Greenhouse Gas (GHG) and energy analysis of a bioethanol oriented biorefinery based on wood

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Summary

A great fraction of worldwide energy carriers and material products come from fossil fuel refinery. This strong dependence on fossil fuels results on the intensive use and consumption of petroleum derivatives which, combined with diminishing petroleum resources, causes environmental and political concerns. There is clear scientific evidence that emissions of greenhouse gases (GHG), such as carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O), arising from fossil fuel combustion and land-use change as a result of human activities, are perturbing the Earth's climate.

In order to simultaneously reduce the dependence on oil and mitigate climate change in transport and chemical sectors, alternative production chains are necessary. The replacement of oil with biomass as raw material for fuel and chemical production leads to the development of biorefinery complexes. In biorefinery, almost all the types of biomass feedstock can be converted to different classes of biofuels and biochemicals through jointly applied conversion technologies. Among the possible biomass raw materials, lignocellulosic feedstocks are particularly important, as they are widespread and locally available for many countries and they have great potentials for producing large quantities of biofuels and biochemicals (thanks to their chemical composition).

This case study deals with a the estimation of the greenhouse gas emissions of a biorefinery system based on forest softwood residues which produces bioethanol, other bioenergy carriers (electricity, heat, biomethane) and chemicals (phenols) using Life Cycle Assessment (LCA) methodology. This biorefinery system is compared with a reference system producing the same products from fossil sources. The assessment also includes an estimation of GHG emissions caused by a change in forest management (i.e. residues which are not anymore left in the forest but are collected and used as raw materials in biorefinery). Since climate change mitigation and energy independence are the main driving forces for future biorefineries, results focus on GHG emissions and cumulative primary

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energy demand. Different allocation methods are used and compared to share the total GHG emissions of the biorefinery system among the co-products.

Results reveal that by 2050 the biorefinery system saves approximately 40% of cumulative GHG emissions when compared with a fossil reference system, while non-renewable energy savings are about 84%. Forest management change is the main responsible for total GHG emissions of the biorefinery system (79%), followed by production of raw materials (15%).

1. Introduction and aim

A great fraction of worldwide energy carriers and material products (especially high value chemicals) come from fossil fuels, both from oil and natural gas refinery. However, there is concern about the on-going price increase of fossil resources (i.e. energy security) and their environmental impacts (i.e. climate change). Alternative solutions able to reduce the consumption of fossil fuels should be promoted. While electricity and heat can be provided by a variety of renewable alternatives (wind, sun, water, biomass and so on), the alternative for production of transportation fuels and chemicals is currently biomass, the only C-rich material source, , besides fossils fuels, available on the Earth.

Therefore the term of “biorefinery” is gaining importance in the scientific and industrial community: a biorefinery is a facility that integrates biomass conversion processes and equipment to produce transportation biofuels, power and chemicals from biomass. The biorefinery concept is analogous to today’s petroleum refinery, which produce multiple fuels and products from raw oil. Thus, the gradual deployment of biorefinery infrastructures is encouraged by the need for a secure energy supply, a reduction of fossil CO_2 emissions and a revitalization of rural areas.

Possible future feedstocks for biorefineries are lignocellulosic materials, organic residues, dedicated energy crops, vegetable oils and other biological matter. Among the other alternatives, lignocellulosic feedstock biorefinery is of specific interest: the raw materials are widespread (wood, straw, paper waste…), cheap and their possible conversion products (e.g. bioethanol) have a good position in the biobased product market. Furthermore, their main advantage is that, compared to conventional starch and oilseed crops, woody crops can supply much more biomass per hectare of land since the entire biomass growth can be used as feedstock. In fact, over the next 10-15 years, it can be expected that lower cost residue and waste sources of cellulosic biomass will provide the first input to biorefinery industries (Perlack et al 2005, Cherubini 2010)

Lignocellulosic biomass is mainly composed of cellulose, hemi-cellulose and lignin. Both the cellulose and hemicellulose fractions are polysaccharides (long chains of C6 and C5 sugars) that can be fermented to ethanol after suitable pre-treatment and hydrolysis. The remaining, non-fermentable lignin fraction (a polyphenol) can undergo thermochemical processes such as gasification or pyrolysis (to produce fuels or chemicals), or combusted to be converted to electricity and heat (used for the ethanol production process and for export to the public grid).

With all this, the aim of this study is to compile a Life Cycle Assessment (based on GHG and energy balances, since climate change mitigation and energy independence are the two driving forces for developing biorefinery systems) of a lignocellulosic biorefinery which produces bioethanol, phenols, electricity and heat from wood forest residues. The main goal is the quantification of the possible GHG and energy savings which are achievable by a biofuel oriented biorefinery system when compared with a fossil reference system producing the same products from fossil sources. Results include forest management change effects caused by collection of residues in the forest (while in the reference system they are left on the ground). In addition, several allocation approaches are used to share the total GHG emissions of the biorefinery among the different co-products, and final results are compared.

2. Goal and Scope

This work deals with the attributional LCA of a biorefinery system which produces:

- Bioethanol as biofuel for the transportation service;
- Electricity from Combined Heat and Power (CHP, from combustion of lignin and process residues);
- Heat from CHP;
- Heat from biomethane (produced from anaerobic digestion of wastewaters); and
- Phenols, extracted from lignin.

The market-mediated impacts are not considered in this study. Nevertheless, indirect effects from the use of the biomass will be minimal since in absence of the bioenergy system it remains on the forest floor to decay.

According to the classification method for biorefinery systems, this concept can be labelled:

C5/C6 sugars, biogas, lignin/pyrolytic oil biorefinery for bioethanol, electricity and heat and chemicals from lignocellulosic residues

This system is a combination of several conversion technologies which are jointly applied in order to produce biofuels and material products from lignocellulosic biomass, within a biorefinery approach. The biorefinery system is compared with a fossil reference system which produces the same amount of products / services from fossils fuels:

- Gasoline for the transportation service;
- Electricity from natural gas (average among Austrian power plants);
- Heat from heavy oil (as alternative to heat from CHP);
- Heat from natural gas (as alternative to heat from biomethane); and
- Conventional phenols from oil refinery.

The reference use for forest residues is assumed to be “residues left in the forest”, where a natural decomposition occurs.

The biorefinery has been designed with a fixed annual consumption of biomass (530 Kt_{dry}/a). The functional unit of the assessment is the $biomasst_{dry}$ of pellets of forest softwood residues. All emissions, energy use will be expressed in units/t_{dry}.

It is assumed that the biorefinery will start operation in 2012. Since the emissions from the bioenergy system vary with time, results are presented for 2020, 2032, 2050 and 2100. The first year, 2020, was selected because it coincides with the EU bioenergy policy targets. 2030 was selected because the biorefinery would be in operation for 20 years. The other two years, 2050 and 2100, were chosen to coincide with long-term global GHG targets.

This study is modelled by means of the LCA software tool SimaPro 7 (http://www.pre.nl/simapro/default.htm) and selected literature references are used to estimate input flows and specific emissions. Effects due to forest management change are estimated by means of a dedicated software tool (CO_2FIX, http://www.efi.int/projects/casfor/). Since climate change mitigation and energy independence are the main driving forces for future biorefineries, results focus on GHG emissions (CO_2, CH_4 and N_2O) and cumulative primary energy demand, divided into non-renewable (fossil and nuclear), renewable (biomass) and other renewable (mainly hydropower). Total GHG emissions of the biorefinery system are then allocated to the products using different allocation criteria and the results are finally compared.

2.1 System boundaries and fossil reference system

In Figure 1, the simplified system boundaries for the biorefinery and fossil reference system are shown. The biorefinery chain starts at the top of the diagram with carbon fixation from the atmosphere via photosynthesis, which is stored in vegetation. At the end, the biorefinery system supplies products and services. All input and output flows occurring along the full chain, for harvesting the residues, processing the feedstock into biofuel, transporting and storing of feedstocks, distributing and final use of biofuels are accounted for using a life cycle perspective. By contrast, the fossil reference system starts with consumption of non renewable sources (i.e. fossil oil and natural gas), and its main life-cycle stages are the following: extraction and conveyance of raw materials, production of the raw fossil fuel, refining, storage, distribution and combustion. The reference system also includes a reference use for the biomass, i.e. left in the forest where a natural decomposition occurs. This forest management change may cause a reduction in the carbon stocks of the forest and a consequent emission of CO_2. Such a decrease of carbon though not “technically” land use change (LUC) is equivalent to LUC. It is estimated in the section 2.3.

The fossil alternatives to the biorefinery products are listed in Table 1. In the right part of the table, their specific GHG and energy factors are reported. These values, calculated by means of the LCA software tool SimaPro 7.1 (the source is shown in the database column), are used for quantifying the GHG emissions and cumulative energy demand of the fossil reference system.

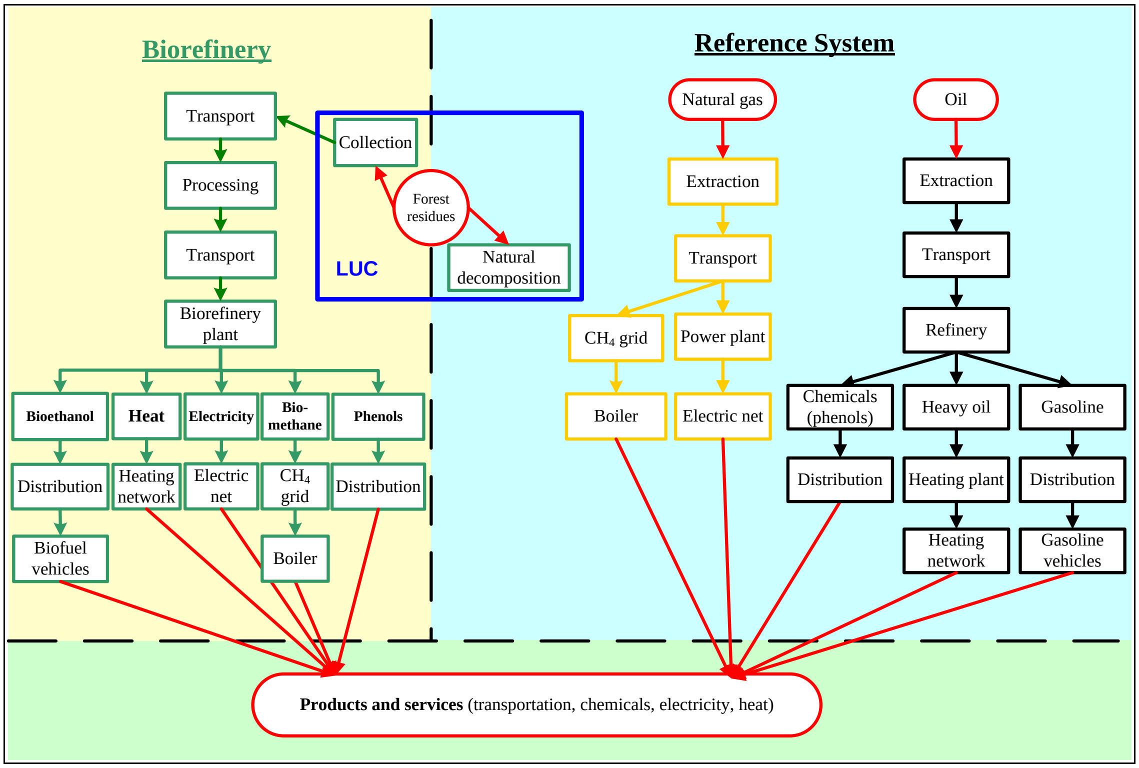

Figure 1 System boundaries of the biorefinery and fossil reference system.

Table 1 Table showing the factors for GHG emissions and cumulative energy demand for the conventional (fossil) alternative of the biorefinery products used in the fossil reference system.

Biorefinery product	Fossil alternative	Unit	GHG emissions g CO_2-eq./unit	Total energy MJ/unit[a]	Database
Bioethanol	Transportation (gasoline)	km	198	3.14	Ecoinvent
Electricity from CHP	Electricity from natural gas[b]	kWh	731	11.9	ETH-ESU 96
Heat from CHP	Heat from oil[c]	kWh	425	5.88	ETH-ESU 96
Heat from biomethane	Heat from natural gas[d]	kWh	289	4.85	ETH-ESU 96
Phenols	Conventional phenols	g	3.5	0.12	Ecoinvent

[a] Mainly fossil energy (99%)

[b] Average among natural gas power plants in Austria

[c] Industrial furnace S Europe

[d] Industrial furnace > 100 kW

2.2 Production of forest softwood pellets

Lignocellulosic biomass (as softwood) is mainly made of three components: cellulose (C6 polymers), hemicellulose (C5 and C6 polymers) and lignin (phenol polymer). Chemical composition of softwood is reported in Table 2.

Table 2 Composition of the lignocellulosic feedstocks used as raw materials (Hamelinck et al., 2005).

Parameter	**Unit (dry)**	**Softwood**
Water	%	10.0
LHV	MJ/kg	19.6
Cellulose	%	44.5
Glucan (C6)	%	44.5
Hemicellulose	%	21.9
Xylan (C5)	%	6.30
Arabinan (C5)	%	1.60
Galactan (C6)	%	2.56
Mannan (C6)	%	11.4
Lignin	%	27.7
Acids	%	2.67
Extractives	%	2.88
Ash	%	0.32
C	%	50.3
H	%	5.98
O	%	42.1
N	%	0.03
S	%	0.01

Forest residues include biomass not harvested or removed from logging sites in commercial forests, as well as material resulting from forest management operations such as pre-commercial thinnings and removal of dead and dying trees.. Harvesting plans on private and public forests require some form of residue management, which results into collection and use for biorefinery purposes. The drawback of this biomass source is that it is generally more cost expensive to be supplied than other biomass sources (Morris, 1999).

The yield of forest softwood residues can significantly vary, according to geographical location and wood species. Collecting residues from forests requires 2.57 L diesel/m^3 of residues (Johnson et al., 2002) and 35.3 g of lubricants/m^3 of residues (NREL, 2009). These values account for hand-felling and cable yarding to a landing site, without transportation. This wood is then assumed to be transported for 40 km (round trip, 16 t lorry) to a facility where wood is firstly dried from 50% to 10% water content (2.26 GJ/t of water removed (Smeets et al., 2009), assumed to be supplied as heat from a natural gas boiler) and then pelletized (86.11 kWh$_e$/ton (Gemis 2009), from Austrian electric grid).

Pellets are then transported for 100 km to the biorefinery plant with 40 ton-capacity trucks. This assumption represents the upper limit of transport distance under an economic feasibility perspective (Narodoslawsky et al. 2008). GHG emissions and primary energy demands for producing and transporting pellets of forest softwood residues to the biorefinery gates are reported in Table 3. These figures do not include emissions caused by forest management change, which are estimated in the following section

Table 3 GHG emissions (without emissions from forest management change) and cumulated primary energy demand for supplying 530 kt of pellets of forest softwood residues to the biorefinery plant (including transport).

	Unit	Amount
GHG emissions		
Total	kg CO_2-eq./t_{dry}	100
CO_2	kg CO_2-eq./t_{dry}	94.5
N_2O	kg CO_2-eq./t_{dry}	1.35
CH_4	kg CO_2-eq./t_{dry}	3.64
Cumulated primary energy demand		
Total	MJ/t_{dry}	23.6
Non renewable[a]	MJ/t_{dry}	1.53
Renewable (biomass)[b]	MJ/t_{dry}	21.8
Other renewable[c]	MJ/t_{dry}	0.31

[a] Fossil and nuclear

[b] Including the energy content of the feedstock

[c] Mainly hydropower

2.3 GHG emissions from forest management change

GHG emissions arise from the land use sector when a change in forest management causes a decrease in carbon stocks. Forest residues, comprised of branches and foliage, are created after management activities such as thinning and final harvest. The forest for the case study is assumed to be in the *Bruck an der Mur* forest region, approximately 75 km north of Graz (Figure 2)

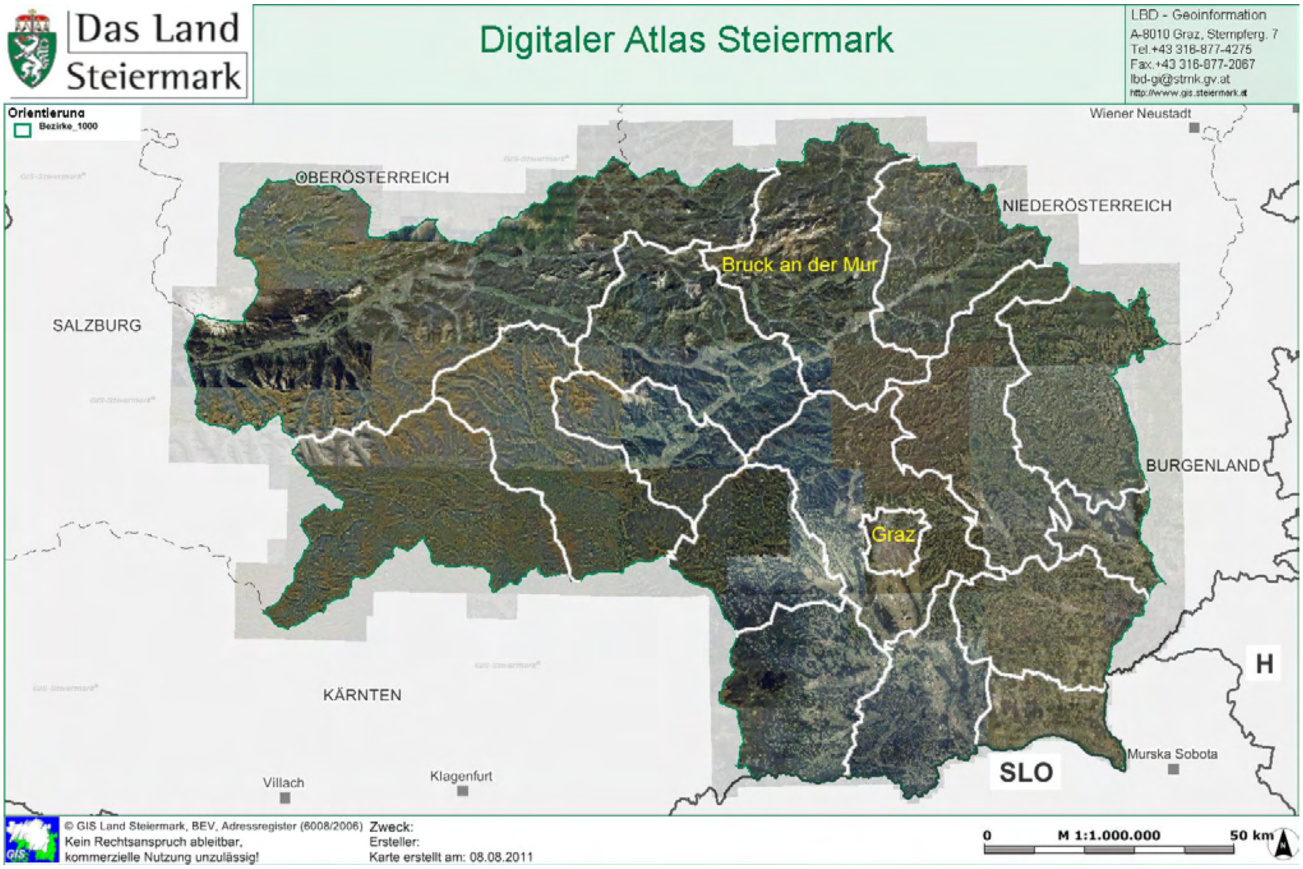

Figure 2: Satellite image of Styria showing the location of the Bruck an der Mur region

In the reference system, all forest residues are assumed to remain in the forest after thinning and final harvest operations, where they decompose. The rate of decomposition is dependent on temperature, precipitation and amount of lignin in the decomposing material. Based on the model of Moore et al. (1999), the average decomposition rates for needles and woody material are 16.6% and 7.5% per year respectively for the study area (average temperature 8.1°C, annual precipitation 795 mm). This is equivalent to an average lifetime of 5.5 years for needles and 12.8 years for woody material. The amount of branches and foliage at harvest is estimated as 14% and 11% of the total tree biomass respectively (JRC 2010). This is equivalent to branches and foliage being 18.8% and 15.3% of stem biomass respectively. The tree biomass at harvest comes from the Austrian forestry yield tables (Marschall 1992),

CO2FIX uses as complicated model, YASSO (Tuomi et al 2008), to distribute decomposing material within compartments of non-living biomass. It does not model the total soil organic carbon (ie the stable components). The compartments it uses are not the standard IPCC definitions of dead wood, litter and soil pools. In CO2FIX, only the fresh components are considered as dead wood and litter. All other material is considered decomposing and part of the soil pool.

Figure 3 shows the net carbon stock changes (reference – project) per hectare and for a forest that supplies the required 530 kt of biomass per year. The first stand is harvested in 2012 and then again 90 years later. In the reference system, the harvest residues are left on site. In the project system, they are removed for bioenergy. The stump is not removed. There is no difference in wood product generation between the reference and project systems. The stand is also thinned every 10 years after 2032, but it is assumed that the thinning residues are left on site in both systems.

Table 4 lists the area required and the amount of biomass consumed by 2020, 2050 and 2100. This table also lists the cumulative emissions and average emission intensity (t CO_2 / t dry biomass) during these years. It is very unlikely that the biorefinery will exist for 100 years. However, the cumulative emissions and emission intensity are shown for the first 100 years because the annual emissions due to the forest management change are very high during the first 20 years. They decrease as the ecological system adjusts to the new forest management system and reach dynamic equilibrium after approximately one harvest rotation (see Figure 3). The average carbon stocks by pool for the bioenergy and reference system are shown in Table 5

Table 4: Area required, biomass consumed, forest management change emissions and emission intensity – class 10[3]

		Unit	Amount
Area			
	Annual	ha/a	5,232
	Total	ha	470,850
Cumulative biomass consumed			
	2020	Mt_{dry}	4.77
	2050	Mt_{dry}	20.67
	2100	Mt_{dry}	47.17
Cumulative emissions from forest management change			
	2020	Mt CO_2-eq.	3.87
	2050	Mt CO_2-eq.	6.65
	2100	Mt CO_2-eq.	8.78
Average emission intensity from forest management change			
	2020	t CO_2-eq./t_{dry}	0.812
	2050	t CO_2-eq./t_{dry}	0.322
	2100	t CO_2-eq./t_{dry}	0.186

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[3] In Austria, forest yield tables are distinguished by species, location and class. Class refers to the mean annual increment (in m3/year) after 100 years.

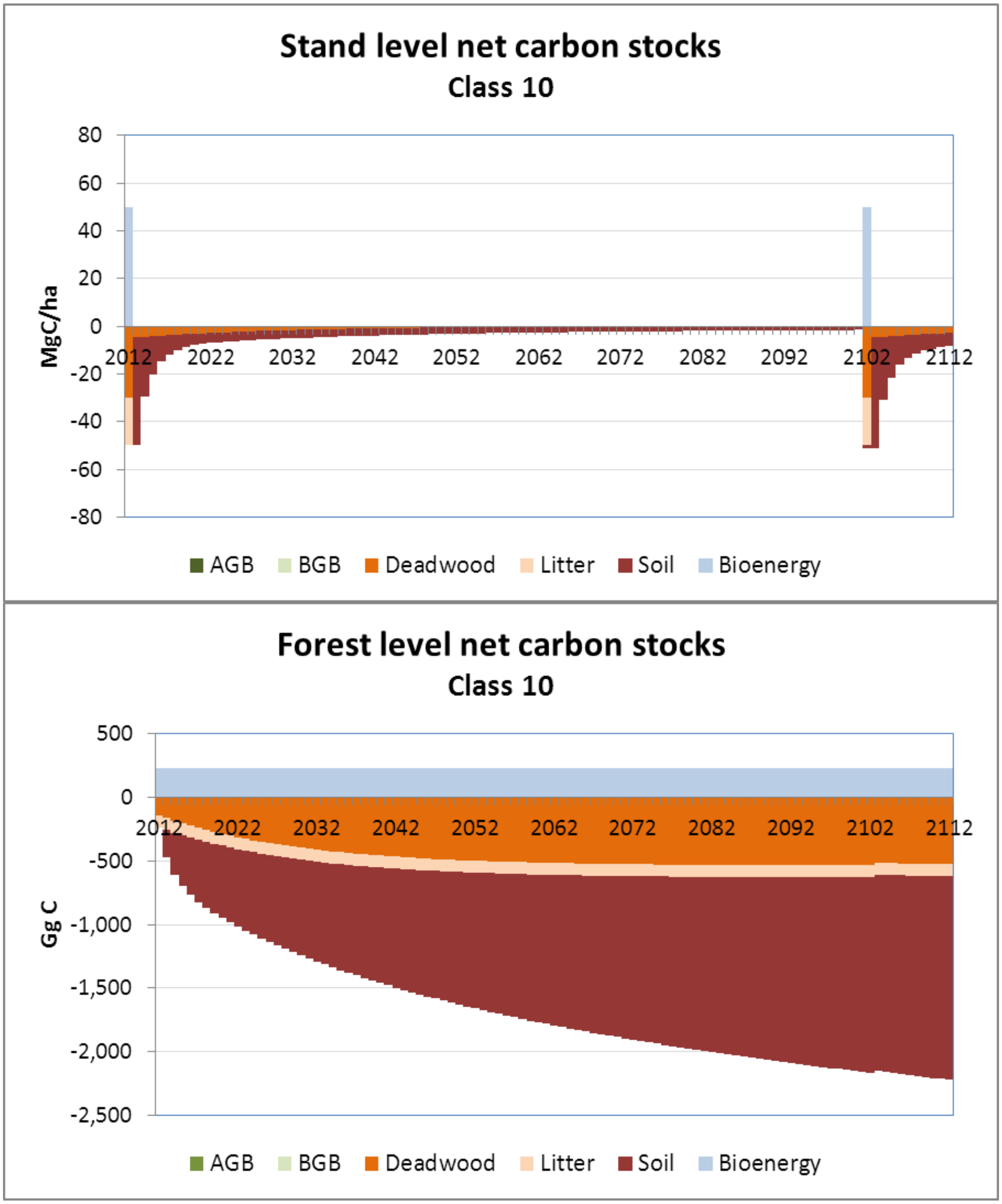

Figure 3: Net carbon stock changes (reference – project) per hectare in a single stand (top) and a forest (bottom) that provides 477,000 t biomass / year. The first stand is harvested in 2012 and then again 90 years later. In the reference system, the harvest residues are left on site. In the project system, they are removed for bioenergy. The stump is not removed. There is no difference in wood product generation between the reference and project systems. The stand is also thinned every 10 years after 2032, but the thinning residues are left on site in both systems. AGB = above ground biomass (stem, branches and foliage). BGB = below ground biomass (roots). Since only residues are being removed, there are no net carbon stock changes in AGB and BGB,

Table 5: Average carbon stocks at dynamic equilibrium – class 10

	Unit	Biorefinery system	Fossil reference system
Biomass			
Total	tC/ha	212	218
Stems, branches, foliage	tC/ha	100	100
Roots	tC/ha	29	29
Deadwood	tC/ha	24	25
Litter	tC/ha	5	6
Soil	tC/ha	54	58
Products			
Total	tC/ha	4.9	4.3
Wood products	tC/ha	4.3	4.3
Bioenergy	tC/ha	0.6	0.0

Note: Biomass in the fossil reference system is for the unused residues left on the surface

2.4 Steps in biorefinery

After collection and processing (i.e. drying and pelletizing), pellets of forest residues are transported to the biorefinery plant where are converted to bioethanol, bioenergy and biochemicals (e.g. phenols). The conversion steps to which this raw material is subjected are the following (see process scheme in Figure 4):

- Pretreatment (uncatalyzed steam explosion) of the raw material in order to depolymerize hemicellulose and separate lignin (Sun & Cheng 2002; Lynd 1996);
- Enzymatic cellulose hydrolysis to glucose monomers (Palmqvist & Hahn-Hägerdal, 2000; Hamelinck et al. 2005);
- Fermentation and distillation of sugars to bioethanol (Hamelinck et al. 2005);
- Anaerobic digestion of wastewaters (Berglund & Börjesson 2006; Romano & Zhang 2008);
- Flash pyrolysis of lignin (20%) followed by phenol separation from the resulting pyrolytic oil (Zhang et al. 2007; Meister 2002);
- Final combustion (for heat and power production) of process residues, fraction of lignin that is not pyrolyzed (80%), pyrolytic char and the remaining pyrolytic oil after phenol extraction (Senneca 2007; Gani & Naruse 2007).

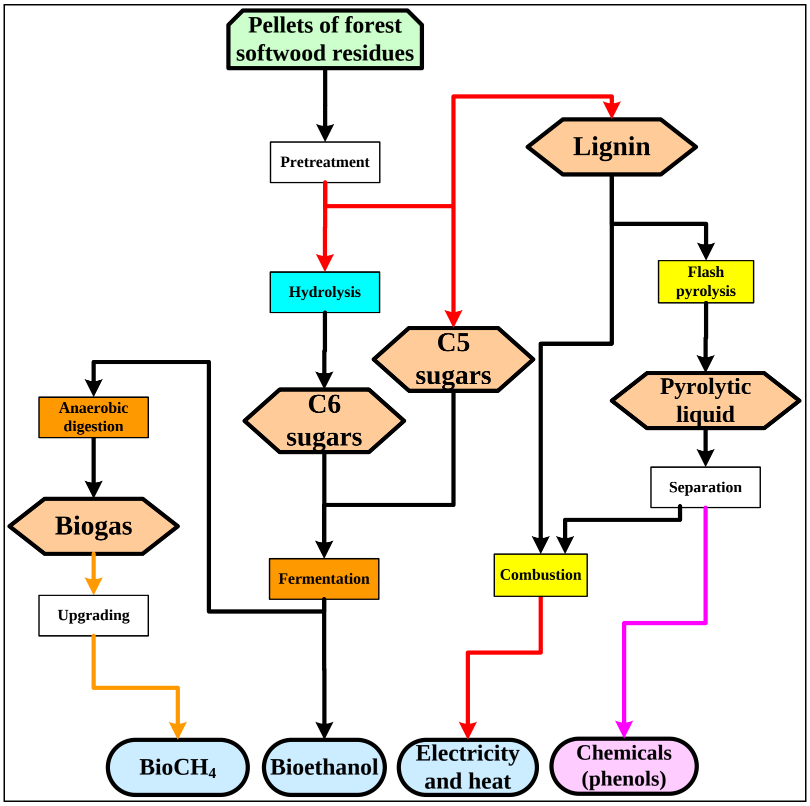

Figure 4 Main process steps of the biorefinery plant.

The feedstock undergoes an uncatalyzed steam explosion, which occurs at a temperature of 160-260 °C, with a reaction time of 2 minutes. During this stage, the C5 sugars in hemicellulose are hydrolyzed to xylose and arabinose with an efficiency of 85% (Hamelinck et al. 2005); arabinose is assumed to have the same conversion efficiency of xylose. Cellulose is hydrolyzed to glucose in the following enzymatic step with an efficiency of 90%; the remaining C6 polymers, galactan and mannan, are hydrolyzed to galactose and mannose with an efficiency of 82% and 89% respectively (Hamelinck et al. 2005). A fraction of cellulose (9%), is set aside for bacteria cultivation. All the sugar monomers are then sent to fermentation while all the residues, together with lignin, undergo thermochemical treatment (combustion or pyrolysis). Concerning sugar fermentation, which occurs in a Simultaneous Saccharification and Co-Fermentation mode (SSCF, with simultaneous fermentation of C5 and C6 sugars), ethanol conversion yields are 92.5% from C6 sugars and 85% from C5 sugars on a molecular basis (Hamelinck et al. 2005).

Bioethanol is finally distilled with an efficiency of 98%. Residues of these two steps are in water solutions and are anaerobically digested in order to produce biogas. These wastewaters have a total dry matter content of 85 $\mathrm{kt_{dry}}$ per 530 kt biomass input and generate biogas at an average rate of 6

GJ/t_{dry} (Berglund & Börjesson 2006). The produced biogas has a higher heating value of 24 MJ/m^3 and methane content of 60% (Alzate & Toro 2006). Methane emissions to the atmosphere during digestion are estimated to be 3.47 mg/MJ and the upgrading of biogas to biomethane (having a CH_4 content greater than 97%) by removing impurities and CO_2, needs 5% of the energy content of the biogas itself (Gemis 2009). The remaining digestate has fertilization properties and it is assumed to be dried up to 10% water content, transported for 150 km and then applied to the field. It is assumed that the digestate has a N content equal to that of the original biomass raw material, and it is applied on the field at a rate of 115 kg N/ha. However, due to large uncertainties, no environmental benefits are assumed from digestate use.

Concerning thermochemical treatments, 20% of lignin is subjected to flash pyrolysis, resulting in 75% pyrolytic oil (higher heating value 16 MJ/kg) and 25% charcoal (higher heating value 14 MJ/kg) (Zhang et al. 2007). The pyrolytic oil is a mix of different chemical compounds having an average phenol content of 32.3% (Zhang et al. 2007), which can be recovered at an efficiency of 50% (Scholze 2002).

The remaining pyrolytic oil after phenol separation is combusted with charcoal and the remaining lignin fraction (higher heating value 22.9 MJ/kg) and other residues (mainly unconverted sugar polymers, with a higher heating value assumed equal to 15.6 MJ/kg) to generate electricity and heat with an efficiency of 25.5% and 44%, respectively (De Feber & Gielen 2000). Ashes are disposed of in a monitored landfill.

This biorefinery system has an electricity demand of 0.83 GJ/t dry feedstock (Hamelinck et al. 2005), plus 0.03 GJ/GJ pyrolytic oil produced in flash pyrolysis (De Feber & Gielen 2000) and 0.54 GJ/t dry matter in wastewater for biogas production and upgrading to biomethane (Suh & Rousseaux 2001; Gemis 2009). The heat demand of the plant is 0.40 GJ/GJ bioethanol produced (De Feber & Gielen 2000) and 110 MJ/t dry matter in wastewaters (Berglund & Börjesson 2006). These energy needs are completely met by heat and power produced by combustion of lignin and residues. Information concerning auxiliary material used comes from Hamelinck et al. 2005. CH_4 and N_2O emissions from combustion of process residues for generation of electricity and heat are also estimated.

2.5 Distribution and final use

Bioethanol is distributed to fuelling stations (transport distance assumed: 100 km) where it is used to fuel passenger cars. Emissions for combustion of bioethanol in cars (i.e. CH_4, N_2O) are also estimated (Halleux et al. 2008). Biomethane is fed to the national natural gas grid, by which it is delivered to final applications where it can replace natural gas in all its existing applications. It is assumed that biomethane will be burnt in a boiler and the resulting emissions are estimated. It should be noted that, since the combustion of these biofuels (e.g. lignin and residues, bioethanol and biomethane) releases CO_2 which has a biological origin, it is not accounted for as a GHG. Phenols are transported for 50 km to their final application. It is assumed that no carbon storage in products occurs, i.e. all the carbon is released to the atmosphere within the time of the functional unit (one year).

2.6 Allocation

Allocation in LCA is carried out to attribute shares of the total environmental impact to the different products of a system. This concept is extremely important for biorefinery systems, as multiple energy and material products are produced. Scientific LCA publications show benefits and disadvantages of several allocation methods (Curran 2007; Ekval and Finnveden, 2001; Frischknecht 2000; Wang et al., 2004), but the issue of the most suitable allocation procedure is still open.
The ISO standards suggest avoiding allocation by expanding system boundaries, when possible. This method relies on the expansion of the product system to include the additional functions related to the co-products. This procedure (called substitution method or system expansion) has the advantage to avoid allocation issues while has the disadvantage to make the system too complex, especially if multiple co-products are present (like in biorefineries). The identification of one of the output as the main product is an arbitrary choice and can be a difficult decision in biorefinery systems, where multiple useful and valuable outputs are co-produced.

In this assessment, the main product is assumed to be bioethanol and the environmental benefits of co-products are assumed as credits, calculated thanks to the fossil reference systems. These credits (i.e. the GHG and fossil energy saved by the co-products) are then subtracted to the total GHG emissions and energy consumption of the whole system; the resulting environmental burdens are completely assigned to the main product.

Allocation methods can use the physic/thermodynamic characteristic or economic value of products for sharing the total GHG emissions among the different products. In this study, in addition to the substitution method, the following allocation procedures are used and compared:

- Energy
- Exergy
- Economic
- New method based on the shares of GHG avoided when compared with a fossil reference system.

The specific factors used for the energy allocation criteria are product higher heating values[4]. The energy content of phenols was estimated by means of the Dulong's formula (HHV = 33950 C + 144200 (H – O/8) + 9400 S [kJ/kg]; average formula assumed: $C_{8.6}H_{11.2}O_{1.4}$). Allocation based on energy content of products can be easily carried out but its application is inconsistent (i.e. lacking of a proper logical relation) and results into misleading conclusions if there are some products which are not used as energy carriers (e.g. chemicals).

Allocation based on exergy overcomes this inconsistency but can be problematic to be applied because of the difficulties for estimating the exergy content of substances (especially new bio-based products). In this study, exergy content of products are collected from a specific database (Ayres et al., 1996).

Allocation based on economic values focuses on external characteristics of the products and has the disadvantages that do not take into account the environmental perspective and the physical properties of the products, because is based on their "value" in human societies. In addition, market values of products can fluctuate according to the reference year, production chain and geographical location (Ekvall, 2001). Economic values of products have been estimated from an internet search.

In Table 6, the specific factors used for the energy, exergy and economic allocation are listed.

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[4] Note: higher heating values are not commonly used for energy allocation currently. At the time of the study the use of lower or higher heating values was still under debate.

Table 6 Specific factors for biorefinery products used for allocation based on energy, exergy or economic value.

	Energy allocation		Exergy allocation		Economic allocation	
	Unit	**Energy content**	**Unit**	**Exergy content**	**Unit**	**Economic value**
Transportation biofuel						
Bioethanol	MJ/kg	27	MJ_{ex}/kg	29	€/kg	1.2[a]
Other bioenergy carriers						
Biomethane	MJ/kg	35	MJ_{ex}/kg	52	€/GJ	7.62[b]
Electricity	MJ/MJ	1	MJ_{ex}/MJ	1	€/GJ	33.33[c]
Heat	MJ/MJ	1	MJ_{ex}/MJ	0.6	€/GJ	7.08[b]
Biochemicals						
Phenols	MJ/kg	32	MJ_{ex}/kg	33	€/ton	610[d]

[a] Calculated on the basis of gasoline price in Austria (0.95 €/L)

[b] Price based on energy content of replaced natural gas

[c] Average electricity price for households in Austria

[d] Average price of phenols in the EU

The new allocation method is based on the fact that environmental burdens are shared among co-products according to the respective shares of the fossil counterparts in the total GHG emissions of the fossil reference system. For instance, if gasoline contributes for 80% to total GHG emissions of the fossil reference system, 80% of total GHG emissions of the biorefinery system will be assigned to bioethanol (which is assumed to replace conventional gasoline). The main advantages of this new allocation method are the following:

- environmental burdens are assigned according to the effective GHG savings of the products, thus giving more importance to those products which are responsible of the largest savings;
- it is not necessary to choose a main product; and
- it can be applied indifferently to energy or material products.

For more information on the allocation method please see Cherubini et al, 2011.

3. Results

The investigated biorefinery system, which uses 530 kt of pellets from forest softwood residues as raw materials annually produces the following amounts of final products/services per year (values are calculated in accordance with feedstock composition and conversion efficiencies):

- Transportation service with bioethanol: 1338 millions of km (i.e. 121 kt);
- Electricity from combustion of lignin and residues (CHP): 350 TJ;

- Heat from combustion of lignin and residues (CHP): 139 TJ;
- Heat from combustion of biomethane: 256 TJ; and
- Phenols: 2.40 kt.

Values for electricity and heat are net, i.e. the energy required to feed the biorefinery plant is already subtracted. The fossil reference system used to compare the final results produces the same products / services from non renewable sources.

3.1 GHG balance

Results of the GHG balance are reported in Table 7.

Table 7 GHG balance of the biorefinery and fossil reference system.

	Unit	**Biorefinery**				**Fossil reference system**
		2020	**2032**	**2050**	**2100**	**(all years)**
Average annual GHG emissions						
Total	kt CO_2-eq./a	435	256	176	104	381
CO_2	kt CO_2-eq./a	430	251	171	99	357
N_2O	kt CO_2-eq./a	1.45	1.45	1.45	1.45	7.42
CH_4	kt CO_2-eq./a	3.70	3.70	3.70	3.70	16.1
Average annual GHG savings						
per year	kt CO_2-eq./a	no savings	125	205	277	
per year	%	no savings	33%	54%	73%	
per input biomass	kg CO_2-eq./t_{dry}	no savings	236	387	523	

The emissions from the biorefinery are time dependent (Figure 5 and Table 7). The biorefinery system releases more GHG emissions than the fossil reference system until 2025, but the longer the system is in operation, the more GHG emissions are saved.. Since the biorefinery system releases more GHG emissions than its fossil energy counterpart until 2025, it cannot help with the EU bioenergy target in 2020. However, the average annual savings during the first 20 years are 125 kt CO_2-eq./a, or 33% of the fossil emissions during the same period.

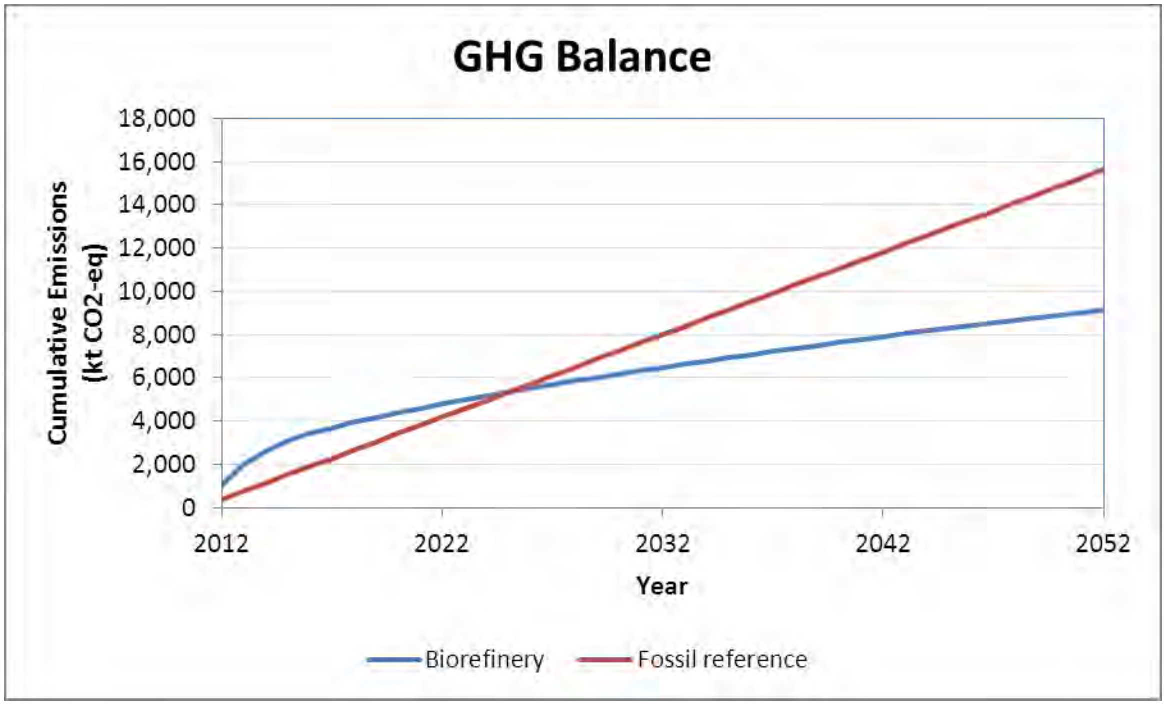

Figure 5: The cumulative emissions of the biorefinery and fossil reference systems. After 2025, the bioenergy system produces less emissions than the comparative fossil reference system.

Contributions to total GHG emissions of the biorefinery system are reported in Figure 6 (expressed as % of total emissions). The contribution from forest management change decreases over time as the land stabilizes to a new dynamic equilibrium. In 2020, the emissions due to forest management change are 89% of total emissions. By 2050, this has decreased to 79%, and by 2100, forest management contributes only 72% to total emissions. The contributions from other portions of the production chain are relatively minor, with the exception of the production of the pellets. By 2100, this activity accounts for 13% of total emissions. The category “other” includes losses of CH_4 during biogas formation and biomethane handling and treatment of waste and wastewaters.

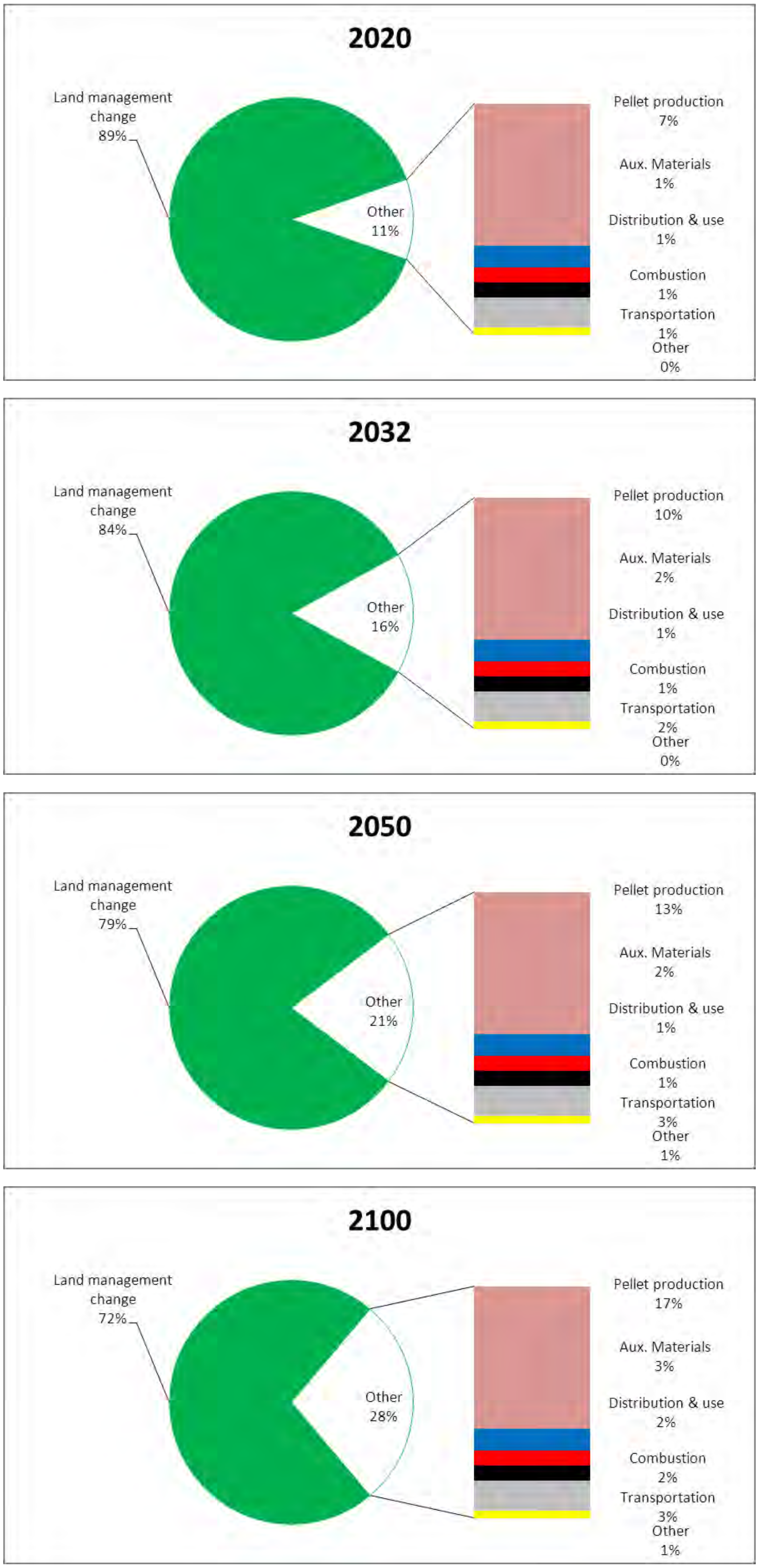

Figure 6 Contributions to total GHG emissions of the biorefinery system.

GHG emissions from pellet production have three main contributors:

- Collection of residues in the forest (35%)
- Transport of the residues from the forest to the pelletizing facility (31%)
- Energy required to produce pellets (34%).

Concerning the fossil reference system, total GHG emissions have the following contributions:

- 69.5% Gasoline,
- 18.6% Electricity,
- 4.30% Heat from oil,
- 5.39% heat from natural gas,
- 2.19% Phenols.

3.2 Energy balance

The energy balances of the biorefinery and fossil energy system are illustrated in Figure 7. Results reveal that the biorefinery system requires a higher cumulative primary energy demand than the fossil reference system (11.4 PJ/a against 6.4 PJ/a), but it is mainly constituted by renewable energy (90%, the energy content of the feedstock) and consistent non renewable energy sources can be saved (5.32 PJ/a, equal to 84% of total non renewable energy used by the fossil reference system). A ton of dry raw material can save approximately 11.2 GJ of non renewable energy.

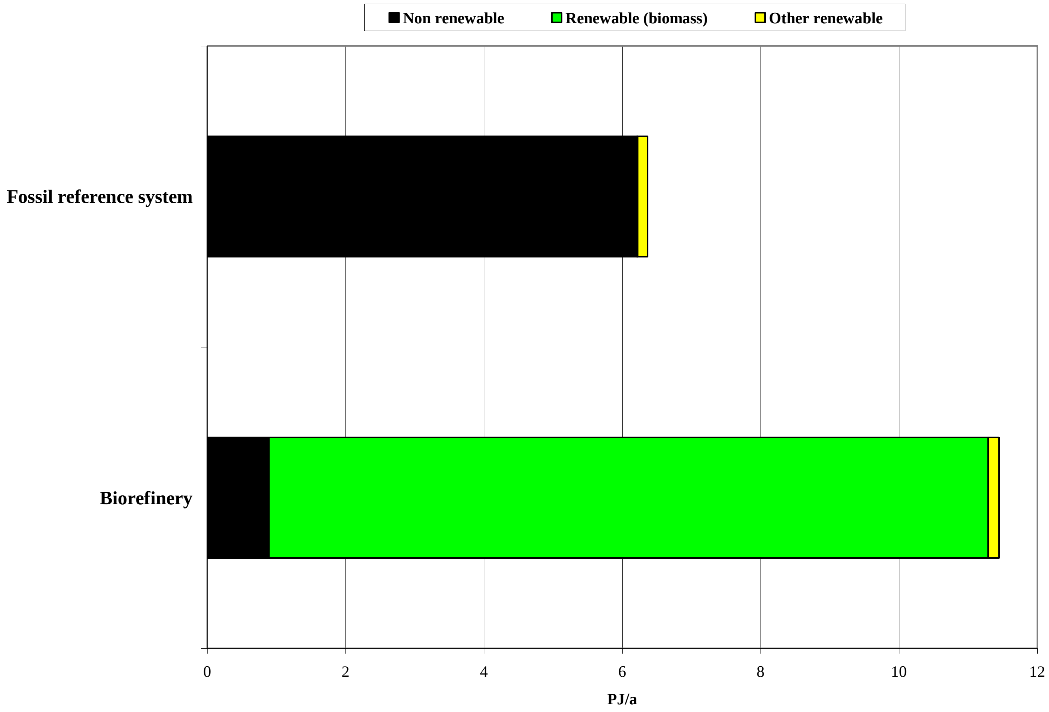

Figure 7 Cumulative primary energy demand of the biorefinery and fossil energy system.

3.3 Allocation

Several allocation procedures are here applied in order to share the total GHG emissions of the biorefinery among the different products. The allocation criteria are based on energy content, exergy content, economic value of products and on the new method (based on the emission shares in the fossil reference system). An attempt to avoid allocation through substitution method was also performed. Results are shown in Figure 8, where average annual GHG emissions of the biorefinery systems are allocated to products (for the sake of brevity, allocation of the primary energy demand is not reported but can be estimated using the results of the energy balance section).

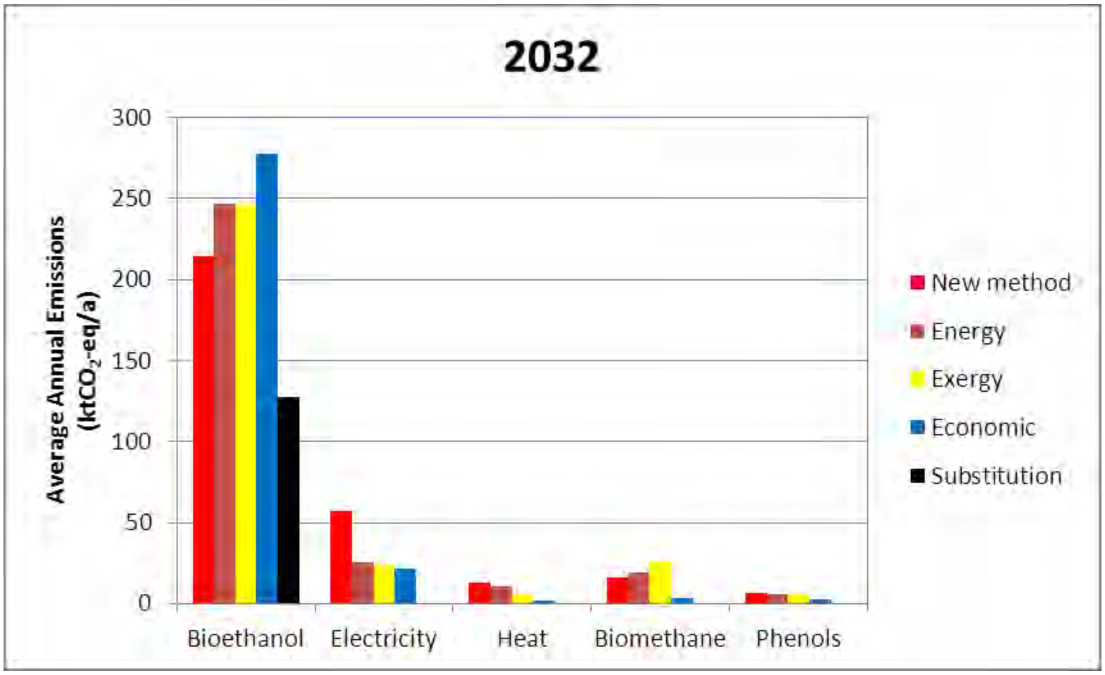

Figure 8 Comparison of average annual emissions by 2032 among different allocation methods.

Besides substitution method which uses a different approach, all the allocation criteria lead to relatively similar results for bioethanol and phenols, while for other products differences are larger. Allocation based on energy and exergy content of products show similar results for almost all the products, while allocation based on economic values increases the shares of the transportation biofuel, while decreasing the environmental burdens assigned to electricity, heat and biomethane.

The "new allocation method" assigns higher emissions to electricity and heat, because these energy carriers are assumed to replace natural gas derived electricity and oil derived heat, which have a relevant contribution to the total GHG emissions of the fossil reference system. This new method is particularly important when GHG savings are the main task of the study, because it assigns more importance to those products which save more GHG emissions.

The specific GHG emission factors for each biorefinery product, according to the allocation procedure, are listed in Table 8. These data are obtained by dividing the results of the previous figure by the quantity of product produced. Therefore, in Table 8, GHG emissions per unit of product, i.e. km driven for biofuels, kWh for electricity and heat, g for phenols and GJ for heat from biomethane, are reported. For instance, these factors can be applied in a LCA if these products are used as auxiliary materials in a future biobased society. Results of these biomass derived products and services can be compared with those derived from oil refinery. For instance, driving a car fuelled with bioethanol (184 g CO_2-eq./km, energy allocation) instead of conventional gasoline (198 g CO_2-eq./km), saves approximately 7% of CO_2-eq. emissions; savings will be up to 19% if the new allocation method is used.

Table 8 Specific factors of GHG emissions to 2032 of the biorefinery products according to different allocation methods.

		New method	Energy	Exergy	Economic	Substitution method
Bioethanol	g CO_2-eq./km	161	184	184	207	95
Electricity from CHP	g CO_2-eq./kWh	591	271	251	229	-1,139
Heat from CHP	g CO_2-eq./kWh	345	271	151	48.7	-663
Heat from biomethane	g CO_2-eq./kWh	64.9	75.8	105	14.5	-125
Phenols	g CO_2-eq./MJ	2.82	2.40	2.31	1.17	-5.43

4. Sensitivity analysis

The allocation results showed in the previous table are an example of how differences in methodological assumptions may lead to diverging results in LCA case studies. Besides allocation, further uncertainties are attributable to differing data sets, including data sources and ages, input parameter values, system boundaries, fossil reference system and others. Therefore, in order to understand the wide variation between LCA results for apparently similar systems, investigation into numerical input assumptions is required as well as into the calculation methodologies that were used to generate the results.

LCA results based on selected single values and simple allocation may significantly increase the risk of drawing misleading conclusions. Some of the key parameters vary widely between different systems and locations, and many are subject to remarkable uncertainties. Thus there is a high probability that the true energy balance and GHG emissions for a specific system will be different from the ‘single value results’. This is the reason why a sensitivity analysis should be always integrated into the final results, in order to take into account the most important assumptions and variables.

Sensitivity analysis is a systematic procedure for estimating the effects of variations in key parameters to the outcome of the assessment, with the aim to establish a required degree of confidence in the results of the study relative to its overall goal. In particular, the objective of this step is to review the results of the analyses, identify the parameters which have the largest influence on the final results and check the accuracy of those data. These key parameters are then changed according to different data sources or assumptions, and the outcomes compared.

Sensitivity to yield class

In this study, the parameter with the largest influence on the final results is forest management (see Figure 6). To investigate the sensitivity of the results of the assumptions made for the assessment of forest management change we derived six different CO_2FIX models (Table 9).

Table 9: Model descriptions for sensitivity analysis of forest management change

Model No.	Yield class	Location	Elevation (masl)	Annual precipitation (mm)	Average annual temperature (°C)
1	8	Bruck an der Mur	482	795.2	8.1
2	10	Bruck an der Mur	482	795.2	8.1
2a	10	Afflenz	780	885.8	6.3
2b	10	Mariazell	875	1081.3	6.1
2c	10	Mürzzuschlag	700	1035.2	6.2
3	12	Bruck an der Mur	482	795.2	8.1

The variation in results by yield class (models 1, 2, and 3) is shown in Table 10. It shows that the number of hectares required varies in response to the yield class. However, the emissions per tonne of biomass are insensitive to yield class. This can be expected because the same amount of biomass is extracted in all cases and the emissions depend on the decay rate of the biomass if it had been left on site. Since the area required is sensitive to yield class but the emissions are not, the loss of biomass over a cycle on a per hectare basis is also sensitive to yield class.

The variation in results by climate (models 2, 2a, 2b, 2c) is shown in Table 11. The net emissions during a period of time from combusting biomass, that would have decayed, are a function of the decay rate of the biomass. As a generality, the faster the decay rate, the less are the emissions. In the extreme, if 100% of the material decays in less than one year, then there are no net annual emissions caused by combusting the material. At the other extreme, combusting biomass than never decays causes 100% emissions.

Table 10: Variation in forest management change components by yield class

	Class 8			Class 10			Class 12		
Area required									
Annual (ha/a)	6,420			5,232			4,399		
Total (ha)	577,824			470,850			395,884		
	2020	**2050**	**2100**	**2020**	**2050**	**2100**	**2020**	**2050**	**2100**
Forest management change emissions									
Total Emissions (Mt CO_2)	3.87	6.65	8.77	3.87	6.65	8.78	3.87	6.65	8.77
Emissions intensity (t CO_2/t dm)	0.811	0.322	0.186	0.812	0.322	0.186	0.812	0.322	0.186
	Reference (tC/ha)	**Project (tC/ha)**	**Net (tC/ha)**	**Reference (tC/ha)**	**Project (tC/ha)**	**Net (tC/ha)**	**Reference (tC/ha)**	**Project (tC/ha)**	**Net (tC/ha)**
Cycle average biomass									
Stems, branches, foliage	78.8	78.8	0.0	100.4	100.4	0.0	122.0	122.0	0.0
Roots	22.7	22.7	0.0	28.9	28.9	0.0	35.2	35.2	0.0
Deadwood	19.9	18.8	-1.0	25.3	24.0	-1.3	30.7	29.2	-1.5
Litter	4.4	4.2	-0.2	5.6	5.4	-0.2	6.8	6.5	-0.3
Soil	45.5	42.1	-3.4	57.9	53.8	-4.1	70.4	65.5	-4.9
Total	**171.2**	**166.6**	**-4.6**	**218.1**	**212.5**	**-5.6**	**265.0**	**258.4**	**-6.7**
Products	3.4	3.4	0.0	4.3	4.3	0.0	5.2	5.2	0.0
Bioenergy	0.0	0.4	0.4	0.0	0.6	0.6	0.0	0.7	0.7
Total including products	**174.6**	**170.5**	**-4.1**	**222.4**	**217.3**	**-5.1**	**270.2**	**264.2**	**-6.0**

Table 11: Variation in forest management change components by climate

	Class 10 – Bruck an der Mur			**Class 10 - Afflenz**			**Class 10- Mariazell**			**Class 10- Mürzzuschlag**		
Area required												
Annual (ha/a)	5,232			5,232			5,232			5,232		
Total (ha)	470,850			470,850			470,850			470,850		
	2020	**2050**	**2100**	**2020**	**2050**	**2100**	**2020**	**2050**	**2100**	**2020**	**2050**	**2100**
Forest management change emissions												
Total Emissions (Mt CO_2)	3.87	6.65	8.78	3.91	6.72	8.87	3.71	6.35	8.33	3.71	6.34	8.31
Emissions intensity (t CO_2/t dm)	0.812	0.322	0.186	0.820	0.325	0.188	0.779	0.307	0.177	0.777	0.307	0.176
	Reference (tC/ha)	**Project (tC/ha)**	**Net (tC/ha)**	**Reference (tC/ha)**	**Project (tC/ha)**	**Net (tC/ha)**	**Reference (tC/ha)**	**Project (tC/ha)**	**Net (tC/ha)**	**Reference (tC/ha)**	**Project (tC/ha)**	**Net (tC/ha)**
Cycle average biomass												
Stems, branches, foliage	100.4	100.4	0.0	100.4	100.4	0.0	100.4	100.4	0.0	100.4	100.4	0.0
Roots	28.9	28.9	0.0	28.9	28.9	0.0	28.9	28.9	0.0	28.9	28.9	0.0
Deadwood	25.3	24.0	-1.3	25.7	24.4	-1.3	23.1	22.0	-1.2	23.0	21.9	-1.2
Litter	5.6	5.4	-0.2	5.6	5.4	-0.2	5.6	5.4	-0.2	5.6	5.4	-0.2
Soil	57.9	53.8	-4.1	58.4	54.3	-4.1	55.2	51.2	-3.9	55.0	51.1	-3.9
Total	**218.1**	**212.5**	**-5.6**	**219.1**	**213.4**	**-5.7**	**213.2**	**207.9**	**-5.3**	**213.0**	**207.7**	**-5.3**
Products	4.3	4.3	0.0	4.3	4.3	0.0	4.3	4.3	0.0	4.3	4.3	0.0
Bioenergy	0.0	0.6	0.6	0.0	0.6	0.6	0.0	0.6	0.6	0.0	0.6	0.6
Total including products	**222.4**	**217.3**	**-5.1**	**223.4**	**218.3**	**-5.1**	**217.5**	**212.7**	**-4.8**	**217.3**	**212.5**	**-4.8**

Sensitivity to climate

Using equations for decomposition rate as a function of climatic parameters (temperature and annual precipitation) from Moore et al (1999), we find for woody material that the annual decomposition is greatest for Mariazell and lowest for Afflenz. The models (Table 11) show the expected relationship. The emissions are highest for the site with the lowest amount of annual decomposition (i.e. slowest decay rate). However the variability is quite small. The standard error of the mean total emissions is about 1.5% of the mean total emissions.

Sensitivity to management

Two assumptions on forest management were made to create the previous models: These assumptions are:

1) Final felling rotation period; and
2) No collection of thinning residues for bioenergy.

Table 12 shows the variation in results if the stands had a final felling every 80 years instead of every 90 years. With shorter rotations, there is 8.5% less biomass to harvest at final felling. Hence more area of forest is required. The stands with no harvesting of forest residues also have less biomass with the 80-year rather than the 90-year rotation and hence there are fewer emissions when the biomass is used for energy. However the difference is small. The emission intensity decreases by approximately 1%. It is important to note that this result does not mean the forest should be converted to an 80-year rotation.

Table 13 shows the variation caused by the assumption on the use of thinning residues for biomass. Fewer hectares of forest are required to produce the required amount of biomass since in addition to the residues from final felling. The thinning residues are 33% of the total residues removed from the forest per year. However the emission intensity does not vary if the thinning residues are used since the emissions are dependent on the amount of biomass removed per year and not whether they come from the final felling or not.

Table 12: Variation in forest management change components by rotation length

	90-year rotation			**80-year rotation**		
Area required						
Annual (ha/a)	5,232			5,400		
Total (ha)	470,850			486,026		
	2020	**2050**	**2100**	**2020**	**2050**	**2100**
Forest management change emissions						
Total emissions (Mt CO_2)	3.87	6.65	8.78	3.85	6.58	8.69
Emissions intensity (t CO_2/t dm)	0.812	0.322	0.186	0.807	0.318	0.184
	Reference (tC/ha)	**Project (tC/ha)**	**Net (tC/ha)**	**Reference (tC/ha)**	**Project (tC/ha)**	**Net (tC/ha)**
Cycle average biomass						
Stems, branches, foliage	100.4	100.4	0.0	89.9	89.9	0.0
Roots	28.9	28.9	0.0	26.5	26.5	0.0
Deadwood	25.3	24.0	-1.3	22.7	21.3	-1.3
Litter	5.6	5.4	-0.2	5.4	5.2	-0.3
Soil	57.9	53.8	-4.1	52.6	48.5	-4.1
Total	**218.1**	**212.5**	**-5.6**	**197.2**	**191.5**	**-5.7**
Products	4.3	4.3	0.0	4.4	4.4	0.0
Bioenergy	0.0	0.6	0.6	0.0	0.6	0.6
Total including products	**222.4**	**217.3**	**-5.1**	**201.6**	**196.5**	**-5.1**

Table 13: Variation in forest management change components by thinning management

	Removal at final harvest residues only			**Removal at thinning and final harvest residues**		
Area required						
Annual (ha/a)	5,232			3,518		
Total (ha)	470,850			316,579		
	2020	**2050**	**2100**	**2020**	**2050**	**2100**
Forest management change emissions						
Total Emissions (Mt CO_2)	3.87	6.65	8.78	3.87	6.65	8.78
Emissions intensity (t CO_2/t dm)	0.812	0.322	0.186	0.812	0.322	0.186
	Reference (tC/ha)	**Project (tC/ha)**	**Net (tC/ha)**	**Reference (tC/ha)**	**Project (tC/ha)**	**Net (tC/ha)**
Cycle average biomass						
Stems, branches, foliage	100.4	100.4	0.0	100.4	100.4	0.0
Roots	28.9	28.9	0.0	28.9	28.9	0.0
Deadwood	25.3	24.0	-1.3	25.3	23.5	-1.8
Litter	5.6	5.4	-0.2	5.6	5.2	-0.4
Soil	57.9	53.8	-4.1	57.9	51.8	-6.1
Total	**218.1**	**212.5**	**-5.6**	**218.1**	**209.9**	**-8.2**
Products	4.3	4.3	0.0	4.3	4.3	0.0
Bioenergy	0.0	0.6	0.6	0.0	0.8	0.8
Total including products	**222.4**	**217.3**	**-5.1**	**222.4**	**215.0**	**-7.4**

5. Conclusions

The use of forest softwood residues in biorefinery has the potential to co-produce bioenergy and chemicals which are currently produced by oil refinery, thus providing savings in GHG emissions and the use of non-renewable energy sources. The biorefinery system depicted in this work produces bioethanol, electricity, heat and phenols from lignocellulosic biomass and is compared with a reference system producing the same amounts of products from fossils.

The GHG balance reveals that the biorefinery system has lower total emissions than the fossil reference system after 13 years. Before this time, the biorefinery system has more emissions because the land is in transition from a dynamic equilibrium with thinnings on site, to one with thinnings removed. The change in forest management cause a loss of carbon stocks in dead wood, litter and soil organic carbon and this is the major source of GHG in the biorefinery system (approximately 70-90% of total emissions depending on year). However, after 13 years, the biorefinery system produces less GHGs that the fossil reference system, and the difference between the two systems continues to increase with time so that after 50 years the biorefinery system has 55% of the emissions of the fossil reference system.

Concerning the energy balance, even if the biorefinery requires more total energy than the fossil reference system, it is mainly made of renewable energy (the energy content of the feedstock) and non-renewable energy sources are saved (84%).

In order to share the total GHG emissions of the biorefinery among the different co-products, several allocation procedures were applied. An attempt to avoid allocation through substitution method was developed and then allocations based on energy content, exergy content and economic value of outputs were compared with a new allocation method based on the shares of the total GHG emissions of the fossil reference system. All allocation methods are finally compared and the specific GHG emission factors (g CO_2-eq./unit) of each product are calculated. These factors can be applied in future LCA case studies, when biorefinery systems will be the basis of the “so-called” bio-economy, where biorefinery products are regularly used by customers or as auxiliary materials in production processes.

Acknowledgements

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