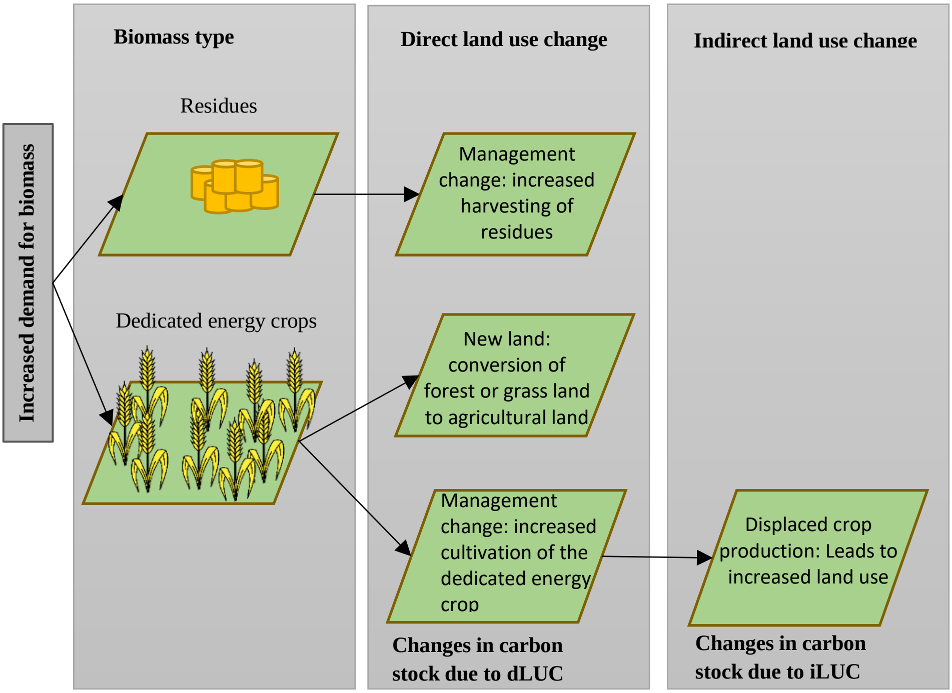

Figure 15. Illustration of direct (dLUC) and indirect (iLUC) land use changes associated with biomass production of residues and dedicated energy crops.

5.2 Effect of methodological choices in LCA

5.2.1 Functional units

The functional unit defines and quantifies the function(s) of the product under study (ISO, 2006a). The choice of functional unit is closely connected with the aim of the study, and different types of functional units are suitable for different research questions (Paper I). Paper I identified four different categories of functional units: *use of feedstock*, *single product*, *function of single product* and *multifunctional*. Two of these types of functional units were used here, *use of feedstock FU* and *single product FU*. The single product FU used in Papers II and V allows for comparison between products with the same function, *i.e.* ethanol produced from different types of feedstock or biodiesel produced in different processes. When making these comparisons, it is important to remember that other methodological choices, such as type of LCA (ALCA or

CLCA), allocation methods, data choice *etc*. are important to consider. In addition, selecting a single product FU for a multifunctional biorefinery system involves handling co-products using allocation by partitioning (Papers II and V) or by system expansion (Method I (ISO) in Paper II).

The *use of feedstock FU* can be used to assess the best use of land or biomass (Paper I). In Paper III, one hectare of faba bean cultivation was used as the FU, in order to assess the consequences of different uses of (the same) hectare of faba beans. In Paper V, a different type of *use of feedstock FU*, one kg of straw was used. Using this FU allowed for comparison of the biorefinery performance as a whole (the combination of products), which could then also be compared with a reference system with the same amounts of conventionally produced products (*i.e*. Earles *et al.*, 2011; Cherubini & Jungmeier, 2010; Cherubini & Ulgiati, 2010), here called GHG substitution potential (see section 4.2.4).

Figure 16 shows the GHG substitution potential for the FU one kg straw for the biorefinery systems studied in Papers II and V. The transport service that can be provided by the products ethanol, biodiesel and biogas was calculated (Table 3) and was assumed to replace the same distance using petrol as a reference. Potential substitution of petrol was calculated based on MJ/km for the respective fuels in relation to petrol (Huss *et al.*, 2013), while climate impact for fossil fuels was assumed to be 83.8 g CO_2/MJ (EC, 2009). Some of the results in Figure 16 are included in Figure 11 and Table 3, but are presented in a different way in Figure 16.

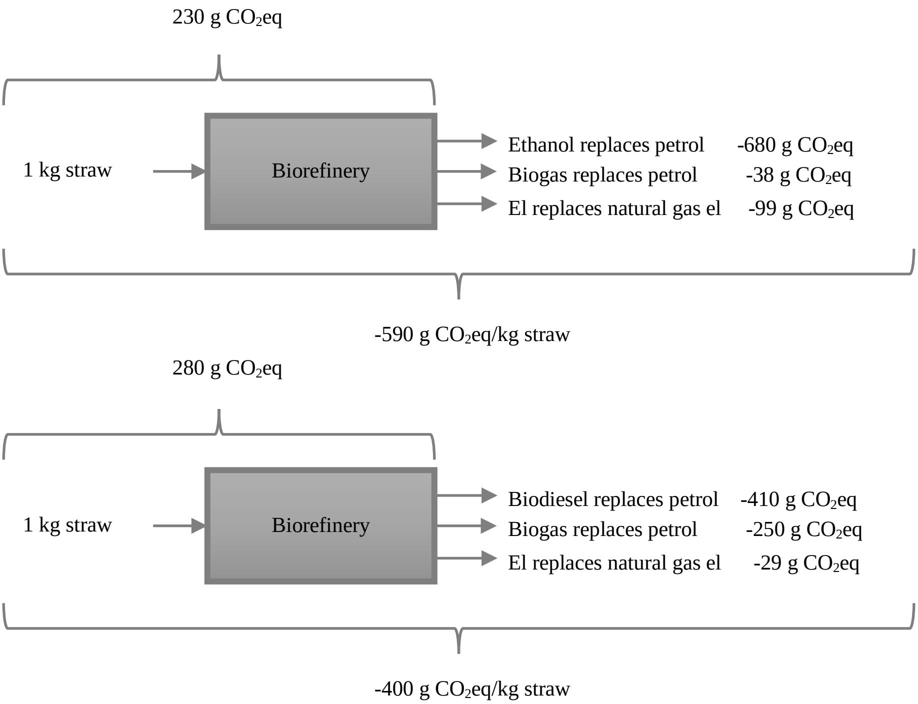

Figure 16. Greenhouse gas (GHG) substitution potential for ethanol (Paper II) and biodiesel (Paper V) from straw. el = electricity.

This way of presenting results shows the benefit of co-products such as electricity or feed without using the system expansion (substitution) that is commonly employed in LCA. The use of substitution can sometimes result in relatively strange results, such as when a single product is associated with a negative environmental impact when its co-products are assumed to replace heavily polluting products (see energy balance results for ethanol from forest residues, Fig. 2 in Paper II). This can be difficult to interpret. The advantage of using a reference system to calculate GHG substitution potential is that it is easy to interpret and that it allows for comparison of different biorefinery concepts and sets of co-products. However, similarly to the use of system expansion, it is associated with a rather arbitrary selection of reference product (or avoided product). To improve evaluation and comparison with existing systems and products, it may be good idea to use several reference systems, *e.g.* Brander (2017) used several scenarios with different marginal technologies in a CLCA.

5.2.2 Handling multifunctionality

How to handle multifunctionality is one of the most discussed issues within LCA methodology (Finnveden *et al.*, 2009). Paper I recommends allocation by partitioning, *i.e*. allocation based on *e.g*. energy content is applicable mainly for ALCA studies, while system expansion is applicable mainly for CLCA studies. However, divergence from these rather general recommendations may be necessary for several reasons (Paper I). In Paper II, two different methods, substitution (Method I (ISO)) and allocation based on LHV (Method II (RED)), were used. Choice of method for handling multi-functionality clearly influenced the results (see Figs. 2 and 3 in Paper II), as also shown by *e.g*. Sandin *et al.* (2015) and Xie *et al.* (2011). The fact that this choice is very important for the results calls for careful consideration of what method to use in relation to the aim of the study (Paper I) and the decision context (Sandin *et al.*, 2015). Paper I also recommends that the same allocation method be used for the biorefinery feedstock in cases where this originates from a multifunctional system (a similar recommendation is made by Sandin *et al.* (2015)). This is especially important for studies using system expansion (Sandin *et al.*, 2015), since it is reasonable to consider indirect effects using system expansion for feedstock supply when this method is used for co-product production. If, for example, the co-products are assumed to replace equivalent products, but the impact from feedstock production is allocated based on mass, one part of the system is credited for producing co-products (*i.e*. replacing equivalent production) while indirect effects, such as alternative uses of the feedstock, are not included.

How system expansion should be carried out for residues depends on whether the residue is fully utilised or not (Weidema *et al.*, 2009). In Papers II, IV and V it was assumed that straw and forest residues were not fully utilised. Straw and forest residues are dependent co-products, meaning that the production volume is not determined by the demand for the co-product, but by the demand for the main product (timber and cereals in the case of forest residues and straw, respectively). According to Weidema *et al.* (2009), the following processes should then be considered when performing system expansion for not fully utilised dependent co-products: (i) the intermediate treatment of the co-product (*i.e*. harvesting and chopping the biomass); (ii) 'waste treatment' of the co-product, which was assumed to be the alternative treatment when the straw forest residues are not harvested, *i.e*. SOC changes due to residue harvesting were included here; and (iii) where the dependent co-product is used, *i.e*. the biorefinery process. This corresponds to how straw and forest residues are handled in Paper II (Method II (ISO)), straw in Paper V and the forest residues in the avoided process in the biorefinery scenario in Paper III. It also corresponds to the direct land use effects when harvesting straw, which according to Paper I should always be included.

If, on the other hand, the residues are fully utilised, increased demand for the residues would increase the demand for a different product (Weidema *et al.*, 2009), which in this case could be a different kind of biomass resource.

5.2.3 The RED method used on biorefineries

Use of LCA for policy applications is associated with several challenges, as described in section 3.3.2 of this thesis. Whether or not current LCA methodology is suitable to use for policies in general and how it should be developed to better suit that purpose was beyond the scope of this thesis. Interesting discussions on this can be found elsewhere (*e.g*. McManus & Taylor, 2015; McManus *et al.*, 2015; Plevin *et al.*, 2014). This section focuses on the use of the current RED methodology on emerging biorefinery systems that use residues as feedstock.

The RED promotes the use of biofuels produced from for residues, waste, non-food cellulosic materials and algae (EC, 2009). These type of fuels are likely to be produced in emerging biorefinery systems, with multiple co-products and using residues as feedstock, to a greater extent than has been the case to date. Handling of system boundaries and functional unit in the RED are discussed below.

System boundaries

Increased use of residues can have effects on SOC that have a potentially large influence on the overall climate impact of the fuel (Papers II and V). In the current RED (EC, 2009) and the proposal (EC, 2017b)[3] residues (including straw and forest residues) are considered to be free from environmental impact up to harvest. Figure 17 shows how a change in system boundaries to include SOC effects would affect the overall climate impact of ethanol produced from straw and forest residues (Paper II) and biodiesel produced from straw (Paper V). The climate impact was calculated in the same way as in Table 3. Applying the RED

[3]The proposal is currently under discussion.

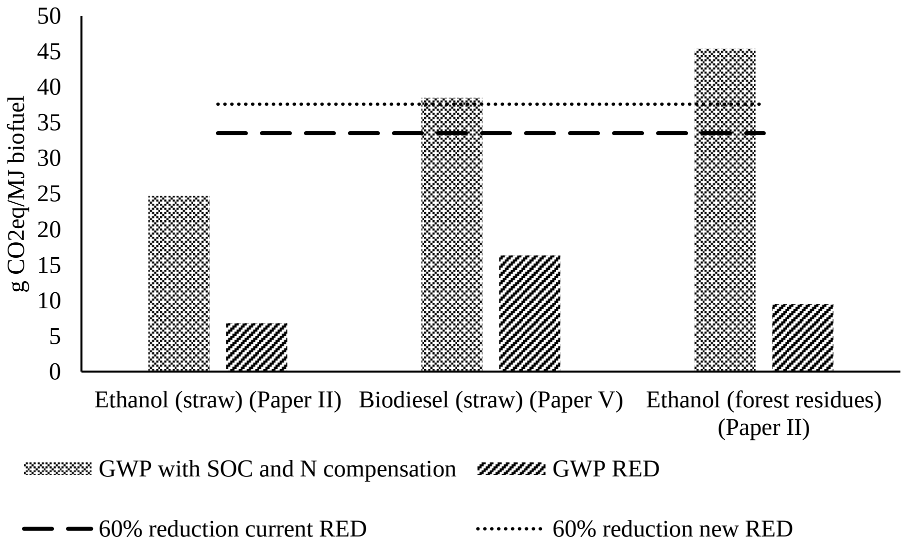

Figure 17. Global warming potential (GWP) for ethanol and biodiesel produced from straw, including soil organic carbon (SOC) and nitrogen replacement and the Renewable Energy Directive (RED) calculation method. 60% reduction from a fossil fuel reference represents the current fossil reference in the RED (83.8 g CO_2/MJ) and in the forthcoming version (94 g CO_2/MJ).

methodology, climate impact reductions (compared with the current fossil fuel reference in the RED, 83.8g CO_2eq/MJ), were found to be 92% for straw-based ethanol, 89% for forest residue-based ethanol and 81% for biodiesel produced from straw. When SOC changes and nitrogen compensation were included, the corresponding reductions were 71% for straw-based ethanol, 46% for forest residue-based ethanol and 54% for biodiesel produced from straw. The reduction targets shown in Figure 17 are relative to the RED today (83.8g CO_2eq/MJ), and the suggested fossil reference (94 g CO_2eq/MJ) in the proposal for the new RED (Edwards, 2017).

Efforts to use residues for biofuel production can have large effects on how agricultural and forest land is managed. Therefore it is important to further study and evaluate these effects and to somehow consider the effects of residue harvesting in policy making.

Handling of multifunctionality

In the RED method, allocation is based on LHV of the products, which means that energy is the determining characteristic of all products (Paper I). For biorefineries this might be problematic, especially when not all co-products are produced for energy purposes (Paper I; Cherubini *et al.*, 2011; Gnansounou *et al.*, 2009). In policy applications, it is difficult to avoid allocation between co-products, and for comparability reasons it is unavoidable to have some type of standard on how the allocation should be done. This is a great challenge when using LCA as a policy instrument. It is important that the standards are applicable to the systems they are assessing, which is challenging with the diversity of biorefinery systems now being proposed. In the case of the RED, the use of energy allocation perhaps favours production of high energy outputs (more production output over which to allocate the environmental burden), and disfavours production of chemicals, protein feed *etc*. that can have a lower energy content.

5.3 Impacts due to increased biomass harvesting

5.3.1 Effects of increased biomass harvesting on climate impact

In Papers II, III and V, impacts on SOC due to residue removal and compensation (mineral fertiliser production) for the nitrogen removed with the harvested residues were included in the assessment. Paper III also included effects on N_2O emissions and nitrogen leaching over the whole crop rotation. In this section, results and overall impact from increased biomass harvesting are discussed. Methods for modelling SOC changes and for accounting for changes in biogenic carbon pools in climate impact assessments are further discussed in section 5.3.3.

Soil organic carbon changes

In line with earlier studies (see *e.g*. Whittaker *et al.*, 2014; Cherubini & Ulgiati, 2010), it was found that SOC changes due to residue harvesting were highly influential for the overall climate impact (Papers II, III and V). The SOC changes contributed 59% (straw-based ethanol) and 73% (forest residue-based ethanol) of total GWP (Figure 11). For biodiesel (base case, Paper V), SOC changes contributed 48% to total GWP, while SOC changes due to whole crop harvesting of faba beans were responsible for 36% of the total GWP (Paper III). In all of the above results, SOC changes were estimated using a SOC model and average changes over 100 years (104 years in Paper III) were calculated (except for forest residues, for which literature values were used (Paper II)). The potential climate impact was calculated using GWP_{100} (as described in section 4.3.1).

Returning part of the process residues from the biorefinery to the field or forest site could be one option to mitigate the impact on SOC. This would return

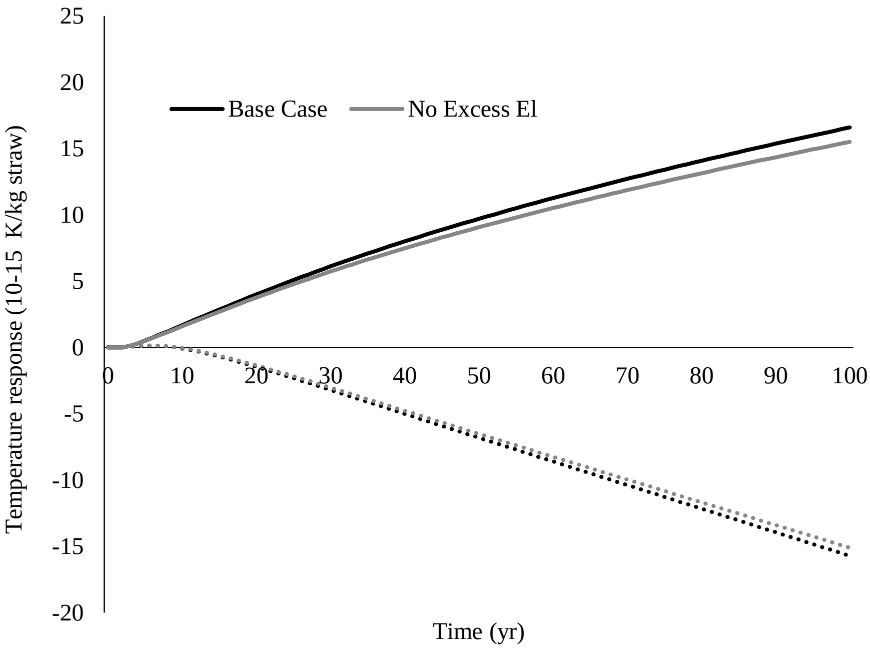

Figure 18. Temperature response from process emissions, including harvesting, processing and soil organic carbon (SOC) changes due to straw harvesting for 1 kg straw (solid lines) and potential avoided warming from replacement of equivalent products for each scenario (biodiesel and biogas replaced diesel and electricity replaced natural gas electricity) (dotted lines). Reference system: 1MJ biodiesel was assumed to equal 1 MJ fossil diesel, 1 MJ biogas to equal 0.82 MJ fossil diesel and 1 MJ electricity to equal 1 MJ natural gas electricity.

nutrients and some of the carbon in the biomass, potentially decreasing the effect of SOC losses and nutrient replacement. Paper V investigated to what extent this effect could be mitigated by returning part of the lignin to the soil. Figure 18 shows the time-dependent global mean surface temperature change comparing the base case with no lignin recycling and the scenario with no excess electricity and instead recycling the lignin to the field. The results show that the impact (solid lines) was higher in the base case due to the higher SOC emissions, although on accounting for the potential substitution (dotted lines) the potential avoided warming was higher for the base case than for the no excess electricity scenario. This is because the excess electricity replaced natural gas electricity. By replacing electricity produced from natural gas, more GHG emissions were avoided than when returning the lignin to the field (Paper V). The difference between the scenarios was small, due to the relatively small fraction of lignin residue that could be returned. Almost all of the lignin was used to satisfy the electricity demand in the plant.

Nutrients removed with the biomass

The impact of nutrient removal with crop residues was only assessed in this thesis for nitrogen removal (Papers II, III and V). The overall contribution varied from 5% (forest residue-based ethanol, using calculation Method I (ISO), Paper II) to 12% (straw-based ethanol (Paper II)) of total GWP per MJ fuel. In fact, numerous nutrients are removed with crop residues, including potassium and phosphorus, which are significant plant nutrients. In Papers II, III and V, part of the biomass was assumed to be burned in the CHP plant, and in that case any potassium and phosphorus present will mainly be recovered in the ash (as opposed to nitrogen), and can thus be returned to the production site (Obernberger *et al.*, 1997).

In Papers II and V, it was assumed that all nitrogen removed with the residues was replaced with mineral nitrogen. As discussed in Paper II**,** it is not likely that all the nitrogen removed will need to be replaced. One alternative method to estimate the nitrogen compensation required is to relate nitrogen requirement to expected decreases in grain yield, as done by Gabrielle and Gagnaire (2008). Nitrous oxide emissions were estimated based on the method in IPCC (2006). When assuming that all nitrogen removed is compensated for, as was done in Papers II and V, the nitrous oxide emissions are the same for residue removal and non-residue removal, as nitrogen added in the form of crop residues and fertilisers has the same emissions factor according to IPCC (2006).

In Paper III, a decrease in the nitrogen effect to the following crop in the crop rotation was assumed when more of the legume biomass was harvested for the biorefinery. Instead of compensating for all nitrogen removed with the biomass, it was assumed that roughly 36% (Nyberg & Lindén, 2008) of the nitrogen in the biomass contributed to the decrease in nitrogen fertiliser demand. When applying this method, harvesting of crop residues decreased nitrous oxide emissions compared with return of crop residues.

5.3.2 Carbon stock changes over time in biofuel LCAs

Biogenic carbon fluxes, including SOC changes, are difficult to handle in LCA. This is because the emissions vary from year to year (as described above).

In Papers II, III and V, average SOC changes over a selected period were calculated. This involved an arbitrary selection of number of years over which the SOC was distributed. This choice was important for the results, *e.g.* in Paper V, GWP of 1 MJ biodiesel increased by 51% if 10 years were used instead of 100 years (base case). Figure 19 shows how this choice can influence the results for straw-based fuels. The average SOC over 10 years (year 1 is when the

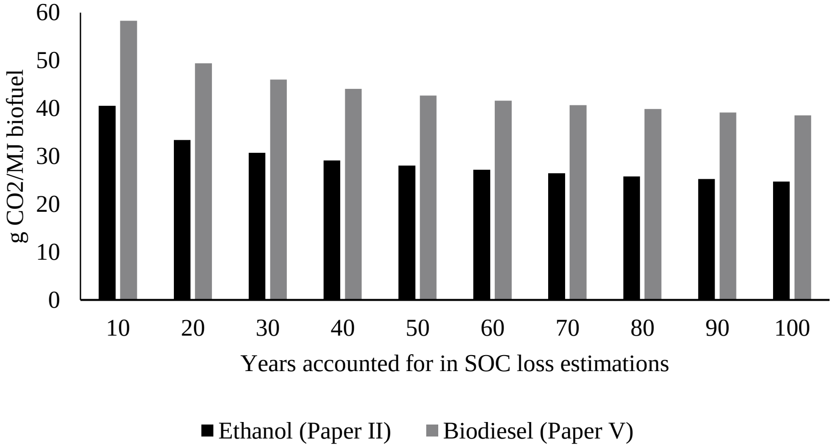

Figure 19. Climate impact per MJ ethanol (paper II) and biodiesel (Paper V), calculated with the Renewable Energy Directive (RED) methodology (allocation based on lower heating value) but including soil organic carbon (SOC), estimated as average SOC losses over 10-100 years.

management change started) was around 280 g CO_2 losses/kg straw, while average SOC losses over 100 years were 130g CO_2/kg straw, corresponding to approximately 76 and 37 g C/kg straw, respectively. These values can be compared against previously published estimates on SOC losses due to straw harvesting, for example: 108 g C/kg straw over 20 years and 50 g C/kg straw over 100 years (Powlson *et al.*, 2011), 50-100 g C over 30 years (Gabrielle & Gagnaire, 2008) and 40 g C/kg straw over 20 years (Cherubini & Ulgiati, 2010).

For estimating the climate impact of systems with changes in biogenic carbon stocks, a climate impact model such as the time-dependent climate model (Ericsson *et al.*, 2013) used in Paper V can be especially useful. The advantages of the time-dependent climate model compared with a single score climate impact indicator such as GWP (used in Papers II, III and V) are: First, the choice of two arbitrator time horizons is partly avoided, *i.e*. the time horizon for the cumulative radiative forcing used to estimate GWP (commonly 100 years) and the time over which SOC changes are allocated. Second, using the time-dependent climate model gives the shape of the climate impact over time, providing additional information that can be an important complement to the use of *e.g*. GWP (Ericsson *et al.*, 2013).

5.3.3 SOC modelling

Soil organic carbon changes due to management changes often have a large impact on the results of LCA studies. Despite this, there is no agreement in the

LCA community on how to estimate SOC changes included in LCA assessments (Goglio *et al.*, 2015).

To include SOC changes in LCA, the effect on SOC due to land use changes or management changes is often modelled (Goglio *et al.*, 2015). This is because SOC changes occur over long periods and therefore measuring them is time-consuming and costly. Although some long-term field studies have measured SOC effects due to management changes (see *e.g*. Kätterer *et al.*, 2011), there is a lack of data, making modelling necessary in many cases. However, SOC changes are difficult to model, as SOC can depend on many factors, such as conditions at the site, *e.g*. climate and soil type (Cowie *et al.*, 2006). In addition, formation of SOC is complex and to some extent unknown (Schmidt *et al.*, 2011). For example, there is evidence that persistence of organic matter is largely governed by complex interactions between organic compounds and the environment, and that decomposition rate is less dependent on chemical structure of the biomass than previously believed (Schmidt *et al.*, 2011). Regarding impacts on SOC formation from crop residues, it has been found that belowground residues (roots) contribute more to SOC formation than aboveground residues (Kätterer *et al.*, 2011). On sandy soil in particular, aboveground residues may contribute little to the SOC pool (Poeplau *et al*., 2015). Future models need to incorporate new scientific results to better predict SOC formation (Schmidt *et al.*, 2011). In short, the challenge with modelling SOC arises not only in LCA studies. However, since SOC changes have been shown to have large impacts on the results, the uncertainty associated with SOC estimates should be discussed and preferably analysed in sensitivity analysis (Paper V).

6 Outlook

6.1 Biofuels and environmental impact conflicts

Climate impact, as studied in the present thesis, is one of the two core planetary boundaries identified by Steffen *et al.* (2015), together with biodiversity loss, due to their large importance for the Earth's system. It is clearly vitally important to lower the human impact on the global climate. This thesis showed that biofuel produced in emerging biorefineries from agricultural and forestry residues can have a smaller climate impact than fossil fuels. However, production of biofuels may have other environmental impacts, including impacts on biodiversity. A full sustainability assessment of biorefinery systems and biofuels needs to account for all relevant environmental, social and economic impacts related to the whole production chain. The following section focuses on conflicting environmental impacts in biofuel production.

Many studies on first-generation and second-generation biofuels report lower GWP and fossil energy use compared with fossil fuels (Borrion *et al.*, 2012; von Blottnitz & Curran, 2007). For other environmental impacts, such as eutrophication potential, acidification potential, ozone depletion and human toxicity, previous studies have reached differing conclusions, with some reporting a decreased impact and some an increased impact compared with fossil fuels (Borrion *et al.*, 2012; von Blottnitz & Curran, 2007).

Several studies have shown that there are environmental conflicts. For example, a study on switchgrass ethanol showed lower impact with respect to GWP, abiotic depletion and ozone layer depletion potential, but a higher impact than petrol for other impact categories assessed, including photochemical oxidation potential, human toxicity potential, eco-toxicity potential, acidification potential and eutrophication potential (Bai *et al.*, 2010). Using straw or maize stovers as biorefinery feedstock has been found to have a lower impact than a fossil reference system for all impact categories (including human toxicity, GWP, ozone depletion acidification *etc*.) except eutrophication, where the impact was higher for both maize stovers and straw.

There are concerns that biofuel production may affect biodiversity negatively. Land use change is one of the main drivers of biodiversity change (MEA, 2005), which is why land use change has been a frequent focus in studies on biodiversity impacts due to bioenergy production. However, not all land use change leads to negative impacts on biodiversity. A review by Immerzeel *et al.* (2014) showed that impacts from bioenergy production on biodiversity are mostly negative but that there are several trade-offs, for example perennial crops such as energy grasses or short-rotation woody crops such as willow can potentially increase biodiversity on field scale in agroecosystems dominated by annual crops.

Intensification of cultivation can lead to negative effects on biodiversity. Pedroli *et al.* (2013) identified increased pressure on land, leading to intensification of current agricultural or forestry production and conversion of habitat-rich land for the production of bioenergy, as threats to biodiversity, as a consequence of bioenergy production.

Second-generation biofuels from *e.g.* energy grasses and crop residues can potentially be produced on marginal land or do not require extra land (residues) and are therefore likely to have lower impacts on biodiversity (Koh & Ghazoul, 2008). In a review by Immerzeel *et al.* (2014), it was concluded that second-generation biomass tends to be less negative for biodiversity than first-generation crops. However, residue harvesting can have a negative effect on biodiversity. For example, harvesting of forest residues may reduce biodiversity due to decreased amounts of dead wood and impacts on soil biodiversity in forests (Pedroli *et al.*, 2013). A study by Degens *et al.* (2000) showed that decreased soil organic carbon can negatively influence soil microbial diversity. This implies that increased withdrawal of crop residues may impact soil biodiversity negatively.

There are several drivers of biodiversity loss, of which climate change is one important factor (MEA, 2005). Consequently, biofuels can contribute to biodiversity loss mitigation by mitigating climate change.

Local air quality

The effect on urban air quality from diesel emissions is increasingly being discussed. Compared with diesel, biodiesel has been shown to decrease emissions of hydrocarbons, carbon monoxide and fine particle matter, while increasing emissions of nitrogen oxides (Robbins *et al.*, 2011). These results refer to tailpipe emissions, which of course are interesting for local air quality. In a life cycle perspective, looking not at only tailpipe emissions but also emissions from biomass production and processing, Wu *et al.* (2006) found that cellulosic fuels produced from switchgrass increased emissions of volatile organic compounds and nitrogen oxides. However, when looking at where these emissions occur, those authors found that emissions of nitrogen oxides, volatile organic compounds and particles decreased in urban areas compared with diesel and petrol for nearly all cellulosic fuels studied.

6.2 Future potential for biofuel production from residues

In 2016, the transport sector in Sweden used approximately 95 TWh for domestic transport alone, while total energy use for transport was 129 TWh (SEA, 2017b). Looking at the future biomass potential values for Sweden presented in section 3.2.1 of this thesis, the biorefinery producing ethanol and biogas (Paper II) could produce 26-33 TWh in a short-term perspective (assuming 47% conversion to ethanol and biogas). The biorefinery concept producing biodiesel and biogas (Paper V) as transportation fuels could produce 22-27 TWh in a short-term perspective (assuming 40% conversion to biodiesel and biogas). Consequently, in the short term, the biorefinery concepts assessed in Paper II could supply a maximum of one-third of the total Swedish energy demand for domestic transport, if all available lignocellulosic biomass were used.

Temperature responses from using 1 kg straw in the biorefinery concept producing biodiesel, biogas and electricity was assessed in Paper V. If all available straw globally were used in this biorefinery concept, the temperature response would be considerable. Searle and Malins (2015) estimated that 10% of all crop residues globally could be used for bioenergy. This corresponds to 460 million tons, or 8 EJ per year. Using all of these crop residues could supply approx. 3.4 EJ, corresponding to approx. 3% of the 110 EJ used for transport globally (U.S. Energy Information Administration, 2016). Extrapolating the results in Paper V to cover global production, the temperature response would be approx. 0.007 degrees K lower in 100 years compared with using fossil fuels. Assuming that 50% of all crop residues could be used as biorefinery feedstock (in the biorefinery concept presented in Paper V), approximately 15% of the current energy use for transport could be supplied, with 0.04 degrees K lower temperature response than the reference system with fossil fuels. These calculations are of course very simplified. For example the SOC changes due to straw harvesting assumed Swedish conditions. However, the calculations indicate that substantial amounts of bioenergy could be produced from crop residues only, with considerable climate mitigation potential compared with the use of fossil fuels. It should be noted that crop residues have been estimated to be of lower importance when it comes to biomass potential for bioenergy production globally (Hoogwijk *et al.*, 2005) and in Sweden (Börjesson *et al.*, 2013a). Therefore, other sources of lignocellulosic materials such as grasses or short-rotation forestry could significantly increase the potential.

6.3 The role of biofuels for a sustainable transport sector

Biofuels, particularly advanced biofuels, are important in order to decrease climate impact and fossil fuel dependency in the transport sector (EC, 2017a; EC, 2013). The main advantage with liquid biofuels is that several can be used in low blends with fossil fuels, which means that they can be used within the existing infrastructure and vehicle fleet, and thus directly replace fossil fuels. Some biofuels, such as HVO, can be used in diesel engines in any blend. Biofuels are also the main renewable alternative to fossil fuels in aviation and marine transport (EC, 2013).

However, biomass is projected to be used not only for fuel production, but also for materials and chemicals. Thus to reach a fossil fuel-free transport sector, biofuels from lignocellulosic biomass will have to be combined with decreased energy use in the transport sector and other energy sources such as electricity, as discussed below.

Currently, there is much interest in electric vehicles (EC, 2017a; REN21, 2017). Today, electricity use in the transport sector is around 1% of the total energy use for transport in Europe, whereas biofuels supply around 4% of the energy used in transport (EU, 2016). Use of both electricity and biofuels is predicted to increase within the transport sector (EU, 2016). There are several advantages with electric vehicles, such as decreased local air pollution. However, the climate impact gain will depend on the electricity grid mix (EC, 2017a).

The following section compares the use of biofuels or electricity from straw. For comparison, the driving distance (passenger vehicle) using 1 kg straw was calculated for the biorefinery systems in Paper II and V and for electricity production from straw. It was assumed that an electric vehicle uses 33% of the energy used by a car running on petrol, and that the conversion efficiency from biomass to electricity is 33%[4]. The results indicate that the driving distance is almost twice as long if the biomass is converted to electricity compared with the biorefinery systems in Papers II and V. This is primarily because of the high efficiency of electric engines. However, for electricity to be a viable option for the road transport sector, a large change in the vehicle fleet towards more electric vehicles and infrastructure for charging the vehicles are needed.

It is important to highlight that, in order to reach climate goals, several different fuels will be needed, in combination with efficiency improvements (EC, 2013) and changes in travel habits. As mentioned earlier, biomass is a renewable resource that can replace fossil resources, not only in the transport sector but also to produce materials and chemicals. When it comes to materials and chemicals, biomass is the main alternative.

[4]Calculated based on three comparable car models using either petrol or electricity. The average energy use for the electric version of these models was found to be 33% of that of an equivalent car running on petrol. Data from: www.bilsvar.se were used. Conversion efficiency from biomass to electricity was assumed to be 33%. Energy use for driving a passenger vehicle was assumed to be: petrol 142.4 MJ/100km, biodiesel 118.5 MJ/100km, ethanol: 137.9 MJ/100km, biogas 145.1 based on (Huss *et al*., 2013) and electricity 47 MJ/100km.

7 Conclusions and future research

7.1 Conclusions

7.1.1 Climate impact and energy balance

The conclusions presented in this section refer to the first specific objective of this thesis work, which was to evaluate three biorefinery systems in a climate impact and energy balance perspective. The conclusions are given for each biorefinery system separately.

Ethanol, biogas and electricity production from lignocellulose

- The results in Paper II were updated using new data for SOC changes, nitrogen replacement and enzyme production. The new results using the ISO method showed that the climate impact for lignocellulosic ethanol ranged between 11.3 g CO_2eq/MJ for straw-based ethanol and 3.3 g CO_2eq/MJ for forest residue-based ethanol.
- Applying the RED methodology, the climate impact reduction compared with fossil fuels was 92% and 89% for straw- and forest residue-based ethanol, respectively. When SOC changes and nitrogen compensation were included in the RED calculations (RED+SOC), the corresponding reduction potential was 71% and 46%, respectively.
- Primary energy use was between -0.83 and 0.14 for MJ_{prim}/MJ ethanol.
- SOC changes had a large influence on the results, as did biorefinery inputs including enzymes (the straw scenario) and nitrogen (the forest residues scenario).

Biodiesel, biogas and electricity production from lignocellulose

- Applying the RED methodology, the reduction in climate impact compared with fossil fuels was 81%. When SOC changes and nitrogen compensation were included (RED+SOC), the reduction potential was 54% (base case).
- Primary fossil energy use was 0.33-0.80 MJ_{prim}/MJ biodiesel.
- Strain development for oleaginous yeast should aim for shorter residence times for lipid accumulation, since this step requires energy for agitation and aeration.
- SOC changes and biorefinery inputs including enzymes and ammonia had large effects on the results.
- Returning lignin from the biorefinery process to the field to mitigate SOC changes was not preferable in a climate perspective when the alternative use of the lignin was combustion in a CHP plant to produce electricity, replacing natural gas electricity.

Ethanol, protein feed and briquette production from faba beans

- Processing whole faba beans in a green biorefinery increased the climate impact by 25% compared with a base case where faba beans were used as dairy cow feed. Land use and primary energy use decreased by 20% and 100%, respectively.
- In a climate impact perspective, ethanol production from starch extracted from faba beans was not beneficial when the starch was replaced by marginal grain in dairy cow feed.
- Therefore, maintaining the current use of faba beans for feed, while exploring other uses of faba bean crop residues, might be interesting to improve the climate impact. However, when harvesting the whole crop, it is important to consider the effects on SOC.

7.1.2 LCA methodology

The conclusions presented in this section refer to the second specific objective of this thesis work, which was to analyse the effects of different methodological choices in LCA studies on biorefinery systems.

- Choice of method to handle co-products and system boundaries (*i.e.* inclusion of upstream impacts in the form of SOC changes) both proved to have a large influence on the results.
- Comparison between LCA results is difficult. Using a *use of feedstock FU* in combination with a reference system (producing the same

functions as the biorefinery) enabled comparison of different biorefinery designs and biorefinery systems using the same biomass.

- Use of time-dependent modelling or GWP did not change the ranking between different scenarios. However, the time-dependent model provided additional information about climate effects over time, which could be important especially in relation to climate target deadlines.

7.1.3 General conclusions

Below, general conclusions in relation to the three themes of the thesis (see section 2.2) are presented.

Greenhouse gas emissions and energy balance of biorefinery systems and products

This thesis showed that lignocellulosic ethanol and biodiesel produced from straw and forest residues in emerging biorefinery systems have a lower climate impact and a more beneficial energy balance than fossil fuels. The biorefinery producing ethanol and co-products from straw showed a lower climate impact and more beneficial energy balance than the biorefinery producing biodiesel and co-products from straw. However, biochemical biodiesel production from lignocellulose is a very new concept and much less research has been conducted on process and strain development of oleaginous yeast, making comparison between the two biorefinery systems difficult.

The consequential LCA showed that when the biorefinery feedstock is a form of biomass that is currently used for other purposes, such as feed, there can be indirect effects that affect the overall climate performance of the system.

LCA methodology for biorefineries

Within LCA, it is well known that methodological choices affect the results of the study. For studies on biorefineries, methodological choices regarding functional unit, impact allocation and handling of biogenic carbon changes are especially crucial. Although methodological choices are essentially guided by the goal of the study, use of different functional units and allocation methods and different methods to estimate the climate impact from changes in biogenic carbon stocks can give different insights into the system under study, and can therefore be recommended for future LCA studies on biorefinery systems. LCA is a tool for assessing the potential environmental impact of products and services. It is also a useful tool for *learning* about the potential environmental impact of different production systems, for which using different methods is very valuable. This is needed to interpret, evaluate and compare LCA results.

An increasing share of biofuels can be expected to be produced in biorefineries and from lignocellulosic biomass. Therefore policy instruments, such as the RED, will need to become compatible with these systems. This will involve careful consideration of allocation methods and the handling of residues from agricultural and forestry in the RED. Including upstream impacts from harvesting residues in the RED should be considered. When SOC changes and nitrogen compensation due to residue harvesting were included in the RED calculations in this thesis, the biodiesel produced from straw and the ethanol produced from forest residues did not meet the 60% reduction target stipulated in the RED.

Influence of increased biomass harvesting on LCA results

The introduction of biorefineries can result in increased recovery of biomass from forestry and agriculture. As already mentioned, SOC changes proved to have large impact on climate impact in this thesis. However, it is important to remember that effects on SOC changes are difficult to predict, resulting in large uncertainties. In addition, LCA methodology is currently under development to handle timing of emissions and there is no consensus on how to deal with this issue. SOC changes are important not only for the climate impact of the system, but also for the sustainability of agricultural systems as a whole. Therefore, this issue deserves special attention within LCA.

7.2 Future research

To improve climate impact assessments of biorefinery systems using residues as feedstock, it is important to improve SOC change estimations and to find methods to deal with changes in biogenic carbon stocks in climate impact assessments. Furthermore, in order to assess the sustainability of biorefinery systems in a broader perspective, inclusion of more, and more relevant, environmental impact categories and social and economic assessments is needed.

The use of oleaginous yeast to produce valuable products from lignocellulosic biomass is relatively poorly described in the literature. In a systems perspective, more research is needed on the effects of different co-products, including food or feed, chemicals and pigments (such as carotene). Production of HVO from the lipids could be interesting in a Swedish perspective, as this application has increased significantly in recent years.

For all biorefinery systems studied in this thesis, there are several alternative process routes for the intermediate products. This thesis focused on transportation fuels, where the use of fossil fuels is generally high. However, there could be great potential for substitution of fossil resources when producing materials such as plastics or chemicals.

To evaluate the best use of biomass, alternative processes and process designs should be studied from an environmental perspective.

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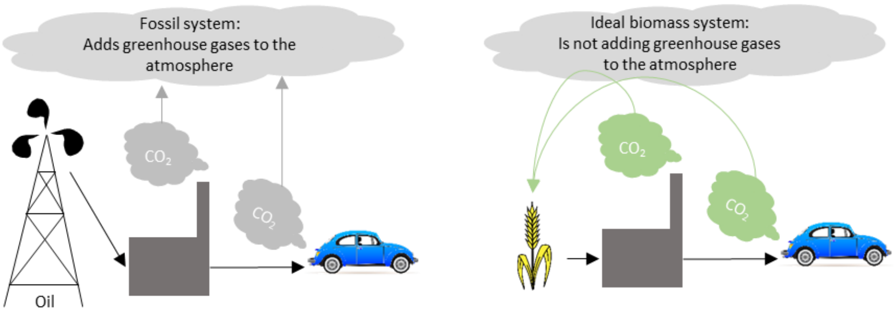
Popular science summary

Background

Climate change caused by increased concentrations of greenhouse gases in the atmosphere is one of the greatest environmental problems facing the planet. Use of fossil resources is the single largest source of emissions, but other activities such as deforestation and food production also cause greenhouse gas emissions.

Fossil resources are used in a variety of sectors, such as for transport, materials and chemical production and electricity and heat production. In order to counteract climate change, it is important to find alternatives to fossil resources. Biomass is a renewable resource that can be used to produce *e.g.* liquid fuels, materials such as plastics, chemicals, electricity and heat. Therefore, biomass can be a viable alternative to fossil resources in many sectors. Biomass is a renewable resource that is said to be carbon-neutral because the carbon dioxide it releases was taken recently from the atmosphere during growth of the biomass. In contrast, fossil fuels have been formed for millions of years, thus adding greenhouse gases to the atmosphere (see diagram).



Combustion of fossil fuels adds greenhouse gases to the atmosphere. In an ideal biomass system, the carbon dioxide released during combustion is absorbed by biomass as it grows.

However, energy and resources are used in the production of biomass, which gives rise to emissions of climate gases. In order to produce biomass, land is also needed, which can lead to land use changes, for example deforestation, in order to increase the production area. Land use changes can alter the stocks of carbon bound in biological materials (plants and soil). Reducing the amount of carbon bound in biomass and soil leads to increased concentrations of greenhouse gases in the atmosphere.

Most biomass used today in order to produce biofuels for the transport sector is grown primarily for energy, for example, wheat or maize for ethanol or rapeseed for biodiesel. This type of biofuel production has been criticised for using land that could otherwise be used for animal feed or food production. One alternative is to use by-products from agriculture (*e.g.* straw) and forestry (forest residues, tops and branches), so-called lignocellulosic material. The advantage of using by-products is that no extra land is needed to produce the biomass. However, increased harvesting of straw and forest residues leads to less biomass being added to agricultural and forestry systems. This can lead in turn to a reduced amount of carbon in the soil (resulting in carbon dioxide emissions). Lignocellulosic materials can be transformed into several of the biofuels used today, including ethanol and biodiesel. The transformation of lignocellulosic materials into liquid fuels (and other products) has several similarities with the current production of crop-based fuels. However, there are some differences, *e.g.* the lignocellulosic material needs to be pre-treated in several steps. Production plants that co-produce several valuable products, such as biomass fuels, materials and chemicals, are called biorefineries.

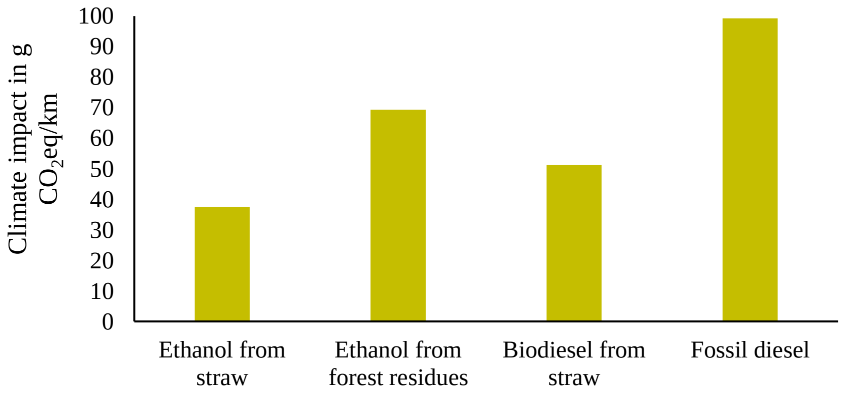
What did I do?

The purpose of this thesis work was to increase knowledge about the climate impact and energy balance of three new types of biorefinery systems, in particular biorefineries that produce fuels together with other products and biorefineries that use agricultural and forestry by-products as feedstock. The three systems were: 1) Co-production of ethanol and biogas from straw and forest residues (tops and branches); 2) co-production of ethanol, protein feed and solid fuel from faba beans (whole plant); and 3) co-production of biodiesel, biogas and electricity from straw.

In order to estimate the environmental impact of these biorefinery systems, life cycle assessment (LCA) was used. LCA is a method that is now widely used to estimate the environmental impact of a variety of products and services, including bioenergy. It is also used in biofuel policies such as the EU Renewable Energy Directive. This thesis examined how LCA can be used to estimate the environmental impact of biorefineries and the methodological difficulties with these types of studies.

What did I discover?

The climate impact of an ethanol-fuelled (ethanol produced from straw) passenger car was 62% lower than that of a fossil-fuelled car. The climate impact was 30% lower if, instead of ethanol produced from forest residues, the car was instead powered by straw-based biodiesel, for which the climate impact was 48% lower than for fossil diesel (see diagram). Effects on soil organic carbon were included in the climate impact assessment. Comparing the two biorefinery systems using straw as feedstock, the biorefinery system which produced ethanol, biogas and straw electricity had a higher climate gain than the biorefinery system which produced biodiesel, biogas and electricity.



Climate impact of ethanol and biodiesel produced from lignocellulosic material compared with that of fossil diesel.

Today faba beans are used mainly as an animal feed. The study on faba beans as a biorefinery input showed that using the crop for this purpose, instead of as feed, could have indirect effects on feed demand, which can be important for the results. Using whole faba beans as biorefinery feedstock increased the climate impact compared with using the beans themselves as feed and returning the rest of the plant to the field.

The aspect that proved to be very important for the climate balance of all biorefinery systems was a decrease in the amount of carbon in the soil due to increased harvesting of straw and forest residues and harvesting of the whole faba bean crop.

In LCA studies on biorefineries, several methodological choices are important for the results, particularly: allocation of environmental impacts between biorefinery products, selection of a functional unit describing the function of interest to the study, and how changes in the biogenic carbon stocks are handled. In order to interpret the results of LCA studies of various biorefinery systems, it is important to be aware that the method can be important for the results.

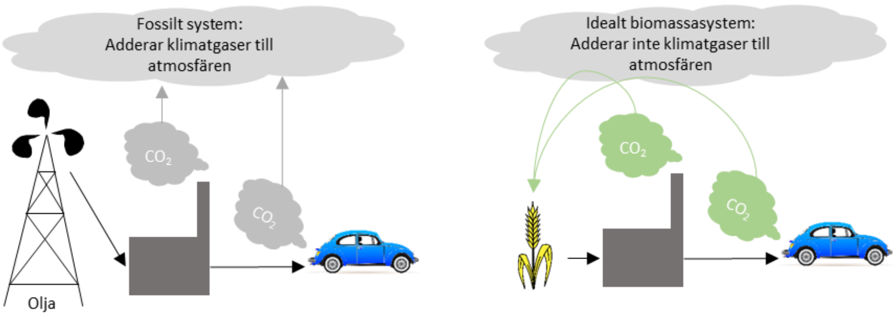
Finally, the results showed that ethanol produced from straw and forest residues and biodiesel from straw had a lower climate impact and better energy balance than fossil fuels and diesel. Therefore, biofuels produced from straw and forest residues can play an important role in reducing the climate impact of the transport sector. However, in order to get the best possible climate benefit, it is important to take into account changes in soil organic carbon as a result of increased harvesting of straw and forest residues, and to design systems with less potential impact on soil carbon stocks.

Populärvetenskaplig sammanfattning

Bakgrund

Klimatförändringen som orsakas av den ökade koncentrationen av växthusgaser i atmosfären, är en av vår tids största miljöproblem. Användningen av fossila resurser är den enskilt största utsläppskällan, men även andra aktiviteter så som avskogning och matproduktion orsakar utsläppen av växthusgaser.

Fossila resurser används inom en mängd sektorer, t.ex. för transporter, material och kemikalieproduktion samt för produktion av el och värme. För att motverka klimatförändringen är det viktigt att hitta alternativ till fossila resurser. Biomassa är en förnyelsebar resurs som kan användas för att producera t.ex. flytande bränslen, plast, kemikalier, samt el och värme. Därför kan biomassa vara ett alternativ till fossila resurser i många sektorer. Biomassa är en förnyelsebar resurs och sägs vara koldioxidneutral. Detta eftersom den koldioxiden som släpps ut nyligen har tagits upp från atmosfären under biomassans tillväxt. Detta kan jämföras med fossila bränslen som har bildats under miljontals år, och därmed ger ett tillägg av klimatgaser till atmosfären (se figur).



Förbränningen av fossila bränslen adderar klimatgaser till atmosfären, i motsatts till ett idealt biomassasystem där lika mycket koldioxid som släpps under förbränningen tas upp av biomassan när den växer.

Produktionen av biodrivmedel är dock inte koldioxidneutral. I produktionen av biomassa används energi och resurser, vilket ger upphov till utsläpp av klimatgaser. För att producera biomassa behövs också mark, vilket kan leda till markanvändningsförändringar, t.ex. avskogning, för att öka produktionsarealen. Den förändrade markanvändningen orsakar förändringar i de pooler av kol som finns i biologiskt material, både växter och mark. Att minska mängden kol bundet i biomassa och mark leder till en ökad koncentration av växthusgaser i atmosfären.

För att producera biodrivmedel för transportsektorn används idag mest grödor som odlas enbart för energiändamål, t.ex. vete eller majs till etanol eller raps till biodiesel. Denna typ av biodrivmedelsproduktion har fått kritik för att mark som annars används till foder- eller matproduktion tas i anspråk för att producera drivmedel. Ett alternativ är att istället använda vissa biprodukter från jordbruk (t.ex. halm) och skogsbruk (skogsrester), så kallat lignocellulosamaterial. Fördelen med att använda biprodukter är att ingen extra mark behövs för att producera biomassan. Ett ökat uttag av halm och skogsrester leder dock till att mindre biomassa tillförs jordbruks- och skogsbrukssystemen. Detta kan bland annat leda till minskad mängd kol i marken (vilket leder till utsläpp av koldioxid). Lignocellulosamaterial kan omvandlas till flera av de biodrivmedel som används idag, bland annat etanol och biodiesel. Omvandlingen av lingnocellulosamaterial till flytande bränslen (och andra produkter), har flera likheter med dagens produktion av grödbaserade bränslen. Dock finns det några skillnader, t.ex. så behöver lignocellulosamaterialet förbehandlas i flera steg. Anläggningar som samproducerar flera värdefulla produkter så som bränslen, material och kemikalier från biomassa, benämns med ett namn som *bioraffinaderier*.

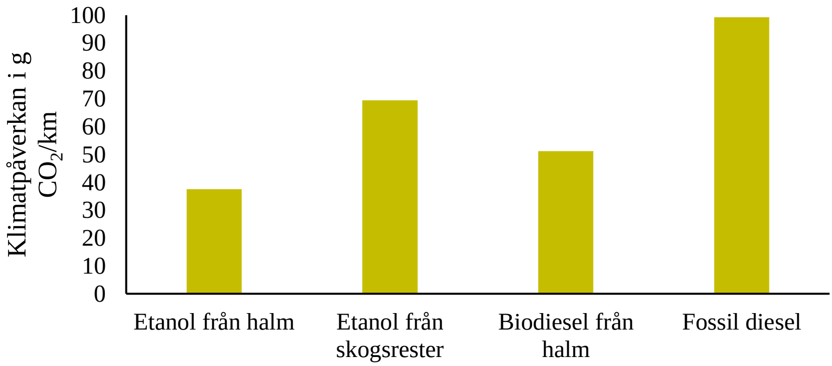
Vad har jag gjort?

Syftet med denna avhandling var att öka kunskapen om klimatpåverkan och energibalanser för tre nya typer av bioraffinaderisystem. Särskilt fokus var på bioraffinaderier som producerar drivmedel tillsammans med andra produkter, samt bioraffinaderier som använder biprodukter från jord- och skogsbruk som råmaterial. De tre studerade systemen var: 1. Samproduktion av etanol, biogas och el från halm och skogsrester (toppar och grenar), 2. Samproduktion av etanol, proteinfoder och fast bränsle från åkerböna (hela växten), 3. Samproduktion av biodiesel, biogas och el från halm.

För att uppskatta miljöpåverkan från bioraffinaderisystemen användes livscykelanalys (LCA). LCA är en metod som numera är flitigt använd för att uppskatta miljöpåverkan av en mängd olika produkter och tjänster, inklusive bioenergi. Metoden används även inom biobränslepolicys som t.ex. EU's förnyelsebart direktiv. Syftet med denna avhandling var även att bidra till diskussionen LCA som verktyg för att uppskatta miljöpåverkan av bioraffinaderier och vad det finns för metodologiska svårigheter.

Vad kom jag fram till?

Klimatpåverkan för en etanoldriven (etanol producerad från halm) personbil var 62 % lägre än för fossila bränslen och 30 % lägre om etanolen produceras från skogsrester. Om bilen istället drivs med biodiesel producerad från halm var klimatpåverkan 48 % lägre än med fossil diesel (se graf). Inverkan på markkol är medräknat. Vid en jämförelse av två bioraffinaderisystem som använde halm som råvara visade det sig att bioraffinaderisystemet producerade etanol, biogas och el från halm hade en högre klimatvinst än bioraffinaderisystemet som producerade biodiesel, biogas och el.



Klimatpåverkan för etanol och biodiesel producerad av lignocellulosamaterial, jämfört med fossil diesel. Markkolseffekter är medräknade.

Åkerböna används idag till framförallt foder. Studien på åkerböna som bioraffinaderiråvara visade att användningen av en gröda som idag har en användning, som t.ex. foder, som bioraffineriråvara kan ha indirekta effekter på efterfrågan på foder. Detta visade sig vara viktigt för resultatet. Att använda hela åkerbönan som bioraffinaderiråvara ökade klimatpåverkan, jämfört med att använda själva bönan som foder och återföra resten av växten till marken.

Den aspekt som visade sig vara mycket viktigt för klimatbalansen för alla studerade bioraffinaderisystem den minskade mängden kol i marken på grund av ett ökat uttag av halm och skogsrester, samt på grund av helskörd av åkerböna.

För LCA-studier på bioraffinaderier är flera metodval viktiga för resultatet: fördelningen av miljöpåverkan mellan bioraffinaderiets produkter, val av funktionell enhet som beskriver den funktionen som är intresset för studien, och hur förändringar i mängden kol bundet i biomassa och mark hanteras. För att tolka resultaten från LCA-studier av olika bioraffinaderisystem är det viktigt att vara medveten om att val av metod kan vara betydelsefullt för resultatet.

Slutligen, resultaten visade att etanol producerad från halm och skogsrester, samt biodiesel från halm, hade lägre klimatpåverkan och bättre energibalanser än fossil bensin och diesel. Därför kan biodrivmedel producerade från halm och skogsrester spela en viktig roll i att minska klimatpåverkan från transportsektorn. För att få bästa möjliga klimatnytta, är det dock viktigt att ta hänsyn till markkolsförändringar till följd av ett ökat uttag av halm och skogsrester, och utforma system med minsta möjliga effekt på markens kollager.

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