Biorefinery Systems for Energy and Feed Production

Greenhouse Gas Performance and Energy Balances

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Abstract

The current dependence on fossil fuels is problematic because these are a finite resource and because their combustion causes environmental degradation. Biorefineries produce a variety of valuable products from biomass that can replace products from petroleum refineries. Biorefining can also introduce new process technologies and increase the use of biomass resources traditionally not utilised for industrial purposes. Although biomass is considered to be a renewable resource, biomass production and processing are associated with environmental impacts.

This thesis examined the environmental impacts of biorefinery systems and products by studying the climate impact and energy balance of two innovative biorefinery systems from a life cycle perspective. These systems were: (1) co-production of ethanol, biogas, electricity and heat in a lignocellulosic biorefinery and (2) processing of faba beans in a green crop biorefinery producing ethanol, protein feed and briquettes. Life cycle assessment methodology concerning biorefinery systems and biomass utilisation was also examined.

The analysis showed that increased residue harvesting from agriculture and forestry had a potentially high impact on overall greenhouse gas (GHG) performance, mainly due to soil organic carbon (SOC) changes. Ethanol from the lignocellulosic biorefinery gave GHG savings of 51-84% compared with a fossil fuel reference and used between -0.71 and 0.20 MJ fossil energy per MJ ethanol. Biorefinery processing of whole faba beans marginally decreased the climate impact (-2%), while primary fossil energy use (-119%) and land use (-20%) decreased significantly compared with reference use of the beans as dairy cow feed. On balance, it was concluded that ethanol production from faba bean is not favourable in a climate perspective. The results for GHG performance and energy balance varied significantly depending on method choices, the most influential being handling of multi-functionality and system boundaries, *i.e.* inclusion of upstream impacts in the form of SOC losses.

Keywords: biofuels, ethanol, biogas, animal feed, LCA, calculation methodology. green biorefinery, lignocellulosic biorefinery, crop residue recovery

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“You are not Atlas carrying the world on your shoulder. It is good to remember that the planet is carrying you”

Vandana Shiva

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 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.

II Karlsson, H., Ahlgren, S., Strid, I. & Hansson, P.-A. Faba bean for feed or biorefinery feedstock? Greenhouse gas and energy balances of different applications (submitted).

Papers I is reproduced with the permission of the publisher.

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

I Planned the study together with the co-authors. Was primarily responsible for data collection, impact assessment and for writing the paper, all with input from the co-authors.

II Planned the study together with the co-authors. Was primarily responsible for data collection, impact assessment and for writing the paper, all with input from the co-authors.

Abbreviations

ALCA	Attributional life cycle assessment
CHP	Combined heat and power generation
CLCA	Consequential life cycle assessment
EJ	Exajoule
FU	Functional unit
GHG	Greenhouse gases
Ha	Hectare
ISO	International standardization organization
LCA	Life cycle assessment
MJ	Megajoule
RED	Renewable energy directive
SOC	Soil organic carbon
TWh	Terawatt hour

1 Introduction

Fossil resources are used for the production of multiple products which are crucial for modern society, including transportation fuels, chemicals and plastics. Globally, 57% of the oil consumed is used for transport (OPEC, 2012) and the transportation sector is largely fossil-based, with 97% of the energy used originating from fossil fuels (IEA, 2013). The industrial sector uses 28% of the global oil supply, with the petrochemical sector alone using 11% (OPEC, 2012). The dependency on fossil fuels is problematic, as fossil resources are finite and, in addition, the use of these resources causes severe environmental problems and is a major driver for one of the most severe enviornmental problems of our time, global climate change (IPCC, 2007). Therefore society needs to move away from its dependency on fossil resources. In a bio-economy, biomass will replace fossil resources as the main source for fuels and materials (Keegan *et al.*, 2013). Therefore, biomass will be used not only for feed and food, but also for fuels, chemicals and materials (Dale & Kim, 2010), which will increase the demand for biomass. Biomass is a renewable resource but not unlimited, and hence efficient utilisation of available resources is important. For efficient utilisation of biomass and co-production of products such as energy, food and chemicals, the biorefinery concept is gaining increased interest (IEA, 2009; Kamm *et al.*, 2007).

Biorefineries process biomass into a wide range of products that can replace the products from fossil refineries, including fuels, materials and chemicals, and can therefore play a central role in replacing fossil refineries.

The biorefinery concept introduces new technologies for the treatment of biomass, enabling a diverse variety of products to be produced from biomass. Moreover, the introduction of biorefineries can change the utilisation of biomass, so that forms of biomass previously considered a residue (straw and forest residues) could find new uses. Therefore, the incentive to harvest a larger proportion of the biomass produced in agriculture and forestry may increase.

Although biomass is considered to be a renewable resource, biomass production and processing is associated with environmental impacts from *e.g.* inputs to production and processing (such as energy, nutrients, enzymes *etc.*), transport and soil emissions. In addition, increased demand for land for bioenergy production may lead to indirect land use effects such as land transformation, with negative impacts on the environment. Consequently, the environmental impacts of biomass production, new process techniques and the changed utilisation of biomass need to be assessed in a life cycle perspective. The life cycle perspective can be used to evaluate the environmental performance of the biorefinery system and products, improvement options, alternative biomass uses and process designs. This evaluation can include systematic comparison of biorefinery systems and products with conventional production systems.

Life Cycle Assessment (LCA) is a method to evaluate the potential environmental impact of products and services (ISO, 2006a). LCA is being increasingly used for environmental evaluation of processes and products in research, but also for policy applications. Hence, method development and evaluation of LCA are important.

2 Objectives

The overall aim of this licentiate project was to increase understanding of the climate impact and energy balance of biorefinery systems and biorefinery products. Specific objectives of the work were to:

- Evaluate the climate impact and energy balance of two different biorefinery systems: (1) Co-production of ethanol, biogas, electricity and heat in a lignocellulosic biorefinery; and (2) processing of faba beans in a green crop biorefinery producing ethanol, protein feed and briquettes. The method used was LCA.
- Contribute to the discussion on LCA methodology concerning biorefinery systems and biomass utilisation by determining how different methodological approaches within LCA for determining the total environmental impact (focusing on climate impact) affect evaluation of the actual system and in relation to other production systems.

3 Background

3.1 Biorefineries

Multiple definitions of the term ‘biorefinery’ have been proposed, but a common feature of all definitions is that biorefineries are facilities producing a spectrum of products from biomass. A commonly cited definition is that 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).

3.1.1 Biorefinery feedstock

The feedstock used in biorefineries can be agricultural crops, forestry products, residues from agriculture and forestry, organic wastes *etc*. Biomass availability, especially for the bioenergy sector, has been estimated in many previous studies (see *e.g*. BEE, 2011; Berndes *et al.*, 2003). The resulting estimates on the future availability of biomass for energy purposes depend on multiple factors, such as projections on future land availability. This depends in turn on competing markets for biomass, land degradation and climate change, as well as assumptions regarding productivity (yield), population growth, diets, *etc*. (Hallström *et al.*, 2011). In addition, there are a number of restrictions to biomass harvesting, including economic, technical and ecological restrictions. Different assumptions regarding these factors result in diverging estimates of biomass potential, making the different estimates difficult to compare (Börjesson *et al.*, 2013). A distinction is made between theoretical, technical, economic, implementation and (sometimes) sustainable biomass potential. The theoretical potential estimates all available biomass, whereas the others estimate biomass potential with technical, economic, social or environmental restrictions (BEE, 2011).

Börjesson *et al.* (2013) reviewed a number of studies on the potential for increased biomass withdrawal for biofuel production in Sweden, some values from which are presented below. Note, however, that due to differences in the studies reviewed, these values should be regarded with caution and merely provide an indication of the magnitude of biomass availability. The approximate biomass potential reported was 55-70 TWh per year in a short time perspective, with the largest potential originating from stump harvesting and forest residues and with straw representing approx. 4 TWh. In a longer perspective (30-50 years), the potential was estimated to be 80-100 TWh or higher (180-190 TWh), with the higher potential representing a future scenario with increased stem wood production and partial fertilisation of forest land (Börjesson *et al.*, 2013). Estimates for future biomass potential in bioenergy in Europe also vary greatly, with a recent review reporting a variation from 2.8 EJ to 23.8 EJ for 2020 (BEE, 2011).

Biomass potential studies often focus on the available biomass for the bioenergy sector, but emerging industries for bio-material and bio-chemical production will compete for the biomass resources in the future. This is often not considered in biomass availability studies (Smeets *et al.*, 2010 *cit.* Keegan *et al.*, 2013; Berndes *et al.*, 2003). The novel uses of biomass in combination with traditional uses for food, feed, building material, pulp and paper *etc.* will increase the pressure on the biomass resources (Keegan *et al.*, 2013). Efficient use of available biomass resources is therefore crucial in meeting the future demands for biomass.

3.1.2 Biomass for non-food purposes

In recent years the sustainability of using conventional food crops for biofuel production has been questioned. The main concerns are competition with food production (Escobar *et al.*, 2009) and greenhouse gas (GHG) emissions due to direct and indirect land use changes (LUC and iLUC) (Fargione *et al.*, 2008; Searchinger *et al.*, 2008). As an alternative, lignocellulosic materials such as residues from agriculture and forestry have been identified as promising feedstock for sustainable biofuel production (Tilman *et al.*, 2009).

Biofuel production currently uses approximately 65% of the vegetable oil produced in the EU, 50% of Brazilian sugarcane and 40% of US maize (OECD-FAO, 2012). In 2008, around 2% (33.3 million ha) of global arable land was used for biofuel production, and these figures do not consider the impact from by-products to biofuel production, for example protein feed from bioethanol (grain) or biodiesel (rapeseed) production (Fargione *et al.*, 2010). The International Energy Agency predicts that by 2050, 100 million hectares will be needed to supply 27% of transportation fuels (65 EJ feedstock will be required) (IEA, 2011). This corresponds to approx. 6% of the arable land projected by the FAO to be available in 2050 (Hallström *et al.*, 2011).

3.1.3 Biomass composition and implications for biorefinery processing

Petroleum is a rather homogeneous feedstock, while biomass is much more variable in composition. This has both positive and negative implications for biorefineries compared with petroleum refineries. One advantage is that biorefineries can produce a larger set of products, including fuels, electricity, steam, chemicals, monomers and polymers, fertilisers *etc*. and, notably, animal feed and human food. A disadvantage is that biorefineries require more complex technologies, for example to separate the substances (Dale & Kim, 2010). Biomass contains carbohydrates, lignin, proteins, fats and, in addition, a number of substances such as vitamins, dyes, flavours and aromatic essences with different chemical structures. Seperation into main groups of substances is a first essential step in biorefinery processing. These groups of substances can then be further processed into a wide range of different end-products (Kamm & Kamm, 2007).

3.1.4 Biorefinery concepts

The term biorefinery include many types of biorefinery concepts using different types of feedstock and technologies and producing different products. Biorefineries have been categorised in different ways (see *e.g.* Cherubini *et al.*, 2009; Kamm & Kamm, 2004). 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 classification suggested by Kamm and Kamm (2007) is applicable. This comprises four biorefinery concepts:

- Lignocellulosic biorefineries, using lignocellulosic feedstock such as straw, wood and grass
- Whole crop biorefineries, using whole crops such as 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.

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

The two biorefinery concepts examined in this licentiate thesis, a lignocellulosic biorefinery and a green biorefinery, are further described in the following sections.

3.1.5 Lignocellulosic biorefinery

The lignocellulosic biorefinery is considered to be one of the most promising biorefinery concepts for two reasons: the low cost of the feedstock and the strongly established position of the output products on existing markets and possibly also in future bio-based markets. Lignocellulosic feedstock may include *e.g.* straw, wood, grass, paper-waste *etc.*, which are relatively inexpensive products (Kamm & Kamm, 2007).

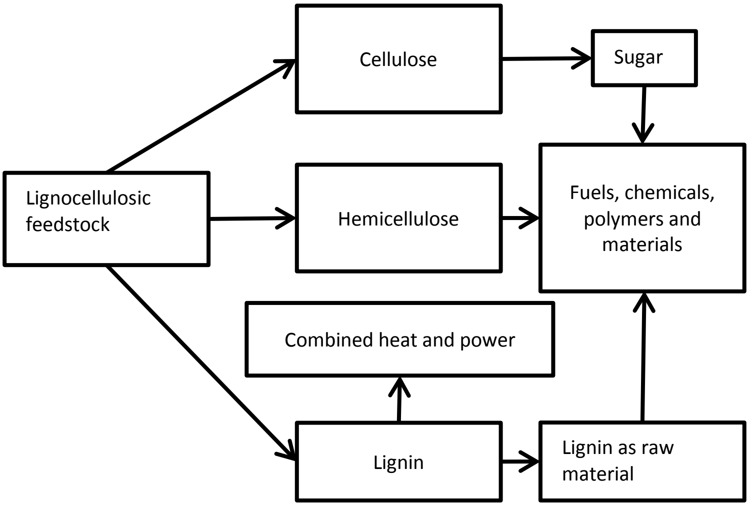

Figure 1. Schematic description of the lignocellulosic biorefinery concept, modified from Kamm & Kamm (2007)

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 1). Cellulose can be hydrolysed to glucose using enzymes or 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). Thermochemical hydrolysis is usually carried out first to hydrolyse hemicellulose, after which cellulose is commonly hydrolysed using an enzymatic hydrolysis method (Olofsson *et al.*, 2008). The sugars can then be used for *e.g.* ethanol production via fermentation 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, and most of this lignin is burned for the production of heat and electricity. 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).

3.1.6 Green biorefinery

In a green biorefinery, fresh or ensiled biomass is utilised to produce a variety of high value products (Kromus *et al.*, 2010). Green crops (for example 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 t/ha (temperate climate) (Kromus *et al.*, 2010). Consequently, there is great potential to produce large amounts of protein and organic material that can be further processed.

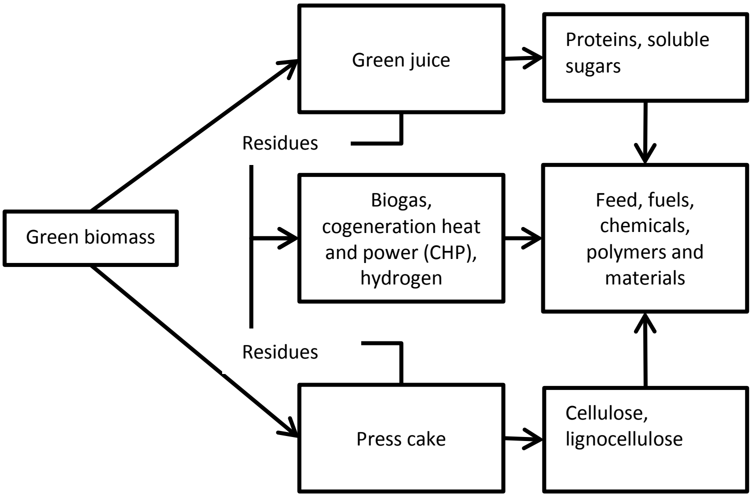

Figure 2. Schematic picture of a green biorefinery, modified from Kamm and Kamm (2007)

The processing of green crops can convert the protein they contain, which is conventionally only accessible by ruminants, into a form that is accessible by 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 2). Many processing options and different end-products are possible. 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).

Since the green biorefinery system is based on fresh biomass, the biomass has to be conserved in order for the biorefinery to run during the whole year and not only during the harvesting season. Furthermore, decentralised processing has been proposed, with initial processing taking place close to the biomass source, while further processing is performed at a central plant, to take advantage of economies of scale while benefiting rural economies and lowering the biomass transportation costs (Kromus *et al.*, 2010; Kromus *et al.*, 2004).

3.2 Life cycle assessment (LCA)

Life Cycle Assessment is an environmental systems analysis (or environmental assessment) tool. Other environmental systems analysis tools include Environmental Impact Assessment (EIA), Ecological Risk Assessment (ERA) and Material Flow Analysis (MFA). In general, environmental systems analysis tools examine social, technical and natural systems and the links between these systems (Baumann & Tillman, 2004).

LCA is an established method to quantify 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. 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 the ISO standards ISO 14040 and ISO 14044 (ISO, 2006a; ISO, 2006b). Several steps are included in LCA, including, Goal and Scope Definition, Inventory Analysis, Impact Assessment and Interpretation (Figure 3). In the Goal and Scope Definition the aim of the study is stated as well as specifications for the modelling. In the Inventory Analysis step, data on resource use and emissions are collected. In the Impact Assessment step, individual emissions and resource use are grouped into different environment impact categories, facilitating the interpretation of the life cycle inventory results. This is done by applying impact assessment methods. Interpretation of the results is done with regard to the initial aim of the study, the data used and impact assessment method used. As the arrows in Figure 3 indicate, LCA is an iterative process. The specifications used for the modelling work presented in this thesis are described in section 4.1.

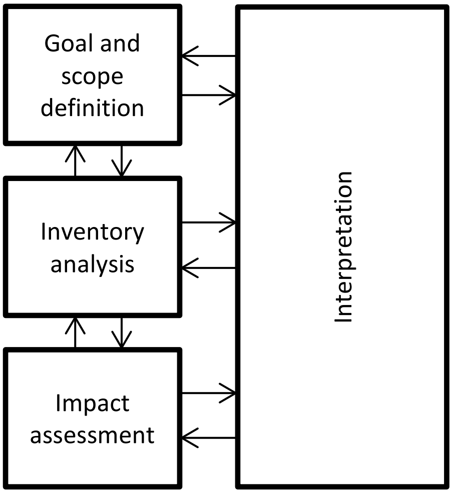

Figure 3. Schematic figure of the life cycle assessment methodology

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 a common application is product declarations (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.

3.2.1 LCA of biofuels and biorefineries

LCA has been widely applied to assess the environmental impact of biofuels (see *e.g.* Wiloso *et al.,* 2012). However, there are still some methodological issues to be resolved, *e.g.* the results can vary significantly, not only due to production system differences, but also depending on methodological choices (Börjesson & Tufvesson, 2011; Gnansounou *et al.*, 2009). For evaluation of biofuels this creates uncertainty and prevents comparison between different feedstocks and technologies.

In the specific case of LCA studies on biorefinery systems, Ahlgren *et al.* (2013) identified seven key issues: (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) choice of data, (6) land use and (7) 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, allocation issues (*i.e.* partitioning of the environmental impact between co-products) and choice of data, while others relate exclusively to the use of biomass, in particular land use issues, including indirect land use changes, and biogenic carbon and timing of emissions. Biogenic carbon is commonly considered to be climate-neutral, as it is assumed that the carbon from CO_2 sequestered during growth of the biomass is equal in quantity to the carbon released during use of the biomass. 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 is especially relevant for biomass systems with long rotation times.

A biorefinery is by definition a multi-functional system, meaning that several products are produced in the same production plant. When analysing biorefineries using LCA, two of the general key issues are 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 producing several different products, it can be difficult to identify one main product or function. Second, an allocation or multi-functionality problem arises when more than one product or service share or partly share a production system. The general principles for handling multi-functionality problems are further explained below. The fact that biorefineries do not produce a main product, but rather a set of valuable co-products with different functions and physical attributes, can complicate the handling of multi-functionality problems (Ahlgren *et al.*, 2013).

3.2.2 Handling multi-functionality in LCA

The principles for handling multi-functionality problems are specified in the ISO standards. According to ISO (2006b), allocation of the environmental impact between co-products should be avoided if possible. 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.

System expansion can be carried out in one of two ways (Figure 4). The environmental impact of the selected main product can be calculated as the emissions from the main production system minus the avoided emissions from the use of the by-products. This application of system expansion is sometimes called substitution. Alternatively, in comparative studies one approach is to expand the system boundaries to include other functions, so that the system under study encompasses the same multiple functions (Guinée *et al.*, 2002). This approach is sometimes called system enlargement (JRC, 2010). Figure 4 shows these two approaches to system expansion applied in the case of an LCA comparing production of product A in systems 1 and 2. In the substitution approach, the system boundary is expanded and the environmental impact of product B from system 3 is subtracted from system 2. In the system enlargement approach, to make systems 1 and 2 comparable system 3 is added to system 1.

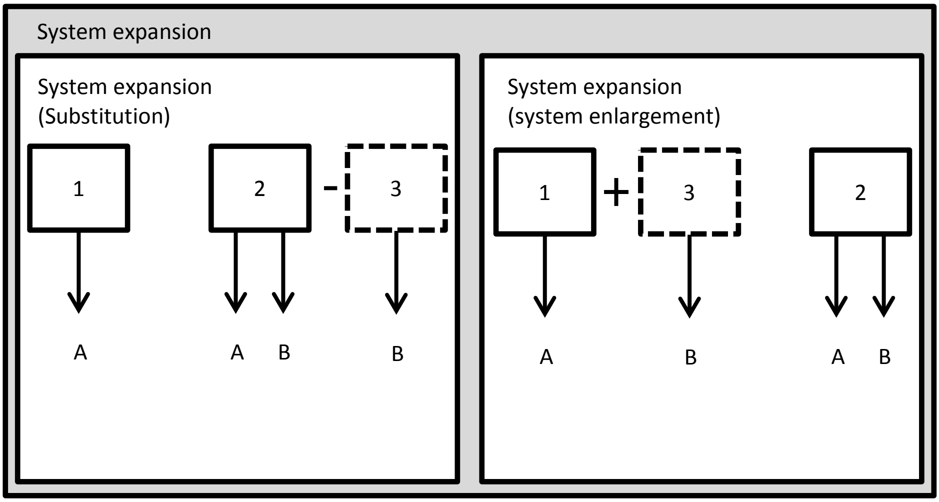

Figure 4. Two different approaches to system expansion in LCA, modified from Ahlgren et al. (2013).

System expansion is generally used to handle multi-functionality problems in CLCA studies. As described above, CLCA is applied when aiming to assess the impact of changes, and thus the technologies affected by the change should be included. These technologies are often called marginal technologies. Changing from one production system to another results in increased demand for some products and decreased demand for others. If the change can be categorised as small (as is often the case), marginal technologies are affected by a small change in demand (Weidema *et al.*, 1999). Small changes are identified as changes that do not affect the whole market (Weidema, 2003).

When performing system expansion to solve a multi-functionality problem, the marginal technologies identified may also be a multi-functional process, *i.e.* the attempt to solve the multi-functionality problem introduces a new multi-functionality problem. When using system expansion and the marginal product identified originates from a multi-functional process, it is important to distinguish between combined and joint production. In combined production the output volumes can be independently varied. The process can then be divided into sub-processes and the allocation can be based on physical relationships between the products. In joint production the output volumes of the co-products are fixed. For joint production, a distinction between determining and dependent co-product is needed and is decisive for the processes affected by a change in demand. A determining co-product is the product for which a change in demand affects the production volume, while for a dependent product a change in demand does not affect the production volume (Weidema *et al.*, 2009). This principle is important when handling straw and forest residues in CLCA, since these can be categorised as dependent co-products in joint production systems, as further explained in section 5.2.1.

3.2.3 The Renewable Energy Directive

In the European Union (EU) Renewable Energy Directive (RED) (EC, 2009), 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 by 2020 and includes GHG reduction requirements from a fossil fuel reference including the current requirement of 35% reduction and the forthcoming 50% and 60% reduction requirements by 2017 and 2018, respectively (EC, 2009). The method to calculate the GHG performance 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 included in the RED and these criteria must be met in order for the biofuel to count towards the target. Through implementing reduction requirements, the calculation method in the RED is potentially highly influential for the European biofuel market.

4 System descriptions and method

4.1 System descriptions of the biorefineries studied

The lignocellulosic biorefinery studied in Paper I involved co-production of ethanol and biogas using two different feedstocks: straw or forest residues (tops and branches) (Figure 5).

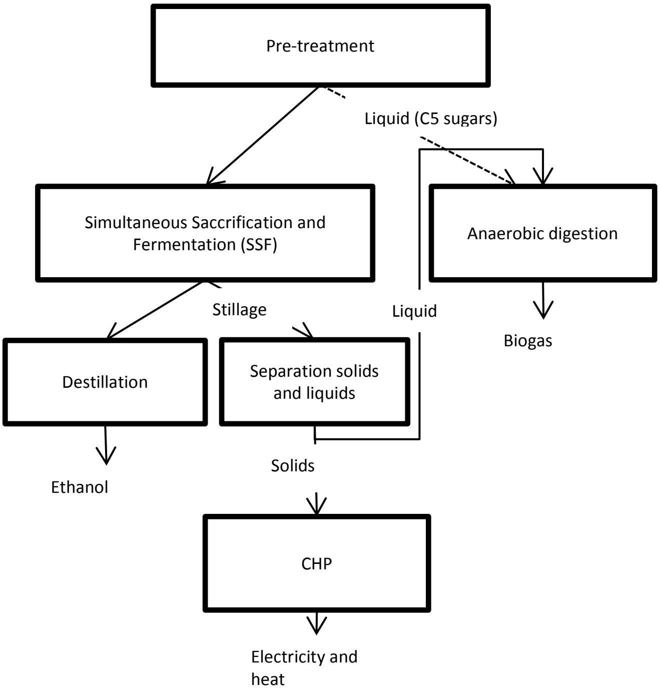

Figure 5. The biorefinery system considered in Paper I. Modified from Börjesson et al. (2013). The dotted arrow represents feeding of pentose sugars from the pre-treatment directly to the anaerobic digestion as opposed to all sugars going to the hydrolysis and fermentation.

The processes basically consisted of pre-treatment followed by simultaneous saccrification and fermentation (SSF) and anaerobic digestion (Figure 5). The pentose sugars can be retained in the pre-treatment and can then either be fed into the SSF (straw scenario) or separated and fed into the anaerobic digestion, as represented by the dotted arrow in Figure 5 (forest residue scenario). The products were ethanol, upgraded biogas, electricity and heat.

The green biorefinery studied in Paper II used ensiled faba beans as feedstock. The whole crop was harvested and processed in the biorefinery (Figure 6). Green crop fractionation was used to separate the green juice and the press cake. The protein in the green juice was separated out and used to produce concentrate animal feed. The starch and protein in the bean was separated out and ethanol was produced from the starch and concentrate protein feed from the protein fraction.

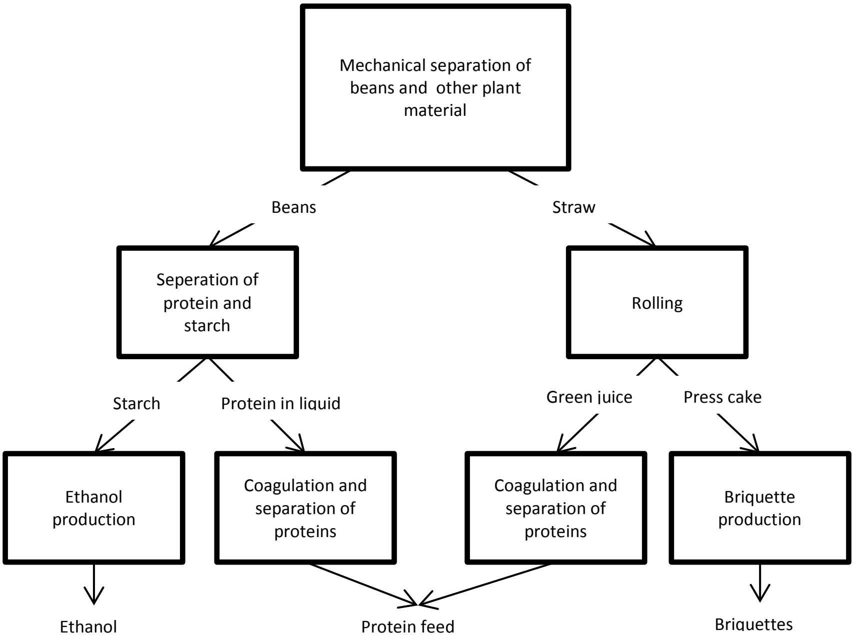

Figure 6. The green biorefinery considered in Paper II.

4.2 LCA method applied

4.2.1 Goal and scope

In LCA the objective of the study, including the intended application of the results and the intended audience, guides many of the methodological choices (ISO, 2006a). The LCA methodology applied in Papers I and II differed (Table 1).

The objective in Paper I was to estimate the GHG performance 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 (EC, 2009). 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 GHG accounting. The LCA calculation method in the RED is similar to ALCA (see section 3.2). To enable comparison between different fuels, the RED has standardised procedures for setting the functional unit (1 MJ biofuel), system boundaries (residues and wastes are cut off from the main system) and allocation (based on the lower heating value (LHV) of the products). In Paper I the RED method was compared with Method I (ISO) using system expansion to avoid allocation in accordance with ISO (ISO, 2006b) in addition Method I (ISO) was including all life cycle steps up to the factory gate, *i.e.* including upstream impacts from harvesting residues, which are not included in the RED.

Table 1. *LCA method applied in Papers I and II*

	Paper I	**Paper II**
Type of LCA	RED/ISO	CLCA
Allocation method	Energy allocation/ system expansion	System expansion
Functional unit	1 MJ LHV ethanol	1 ha faba bean cultivation
Choice of data	Average	Marginal
System boundaries	Cradle to gate[a]	Cradle to gate

[a]In the RED method residues from agriculture and forestry are considered to be 'free"' up to the point of harvest.

The objective of Paper II 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 overall aim was to determine the most environmentally beneficial use of available faba bean production (with regard to the three impact categories assessed). The intended audience was biorefinery owners, faba bean producers and policy makers. 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 should be included, rather than average data. For processes for which a change in demand was induced due to the change from the reference situation, marginal data were identified in Paper II.

The functional unit defines and quantifies the function(s) of the product under study (ISO, 2006a). The single product FU used in Paper I allowed comparison between products with the same function, *i.e.* ethanol produced from different types of feedstock. However, selecting a single product FU for a multifunctional biorefinery system involves handling co-products using allocation by partitioning (Method II (RED)) or by system expansion (by substitution) (Method I (ISO)). The FU selected in Paper II was 1 hectare of faba bean cultivation, an input-based FU that was used to assess the consequences of different uses of (the same) 1 hectare of faba bean.

4.2.2 Scenario descriptions

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

Table 2. *Description of the scenarios used in Papers I and II*

	Scenario	**Description**
Paper I		
	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 II		
	Reference	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)

4.2.3 Accounting for changed use of biomass

The introduction of biorefineries can result in crops traditionally used for food or feed purposes being used as biorefinery feedstock. It could also involve a change in harvesting practices, *e.g.* crop residue harvesting to a larger extent. For increased or changed use of crops, it is relevant to consider the indirect effects on food or feed use (Paper II). For changes in management such as increased biomass harvesting, direct impacts on the production systems are relevant (Papers I and II). In Paper I**,** the recovery of agricultural and forestry residues for use in biorefineries resulted in increased withdrawal of biomass from agriculture and forestry. Effects due to residue harvesting on soil organic carbon (SOC) and compensation for nitrogen (N) removal were included in Paper I (Method I (ISO)).

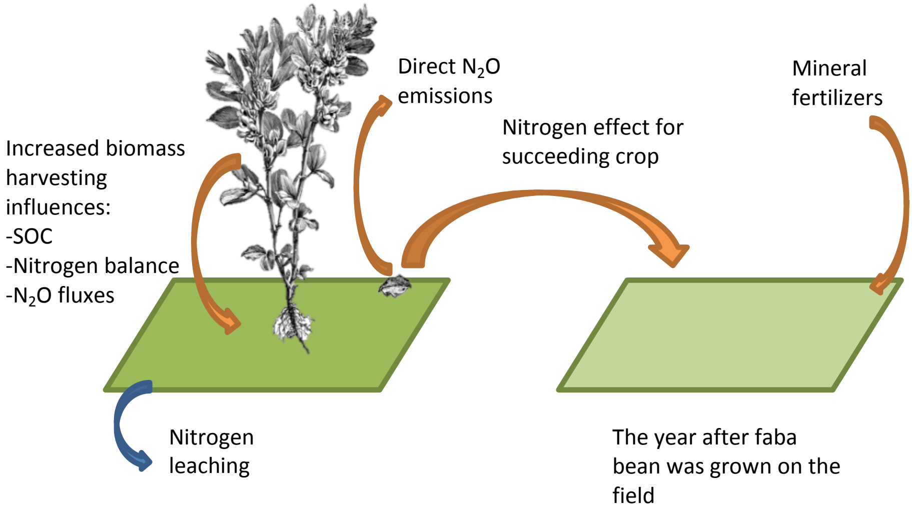

Figure 7. Changes due to whole crop harvesting that was included in the analysis (Paper II) (the picture is modified from http://etc.usf.edu/clipart/).

In Paper II, a change in use of an available faba bean crop was considered, for which the effects of changing from the reference use of faba bean as a protein feedstuff for cattle (scenario 1) to two other types of uses were assessed. The two alternative scenarios were: whole crop harvesting and use as feedstock for a green biorefinery (scenario 2); and whole crop harvesting and use as roughage feed (scenario 3) (see Table 2).

The following changes were considered:

- Changes in the N balance and SOC changes due to whole crop harvesting, as opposed to harvesting only the beans and returning the crop residues to the soil (Figure 7)
- Changes in feed use when the faba beans (only the beans) were no longer used as cattle feed and changes due to the introduction of new feed products, concentrate protein feed in the biorefinery scenario (2) and a protein-rich roughage feed in the roughage scenario (3)
- Changes in demand for feed products can result in indirect land use changes. This was handled by accounting for carbon stock changes due to land transformation. In some situations, changes in production were assumed to be supplied via a yield increase, *i.e.* increased demand was assumed not to result in land transformation
- Changes in production output were handled with system expansion, where the differences in product output were assumed to replace marginal products with equivalent functions
- Changes in product use (artificial fertiliser, diesel, electricity) were handled as increased use or avoided production of marginal products.

5 Results and discussion

5.1 GHG performance and energy balances

5.1.1 Lignocellulosic ethanol co-produced with biogas

Previous LCA studies on lignocellulosic ethanol have primarily focused on stand-alone ethanol production, isolated from other system processes, and have shown that lignocellulosic ethanol is generally more favourable in terms of climate impact and energy balance than first generation ethanol (Wiloso *et al.*, 2012). Paper I assessed the GHG performance and energy balance of ethanol co-produced with biogas and electricity in a lignocellulosic biorefinery. For ethanol produced from straw and forest residues, the GHG savings were 51-84% relative to the fossil fuel reference in the RED (83.9 g CO_2eq per MJ fuel; EC, 2009) and the primary fossil energy use was between -0.71 and 0.2 MJ per MJ ethanol. The results varied depending on calculation method used. However, ethanol produced from forest residues generally had lower GHG emissions than ethanol produced from straw, while no such trend was observed for the energy balance. Significant life cycle steps were enzyme production and SOC changes due to residue harvesting. The impact from residue harvesting is further discussed in section 5.1.3. Enzyme production contributed 13-70% of the climate impact and also contributed significantly to the fossil energy use. Enzyme production is known to be energy-intensive and has been shown to have a significant impact in the life cycle of lignocellulosic ethanol in previous studies (MacLean & Spatari, 2009; Slade *et al.*, 2009). Enzymatic hydrolysis is considered to be one of the most promising methods for hydrolysing lignocellulosic materials (Binod *et al.*, 2011). Apart from the climate impact and energy use of enzyme production, the enzymes themselves are costly. Research aiming to reduce the required enzyme dose for cellulosic ethanol is ongoing, for example by improving enzyme efficiency (Novozymes A/S, 2012) and developing appropriate pre-treatment methods (Yu *et al.*, 2011; Linde *et al.*, 2008).

5.1.2 Feed or biorefinery feedstock? Indirect impact from using traditional feed crops as biorefinery feedstock

Paper II examined two alternative scenarios to the reference scenario (1), namely the biorefinery and roughage scenarios (2 and 3). Changing the use of faba bean from the current use as a cattle feedstuff to processing of the whole plant in a biorefinery decreased the climate impact slightly (-2%), while a more significant decrease was observed for primary fossil energy use (-118%) and land use (-20%) (Figure 8).

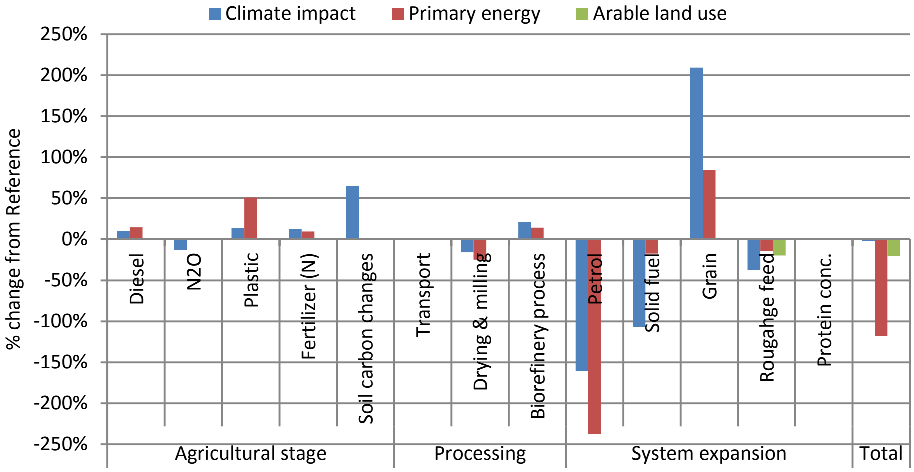

Figure 8. Disaggregated change from the Reference scenario for climate impact, primary energy use and arable land use due to the introduction of the biorefinery.

Apart from the substitution effects when *e.g.* ethanol replaced petrol, indirect changes due to the changed use of the beans as a cattle feedstuff had a great impact on the results (as indicated by the bars for grain and roughage feed in Figure 8).

The climate impact for the agricultural and processing steps increased by 95% relative to the reference (scenario 1). This was primarily due to SOC changes and increased demand for plastics for ensiling, diesel for field operations and nitrogen fertiliser for the subsequent crop. Impacts from whole crop harvesting are discussed in section 5.1.3. In addition, the biorefinery process, with inputs of electricity and other inputs such as enzymes, contributed around 10% of the climate impact during the agricultural and processing stages.

Paper II analysed changes in feed rations for dairy cows induced by the changed use of faba bean. The changes in annual feed use per hectare are shown in Figure 9. The basis of the comparison was that the biorefinery option (scenario 2) should supply the same number of dairy cows with complete feed rations as the reference (scenario 1). When the use of 1 hectare of faba bean changed, 3500 kg faba beans could no longer be used as feed. This was substituted with 2900 kg grain, while the introduction of the new protein feed products from the biorefinery decreased the use of grass/clover roughage by 1400 kg and protein feed concentrate by 10 kg. The largest impact on climate change and fossil energy use was related to the increased use of grain resulting from the withdrawal of the faba beans for feed, for use in production of ethanol from the starch.

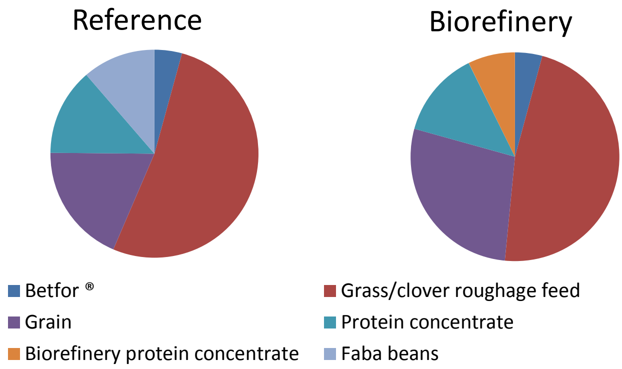

Figure 9. Proportion of the annual use of different feed products in the Reference and Biorefinery.

Studying the impacts of changing the use of biomass is relevant for determining the best use of available resources. It highlights that resources are limited and that current resources have uses that will have to be satisfied in other ways if these resources are to be used for new purposes. However, it implicitly assumes a steady state system, *i.e.* that demand for feed will be the same in future as it is today. In fact, demand for biomass and land will change in the future and developments in agricultural systems and food demand can fundamentally alter the requirements for land. There is general global concern about future increased demand for food and thereby increased demand for land. However, alternative developments are plausible. For example efficiency improvements in the food chain, particularly for animal food production, and dietary changes could potentially decrease the amount of arable land needed for food production in the future (Wirsenius *et al.*, 2010).

5.1.3 Impact from residue harvesting and whole crop harvesting

Biorefinery feedstock may originate from dedicated systems solely producing biorefinery feedstock, or comprise residues from forestry or agriculture. The return of crop residues to the soil is important for maintaining soil quality and reducing soil erosion, and therefore wide-scale harvesting of crop residues has been questioned (Lal, 2008). However, harvesting of the whole crop (*i.e.* including aboveground plant parts) can decrease the competition for land, since the function of food or feed production can be maintained, while the remaining biomass (aboveground crop residues) can be utilised for other purposes. Consequently, intensified biomass harvesting from existing agricultural and forest production systems could decrease land transformation resulting from increased demand for biomass. Land transformation where *e.g.* forest land is converted to agricultural land could lead to substantial SOC losses and other environmental consequences such as loss of biodiversity (Vitousek *et al.*, 1997).

Several consequences of residue harvesting have been included in previous LCA studies on removal of agricultural residues for bioenergy production, including impacts in SOC changes, nutrient requirements, nitrous oxide emissions (see for example Cherubini & Ulgiati, 2010), nitrate leaching and ammonia emissions (Gabrielle & Gagnaire, 2008).

In general, SOC changes due to residue harvesting can have a very significant impact on the GHG balance of lignocellulosic biofuels (*e.g.* Liska *et al.*, 2014; Whittaker *et al.*, 2014) and biorefinery systems (Cherubini & Ulgiati, 2010).

Impacts on SOC and compensation for nitrogen removal (including nitrous oxide emissions and compensation with artificial fertiliser) were included in Papers I and II.

Impact on soil organic carbon

Residue harvesting can influence the SOC content. The amount of SOC depends on carbon input and decomposition rate and this balance will be altered when a larger proportion of the crop is harvested. The loss of SOC can affect the long-term productivity and increase GHG emissions from the system (Cowie *et al.*, 2006). There are three mechanisms through 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.

In the RED methodology (Paper I), it is assumed that residues from agriculture and forestry are 'free' up to the point of harvest (EC, 2010). This means that no impact from the agricultural or forestry system is allocated to the residues, as further discussed in section 5.2.2. The other calculation method in Paper I (Method I (ISO)) included SOC changes. The change was assumed to be 75 g C per kg for straw removal (calculated from Börjesson *et al.* (2010), Nilsson & Bernesson (2009) and yield statistics) and 90 g C per kg for forest residue removal (Lindholm *et al.*, 2011). For a straw removal rate of approx. 60%, this figure corresponds to a loss of 150 kg SOC per hectare and year over 30-50 years. In a recent review of the impact on SOC due to straw removal or incorporation, it was found that estimated SOC changes differ greatly (110-640 kg C per ha and year) (Whittaker *et al.*, 2014). The actual effect will depend on soil type, climate, residue removal rate *etc*. (Cowie *et al.*, 2006). In Paper I, the contribution of SOC changes to total GHG emissions was large, representing 59% and 66% of the total GHG emissions for ethanol produced from straw and forest residues, respectively (Method I (ISO)).

In both Papers I and II, changes in SOC were assessed based on the average change over a specific time period. In Paper II the Introductory Carbon Balance Model (ICBM) (Andrén & Kätterer, 1997) was used to estimate SOC changes (Figure 10).

Paper II compared faba bean cultivation with only harvesting the beans and return of crop residues (scenario 1) with whole crop harvesting and use in a biorefinery (scenario 2). Because the whole crop was harvested, scenario 2 gave lower soil carbon sequestration than the reference (scenario 1). The SOC changes over 104 years (13 crop rotation) when faba bean residues are returned to the soil (scenario 1) are shown in Figure 10. The largest carbon inputs in the crop rotation were during year 2 (faba bean) and year 3 (winter wheat). SOC accumulated faster during the first crop rotation than during later crop rotations.

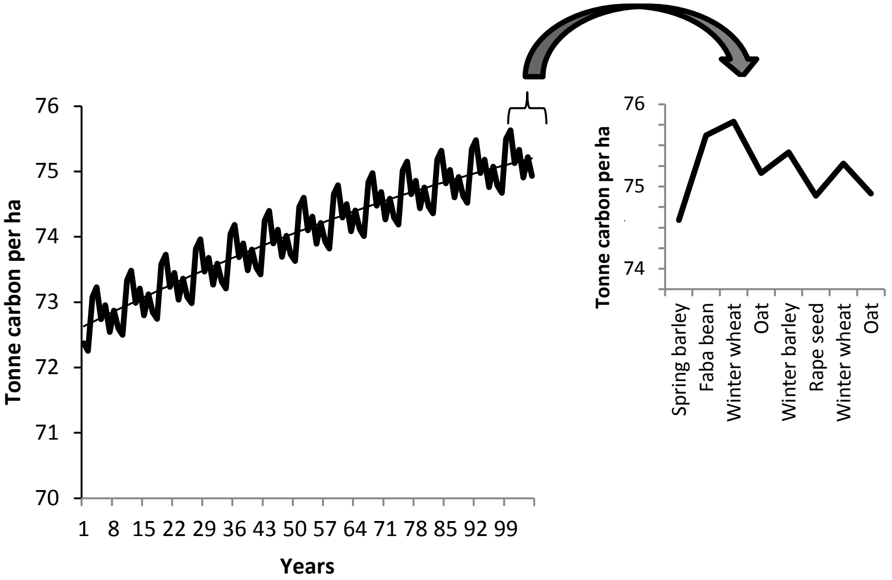

Figure10. Soil organic carbon changes in a crop rotation where faba bean crop residues are returned to the soil, representing the Reference scenario in Paper II. The small figure to the right shows the yearly SOC changes and the crops for each year of the 8 year crop rotation.

When using average values for SOC changes, the time period considered will have an impact on the estimates produced, as longer time perspectives can give a lower average annual change and shorter perspectives a higher annual average change. Carbon stock changes in forestry systems depend on the decomposition rate of the residues. For example, stumps decompose more slowly than forest residues (tops and branches), which gives a larger difference over a longer time between use of the forest biomass as biofuel and leaving the forest biomass on-site to decompose (Lindholm *et al.*, 2011; Repo *et al.*, 2011). Harvesting of stumps is being debated in Sweden but is currently not practised to any great extent. Therefore forest residues (tops and branches) were assumed to be used in the forest residue scenario in Paper I and as marginal solid biofuel in Paper II.

As SOC changes and carbon sequestration in biomass are dynamic processes that occur over long periods, dynamic methods as opposed to the use of average values have been developed to deal with the aspect of timing of CO_2 emissions in biofuel production systems (see *e.g*. Ericsson *et al.*, 2013).

Although there are large uncertainties regarding the magnitude of SOC changes due to residue harvesting, the potential impact on the overall GHG performance is large. Therefore, impacts on SOC cannot be ignored in LCA studies. Although appropriate methods for measuring SOC changes are available, there is an inherent challenge in these measurements due to the spatial variations and long periods involved. One option for estimating SOC changes in bioenergy studies is to use SOC models, or a combination of models and measurements (Cowie *et al.*, 2006). To analyse the importance of input parameters in the modelling, sensitivity analysis can be performed.

Impact from nitrogen removal

The impact of nutrient removal with crop residues was only assessed in this thesis for nitrogen removal (Papers I and II). In fact, numerous nutrients are removed with crop residues, including potassium (K) and phosphorus (P), which are significant plant nutrients. In both Paper I and II, part of the biomass was assumed to be incinerated and in this case any P and K present will mainly be recovered in the ash (as opposed to N), and can thus be returned to the production site (Obernberger *et al.*, 1997). An alternative to incineration of the biomass is to return the digestate or residue to the production site after the biorefinery process. This would return nutrients and also some of the carbon in the biomass, potentially decreasing the effect of SOC losses and nutrient replacements. In order to enable the return of bio-fertilisers from biorefineries, the biorefinery process would have to be designed so that return of the residues is possible (*e.g.* by not adding unsuitable substances).

In Paper I it was assumed that all nitrogen removed from the soil with the residues was replaced with mineral nitrogen corresponding to 8 g N per kg DM straw (mixture of cereal and oilseed straw) and 5 g N per kg DM forest residues. In a recent literature review, nitrogen compensation due to straw recovery was found to range between 3.0 and 19.4 g N per kg DM straw (Whittaker *et al.*, 2014). As discussed in Paper I, 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 yields, as done by Gabrielle and Gagnaire (2008). Those authors found that straw fertiliser value ranged from 1.5-4.5 g per kg DM cereal straw (the N content in straw was 6 g N per kg DM). 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 Paper I, 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 II, a decrease in the nitrogen delivery effect to the following crop 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 N fertiliser demand. When applying this method, harvesting of crop residues decreased nitrous oxide emissions compared with return of crop residues.

Summary of section 5.1.3

In summary, whole crop harvesting had a significant impact on the GHG performance (Paper I and II) and energy balance (Paper II) of the lignocellulosic and green biorefineries studied. However, it did not result in higher overall GHG emissions from the systems analysed compared with the reference systems. It is important to note that a significant share of the potential impact resulted from changes in SOC and that the effect on SOC is difficult to estimate. Hence, the estimates presented involve large uncertainties.

5.2 Implications of methodological choices in LCA studies of biorefineries

5.2.1 Effect of multi-functionality handling in LCA

How to handle multi-functionality problems is one of the most widely discussed methodological aspects of LCA (Finnveden *et al.*, 2009). The fact that biorefineries do not produce a main product, but rather a set of valuable co-products with different functions and physical attributes, complicates the issue of how to handle multi-functionality in this particular case.

Paper I compared two different calculation methods using different methods to handle multi-functionality. Method I (ISO) used substitution and Method II (RED) used allocation based on the LHV of the products. Choice of method for handling multi-functionality clearly highly influenced the results. The total climate impact per MJ ethanol in g CO_2 eq., including co-product substitution effects, estimated using Method I (ISO) shown in Figure 11.

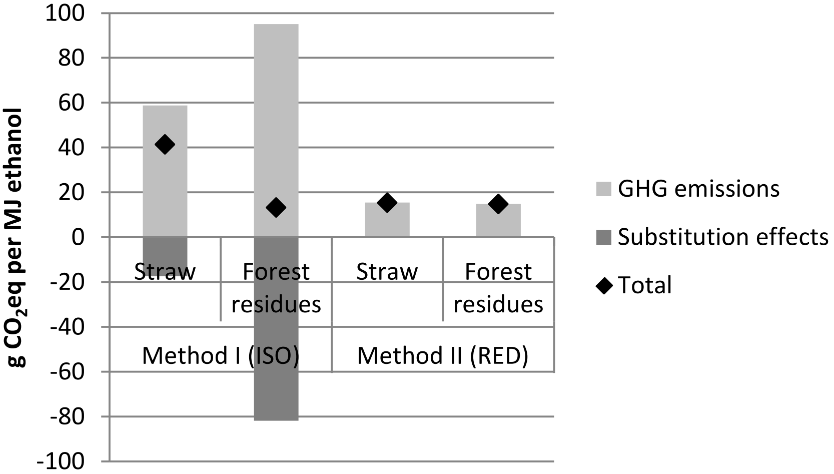

Figure11. Total climate impact per MJ ethanol for ethanol produced from straw and forest residues using two different calculation methods.

One of the main challenges when performing system expansion is that there is uncertainty associated with selection of product system to include, *i.e.* the product system/s affected by changes in the system studied, as this depends on complex market interactions. In addition, this decision can have a significant impact on the results (Gnansounou *et al.*, 2009). However, one advantage with the substitution method is that it can deal with products with different functions and characteristics, for which it may be difficult to find a common characteristic as a basis for allocation. This could be particularly important for biorefinery systems that produce a high variety of products.

The sensitivity analysis in the studies presented in this thesis confirmed that the results were influenced by selection of substituted products (Papers I and II) but also by the choice of main product (Paper I). Similar results for biorefineries have been reported by Cherubini *et al.* (2011). The production mix of the biorefinery also proved to be very important, *e.g.* relatively large production of co-products resulted in significant GHG credits. This was particularly evident for the forest residue scenario in Paper I, where the relatively high production of biogas in relation to the amounts of ethanol produced resulted in large GHG credits and fossil energy replacement. For the energy balance it even resulted in a negative value, *i.e.* more fossil energy was replaced by the co-products than was used in the production of the biofuel (Paper I).

In Paper II, system expansion was used to compensate for differences in production between the reference scenario and the other two scenarios. For the scenarios to be comparable, differences in the product output from the reference scenario were assumed either to replace equivalent products (substitution) or increase the use of certain products (mainly feed products, as faba beans are used as a cattle feedstuff in the reference scenario).

In general, when system expansion is used new multi-functionality problems can be introduced, since the avoided products are often part of another multi-functional system. For example, the protein concentrate that was produced in the biorefinery (Paper II) was assumed to affect the demand for feed grain, grass/clover roughage and protein concentrate consisting of soybean meal. Soybean meal is co-produced with soybean oil and a change in demand for soybean meal was assumed to result in the following: decreased demand for soybean meal resulted in a decrease in production of soybean oil, which in turn was assumed to increase demand for palm oil, increasing the production of palm kernel meal, which in turn slightly decreased the demand for soybean meal and spring barley (Dalgaard *et al.*, 2008). In the paper by Dalgaard *et al.* (2008), this is called the soybean/palm loop. It clearly illustrates the statement by Weidema (2000) that each time the system expansion process is repeated, the economic value and the volume of the displaced product tend to decrease, meaning that at a certain cut-off point it will have little influence on the results.

When the process affected is a joint production system (see section 3.2.3), it is helpful to distinguish between dependent and determining co-products, as these are handled differently when impacts due to changes in demand are assessed. One example is forest residues, which could be categorised as a dependent co-product from forestry (producing the determining co-product of logs), as a change in demand for forest residues is not likely to affect the production of the main product. In Paper II, briquettes from the biorefinery were assumed to replace wood chips from forest residues. It was assumed that wood chips from forest residues are a dependent co-product that is not fully utilised. The following processes should then be included in the assessment (based on the description in Weidema *et al.* (2009)): (i) The production of the dependent co-product (*i.e.* the chopping of the forest residues to wood chips); (ii) 'waste treatment' of the co-product, which was assumed to be the alternative treatment when the forest residues are not harvested, i.e. SOC changes due to residue harvesting were included here; and (iii) the intermediate treatment of the co-product, *i.e.* harvesting and transportation. This corresponds to how straw and forest residues are handled in Paper I (Method II (ISO)) and the forest residues in the avoided process in the biorefinery scenario are handled in Paper II.

One strategy to avoid allocation between the products when studying biorefinery systems is to select a functional unit so that the multi-functionality problem is avoided. A functional unit that avoids allocation between co-products could be *e.g.* 1 tonne of biomass input, 1 biorefinery or a combination of all outputs. The whole biorefinery system and its products are usually compared with a product mix with conventionally produced equivalent products (*i.e.* Earles *et al.*, 2011; Cherubini & Jungmeier, 2010; Cherubini & Ulgiati, 2010; Uihlein & Schebek, 2009). In Ahlgren *et al.* (2013), these types of functional units are called *input FU* (i.e. 1 tonne of biomass, 1 hectare of land) and a *multi-functional FU* (*i.e.* production of 1 kg product of A, 2 MJ of product B, 100 kg of product C). This strategy avoids allocation to some extent and allows comparison with other production systems or a combination of other production systems. For example, the potential benefits of a biorefinery system (producing products A, B and C) compared with conventional systems producing the equivalent products can be assessed. However, a disadvantage could be that a multi-functional FU can in some cases be difficult to communicate and compare with those of other studies, *e.g.* results stating that the potential global warming impact is 100 kg CO_2-eq. for 10 kg of product A, 2 kg of product B and 0.1 kg of product C are not always very useful (Ahlgren *et al.*, 2013). An input FU is suitable for determining the best use for a specific feedstock or land area (Cherubini & Strømman, 2011) or when analysing the use of waste products as feedstock in biorefineries.

Whether these types of functional units (input-based FU and multi-functional FU) are appropriate to use or not depends on the objective of the study. The functional unit should preferably not be selected solely because it avoids allocation, but rather for its applicability as regards the objective of the study.

In the RED method used in Paper I, allocation was based on LHV of the products, which means that energy became the determining characteristic of all products. For biorefineries this might be problematic when not all co-products are produced for energy purposes (Cherubini *et al.*, 2011; Gnansounou *et al.*, 2009). For example, excess heat in the form of warm water has no heating value in the method and is therefore not attributed any environmental burden, even though it may have an energy use in *e.g.* district heating. Furthermore, all energy carriers are considered to be equivalent, irrespective of their application in society and past production history. Another basis for allocation instead of energy content could be used, such as mass or economic outcome. Cherubini *et al.* (2011) suggest that a suitable basis for allocation when dealing with a diverse set of co-products (energy and materials) is an economic or exergy allocation or a hybrid approach of system expansion and allocation.

5.2.2 Assessing biorefinery systems applying the RED

Since the RED has specific rules for calculation of GHG performance for liquid biofuels, it is potentially highly influential for European biofuel production, and thereby for biorefinery processing. As mentioned above, the use of allocation based on LHV in the RED is problematic for the assessment of biorefinery systems that produce diverse products which are not used solely for energy purposes (Cherubini *et al.*, 2011; Gnansounou *et al.*, 2009). In the RED, carbon stock changes due to land use change are only included if the land use change occurs between defined land use categories (Paper I) and not due to management changes to agricultural land (Ahlgren, 2012). Hence, impacts on SOC due to residue harvesting are not included. As shown in Paper I, the impacts from residue recovery can be significant for the overall GHG performance of lignocellulosic ethanol. Including impacts from SOC changes in the RED calculations more than doubled the total GHG emissions in that example.

The results from the sensitivity analysis (Paper I) and the required reduction from a fossil fuel reference stated in the RED are shown in Figure 12.

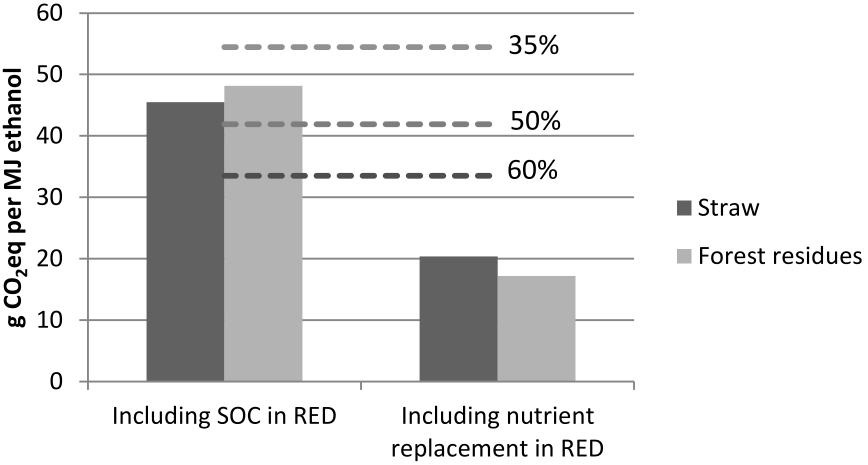

Figure 12. Results from sensitivity analysis Paper I where impacts from SOC changes and nutrient replacement due to residue harvesting were included in the RED calculations. The dotted lines represent the reduction requirements from a fossil fuel reference included in the RED.

When only SOC was included in the calculations, the ethanol produced from straw and forest residues only met the 35% reduction target, and not the forthcoming targets of 50 and 60 % reduction from a fossil fuel reference. Whittaker *et al.* (2014) estimated the GHG performance of straw-based ethanol, including SOC changes and nutrient compensation, and found that the GHG saving relative to fossil fuels was 21-58%. Using Monte Carlo simulation, those authors also showed that straw-based ethanol has a 30% chance of meeting the 35% reduction target.

The handling of agricultural and forestry residues in the RED is based on the assumption that these residues are wastes which have no alternative use (Whittaker *et al.*, 2011). By assigning no upstream impacts from residue harvesting, the RED is encouraging residue use for biofuels. Furthermore, to stimulate the production of biofuels from lignocellulosic materials, such biofuels count double towards the 10% target (Ahlgren, 2012). Increased demand for biomass in a future bio-economy could lead to increased use of residues. Therefore, the main EU policy regulation in the area of biofuels (the RED) should recognise the value of residues in agricultural and forestry and consider including the upstream impacts of residue harvesting.

6 Conclusions

6.1 GHG performance and energy balance

For ethanol co-produced with biogas in a lignocellulosic biorefinery, the following conclusions were drawn:

- Ethanol produced from forest residues generally showed lower GHG emissions than ethanol produced from straw, although the difference was small when Method II (RED) was used
- GHG savings for lignocellulosic ethanol were between 51 and 84% compared with a fossil fuel reference
- Primary fossil energy use was between -0.71 and 0.20 MJ per MJ ethanol
- Enzyme production and SOC changes were most influential for the results.

For changing the use of available faba bean cultivation to whole crop processing in a green biorefinery, the following conclusions were drawn:

- Biorefinery processing of whole faba bean decreased the climate impact (-2%), primary fossil energy use (-119%) and land use (-20%) compared with the reference use of faba beans as a dairy cow feedstuff. The overall decrease in GHG emissions and fossil energy use for the biorefinery was solely due to the substitution effects of the biorefinery products
- The alternative feed use of the whole faba bean plant as roughage (scenario III) significantly increased the climate impact (+290%) and energy use (+150%), but decreased the use of arable land (-130%) due to substitution of roughage
- In a GHG perspective, ethanol production from faba bean starch was not beneficial. Consequently, maintaining the current use of faba beans

> for feed while exploring options for biorefinery processing of the green biomass could be more beneficial for the GHG performance of the system as a whole. Alternatively, a better use (than ethanol production) of the starch could be an option, if the biorefinery wanted to reduce the climate impact further.

In general, the introduction of biorefineries can result in changes in the use of biomass and increased recovery of biomass from forestry and agriculture. This thesis showed that increased biomass harvesting from agriculture and forestry had potentially high impacts on the overall GHG performance of biorefinery products and systems.

6.2 LCA methodology

The results for GHG performance and energy balance varied significantly depending on the calculation method used. The most influential factors were choice of method to handle multi-functionality and system boundaries, *i.e.* the inclusion of upstream impacts in the form of SOC losses.

Since an increasing share of biofuel can be expected to be produced in biorefineries and from lignocellulosic biomass, policy instruments such as the RED will need to become compatible with these systems. This will involve *e.g.* careful consideration of allocation methods and the handling of agricultural and forestry residues in the case of the RED. Consequently, including upstream impacts from residue harvesting in the RED is recommended. The inclusion of SOC changes due to residue recovery should also be carefully considered.

Methodological choices when handling multi-functionality were influential for the results, in particular assumptions regarding technologies affected in system expansion. Biorefineries are by definition multifunctional systems and therefore methods to handle multi-functionality may be particularly important when analysing these systems using LCA, especially for biorefinery systems producing a diversity of products with different functions.

7 Further research

Innovative biorefinery systems to replace petroleum refineries are currently being developed. Evaluation of these systems regarding biomass supply and process requirement for energy and process inputs needs to be conducted from an environmental perspective. This includes evaluation of impacts on the environment resulting from new harvesting management systems that are associated with the introduction of biorefineries, *e.g.* whole crop harvesting or increased recovery of forest residues. Methods for decreasing the uncertainty of SOC impacts and for handling long-term changes in biogenic carbon stocks need to be further developed to include dynamic changes and timing of emissions. In addition, increased biomass withdrawal can have impacts on the long-term sustainability and productivity of agricultural and forestry systems (biological production systems) that should be reflected in the assessment. Relevant environmental impact categories apart from climate impact, energy balances and land use should also be included in the assessments.

There are many alternative process routes for the intermediate products produced in the biorefinery systems evaluated in the present thesis. For example, lignin was merely assumed to be combusted in the systems evaluated, but studies have shown that lignin can be utilised to produce *e.g.* liquid transportation fuels. Furthermore, this thesis focused on the production of transportation fuels, which is interesting from a climate perspective as the transportation sector is currently fossil fuel-based. However there are other products that have the potential to replace fossil resources, *e.g.* bio-based products such as bio-plastics can replace plastics made from fossil resources. Likewise, bio-based chemicals could be associated with large fossil resource replacement potential.

To evaluate the optimal use of available resources, alternative process designs should be included and studied from an environmental perspective.

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