

Lifecycle assessment of methyltetrahydrofuran, methylfuran and butanone as tailor-made fuel components

Accompanying environmental assessment within the
Cluster Tailor-made fuels from Biomass at RWTH
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Abbreviations

2,3-BD	2,3-butanediol ($C_4H_{10}O_2$)
CED n.r.	Cumulative energy demand, non-renewable (fossil, nuclear, primary forest)
CED r.	Cumulative energy demand, renewable (biomass, geothermal, solar, wind)
CHP	Combined heat and power generation
CRS	Cellulose-rich stream, wet solid output from biomass fractionation
FU	Functional unit; reference unit for lifecycle assessment
GWP	Global warming potential
HC	Hemicelluloses
HMF	Hydroxymethylfurfural ($C_6H_6O_3$)
IBA	Isobutanol (C_4H_8O); isomer to MEK
LCA	Lifecycle assessment
LHV	Lower heating value
LRS	Lignin-rich stream, dry solid output from biomass fractionation
LUC	Land use change
MEK	Methyl ethyl ketone, butanone (C_4H_8O)
MF	2-methylfuran (C_5H_6O)
MFF	2-methylfurfural ($C_6H_6O_2$)
MTHF	Methyltetrahydrofuran (as 2- and 3-MTHF) ($C_5H_{10}O$)
OS	Organosolv; biomass fractionation process
PEM	Polymer electrolyte membrane
PGM	Platinum-group metals
PtH	Power-to-heat
PV	photovoltaics
TON	Turn-over number
XRS	Xylose-rich stream, liquid output from biomass fractionation

Summary

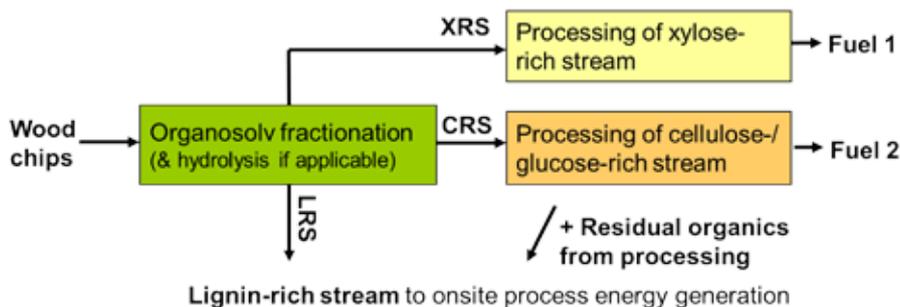
The Cluster “Tailor-made fuels from biomass” at RWTH Aachen University screens a wide variety of potential lignocellulose-based molecules with promising properties as fuel components. For four selected candidates detailed production routes were proposed and analysed based on conceptual process design:

- Ø 2- and 3- methyltetrahydrofuran (MTHF)
- Ø 2-methylfuran (MF)
- Ø 2-butanone (methyl ethyl ketone, MEK)

In internal combustion engines MTHF can serve as a diesel substitute, while MF and butanone are gasoline substitutes.

This report presents the results of a lifecycle assessment (LCA) which was carried out for three potential biorefinery concepts producing these fuels using woodchips as feed. The following illustrates the basic set-up underlying all three concepts.

System set-up of the different biorefinery concepts



Source: Öko-Institut e.V.

The following table summarizes the reaction steps underlying each concept.

Reaction steps for XRS- and CRS-processing in the different concepts

	MTHF concept	MF concept	Butanone concept
XRS-processing (Fuel 1)	Fermentation to itaconic acid, hydrogenation to 3-MTHF	Furfural synthesis, hydrogenation to 2-MF	Furfural synthesis, hydrogenation to 2-MF
CRS-processing (Fuel 2)	Levulinic acid synthesis, hydrogenation to 2-MTHF (& pentanol)	Two-step chemo-catalytic synthesis of 2-MF via methyl furfural	Hydrolysis, fermentation to 2,3-butanediol, dehydration to butanone

Source: Öko-Institut e.V.

The lifecycle assessment illustrates the challenges related to the real implementation of tailor-made fuel production in an integrated complex biorefinery concept. Starting with Organosolv frac-

tionation various conversion and purification steps are included with their concomitant auxiliary material and especially energy requirements. For the four proposed candidates following OS fractionation two synthesis steps are necessary in all cases.

The total cumulative energy demand for production varies between 3 to 4 MJ/MJ_{fuel}, with biomass requirement varying between 2.4 and 2.5 MJ/MJ_{fuel}.

The carbon and energy efficiencies of the three concepts are summarized in following table.

Carbon and energy efficiencies of the three concepts

	MTHF	MF	Butanone
Carbon (kg-C _{fuel} /kg-C _{biomass})	32%	38%	35%
Energy, LHV-basis (MJ _{fuel} /MJ _{biomass})	40%	42%	40%
Energy, LHV-basis (MJ _{fuel} /MJ _{biomass+H2})	36%	40%	39%

Source: Öko-Institut e.V.

The highest carbon and energy efficiency was found for the MF concept. Through a CO-recycle in this concept it is assured that only 1 mol-C/mol-fuel is lost from cellulose processing. MTHF and Butanone concept perform equally well with respect to biomass requirement. However, the MTHF concept relies on additional hydrogen input in a much stronger way. Thus, the total energy efficiency with respect to biomass and hydrogen input is lower. If in the MF concept the pure CO₂ stream was reduced with hydrogen, the carbon efficiency would increase to 42 %, the energy efficiency to 43 % (MJ_{fuel}/MJ_{biomass+H2}) or 41 % (MJ_{fuel}/MJ_{biomass+electricity}). The required hydrogen input would still be smaller than that of the MTHF concept. By TMFB research potential biofuel blends with energy efficiencies > 50 % (referring to biomass and hydrogen input) were identified by model-based product and pathway design. The resulting blends vary concerning hydrogen requirement and complexity. A lifecycle assessment of the most promising candidates will be helpful in order to evaluate the trade-off between process complexity (concomitant process energy requirement) and performance in terms of carbon efficiency and biomass requirement.

The lifecycle impacts differ considerably between the concepts. In general, the MF concept has the lowest values mostly due to the very low energy demand estimated for the CRS processing. However, toxicity concerns were identified for this candidate, which need to be substantiated in order to decide on its suitability as non-hazardous biofuel. In case MF production can be pursued the concept for 2-MF from C5-sugars via furfural needs to be revised. This preliminary LCA already revealed a relevant impact of steam requirement for furfural production even based on the relatively low value reported for the advanced Suprayield process.

For the Butanone concept a relevant contribution of cellulase consumption was found. More research is needed here and other hydrolysis methods should be evaluated. Also alternative water separation concepts could reduce the impact further. Additionally, depending on achievable yields, a combined fermentation of C6 and C5 sugars could be looked at.

In the MTHF concept steam requirement for levulinic acid synthesis is an important driver. It might be investigated whether it can be reduced or better covered when omitting a previous fractionation step. The effort and losses in 3-MTHF production are comparably high. The lower heating value of itaconic acid is rather low so that hydrogen requirement is high. This suggests that in the case of itaconic acid other uses than fuel may be more promising.

Finally, it might be seen whether the OS fractionation, which in all concepts contributes to the impact, can be simplified if no material use of the lignin fraction is envisaged or which conditions justify the effort of fractionation (e.g. higher-value material use of products). A sensitivity analysis for the utilization of the lignin-rich stream as phenol substitute indicates that it would reduce the environmental impact compared to fossil phenol in almost all cases while the impact allocated to the fuels would remain similar, and even be slightly reduced.

At present a relevant fraction of the biomass is consumed on-site process energy generation. In order to substantiate the LCA results more research is needed to determine the gross energy demand of the processes under real conditions (using real instead of pure substrate as feed), as well as their net impact based on a more precise estimate of the energy actually recoverable on-site. If the only product is fuel from the carbohydrate fractions, it appears that viable concepts need to be at least energetically self-sustaining at the biomass quantities required in the analysed concepts.

The use of renewable electricity for hydrogen production and especially directly for process energy reduces the global warming potential. The total cumulative energy demand (CED) remains essentially constant. While also for the eutrophication and acidification potentials the impact of the fully renewable electricity mix is considerably reduced compared to the base mix in 2030, the substitution of natural-gas based hydrogen and heat leads to increased impacts in these categories. For further analyses the question of availability of renewable electricity and hydrogen (also including possible storage losses or transport requirements) should be considered in detail. In order to increase the share of direct renewable electricity the development of flexible production concepts able to respond to fluctuating renewable generation is of interest.

The upstream impact of the biomass becomes relevant when the net impacts of the processes are low, especially in case self-sustaining concepts are reached. At a transport distance of 200 km by lorry, the GWP of transport is similar to that of harvesting and chipping, together amounting to about 10 % of the fossil comparator value (incl. combustion). Depending on the biomass used, there may be additional climate-relevant effects which further increase the upstream GWP. In the case of forest residues soil carbon loss may occur upon removal. Future process development should investigate the utilization of real feedstock focusing on robust processes being able to accept inputs with lower quality and/or varying composition which are less likely to compete with other uses. In general, limitations of biomass potentials have been recognized in recent years so that short-term likely scenarios for national transport limit biofuel amounts to their current volume in Germany. In long-term sustainable scenarios liquid fuels are rather allocated to transport modes where the potential for direct electrification is restricted.

For a complete picture, future LCA work needs to be extended to include the combustion of the fuels taking into account real on-road emissions.

Overall, it has to be taken into account that the processes assessed in this study are in an early stage of development. The focus was on the conversion processes and data for the assessment are mostly based on simulations. The results should hence be taken as indicative and need to be substantiated under real conditions, especially based on the utilisation of real substrate.

Acknowledgments

This work was funded by the German Research Foundation (DFG) within the TMFB Cluster of Excellence EXC 236/2. We would like to thank the responsible researchers at RWTH for the fruitful discussions and information made available concerning the various process steps investigated. Without this supply of background knowledge and process data this work would not have been possible.

1. Introduction and Framework

The Cluster “Tailor-made fuels from Biomass (TMFB)” at RWTH Aachen University aims at developing conversion routes for the production of liquid biofuels while in parallel adapting their properties to the specific needs of the combustion process in order to obtain maximum efficiency of the complete well-to-wheel system. In order to avoid competition with biomass use for food and feed the cluster envisages the use of lignocellulosic feedstock. Since the beginning Oeko-Institut e.V. accompanies the research by assessments from an environmental perspective.

In the second cluster phase the main focus was on the lifecycle assessment of the reference processes proposed by the cluster for the production of four tailor-made fuel candidates: 2- and 3-methyltetrahydrofuran, 2-methylfuran and butanone. The assessments were continuously updated and revised. This report presents the final results taking a look at different energy supply cases and indicating bottlenecks and perspectives for further improvement.

From 2019 the successor of the TMFB Cluster, the Fuel Science Center, will extend its research scope to the conversion of renewable energy and carbon sources in general.

2. Lifecycle assessment of the TMFB reference processes

2.1. Lifecycle methodology

2.1.1. Background and goal of the study

The TMFB-Cluster screens a wide variety of potential lignocellulose-based molecules with promising properties as fuel components. For three selected candidates detailed production routes were proposed and analysed based on conceptual process design:

- Ø 2- and 3- methyltetrahydrofuran (MTHF)
- Ø 2-methylfuran (MF)
- Ø 2-butanone (methyl ethyl ketone, MEK)

In internal combustion engines MTHF can serve as a diesel substitute, while MF and butanone are gasoline substitutes.

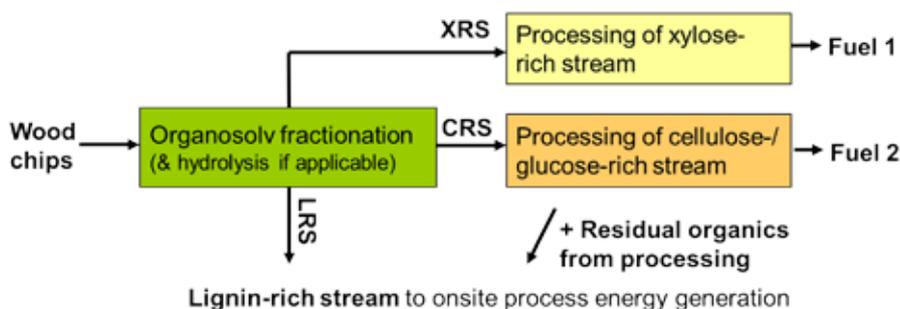
In order to evaluate the performance of the proposed production processes from an environmental point of view, a lifecycle assessment (LCA) was carried out. For this the production routes were integrated into potential biorefinery concepts using woodchips as feed.

As the aim of the accompanying LCA was to give guidance for process development within the TMFB Cluster the main audience of the report are the researchers of TMFB but also other researchers interested in the development of advanced biofuels.

2.1.2. System description and functional unit

The general set-up of the production concepts is illustrated in Figure 2-1.

Figure 2-1: System set-up of the different biorefinery concepts



Source: Öko-Institut e.V.

All concepts are based on an Organosolv fractionation step generating three fractions: the xylose-rich stream (XRS) containing solubilized hemicelluloses, the solid cellulose-rich stream (CRS) as well as a lignin-rich stream (LRS).

While for the MTHF concept the XRS is processed to the fermentation of itaconic acid which is further hydrogenated to 3-MTHF, for the other concepts a conversion to 2-MF via furfural is assumed. The CRS is either converted to 2-MTHF via levulinic acid, or to 2-MF via 2-methylfurfural. For the butanone-concept it is first hydrolysed and then processed to fermentation of 2,3- butanediol which is finally dehydrated to butanone. The different conversion routes are summarised in Table 2-1. It can be seen that in all cases two consecutive reactions steps are necessary to obtain the final product. A detailed description of the processing steps can be found in the corresponding sections in Chapters 2.2-2.4.

Table 2-1: Reaction steps for XRS- and CRS-processing in the different concepts

	MTHF concept	MF concept	Butanone concept
XRS-processing (Fuel 1)	Fermentation to itaconic acid, hydrogenation to 3-MTHF	Furfural synthesis, hydrogenation to 2-MF	Furfural synthesis, hydrogenation to 2-MF
CRS-processing (Fuel 2)	Levulinic acid synthesis, hydrogenation to 2-MTHF (& pentanol)	Two-step chemo-catalytic synthesis of 2-MF via methyl furfural	Hydrolysis, fermentation to 2,3-butanediol, dehydration to butanone

Source: Öko-Institut e.V.

In all cases the lignin-rich stream (LRS) is assumed to be combusted for on-site heat and power generation (CHP) alongside with residual organics from processing. Moreover, a material use of lignin as phenol substitute was considered (see Chapter 2.7).

As the production concepts were developed with the aim of product utilisation in internal combustion engines the **functional unit** is defined as **1 MJ fuel**.

For the MF concept, which yields only 2-MF as a product, no allocation between by-products becomes necessary. The MTHF concept as well as the butanone concept each yield two different substances. Given their intended application as fuel, the most straight-forward allocation parameter

would be their energy contents. For MTHF, the impact is then evenly distributed as 2-MTHF and 3-MTHF have the same lower heating values. For the butanone concept, the heating values of the two fuels differ slightly (butanone 31.5 MJ/kg, MF 31.2 MJ/kg). However, given the general uncertainties related to the present LCA (see Chapter 2.1.5) this difference is insignificant. Thus, for the sake of simplicity the burden is also distributed evenly making the concept equal to a single-output process yielding one “fuel”.

As direct emissions to air only CO₂ is traced in order to determine the carbon efficiency of the concepts. It is assumed to be climate-neutral due to its biogenic origin. Climatic impacts potentially caused in the pre-chain of biomass supply are discussed in a separate Chapter (see Chapter 0). Other direct emissions to air are neglected. The impact of wastewater streams potentially requiring further treatment is neglected.

On the input side all materials and energy required for production need to be considered.

The main **raw material input** is **wood chips**. A lignocellulosic feedstock is explicitly chosen as the basis for all activities of the TMFB cluster in order to avoid direct competition with food. For this study the use of wood was further restricted to forest residues. Some general considerations concerning the availability and sustainability of wood/forest residues for biofuel production are given in Chapter 0.

As **auxiliary materials** ethanol and water are included for the Organosolv process as well as other alcohols, acids or catalysts as required for the specific processes (see Chapters 2.2.1, 2.3.1 and 2.4.1). Their impact is considered “at factory gate”, i.e. the transport of the materials to the biorefinery plant is neglected as a simplification in the present early phase of development where in general uncertainties are high and the focus is on process development. As a further simplification only the operation of the plant is considered, i.e. the impact of plant construction and decommissioning is neglected.

Based on these considerations the LCA results estimate the impact of the operation of the production processes for 1 MJ fuel at factory gate (neglecting the impact of the production plant).

Energy consumption is modelled according to three different energy supply cases as summarised in Table 2-2.

Table 2-2: Energy supply cases

Energy carrier	Base 2030	Base 2030 plus renewable hydrogen	Process energy fully renewable
electricity	German grid mix extrapolated to the year 2030		renewable electricity (PV, wind)
heat / steam	combustion of natural gas		renewable electricity, direct use
hydrogen	by steam reforming of natural gas	by electrolysis based on renewable electricity	

Source: Öko-Institut e.V.

The base reference year for the assessment was set to 2030 so that the electricity mix is extrapolated to this year. For heat and hydrogen production in 2030 natural gas may still form the main energy carrier so that in the base case this source is considered. Electrolytic hydrogen is discussed as a means of storing intermittent renewable energy generation. It may then be used to enhance the carbon efficiency of organo-chemical processes, e.g. in a combined bio- and e-fuel synthesis. For this, the energy supply case “Base 2030 plus renewable hydrogen” is meant to assess the performance of the concepts in case renewable hydrogen is available as external energy source. A fully renewable energy supply case where all external energy is produced by solar or wind is finally included. It has to be kept in mind that these supply cases do not provide an indication on practical feasibility as they do not take into account actual availability of renewable sources or potential necessities concerning their transport and storage (incl. concomitant losses). These aspects need to be elaborated in further analyses.

2.1.3. Data sources

This chapter gives details on the data sources used for those processes which are common to all three concepts. In particular this includes the biomass upstream impact, the Organosolv fractionation, modelling of the CHP-unit as well as the generic data used for the assessment of the supply chain. The data sources for the concept-specific process steps are detailed in Chapters 2.2.2, 2.3.2 and 2.4.2.

Harvesting and wood chip production is assessed based on the ecoinvent dataset “hardwood forestry, beech, sustainable forest management”. Additionally the Globiom study found that the use of forest residues may be linked to a **soil organic carbon loss** which results from the removed residue no longer decomposing in the soil (Globiom 2015). Based on the modelling approach detailed in Globiom 2015 a value of $17 \text{ g CO}_2\text{.eq/MJ}_{\text{fuel}}$ was determined. This value is included in the present LCA but as it rather has to be seen as an indicative value of potential effects calculated for the use of forest residues under determined conditions no effort was made to adapt it to the actual efficiencies of the production processes under consideration in this study. Some general considerations concerning the availability and sustainability of wood/forest residues for biofuel production are given in Chapter 0.

Woodchip transport to the plant was considered over a distance of 200 km (moisture content 35 %) as the TMFB cluster originally aimed for a very high biofuel production capacity (100 000 tons per year to be produced from the cellulose-rich stream of the Organosolv step, fuel 2 in Figure 2-1). As a simplification it was assumed that the whole distance is done by lorry so that the impact of transport scales linearly with distance. For comparison the woodchip-based ethanol production process in ecoinvent assumes a transport distance of 70 km for a production capacity of 42 000 tons of ethanol per year from all sugars (Jungbluth et al. 2007).

The LCA of the **Organosolv** process (OS) is based on TMFB conceptual process design (Viell et al. 2013). The design is based on both experimental and empirical process parameters and puts an emphasis on solvent recycling and energy integration.

Simulations were carried out for a processing capacity of 50 000 t/a of wood which corresponds to 6 250 kg/h at an operation time of 8 000 h/a. A direct use of **wood chips** is assumed with an underlying composition of beech wood:

- Ø 46 wt.% cellulose
- Ø 21 wt.% lignin
- Ø 27 wt.% hemicelluloses
- Ø 6 wt.% other materials.

The external material and energy requirements are summarized in Table 2-3.

Table 2-3: Organosolv process – net material & energy demand

Input	Unit	MTHF concept	MF concept	Butanone concept
Ethanol	kg/kg _{wood,in}	0.0013	0.0013	0.0013
Water	kg/kg _{wood,in}	0.49	0.49	0.62
Electricity	MJ/kg _{wood,in}	1.5	1.6	1.6
Heat	MJ/kg _{wood,in}	1.4	1.4	1.4

Source: Öko-Institut e.V., based on Viell et al. 2013

The OS fractionation simulated by Viell et al. 2013 includes a cellulose hydrolysis step and yields three products: an aqueous glucose solution, a dry xylose-rich stream and a dry lignin-rich stream. Thus, depending on the process set-up of the biorefinery concept considered in this study (see Figure 2-1 and Table 2-1) some adaptations of the input parameters became necessary.

While for the butanone concept the net water requirement for OS fractionation was directly taken from Viell et al. 2013, a corrected value was calculated for the water content of the CRS to be used directly in the MTHF and MF concepts leading to modified net water consumption (Viell 2018a). Ethanol recovery is equally high in all concepts.

Concerning external energy demand there is no influence of the highly heat-integrated cellulose hydrolysis step (Viell 2018a). However, Viell et al. 2013 consider drying of the XRS in order to make it a marketable and transportable product. In a biorefinery concept drying may not become necessary (or to a lower extent) depending on the actual following processing steps. If XRS drying is omitted, the electricity demand is reduced from 1.6 MJ/kg_{wood,in} to 1.4 MJ/kg_{wood,in}. Heat requirement remains unchanged indicating the good integration of the process (Viell 2018a). Thus for the present assessment the overall energy demand is taken from Viell et al. 2013 if thermochemical processing of the XRS is envisaged (MF and butanone concepts) corresponding to a dry XRS stream. For the MTHF concept where the XRS stream is processed to fermentation a reduced overall electricity demand of 1.5 MJ/kg is used as a rough estimate (no full drying of the XRS). Additional water requirement is, however, neglected.

For **onsite energy generation** a combined heat and power plant (**CHP-unit**) is considered assuming high thermal and electrical efficiencies of 60 % and 20 %, respectively, based on the lower

heating value (LHV).¹ The impact of plant construction and decommissioning as well as any auxiliary material input and emissions other than CO₂ are neglected. While the energy content of the LRS is known, the energy content and separation effort of residual streams is in general not modelled in detail. Side components which are processed through a reaction along with the educt (e.g. residual lignin in the CRS) or reaction by-products may have a negative influence on reaction performance and need to be separated from the product with a corresponding energy requirement. Moreover, harsh reaction conditions may alter their molecular structure. Thus, as a conservative estimate, we considered that only 50 % of their original energy content (LHV) reaches the CHP-unit (see also respective Chapters 2.3.2 and 2.4.2).

For quantification of the **lifecycle material and energy inputs** datasets are consistently taken from **ecoinvent 3.4** based on the “at point of substitution” allocation method (ecoinvent 3.4). For hydrogen from steam reforming of natural gas no ecoinvent dataset is available. Here we refer to the dataset provided by the GaBi database (GaBi 6). All datasets are listed in Annex 1.

The electricity mix for the base case is assessed based on the electricity generation structure in Germany in 2030 as modelled by the study “Climate protection scenario 2050” according to the ambitious reduction scenario reaching a reduction of -95 % GHG emissions in 2050 compared to 1990 (OEKO/ISI 2015, see Annex 2). Completely renewable electricity is modelled as 25 % photovoltaics, 75 % wind energy. As a simplification only two specific technologies are considered based on ecoinvent datasets: PV as “electricity production, photovoltaic, 570kWp open ground installation, multi-Si, DE” (25 %) and wind energy as “electricity production, wind, 1-3MW turbine, on-shore, DE” (with gear box) (75 %).

For renewable heat direct conversion of electricity to high-temperature heat via heating rods is assumed. As a simplification full conversion is assumed, and, as for natural gas, distribution losses are neglected.

For electrolysis an efficiency of 70 % is assumed. For a first indication the inventory of the electrolyser components is taken from Spielmann et al. 2015 which is based on studies from the early 2000s on alkaline electrolyser technology as operated in Reykjavik/Iceland by HydroStatoil.

2.1.4. Impact categories and assessment method

Following the quantification of the material and energy flows for the entire production process and the corresponding auxiliary material and energy demand these are evaluated concerning their contribution to the potential environmental impact of the system under study.

Different forms of resources use and pollutants emissions identified in the life cycle inventory work usually have different potential environmental impacts within impact categories. The potential environmental impact is calculated using characterisation methods that associate a pollutant emission to the relevant impact categories by the use of characterisation factors.

Update PEFCR Guide (EC 2018) lists these default impact categories:

climate change, ozone depletion, ecotoxicity for aquatic fresh water, human toxicity – cancer effects, human toxicity – non-cancer effects, particulate matter/respiratory Inorganics, ionising radiation – human health effects, photochemical ozone formation, acidification, eutrophication – terrestrial, eutrophication – aquatic, resource depletion – water, resource depletion – mineral, fossil, land transformation.

¹ Own calculations based on data of the German Federal Statistical Office suggest current utilisation ratios of 58 % and 17 %, thermal (in industry) and electrical, respectively, based on the lower heating value.

The ILCD-Handbook (ILCD 2010) recommends these impact categories:

climate change, (stratospheric) ozone depletion, human toxicity, respiratory inorganics, ionizing radiation, (ground-level) photochemical ozone formation, acidification (land and water), eutrophication (land and water), ecotoxicity, land use, resource depletion (minerals, fossil and renewable energy resources, water).

As ISO 14044 does not set an obligatory list of impact categories the selection is up to the LCA creator (Klöpffer 2009).

In this LCA the following impact categories are taken into account:

- Ø Global warming potential (GWP)
- Ø Eutrophication potential (EP)
- Ø Acidification potential (AP)
- Ø Photochemical ozone formation (POCP)
- Ø Cumulative energy demand (CED n.r. and CED r.)

The calculation of **global warming potential** in terms of CO₂ equivalents is a generally accepted approach. In addition, the Intergovernmental Panel on Climate Change (IPCC) is a specialist international committee that generates and updates the computational methods and the corresponding values for each climatically relevant substance. The calculation of CO₂ equivalents takes into account the length of time that the gases spend in the troposphere. It is therefore necessary to establish the period of time to be used for the climate model for the purposes of the LCA. ISO TS 14067 [ISO 2013] recommends modelling on a 100-year basis as this is most likely to reveal long-term results of the greenhouse effect. Hence for this study, the GWP 100 is calculated with the characterization factors given in [CML 2016]. The GWP presented in this study for the different biofuel production concepts already includes the CO₂ emissions resulting upon combustion.

Eutrophication means the over-enrichment of a water body or the soil with nutrients. In the context of this study, only terrestrial eutrophication is considered. The eutrophication potential (EP) of nutrient emissions is calculated from the aggregation of phosphate equivalents according to [CML 2016].

Acidification can also occur in both terrestrial and aquatic systems. Emissions of acid-forming waste gases are responsible. Acid formation potentials (AP, [CML 2001]) are calculated with the characterization factors given in [CML 2016].

The impact category **photochemical ozone creation potential (POCP)** concerns the formation of summer smog or ground-level ozone. It describes the formation of ground-level ozone (O₃) in the lower layers of air. Ozone causes damage to forests and other vegetation. At higher concentrations it is toxic to humans (causing irritation of respiratory organs, asthma, coughs and eye irritation) [Schmid et al. 2006]. Ozone formation is a complex process in which an ozone creation potential can be allocated to each hydrocarbon. Exact potentials apply only for a defined environment with a particular light intensity, a particular NO_x concentration and defined meteorological conditions. In this LCA the POCP is calculated from the aggregation of ethene equivalents according to [CML 2016].

The consumption of energy resources is represented by the **cumulative energy demand (CED)**. CED is a measure of the total amount of energy resources used to make a product or provide a

service. It also includes the energy contained in the product itself. The CED identifies all non-renewable and renewable energy resources as primary energy values. For this study their lower heating value was used as the basis in the calculations. No characterization factors are used. This means that the consumption of energy resources is not an impact category based on different impact factors, but a life cycle inventory parameter. The advantage is that it hence does not depend on the uncertainties involved in impact assessment so that according to Klöpffer (1997) the accuracy of the calculation of CED is in general much greater than that of most (emission-based) characterization procedures. In that way it serves as an important sum parameter representing the precautionary principle because it implicitly indicates the environmental interventions due to the energy consumption connected with the system analyzed (Klöpffer 1997). As an example the renewable CED may serve as a proxy for impacts related to the expansion of renewable energy generation, like land or resource use.

A number of methodological concepts have been developed in recent years to characterize **human and ecotoxic effects**. In addition to classical midpoints methods such as CML [Guinée et al. 2002] and EDIP [Hauschild / Wenzel 1998], other methods attempt to estimate potential damage at the endpoint, such as Eco-indicator 99 [Goedkoop / Spriensma 1999] or EPS [Steen 1999]. Or combine both concepts, such as ReCiPe [Goedkoop et al. 2009], IMPACT 2002+ [Humbert et al. 2012]. The USEtox model of the UNEP / SETAC Life Cycle Initiative [Rosenbaum et al. 2011, Rosenbaum et al. 2008, Hauschild et al. 2008] tries to harmonize the different models. USEtox is a rating system for the categories of human toxicity and aquatic ecotoxicity. The model puts very high requirements on the data quality and the number of substances to be considered. Since the data on a large number of human and ecotoxic substances is so poor that it is difficult to derive reliable results, this study does not include human toxicity and ecotoxicity by means of an impact assessment. Specifically, the relevant data were not available for the products of the concepts analysed in this study.

Other impact categories, e.g. respiratory inorganics or land use, should be included in further assessments.

2.1.5. Limitations

The interpretation of the results is limited by the fact that many of the processes are still in an early stage of development. Data are mostly taken from in general optimistic simulations based on the results of lab-scale experiments, in many cases in ideal environments, and on values reported in literature. While optimisations were in many cases carried out within the different steps (Organosolv, CRS-processing, XRS-processing) no optimisation was carried out at the links between them. Partly the available data were completed by own estimates where no specific information was available. Also the energy recoverable onsite could only be estimated. Its quantity is, however, decisive and may differ under real conditions.

The results should hence be taken as indicative as they are still subject to uncertainties in the present early phase of development. They need to be substantiated under real conditions, especially based on the utilisation of real substrate.

2.1.6. Critical review

No critical review was carried out for this study because the main goal of the accompanying LCA work was the identification of levers for process improvement in each production concept. Moreover, all production concepts are in a very early stage of development.

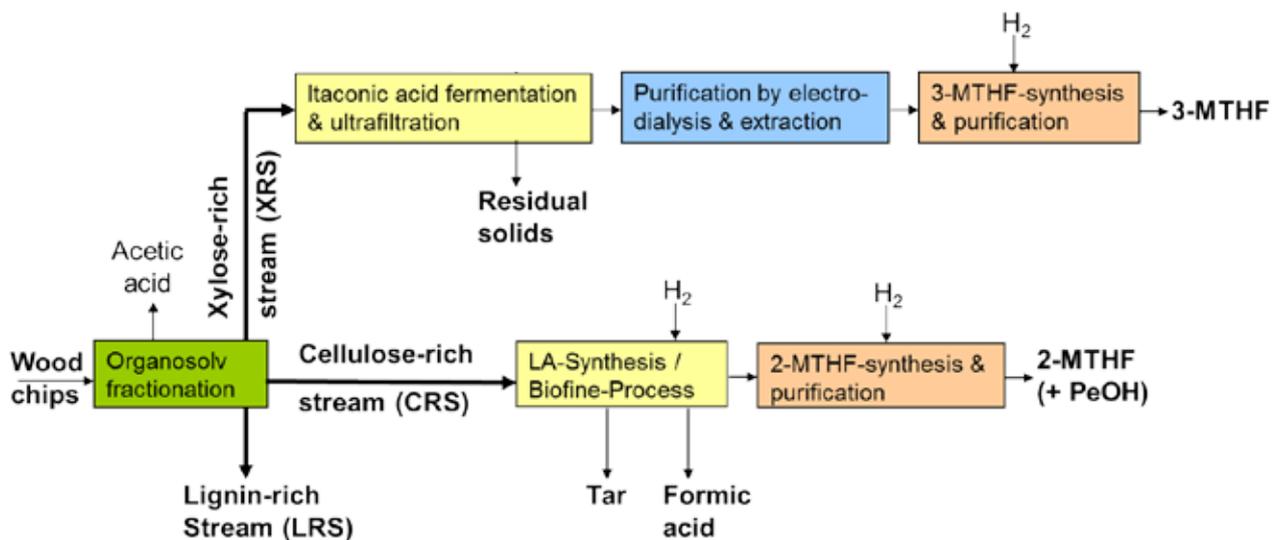
2.2. Lifecycle assessment of the MTHF concept

This chapter first gives a description of the specific set-up of the MTHF concept (Chapter 2.2.1) followed by information on the specific data sources for the concept (Chapter 2.2.2, for description of general sources, the Organosolv fractionation and the treatment of the lignin-rich stream please refer to Chapter 2.1.3). Then the assessment results of the MTHF concept are presented in the form of carbon and energy flows through the processes (Chapter 2.2.3) and lifecycle assessment results (Chapter 2.2.4).

2.2.1. Process description

Based on the Organosolv fractionation (see Chapter 2.1.3) the MTHF concept converts the xylose- and the cellulose-rich streams into 3- and 2-MTHF, respectively. Both pathways proceed via the synthesis of an intermediate acid.

Figure 2-2: MTHF concept



Source: Öko-Institut e.V.

The XRS is directly processed to itaconic acid (IA) fermentation. In order to avoid negative effects on fermentation performance acetic acid needs to be previously separated during Organosolv fractionation. Viell et al. 2013 consider this separation, however, omitting its energy requirement. In all cases only small quantities of acetic acid are produced (< 2% of biomass input according to Viell et al. 2013 [SI]). Any further impact of this removed acid is neglected for the present LCA. No sterilization before fermentation is considered because an industrial scale process should be able to run under non-sterile conditions (Viell 2018a). The process design is based on Ulonska et al. 2014 who analysed different fermentation and downstream-processing concepts for the XRS-route. This work identified the fermentation with *Aspergillus terreus* as advantageous because, even though it requires batch fermentation with comparably long residence times, it shows a considerably better performance concerning product yield. Moreover, the low final pH value of 2 makes downstream processing less energy-intensive. Here, after ultrafiltration, itaconic acid is further purified by electro-dialysis yielding a binary mixture of a high-boiling itaconic acid and water. In order to avoid the

removal of water, IA is then extracted with 3-MTHF after a concentration by reverse osmosis.² Itaconic acid is then hydrogenated to the final product, 3-MTHF, and water which are separated by rectification. The treatment of this water, as well as of the purge stream from electrodialysis containing salts and nutrients is neglected in the present analysis. The residual solids stream from fermentation, consisting of cell mass and lignin, is considered to be burnt on-site for process energy generation.

The CRS is fed to levulinic acid (LA) synthesis according to the Biofine process (Hayes et al. 2006). Here the cellulose is converted thermochemically to levulinic acid, with formic acid and tar as by-products. Also hemicelluloses, if present, can be partially converted to LA via furfural and subsequent hydrogenation. The remaining is also incorporated into the tar as well as the lignin share in the CRS. Under the prevailing process conditions this tar is cracked to a dry char which for this assessment is assumed to be used for on-site process energy generation.

The levulinic acid is then hydrogenated to 2-MTHF and a small quantity of pentanol (PeOH). For the hydrogenation of both itaconic and levulinic acid a catalyst system is required based on a ruthenium-containing precursor complex, a ligand (Triphos) and ionic and/or acidic additives as described in Geilen et al. 2010.

The lignin-rich stream is used for energy generation (see Chapter 2.1.3).

2.2.2. Specific data

The values for **XRS-processing** to 3-MTHF are taken from the TMFB conceptual process design for itaconic acid fermentation, downstream-processing including electrodialysis, reverse osmosis and extraction, and 3-MTHF-synthesis including hydrogenation and purification (Ulonska et al. 2014). The fermentation yields were set 0.4 g_{IA}/g and 0.6 g_{IA}/g from C5-sugars and C6-sugars, respectively. In 2015 an update was issued where the energy requirement for stirring and aeration during fermentation was reduced to 0.3 kW/m³ (formerly 1 kW/m³). The energy requirement used for the present LCA is summarized in Table 2-4.

Table 2-4: Energy requirement of XRS processing incl. updated energy requirement for fermentation

Unit operation	Heat (MJ/MJ _{3-MTHF})	Electricity (MJ/MJ _{3-MTHF})
Fermentation		0.099
Ultrafiltration		0.014
Electrodialysis		0.014
Reverse osmosis & extraction		0.004
Reaction (hydrogenation)	0.228	
Purification	0.008	0.041
Total	0.236	0.172

Source: Ulonska et al. 2014 & 2015

² Also 2-MTHF could be envisaged as an extraction agent, if the presence of excess 3-MTHF proved to negatively influence the subsequent reaction. At this level of detail, the calculations are valid for both cases due to the large molecular similarity of 2-MTHF and 3-MTHF (Ulonska et al. 2014).

For fermentation no nutrient addition is considered.

The calculations for the extraction are based on a published partition coefficient of itaconic acid in 2-MTHF/water of 4.4 (Li et al. 2013) and extrapolated to the process conditions based on internal cluster work. First laboratory results in the TMFB cluster show even higher values for itaconic acid in 3-MTHF/water, 12.14 instead of 4.4, leading to a reduction of 30 % for total energy requirement of XRS processing (Ulonska et al. 2014). For energy requirement at this early stage of development, the analysis only takes the high temperature and pressure for the reaction into account, while an intermediate separation is not considered yet (Ulonska et al. 2014). A stoichiometric hydrogen demand and a yield of 0.96 mol-3-MTHF/mol-IA is considered. The final purification makes use of the heteroazeotrope of MTHF and water and applies vapour recompression for the MTHF column is applied (see also Harwardt 2013).

For the LCA moreover, the net metal requirement for the catalyst was estimated based on the assumption that a turn-over-number (TON) of $1:10^5$ is reached as a mandatory value for industrial operation. Net metal consumption was calculated based on the assumption that the precious PGM-catalyst is recycled at end-of-life with an efficiency of 98 % based on losses found for other platinum group metals by Hagelüken et al. (2005) for Germany. Net metal consumption was then assessed as primary production withecoinvent datasets; Due to the unavailability of primary ruthenium production in the database a proxy for PGM primary production was calculated as a mix of 50% platinum and 50% palladium.

In general the XRS downstream process concept has not been tested experimentally and a validation of the concept is required at laboratory-scale. In particular the reaction to 3-MTHF needs to be tested with real substrate from the fermentation broth to account for the effect of any impurities and remaining water in the itaconic acid. Kinetics should be included in refined process models to identify the optimal process structure and operating point (Ulonska et al. 2014). Hence, Ulonska et al. 2014 consider their report as a very optimistic analysis of the reference process partially based on envisaged performance data.

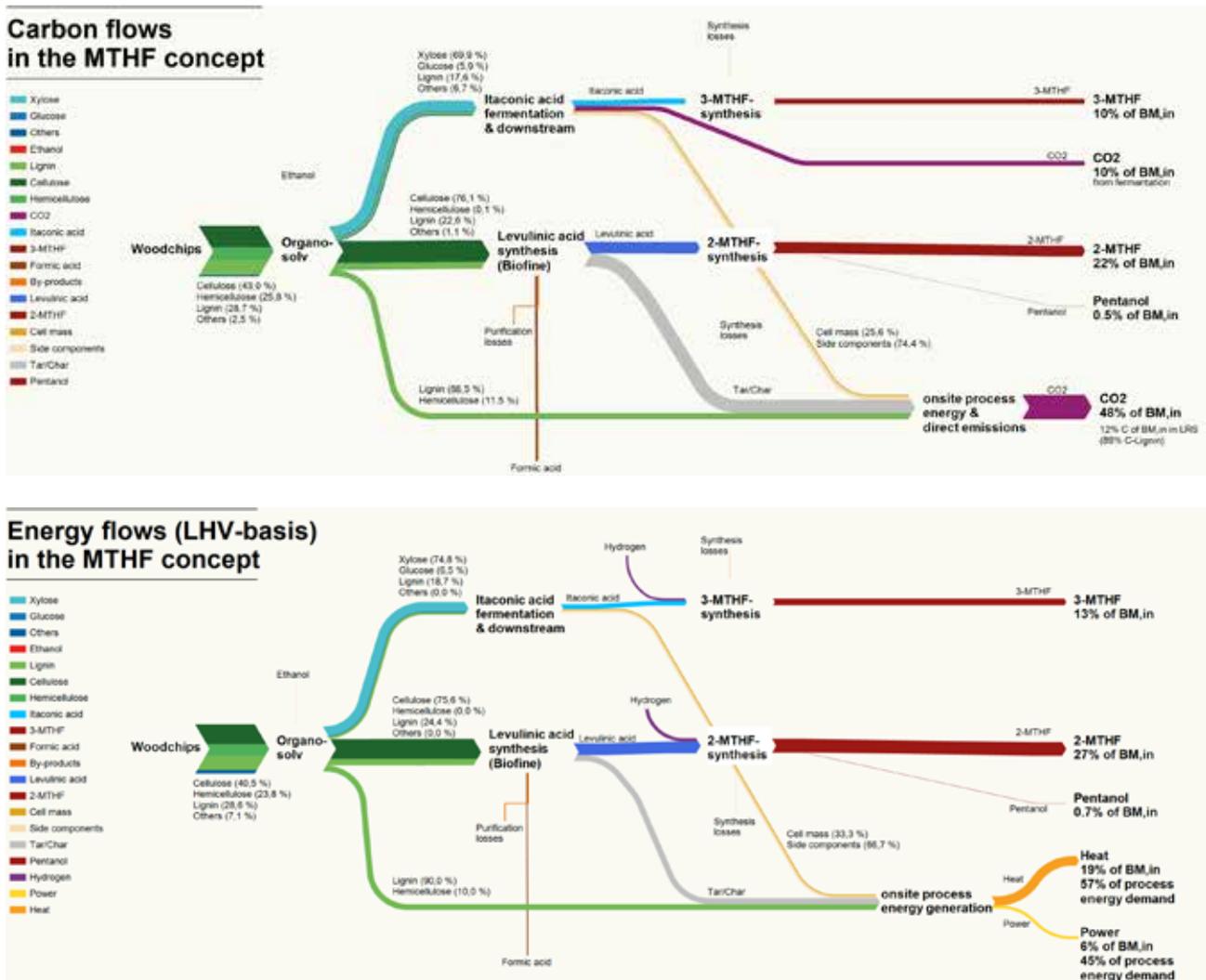
Modelling of **CRS processing** via the Biofine process is based on the yields, auxiliary material demand and gross energy requirements given in Hayes et al. 2006 based on process simulations. The estimate for the energy generated on-site by combustion of the resulting char is based on values found for the processing of paper sludge (Hayes et al. 2006): 0.15 kg-char/kg-feed with a LHV of 25.6 MJ/kg. It can be considered a conservative estimate because of the higher lignin content of the CRS compared to paper sludge. Therefore a sensitivity analysis with a higher value of 0.23 kg/kg at the same LHV was carried out, the effect is discussed in the results sections (Chapter 2.2.3 and 2.2.4). The subsequent hydrogenation of LA to 2-MTHF and pentanol is based on yields of 0.97 mol/mol-LA and 0.02 mol/mol-LA, respectively, as given in Geilen et al. 2010. Stoichiometric hydrogen demand is considered. Energy requirement for hydrogen compression to operating pressure and subsequent product work-up are taken into account based on TMFB conceptual process design data for 2-MTHF synthesis and separation (see also Harwardt 2013). The net metal requirement for the Ruthenium-catalyst is estimated and assessed as for IA hydrogenation.

The simulations for TMFB conceptual process design are carried out for a required output of 100.000 t/a of CRS fuel, and for Hayes et al. 2006 for an output of 133.000 t/a ethyllevulinate (equalling a biomass requirement of 350.000 dry tonnes per year), thus all representing very large-scale processes. Compared to this envisaged large scale, the basic data for the process simulations in many cases still rely on early lab-scale results rather in ideal environments.

2.2.3. Carbon and energy flow analyses

Based on the process set up and material and energy flow modelling described in Chapters 2.2.1 and 2.2.2 the carbon and energy flows through the MTHF concept are calculated (Figure 2-3). The energy flows are indicated based on their lower heating value with respect to dry mass (for onsite process energy generation water content is subtracted).

Figure 2-3: Carbon and LHV-based energy flows in the MTHF concept



Note: in „Energy flows“ only biomass based energy flows and hydrogen input are shown. The MTHF concept moreover requires an external input of heat and electricity (see Table 2-5).

Source: : Öko-Institut e.V.

About one third of the carbon contained in the biomass is converted to fuel, two thirds to CO₂ (about 50 % in CHP or other non-specified direct emissions). The CRS is the biggest fraction from OS separation, resulting in a transfer of 22 % of the carbon to 2-MTHF via the Biofine process and subsequent LA hydrogenation. The main side product from the Biofine step is tar which incorporates side components, mainly lignin, and by-products from the reactions. As a consequence it contains even slightly more carbon than the product levulinic acid. The XRS is smaller and the itaconic acid yield is lower so that only 10 % of the carbon are transferred to 3-MTHF. Based on the present estimate approximately the same quantity is converted to CO₂ during fermentation (IA

losses in downstream processing are neglected in the current assessment). The carbon content of auxiliary materials is comparably negligible, also purification and synthesis losses from hydrogenation as modelled in this assessment do not contribute a major share. 5 % are converted to formic acid in Biofine process. It is, however, not considered as a further valuable output of the process for this early impact assessment.

The energy efficiency of the MTHF concept with respect to biomass input is about 40 %. It is considerably higher than overall carbon efficiency because the external hydrogen addition rises the energy content of the fuel. If hydrogen input is considered in the calculation as an additional energetic input the energetic efficiency is 36 %. The efficiency translates into a biomass requirement of $2.48 \text{ MJ}_{\text{biomass}}/\text{MJ}_{\text{fuel}}$. The quantity of hydrogen for CRS processing is higher because more 2-MTHF is produced. In relative terms, however, more H_2 is needed in 3-MTHF synthesis: 50 % and 30 % of the energy input with the educts come from hydrogen, in 3-MTHF synthesis and 2-MTHF synthesis, respectively.³

In the CHP unit about 20 % of the energy in the biomass is obtained as heat, 6 % as electricity. Only around half of the process energy demand can thus be satisfied onsite. The remaining external energy demand is shown in see Table 2-5. As laid out in Chapter 2.2.2 the energy contained in the char which is sent to CHP from the Biofine process might still be higher. For this a sensitivity analysis was carried out based on the assumption that the respective tar quantities generated from cellulose, hemicellulose and lignin keep their original LHV equalling about $0.23 \text{ kg}_{\text{char}}/\text{kg}_{\text{CRS}}$ with a LHV of 25.6 MJ/kg (compared to $0.15 \text{ kg}_{\text{char}}/\text{kg}_{\text{CRS}}$ in the base case). In this case about 70 % and 54 % of heat and electricity requirement, respectively can be covered on-site. The real LHV of the char is substrate-specific and would have to be substantiated experimentally for the specific feedstock (Hayes et al. 2006).

Table 2-5: External energy demand of the MTHF concept

	MJ/MJ_{fuel}	Comment
Electricity	0.19	
Heat	0.34	distribution losses neglected
Hydrogen	0.31	$0.44 \text{ MJ}_{\text{el}}/\text{MJ}_{\text{fuel}}$ for electrolytic H_2

Source: Öko-Institut e.V.

³ Also in the Biofine process hydrogen can be added to transform hemicellulose sugars into levulinic acid via furfural and subsequent hydrogenation. In this assessment the quantity is negligible due to negligible quantities of hemicelluloses in the CRS.

The carbon and energy efficiencies are summarized in Table 2-6.

Table 2-6: Carbon and energy efficiencies of the MTHF concept

	Carbon	Energy	Comment
in CRS fuel	22%	27%	
in XRS fuel	10%	13%	
overall	33%	40%	2.48 MJ _{biomass} /MJ _{fuel}
overall incl. ext. H ₂		36%	external hydrogen demand included as energy input

Source: Öko-Institut e.V.

2.2.4. LCA results

Following the quantification of the material and energy flows for the entire production process and the corresponding auxiliary material and energy demand these are evaluated concerning their lifecycle environmental impacts. The results are shown in Figure 2-5 and Figure 2-6. The corresponding values are listed in Table 2-7.

2.2.4.1. Main observations for the base case Base 2030

For the base case Figure 2-4 additionally shows the relative contribution of the biomass upstream impact and the impacts related to the TMFB conversion processes differentiating their origin by heat, electricity and auxiliary material input. It can be seen that even with respect to the net impacts, i.e. including onsite energy generation, process energy requirements are responsible for half or more of the impacts. Only for CED r. the main contribution is the energy content of the biomass itself, while for POCP the biomass upstream plays the major role (from machinery used in harvesting).

For GWP and CED n.r. the next biggest contribution comes from auxiliary materials, over 95 % being due to external hydrogen input.

For GWP the contribution of biomass upstream lies in the same range when indirect effects caused by the removal of residual wood from the forest are taken into account according to Globiom 2015. This aspect is discussed along with other considerations on biomass availability and impact in Chapter 2.6. At a transport distance of 200 km, harvesting and transport both contribute similarly to GWP, and together account for approx. 10 % of the net impact.

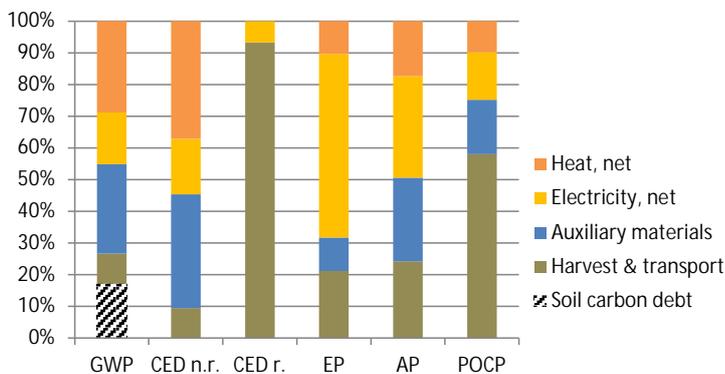
In the base case the net values of the GWP and the non-renewable CED n.r. are similar to that of their fossil comparator diesel (incl. combustion assuming same efficiency). The renewable CED r. essentially represents the energy contained in the biomass: 2.48 MJ/MJ_{fuel} in this concept. The total CED thus amounts to 4 MJ/MJ_{fuel} (see Figure 2-5 a)-d)).

When analysing GWP and CED n.r. with respect to the TMFB conversion steps it is found that the main impact is due to CRS processing the main contribution coming from the heat demand of the Biofine process (62 %), electricity demand plays a minor role (11 %). The remaining is almost exclusively due to the hydrogen requirement (24 %). For the impact of XRS processing hydrogen requirement contributes more than half (53 %), the remaining being in similar parts due to electricity and heat demand. The impact of OS fractionation is dominated by electricity (55 %) followed by heat demand.

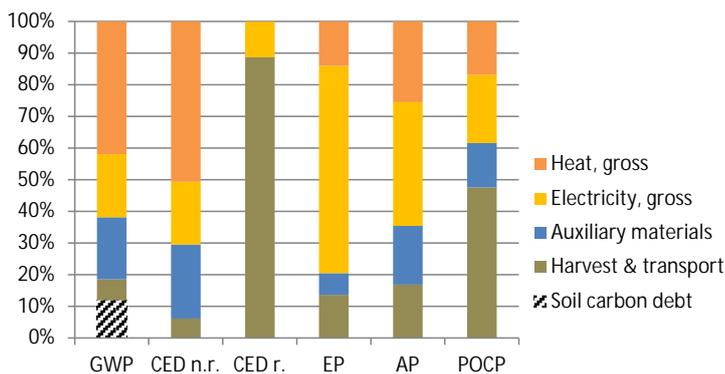
As mentioned in Chapters 2.2.2 and 2.2.3 the energy available for CHP generation from the Biofine char may be underestimated in this study. Therefore a sensitivity analysis was carried out where the quantity was increased by 50 % (0.23 instead of 0.15 kg_{char}/kg_{CRS} at constant LHV, see Chapter 2.2.3). The resulting lower external electricity and heat demand lead to a decrease in net GWP of approx. 10 %. Process energy demand can still not be covered onsite (see Chapter 2.2.3).

For the net EP of the MTHF concept approx. 20 % come from biomass harvesting and transport. Within the TMFB conversion steps electricity causes the main impact: in OS fractionation 91 %, CRS processing 53 % and XRS processing 80 %. For CRS processing heat demand for the Biofine process accounts for a further relevant share (31 %). Also for AP biomass upstream accounts for approx. 20 % of the total impact. Within the TMFB conversion steps OS step is again dominated by electricity (79 %), CRS processing by heat (44%) followed by electricity (22 %) and for XRS processing electricity (48 %) and hydrogen (20 %) are dominant. For POCP with its very high contribution of biomass harvesting the TMFB conversion processes show a similar picture as for AP, but the impact of hydrogen slightly more prominent.

Figure 2-4: Contribution analysis for the different impact categories, Base 2030



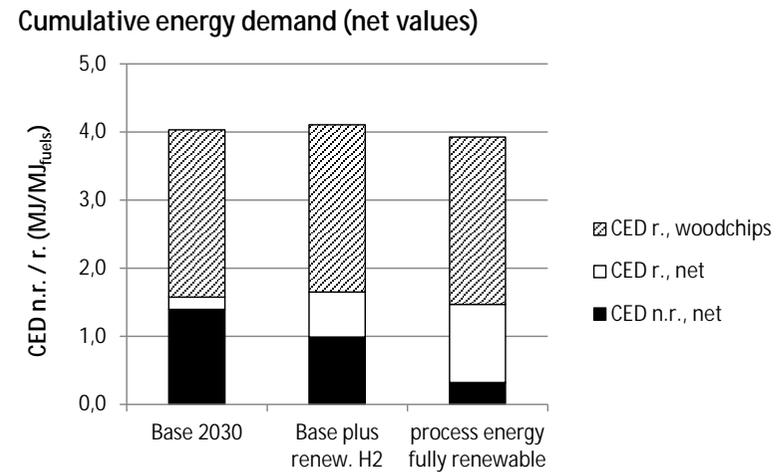
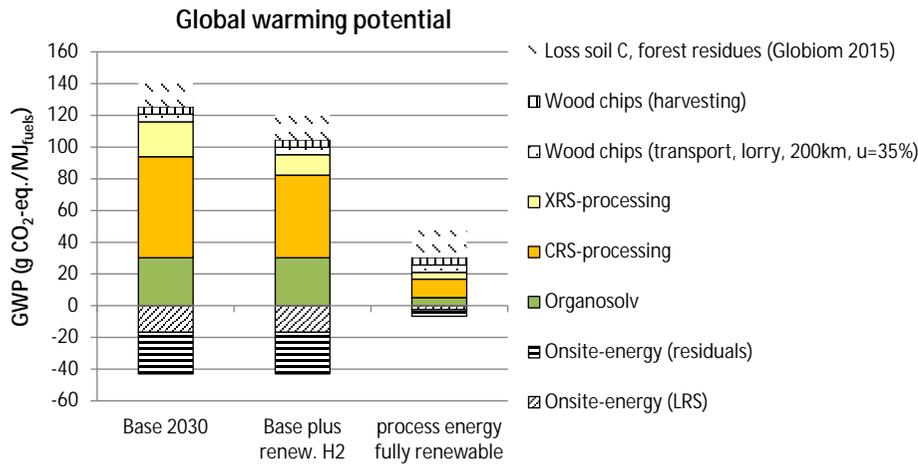
a) Contribution with respect to the **net impact**, i.e. considering external process energy demand only (incl. energy recovery by CHP)



b) Contribution with respect to the **gross impact**, referring to total process energy demand (excl. energy recovery by CHP)

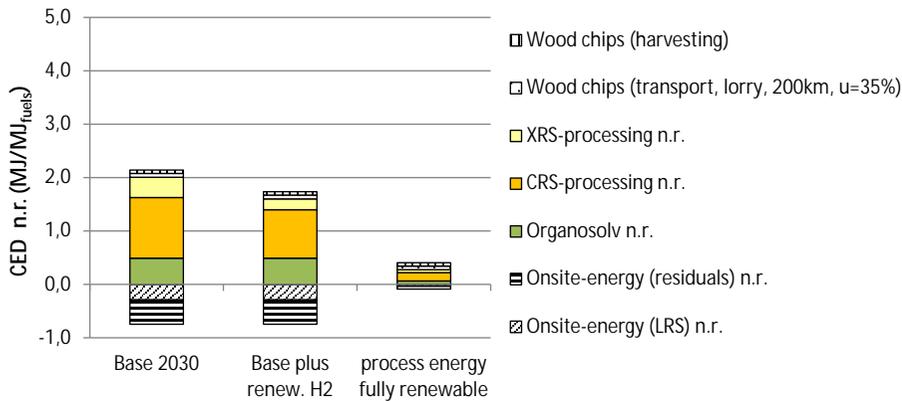
Source: Öko-Institut e.V.

Figure 2-5: Global warming potential (GWP) and cumulative energy demand (CED) for the MTHF concept



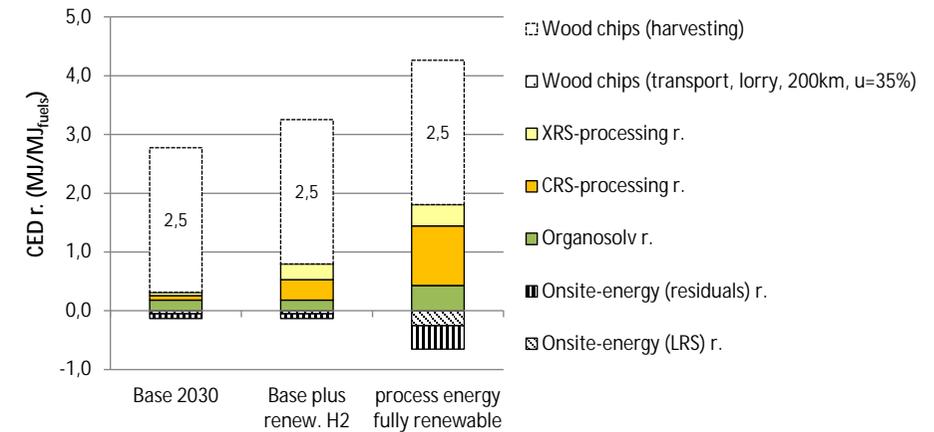
a) Global warming potential*

Cumulative energy demand, non-renewable



b) Total cumulative energy demand, net values

Cumulative energy demand, renewable



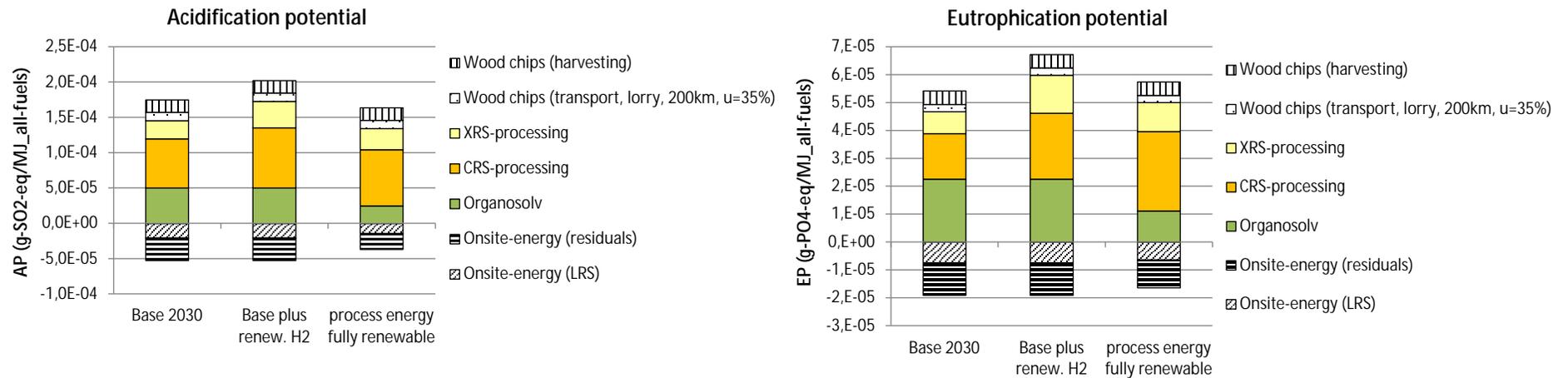
c) Non-renewable cumulative energy demand

d) Renewable cumulative energy demand

* GWP including the CO₂ emissions resulting upon combustion..

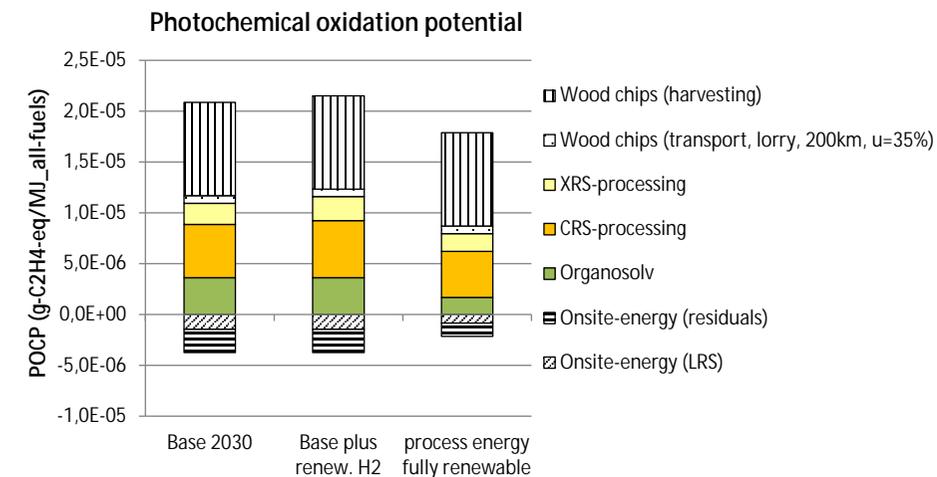
Source: Öko-Institut e.V.

Figure 2-6: Acidification, eutrophication and photochemical oxidation potentials for the MTHF concept



e) Acidification potential

f) Eutrophication potential



g) Photochemical oxidation potential

Source: Öko-Institut e.V.

Table 2-7: Lifecycle impact of the MTHF concept

Impact category \ Result	Base	Base plus re- new. H2	process energy fully renewable
GWP forest residues, soil-C	17	17	17
GWP biomass harvest & transport	9	9	9
GWP TMFB processes, net	73 (64)*	52	14
GWP overall**	99 (90)*	78	40
CED n.r. biomass harvest & transport	0,1	0,1	0,1
CED n.r. TMFB processes, net	1,3	0,9	0,2
CED n.r. overall	1,4	1,0	0,3
CED r. biomass harvest & transport	2,5	2,5	2,5
CED r. TMFB processes, net	0,2	0,7	1,2
CED r. overall	2,6	3,1	3,6
EP biomass harvest & transport	7,4E-06	7,4E-06	7,4E-06
EP TMFB processes, net	2,8E-05	4,1E-05	3,4E-05
EP overall	3,5E-05	4,8E-05	4,1E-05
AP biomass harvest & transport	2,9E-05	2,9E-05	2,9E-05
AP TMFB processes, net	9,3E-05	1,2E-04	9,7E-05
AP overall	1,2E-04	1,5E-04	1,3E-04
POCP biomass harvest & transport	9,9E-06	9,9E-06	9,9E-06
POCP TMFB processes, net	7,2E-06	7,8E-06	5,8E-06
POCP overall	1,7E-05	1,8E-05	1,6E-05

* The values in parentheses represent a sensitivity analysis concerning a potentially higher energy content of the char stream (see Chapters 2.2.2 and 2.2.3). Given that it reduces external energy requirements, it affects all impact categories but for sake of clarity is shown only indicatively for GWP.

** GWP including the CO₂ emissions resulting upon combustion.

Source: Öko-Institut e.V.

2.2.4.2. Changes for renewable hydrogen and a completely renewable process energy supply

[The upstream impact of the biomass supply remains unchanged in all cases.]

The impacts of renewable and natural-gas based hydrogen are shown in Table 2-8 in comparison. Based on these datasets GWP and CED n.r. are considerably reduced for renewable electrolytic hydrogen, but acidification and especially eutrophication potentials are increased. POCP is similar.

Table 2-8: Comparison of impact of renewable and natural-gas based hydrogen

Hydrogen source		GWP g CO ₂ eq/ kg-H ₂	CED n.r. MJ/ kg-H ₂	CED r. MJ/ kg-H ₂	EP kg PO ₄ eq/ kg-H ₂	AP kg SO ₂ eq/ kg-H ₂	POCP kg C ₂ H ₄ eq/ kg-H ₂
Renewable (70% efficiency; 25% PV, 75% wind)	own estimate	2,4E+03	3,0E+01	1,9E+02	5,7E-03	1,5E-02	9,4E-04
Steam reforming of natural gas	GaBi 6	1,1E+04	1,9E+02	3,0E-01	5,9E-04	4,5E-03	6,9E-04
ratio renewable/fossil-based		23%	16%	62991%	970%	337%	137%

Source: Öko-Institut e.V.

Also for heat if a direct use of renewable electricity is assumed the impact of GWP and CED n.r. is reduced considerably compared to the utilisation of natural gas but acidification and eutrophication potentials increase, by factors of 1.2 and 3, respectively. POCP again remains similar. With respect to electricity the impact of the fully renewable mix (25 % PV, 75 % wind energy) shows considerably lower values than the “Base 2030”-mix in all categories: GWP and CED n.r. over 85 % reduction, for AP, EP and POCP around 70 % reduction. CED r. is of course increased. It has to be kept in mind that the “Base 2030”-mix already has a share of almost 70 % renewables in 2030 as it is derived from the ambitious climate protection scenario reaching -95 % GHG emissions in 2050 (OEKO/ISI 2015, KS 95; see Annex 2).

For the “Base 2030 plus renewable H₂”-case the change reflects in the XRS and CRS processing steps where the hydrogen is added. As a relatively high amount of hydrogen is needed for the hydrogenation of levulinic and itaconic acid (3 and 5 mol-H₂/mol-fuel for 2- and 3-MTHF, respectively) the GWP can be reduced compared to the base case by 29 % with respect to wood chip conversion processes only and by 21 % when also including the biomass upstream impact. Also for CED n.r. a reduction is reached while of course CED r. increases. The total CED stays approximately constant due to the relatively small contribution of hydrogen to the overall system demand dominated by woodchips input.

For a completely renewable process energy supply the GWP and CED n.r. of the woodchip conversion processes fall in the same order of magnitude as the upstream biomass impact. CED r. concomitantly increases so that the total CED remains essentially constant. This is remarkable because in general renewable electricity generation is linked to a smaller total CED due to decreased conversion losses (and system boundary definition starting after solar/wind energy is first captured). However, in the present LCA not only direct electricity is substituted by renewable generation but also heat and hydrogen where the total CED remains approx. constant (heat) or is even increased (hydrogen) based on the datasets used.

EP and AP increase for electrolytic hydrogen supply; for all renewable process energy supply the impact sinks again especially in OS fractionation (high electricity demand) but still remains higher in CRS and XRS processing (H₂ and heat demand). The net value for EP thus remains higher than in the base case. For AP it falls back to the value of the base case.

The POCP of the TMFB conversion processes slightly increases for electrolytic hydrogen supply, while for all renewable process energy supply it is reduced. As over half of the POCP is due to the biomass upstream impact the overall value remains about constant.

2.2.4.3. Discussion of upstream impact of renewable energy generation

The reason for the high contribution of renewable energy generation especially in EP and AP is the actual prechain of photovoltaics production. It still contains a high share of coal energy which contributes mainly through mining-related (but also combustion) processes. Another important impact is related to copper requirement which is mined from sulfidic ores. Also the impact of platinum group metal production is among the single highest impacts of the complex PV production.

- Ø For EP the treatment of sulfidic tailings contributes more than 40 % of the total impact of PV-based energy generation. Lignite and hard coal mining together another > 30 %.
- Ø For AP of PV not only the panel (however, with highest impact 37 %), but also electric installation (copper) with 28 % of impact, inverter (aluminium and electricity use in Asia, 21 %) and aluminium-based mounting system (14 %) contribute; within the impact of panel production the silica wafer production is the dominant source.
- Ø Due to the pre-chain of PV production also GWP remains comparably high for a renewable energy process (25 % that of natural gas-based electricity generation by combined-cycle technology), the main impact being the energy-intensive production of the panel/silica wafers (panel > 70 % contribution to GWP, followed by mounting system 16 %, inverter 8 % and electric installation 3 %).
- Ø For the considered wind energy converters (with gear technology) the impact per electricity output is about 5 times lower than for PV in GWP, EP and AP categories. If gear-less wind energy converters are regarded the picture may differ, e.g. if the technology is based on copper coils, the contribution to AP and EP could be higher also depending on recycling rate of the copper used.

The impact of PV is based on the ecoinvent 3.4 database. The lifecycle assessment, also regarding other conformations (e.g. roof installation) and cell chemistries, was carried out by Jungbluth et al. 2012.

Concluding, the upstream impact of renewable energy generation today is still partly based on the use of fossil energy in production. Besides, the mining of technology metals leads to impacts in the categories regarded in this study. In addition, the required space needed for the installation of renewable energy generation plants entails constraints. Thus, even if the transition to renewable energy generation is mandatory from a climate protection perspective, renewable energy is a limited resource and efforts need to be continuously directed at reducing the energy requirement of products and services in order to respect sustainable limits.

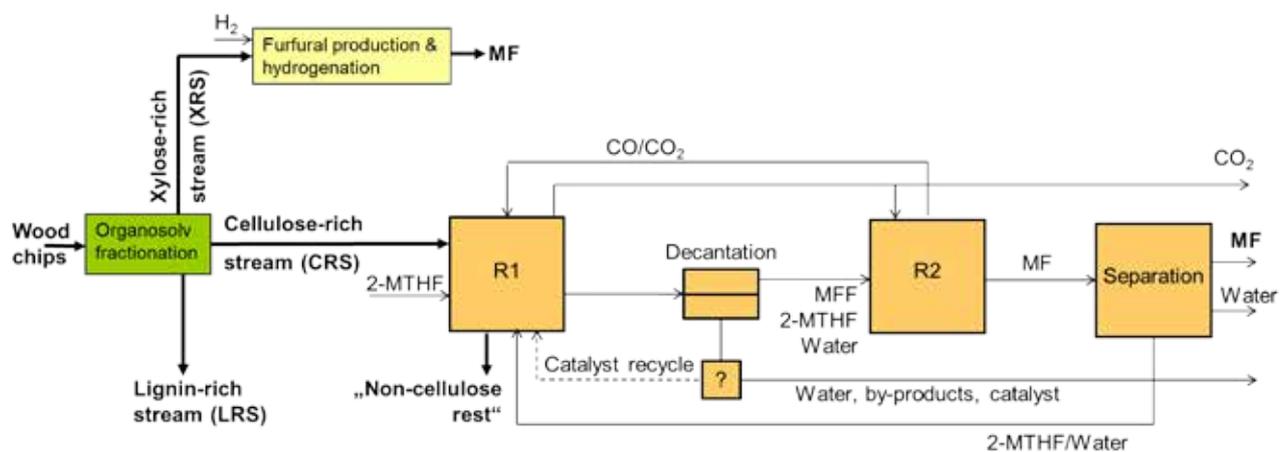
2.3. Lifecycle assessment of the MF concept

This chapter first gives a description of the specific set-up of the MF concept (Chapter 2.3.1) followed by information on the specific data sources for the concept (Chapter 2.3.2, for description of general sources and the Organosolv fractionation please refer to Chapter 2.1.3). Then the assessment results of the MF concept are presented in the form of carbon and energy flows through the processes (Chapter 2.3.3) and lifecycle assessment results (Chapter 2.3.4).

2.3.1. Process description

Based on the Organosolv fractionation (see Chapter 2.1.3) the MF concept converts the xylose- and the cellulose-rich streams into 2-MF.

Figure 2-7: MF concept, detail of the cellulose conversion to MF



Source: Öko-Institut e.V., CRS conversion to MF taken from Redepenning et al. 2015

MF synthesis from cellulose proceeds via two consecutive catalysed reactions. In the first reaction (R1 in Figure 2-7) cellulose is converted to hydroxymethylfurfural (HMF) which reacts with carbon monoxide to form 2-methylfurfural (MFF) and CO₂ (Table 2-9). The yield of this reaction is about 60 % on a molar basis because of by-product formation, probably humins. Secondly, MFF is converted to MF by abstraction of CO assuming full conversion. The CO is recycled to the first reactor and consumed stoichiometrically in the first reaction. Hence, no external carbon or hydrogen source is necessary and overall only 1 mol C per mol MF produced is lost.

Table 2-9: Reaction equation and parameters for the MF concept

	Reaction 1 (R1)	Reaction 2 (R2)
Equation	a) Cellulose => HMF + 2 H ₂ O b) HMF + CO => MFF + CO ₂ c) HMF => byproducts	MFF => 2-MF + CO
Catalyst	PGM complex	PGM complex
Yield (mol/mol)	a) 100%, b) 60 % c) 40%	close to 100 %
Temperature (°C)	180	200
Pressure (bar)	100	200

Source: Redepenning & Viell 2015

The described reactions have been realised at RWTH Aachen by ITMC on lab-scale with pure educts in pure CO or CO₂ atmospheres and reaction volumes below 10 mL. For the whole production process a concept has been elaborated in the TMFB cluster by AVT.SVT which includes the necessary separation steps (see Figure 2-7, Redepenning & Viell 2015).

The **XRS conversion to MF** is currently not in the focus of research at TMFB so that no process simulation was carried out for this part. For the present study it is hence considered on a very rudimentary basis only assuming that MF can be obtained directly from furfural via hydrogenation. According to cluster work, however, MF production from C5 sugars proceeds in a two-step synthesis via furfuryl alcohol (König 2018). It is hence suggested to rather see the assessment of the 2-MF production from the XRS as an estimate for furfural production only, with furfural potentially serving as a platform chemical for further uses. For future assessment of 2-MF production from C5 sugars the correct pathway should be taken into account.

The side streams, beside the LRS, considered to be sent to the CHP-unit are

- ∅ the non-cellulose components in the CRS (mainly lignin)⁴
- ∅ the by-products from MFF-synthesis
- ∅ the non-xylose components in the XRS (lignin, glucose)
- ∅ the by-products from furfural synthesis

Moreover, an essentially pure CO₂ stream is emitted from the first reaction.

2.3.2. Specific data

In order to evaluate the potential process performance of **MF synthesis from cellulose** conceptual process design and process simulations in ASPEN have been carried out in the TMFB cluster. They are based on data from the lab-scale experiments in ideal environments and on estimated property data. For process energy demand they include necessary separation steps and then rely on minimum energy demand calculations disregarding costs. Moreover, the values for heat demand used for the LCA take heat integration into account.

It has to be noted that the simulations are based on pure cellulose as an input. The real CRS from Organosolv fractionation also includes side components, mainly lignin, which may interfere with the reaction. Moreover, at the present early stage of development a mandatory catalyst recycle could not yet be included and product losses of HMF, MFF and MF are neglected. Thus the results of the assessment represent an optimum bound for the investigated process. Real energy requirements will be higher (Redepenning & Viell 2015).

⁴ If real CRS is fed to the first reaction of MF synthesis (reaction of cellulose to MFF via HMF) the non-cellulose components may end up as a solid residue in a similar manner as for the Biofine process where the “tar” can then be separated and used for energy generation. However, part of them may undergo different reactions the products of which end up in the liquid phase and subsequently need to be discarded to the wastewater (along with reaction by-products). This entails additional separation effort and any energetic use of these by-products depends on their concentration in the wastewater. Based on current modelling it is assumed that water evaporation requires only a minor share of the energy contained in the by-products from the first reaction.

The simulation results for the energy requirement are summarized in Table 2-10.

Table 2-10: Energy requirement for MF synthesis from cellulose

Input	MJ/MJ _{MF}	Comment
Electricity	0.24	mainly for the compression of the gas recycle from R2 to R1 and the compression of the recycle of the bottom product from the separation section to R2
Heat	0.38	at 200°C, after heat integration
Cooling	1.24	between 100°C to ambient temperature; not considered in the present assessment, might be used to dry the incoming CRS which is also neglected in the present assessment

Source: Redepenning & Viell 2015

Moreover, 2-MTHF is required as an auxiliary material in the process. Its impact is assessed with the values calculated by the LCA of the MTHF concept (see Chapter 2.2.4).

For the estimate of the environmental impact of the catalysts the same turn-over number (TON) as for the catalyst of 2-MTHF synthesis is assumed ($\text{TON } 10^5 \text{ mol}_{\text{educt}}/\text{mol}_{\text{catalyst}}$) which is necessary to make the process industrially attractive. The net metal consumption for each PGM-catalyst is calculated based on the assumption that at the end of the catalyst life the platinum group metals are recycled at a rate of 98 % which is likely based on their high value and good recyclability (Hagelücken et al. 2005). As for MTHF synthesis it is assessed with a proxy for PGM primary production calculated as a mix of 50% platinum and 50% palladium based onecoinvent 3.4 datasets. No other components of the catalyst complexes are considered for their environmental impact in this screening assessment.

For conversion of the **C5 sugars in the XRS to furfural** the yield as well as steam demand are taken into account. For the yield a higher-end value of $0.5 \text{ kg}_{\text{furfural}}/\text{kg}_{\text{xylose}}$ is used as it is obtained with modern processes (e.g. Biofine, Suprayield). Steam requirement is set at $10 \text{ t}_{\text{steam}}/\text{t}_{\text{furfural}}$ which, compared to current commercial processes, is a rather low value realised by the Suprayield concept (Arnold & Buzzard 2003). This concept uses steam for keeping the reaction medium boiling instead of using it for furfural stripping. Furfural rapidly separates from the boiling medium whereas other components stay in the liquid phase. Thus, furfural yield is significantly increased by avoiding condensation and resinification reactions and a purer product stream is obtained reducing downstream purification requirements (Arnold & Buzzard 2003). The sulfuric acid requirement is estimated based on data given in Huber et al. (2006) and Hoydonckx et al. (2007). No further energy or material requirements are taken into account. For an estimate of the subsequent theoretical direct hydrogenation to 2-MF only stoichiometric hydrogen demand is considered.

With respect to the combustion of side streams the following assumptions are made:

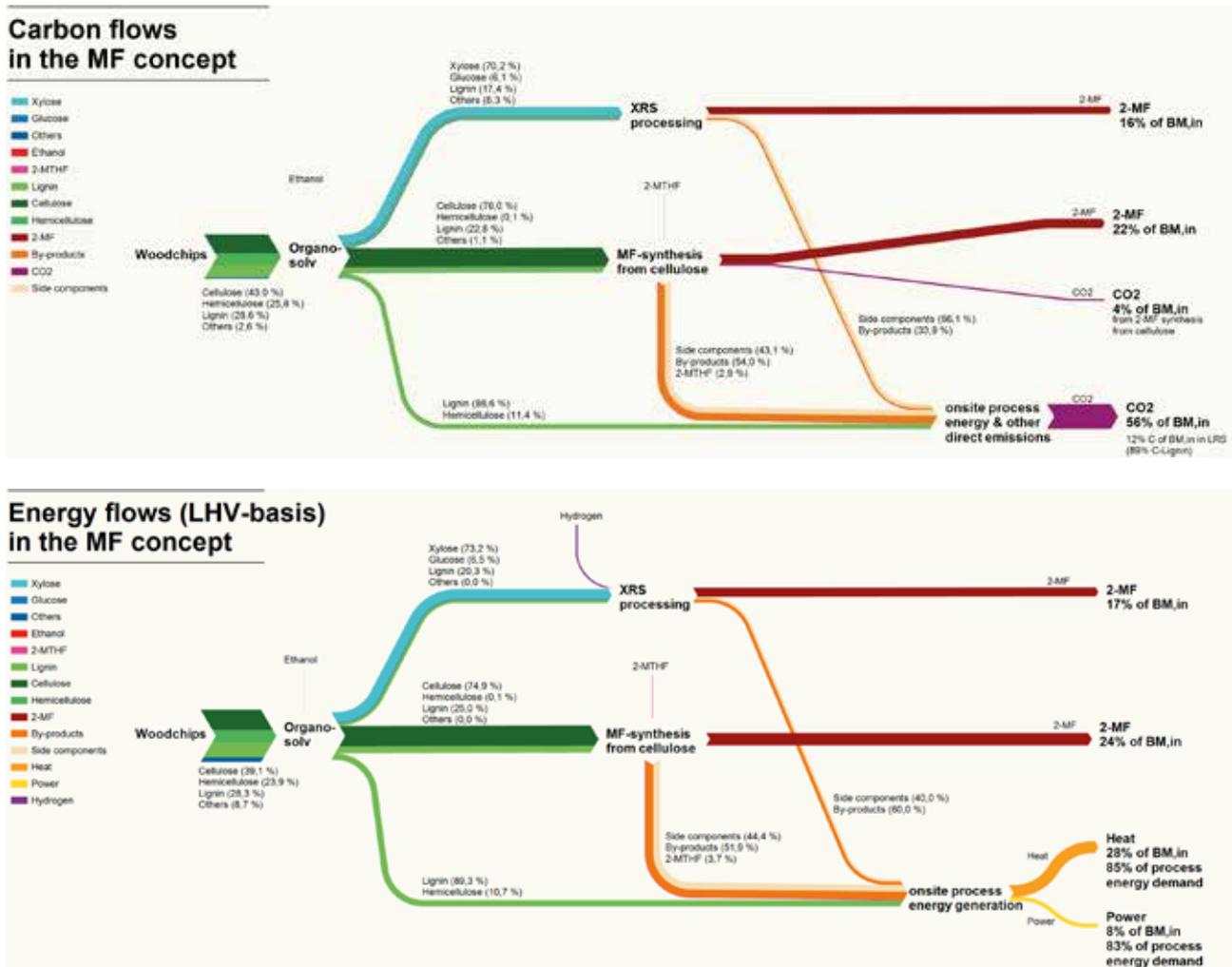
- Ø As the simulation of MF synthesis from cellulose was carried out for pure cellulose, no information is available on the effect and fate of the **non-cellulose components in the CRS**. It is likely, however, that they interfere in some ways, e.g. concerning the reaction yield or entailing additional separation effort. Moreover, their structure may be altered through the process conditions. To still allow for an estimate of their potential contribution to on-site process energy generation the following approach was chosen: the energy content of the non-cellulose components was calculated based on their original lower heating value and then corrected by a factor of 0.5 in order to account for the aspects described above. No residual water content (and corresponding energy loss) was taken into account.
- Ø The energy content of the **by-products from MFF synthesis** was estimated with a lower heating value of 19.5 MJ/kg, based on a carbon content of 55 % and the fact that they are possibly humins which goes together with this C-content. The energy required to evaporate the water contained in the waste stream was subtracted.
- Ø The **by-products from furfural synthesis** make up approx. 20 % by mass as determined via mass balance. From this a carbon content of 40 % was determined via carbon balance. Based thereon a lower heating value of 15 MJ/kg was assumed, comparable to sugars with the same carbon content. Moreover, a correction factor of 0.5 was assumed in the same way as for the non-cellulose rest for the same reasons as described there.
- Ø For the **non-xylose components in the XRS** the same approach as for the non-cellulose components in the CRS was taken for the same reasons. This means that a correction factor of 0.5 was applied to their original LHV. No residual water content (and corresponding energy loss) was taken into account. Anyway, according to the simulation by Viell et al. 2013 the XRS from Organsolv fractionation is dry.

In order to test the effect of the assumed LHV-correction factor of 0.5 a sensitivity analysis was carried out where it was set 1. The effect is discussed in the results sections (Chapters 2.3.3 and 2.3.4).

2.3.3. Carbon and energy flow analyses

Based on the process set up and material and energy flow modelling described in Chapters 2.3.1 and 2.3.2 the carbon and energy flows through the MF concept are calculated (Figure 2-8). The energy flows are indicated based on their lower heating value with respect to dry mass (for onsite process energy generation water content is subtracted).

Figure 2-8: Carbon and LHV-based energy flows in the MF concept



Note: in „Energy flows“ only biomass based energy flows and hydrogen input are shown. The MF concept moreover requires an external input of heat and electricity (see Table 2-11).

Source: Öko-Institut e.V.

Almost 40 % of the carbon is converted to fuel, 4 % are emitted as an essentially pure stream of CO₂ from the reactions occurring from cellulose conversion during CRS processing (see Figure 2-7 and Table 2-9). This pure CO₂ stream might be attractive to be reduced with hydrogen in order to increase the carbon efficiency or it might be sold; these options will have to be taken into account for future assessments. The remaining 56 % are converted to CO₂ in CHP generation or other.

The CRS represents the biggest fraction from the Organosolv step resulting in a transfer of 22 % of the carbon to MF. The XRS is smaller, but carbon efficiency to furfural is higher so that 16 % of the carbon is transferred to MF via this path. It has to be kept in mind, however, that this assessment assumes a direct conversion of furfural to MF based on stoichiometric hydrogenation. The real pathways would probably follow an indirect route via furfuryl alcohol (see Chapter 2.3.1).

The energy efficiency for MF with respect to biomass input is 42 % equalling a biomass input of 2.40 MJ/MJ_{fuel}. The hydrogen input in this case is rather moderate so that even if it is considered in the calculation an overall energy efficiency of 40 % is obtained.

Around 85 % of the energy demand can be satisfied by the combustion of the side streams. The remaining external energy requirement is shown in Table 2-11.

Table 2-11: External energy demand of the MF concept

	MJ/MJ _{fuel}	Comment
Electricity	0.04	
Heat	0.10	distribution losses neglected
Hydrogen	0.08	0.12 MJ _{el} /MJ _{fuel} for electrolytic H ₂

Source: Öko-Institut e.V.

If the correction factor for the energy content of the non-cellulose components in the CRS as well as the non-xylose components and by-products in the XRS (see Chapter 2.3.2) is set “1”, the energy input to the CHP unit is sufficient to produce all process energy onsite. The MF concept becomes self-sustaining and may even produce some excess electricity which would have to be considered as an additional valuable output. However, there are still many uncertainties related to how the processing will behave when real substrate from OS fractionation is used. Moreover, the simulations for pure cellulose processing (Redepenning et al. 2015) already represent an optimistic estimate for the energy requirement of MF production. Also the MF production from furfural was assessed on a rudimentary basis only (see Chapter 2.3.1). As a consequence more research is still needed in order to estimate the net energy requirement under real conditions and further discussion is based on the results including a LHV correction factor of 0.5.

Approximately 12 % of the carbon and LHV in the biomass are incorporated into the LRS, about 90 % of it lignin. A slightly larger share of the lignin in the biomass is transferred into the CRS and needs to be separated in a way during processing (effort not yet included in the assessment). Also the XRS contains some residual lignin. In the base case all residual streams are assumed to be combusted for onsite energy generation. In a sensitivity analysis the material utilization of the LRS as a phenol substitute is regarded (see Chapter 2.7).

The carbon and LHV content of auxiliary materials is comparably negligible.

The carbon and energy efficiencies are summarized in Table 2-12.

Table 2-12: Carbon and energy efficiencies of the MF concept

	Carbon	Energy	Comment
in CRS fuel	21%	24%	
in XRS fuel	16%	18%	
overall	38%	42%	2.40 MJ _{biomass} /MJ _{fuel}
overall ext. H ₂		40%	external hydrogen demand included as energy input

Source: Öko-Institut e.V.

2.3.4. LCA results

Following the quantification of the material and energy flows for the entire production process and the corresponding auxiliary material and energy demand these are evaluated concerning their lifecycle environmental impacts. The results are shown in Figure 2-10 and Figure 2-11. The corresponding values are listed in Table 2-13.

2.3.4.1. Main observations for the base case Base 2030 (see Table 2-2)

For the base case Figure 2-9 additionally shows the relative contribution of the biomass upstream impact and the impacts related to the TMFB conversion processes differentiating their origin by heat, electricity and auxiliary material input. While with respect to the gross values, the dominant impact is from process energy demand, its contribution is at maximum 35 % when onsite energy recovery is considered. For CED r. the main contribution is again the energy content of the biomass itself, while for POCP the biomass upstream plays the major role (from machinery used in harvesting).

For GWP the contribution of biomass upstream reaches about one third of the net impact when soil carbon loss caused by the removal of residual wood from the forest is taken into account according to Globiom 2015. This aspect is discussed along with other considerations on biomass availability and impact in Chapter 2.6. At a transport distance of 200 km, harvesting and transport both contribute similarly to GWP, and together account for approx. 20 % of the net impact. The GWP of auxiliary materials is mainly caused by external hydrogen supply (70 %) even if in the MF concept hydrogen supply is low compared with the MTHF concept.

In the base case the net GWP is thus considerably lower than that of fossil gasoline (incl. combustion assuming the same efficiency), with the impact of the woodchip conversion processes and the upstream impact of biomass in the same range: 20 g.CO₂eq/MJ_{fuel} and 27 g.CO₂eq/MJ_{fuel} (incl. emissions due to soil carbon loss as calculated by Globiom 2015), respectively.

Also the non-renewable CED n.r. is considerably lower, while the renewable CED r. essentially represents the energy contained in the biomass: 2.4 MJ/MJ_{fuel} in this concept. The total CED thus amounts to approx. 3 MJ/MJ_{fuel} (see Figure 2-5 a)-d)).

With respect to the conversion steps the non-renewable CED n.r. shows the same pattern as the GWP: the main gross impact is due to OS fractionation and XRS processing, CRS processing shows only a very small contribution. The main contributor to the impact of OS fractionation is electricity followed by heat demand. For the impact of XRS processing steam demand for furfural synthesis has the highest contribution by far, the remaining is due to hydrogen requirement. For CRS processing about half of the impact is due to process energy requirement. The other half is caused by MTHF make-up requirement which is used as a solvent. Here the values shown for MTHF in Chapter 2.2.4 were used to estimate the impact.

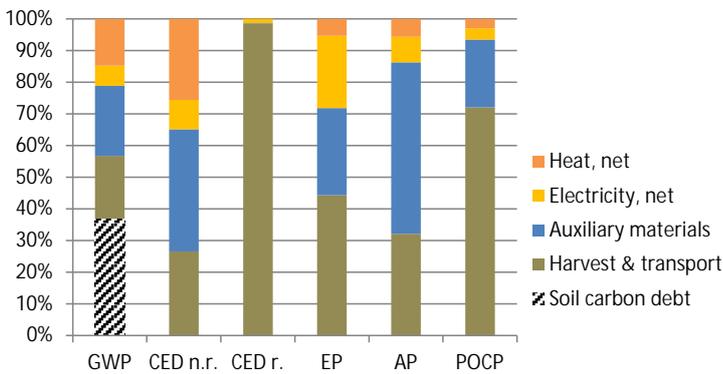
As mentioned in Chapters 2.3.1 and 2.3.2 the energy available for CHP generation from the CRS and XRS side streams may be underestimated by the possibly conservative LHV correction factor of 50 %. When this correction factor was set 1 the MF concept becomes self-sustaining and some excess electricity might even be exported (see Chapter 2.3.3). This results in a very low net GWP of the conversion processes as indicated in Table 2-13 (without considering excess electricity as a further valuable product).

While a promising performance for GWP and CED was found for the MF concept, mainly due to the very low impact of CRS processing (cellulose conversion to MF by simulation, see Chap-

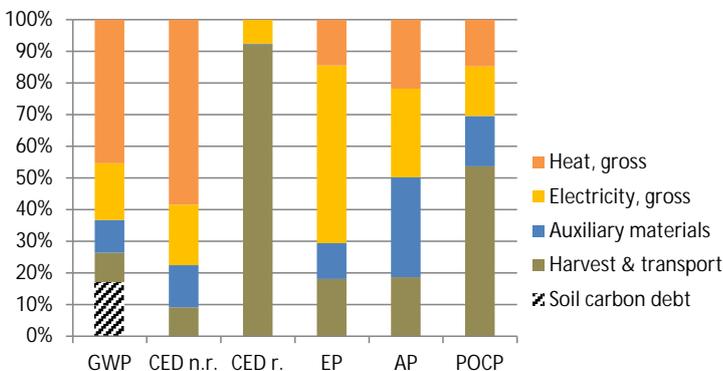
ter 2.3.2), investigation on toxicity aspects of various biofuels within the TMFB Cluster revealed a comparably high toxic potential of this candidate indicating that 2-MF may not be eligible as a non-hazardous biofuel (Heger et al. 2018, Blum et al., submitted).

Concerning AP, EP and POCP the impact of CRS processing is more prominent than for GWP and CED but still the Organosolv step (especially for EP) and XRS processing have higher shares. For all three categories electricity is the main driver in the OS fractionation step. For XRS processing the most relevant contribution comes from heat followed by sulfuric acid used as a catalyst in furfural synthesis. AP and POCP of the CRS processing are dominated by the impact of the net metal demand for the PGM catalysts, while for the EP additionally heat and 2-MTHF requirement play a role.

Figure 2-9: Contribution analysis for the different impact categories, Base 2030



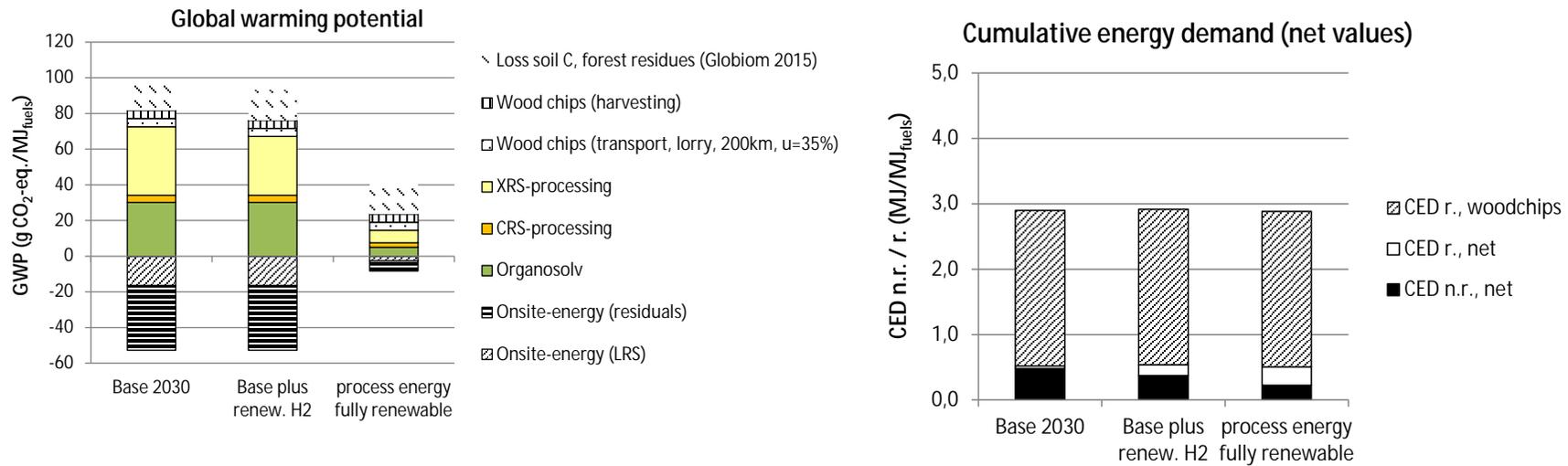
a) Contribution with respect to the **net impact**, i.e. considering external process energy demand only (incl. energy recovery by CHP)



b) Contribution with respect to the **gross impact**, referring to total process energy demand (excl. energy recovery by CHP)

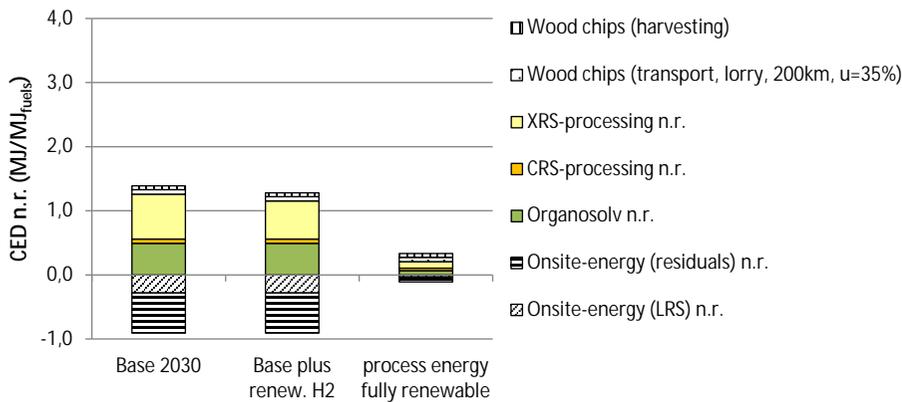
Source: Öko-Institut e.V.

Figure 2-10: Global warming potential (GWP) and cumulative energy demand (CED) for the MF concept



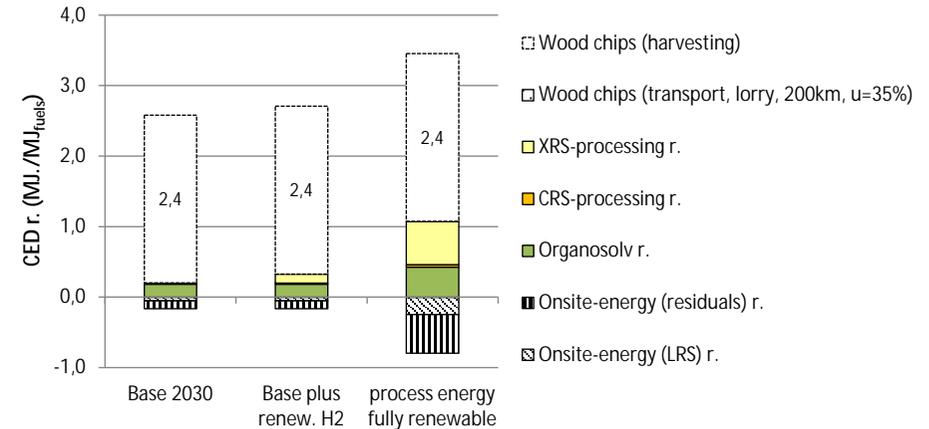
a) Global warming potential*

Cumulative energy demand, non-renewable



b) Total cumulative energy demand, net values

Cumulative energy demand, renewable



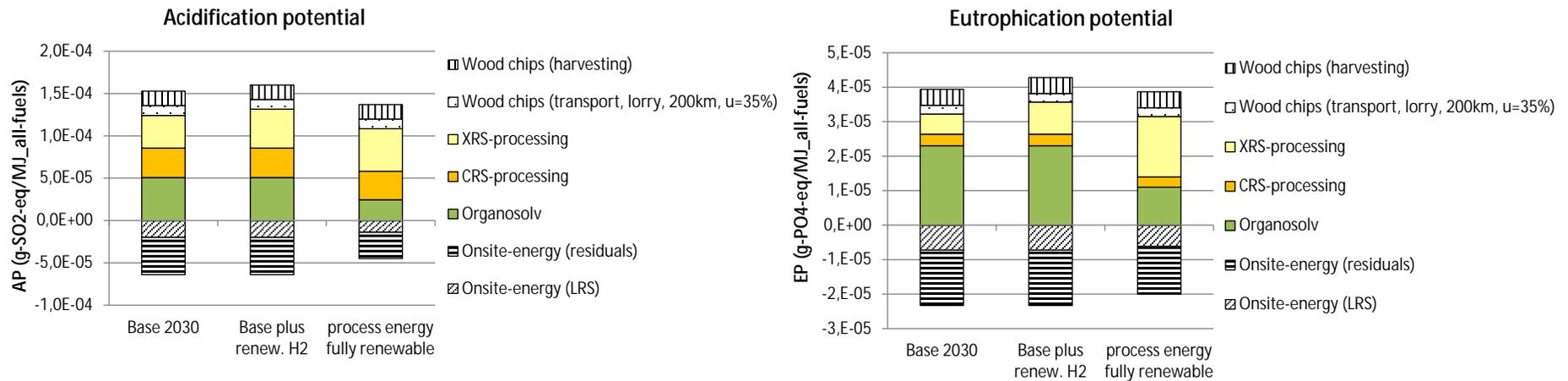
c) Non-renewable cumulative energy demand

d) Renewable cumulative energy demand

* GWP including the CO₂ emissions resulting upon combustion

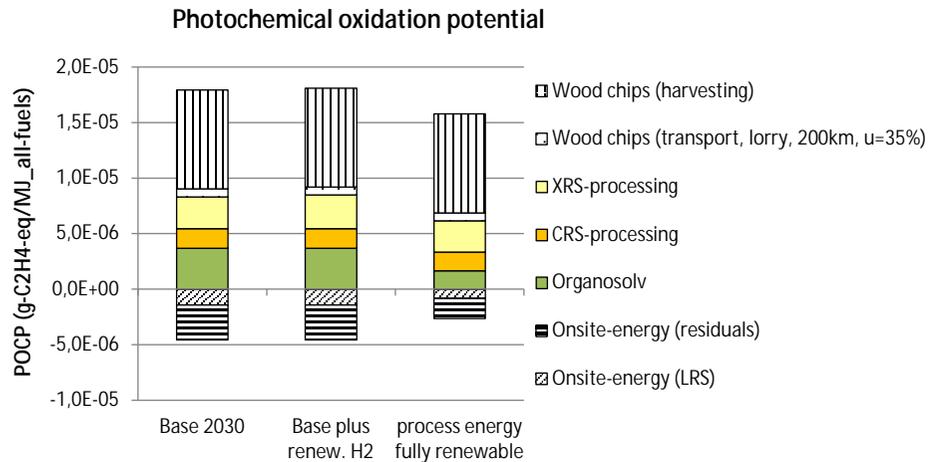
Source: Öko-Institut e.V.

Figure 2-11: Acidification, eutrophication and photochemical oxidation potentials for the MF concept



e) Acidification potential

f) Eutrophication potential



g) Photochemical oxidation potential

Source: Öko-Institut e.V.

Table 2-13: Lifecycle impact of the MF concept

Impact category \ Result	Base	Base plus renew. H ₂	process energy fully renewable
GWP forest residues, soil-C	17	17	17
GWP biomass harvest & transport	9	9	9
GWP TMFB processes, net	20 (4)*	14	6
GWP overall**	46 (30)*	40	32
CED n.r. biomass harvest & transport	0,1	0,1	0,1
CED n.r. TMFB processes, net	0,4	0,2	0,1
CED n.r. overall	0,5	0,4	0,2
CED r. biomass harvest & transport	2,4	2,4	2,4
CED r. TMFB processes, net	0,0	0,2	0,3
CED r. overall	2,4	2,5	2,7
EP biomass harvest & transport	7,2E-06	7,1E-06	7,1E-06
EP TMFB processes, net	9,0E-06	1,2E-05	1,2E-05
EP overall	1,6E-05	2,0E-05	1,9E-05
AP biomass harvest & transport	2,9E-05	2,9E-05	2,9E-05
AP TMFB processes, net	6,0E-05	6,8E-05	6,4E-05
AP overall	8,9E-05	9,6E-05	9,2E-05
POCP biomass harvest & transport	9,6E-06	9,6E-06	9,6E-06
POCP TMFB processes, net	3,7E-06	3,9E-06	3,5E-06
POCP overall	1,3E-05	1,4E-05	1,3E-05

* The values in parentheses represent a sensitivity analysis concerning a potentially higher energy content of the non-cellulose side components of the CRS and the non-xylose side components and by-products of the XRS sent to the CHP unit (see Chapter 2.3.2). Given that it reduces external energy requirements, it affects all impact categories but for sake of clarity is shown only indicatively for GWP. In this extreme case even some electricity might be exported and lead to even lower values. However, this was not considered in this study and real possibilities would have to be explored first.

** GWP including the CO₂ emissions resulting upon combustion

Source: Öko-Institut e.V.

2.3.4.2. Changes for renewable hydrogen and a completely renewable process energy supply

[The upstream impact of the biomass supply remains unchanged in all cases.]

As the renewable energy cases (see Table 2-2) are the same for all concepts the effects go in the same direction in all three concepts (see also discussion for the MTHF concept, Chapter 2.2.4.2). The degree varies with the specific characteristics of each concept.

In general, while utilizing renewable electricity to substitute the Base-2030 mix leads to reduced impacts in all categories except CED r., its utilization to substitute natural gas-based hydrogen production and heat decreases the impacts of GWP and CED n.r., but increases the impacts for EP and AP (by factors of 9 and 3, and 3 and 1.2, respectively; see Chapter 2.2.4.2, Table 2-8). The impact of POCP stays similar.

In the MF concept the change in the “Base 2030 plus renewable H₂”-case reflects only in XRS processing. The overall net GWP is reduced by 12 %, with respect to the conversion processes only the reduction is of 28 %. CED r. increase slightly more than CED n.r. decreases, but as the quantity of hydrogen is rather small, the overall CED remains essentially constant.

The overall net EP and AP increase by 21 % and 8 %, respectively (with respect to the conversion processes only 38 % and 12 %) and POCP stays approximately constant.

If the CO₂ stream from the first reaction (see Figure 2-7) was reduced to a paraffinic fuel this would increase the carbon efficiency from 38 % to 42 % and additionally require 0.16 MJ_{H₂}/MJ_{MF} on a stoichiometric basis, an increase of external H₂-input by a factor of three (cf. Table 2-11). The amount of fuel produced would be 0.13 MJ_{-CH₂-}/MJ_{MF}. Referring to all fuels produced this equals a hydrogen requirement 0.21 MJ_{H₂}/MJ_{MF+-CH₂-} (+160%) and a biomass input of 2.1 MJ_{biomass}/MJ_{MF+-CH₂-} (-12 %). With respect to biomass and hydrogen input the energetic efficiency increases from 40 % to 43 %, when considering the electricity input required for hydrogen production, the energetic efficiency increases from 40 % to 41 % (MJ_{MF+-CH₂-}/MJ_{biomass+electricity}).

For a completely renewable process energy supply the net GWP and CED n.r. of the woodchip conversion processes fall in the same order of magnitude as the impact of biomass harvesting and transport. CED r. concomitantly increases so that the total CED remains essentially constant. This is remarkable because in general renewable electricity generation is linked to a smaller total CED due to decreased conversion losses (and system boundary definition starting after solar/wind energy is first captured). However, in the present LCA not only direct electricity is substituted by renewable generation but also heat and hydrogen where the total CED remains approx. constant (heat) or is even increased (hydrogen) based on the datasets used.

The impact of EP and AP for all renewable process energy decreases again especially due to OS fractionation (high electricity demand) whereas in XRS processing they are still increased (heat and H₂ demand). The overall net values remain higher than in the base case. The POCP is largely dominated by the biomass upstream impact. For the woodchip conversion processes it varies by less than +/- 10 %.

2.3.4.3. Discussion of upstream impact of renewable energy generation

For considerations on the upstream impact of renewable energy generation please refer to Chapter 2.2.4.3.

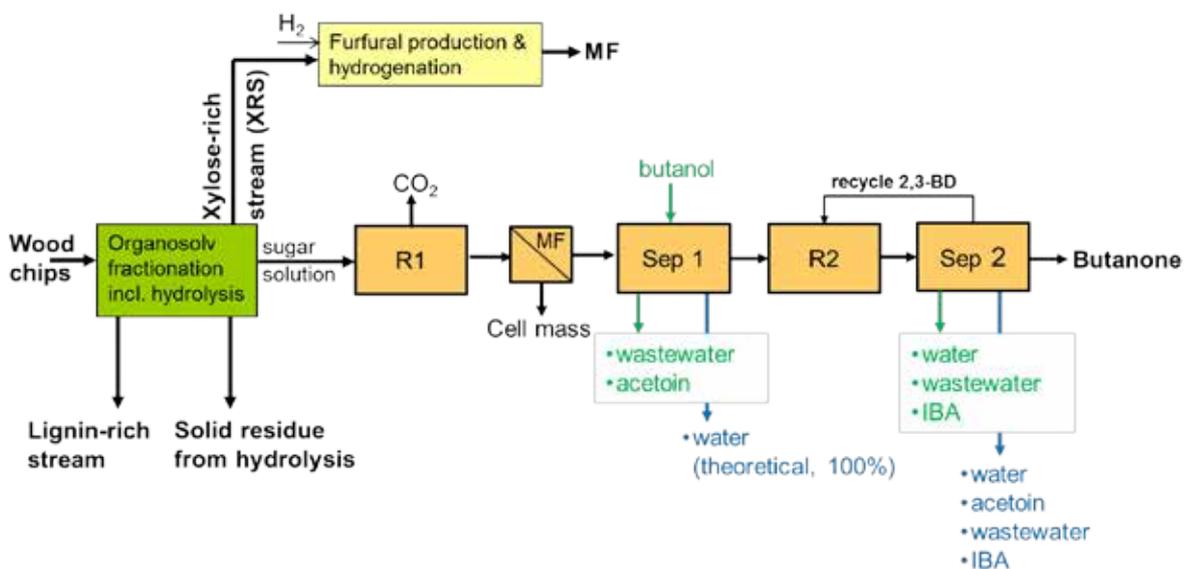
2.4. Lifecycle assessment of the butanone concept

This chapter first gives a description of the specific set-up of the Butanone concept (Chapter 2.4.1) followed by information on the specific data sources for the concept (Chapter 2.4.2, for description of general sources and the Organosolv fractionation please refer to Chapter 2.1.3). Then the assessment results of the Butanone concept are presented in the form of carbon and energy flows through the processes (Chapter 2.4.3) and lifecycle assessment results (Chapter 2.4.4).

2.4.1. Process description

Based on the Organosolv fractionation (see Chapter 2.1.3) the Butanone concept converts the xylose-rich stream into 2-MF and the cellulose-rich stream into butanone as depicted in Figure 2-12.

Figure 2-12: Butanone concept, glucose conversion



Glucose conversion to butanone taken from Penner et al. 2015 acc. to scenarios 3-2 & 4-2:

Green: 3-2	Sep 1:	Sep 2:
Blue: 4-2	3-2: RO, Extraction, Rect. 1, Rect. 2	3-2: 4 rectification steps
	4-2: theoretical separation of 100 % of water before R2 as „lower bound“	4-2: 5 rectification steps, one more for separation of acetoin (after R2)

Source: Öko-Institut e.V., glucose conversion to butanone taken from Penner et al. 2015

The modelling of the processing of the XRS to 2-MF is the same as for the MF concept. For details please refer to Chapters 2.3.1 and 2.3.2.

For the Butanone concept a hydrolysis of the CRS is considered within the Organosolv fractionation step in order to produce fermentable sugars (Viell et al. 2013, Chapter 2.1.3). For the conversion to butanone different process concepts were analysed in the TMFB cluster. Based on preliminary mass and energy balances the following settings were identified as the most promising routes (Penner et al. 2015). They both feature the same reactions but differ for the separation steps (Figure 2-12).

In the first reaction glucose is fermented to 2,3-butanediol which is subsequently converted chemocatalytically to butanone:

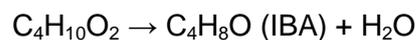
- **R1:** fermentation of 2,3-butanediol (2,3-BD) from glucose as documented in literature (Penner et al. 2015 based on parameters by Ji et al. 2011, Ma et al. 2009)



The equilibrium between acetoin and 2,3-BD is assumed to result in 6 % acetoin and 94 % 2,3-BD. In practice it can be regulated by oxygen supply control.

Depending on the biochemical reactions CO₂, H₂, ethanol, acetate and succinate can be formed as by-products (Ji et al. 2011). The simulations by Penner et al. (2015) consider the formation of CO₂ (2 mol_{CO2}/mol_{glucose,fermented}). No other by-products are considered.

- **R2:** chemo-catalytical reaction based on literature; the reaction produces butanone and its isomer IBA with a 90 % selectivity to butanone (Penner et al. 2015 based on parameters by Multer et al. 2012)



In both scenarios the fermentation is followed by microfiltration (MF) to remove cell mass.

The separation steps differ between the two scenarios:

- **Scenario 3-2** (green in Figure 2-12): The first separation step (Sep 1) is modelled as a real separation including reverse osmosis and extraction for concentration of the 2,3-BD output from fermentation followed by two rectification columns to regenerate the extraction agent and separate water (100 %; incl. some other components) and to separate the by-product acetoin. Butanol is employed as extracting agent. Pure 2,3-BD is processed to R2. The separation after R2 (Sep 2) consists of four rectification columns which purify the butanone product output and separate IBA as well as the water produced in R2. Moreover, non-reacted 2,3-BD is separated and recycled to R2.
- **Scenario 4-2** (blue in Figure 2-12): Based on the finding that the separation of the in this case low-boiling water by rectification is very energy consuming, Scenario 4-2 assumes an idealized separation of 2,3-BD and acetoin from the fermentation broth. After microfiltration the water is separated from the fermentation products without specifying a process, no product losses and energy requirements are taken into account. The strategy is to demonstrate the minimum bound of this process set-up and underline the necessity for research into alternative water separation concepts. It is hence a fictitious scenario. The water-free mixture of 2,3-BD and acetoin is processed to R2. The second separation step is similar to the separation in Scenario 3-2 but with one additional rectification column to separate the acetoin after R2.

Beside the LRS, the side streams which are considered to be sent to the CHP-unit in the Butanone concept are:

- ∅ the solid residue from CRS hydrolysis (residual cellulose, lignin)
- ∅ the cell mass separated after fermentation⁵
- ∅ the non-xylose components in the XRS (lignin, glucose) (same as for the MF concept)
- ∅ the by-products from furfural synthesis (same as for the MF concept)

2.4.2. Specific data

TMFB conceptual process design was carried out for the conversion of the CRS hydrolysate into butanone. The mass flows rely on ASPEN simulations based on parameters from literature and minimum energy requirement is estimated as documented in Penner et al. (2015). As a simplification modelling was carried out for only glucose as input. The hydrolysate from CRS contains > 94% glucose by mass on a dry basis, the main other components being unspecified “others” and residual cellulase (see Viell et al. 2013 [SI]). The assumption of pure glucose conversion hence seems justified, even if side components may have an influence on the performance of the microorganisms and may lead to increased separation efforts. Proof with real substrate is thus still mandatory. The conceptual process designs include the necessary separation steps and then rely on minimum energy demand calculations. The values for heat demand used in for the LCA take heat integration into account.

The simulation results for the energy requirement are summarized in Table 2-14.

Table 2-14: Energy requirement for butanone synthesis from glucose

Input	MJ/MJ _{Butanone}		Comment
	Sc. 3-2	Sc. 4-2	
Electricity	0.026	0.019	stirring of fermenter, reverse osmosis, reaction 2
Heat	0.36	0.14	at 200°C, after heat integration

Source: Penner et al. 2015

The energy requirement for stirring is based on the default value of 0.5 kW/m³ given by Hermann et al. (2007) for anaerobic fermentations. No further energy requirement for aeration is included because TMFB research suggests that power consumption at low oxygen demand is quite similar to the estimate for anaerobic processes (Kreyenschulte et al. 2016). Also electricity requirement for microfiltration was estimated to be small based on the simulated permeate quantities and the default value given by Hermann et al. (2007) and consequently neglected.

As auxiliary materials cellulase for the hydrolysis step and butanol as extracting agent for 2,3-butanediol are needed. Both are assessed based on ecoinvent 3.4. While the ecoinvent dataset for enzymes is based on a rather rough assessment, a detailedly documented LCA on European cel-

⁵ Another option for the cell mass would be its use as feed. Here the energetic use was prioritized due to the need for process energy, this alternative could still be considered in future work.

lulase enzyme production was recently published by Gilpin & Andrae (2017). Table 2-15 shows the results in comparison.

Table 2-15: LCA impact of cellulase

Source	Carbon source	GWP	CED, n.r	CED, r.	EP	AP	POCP
		g CO ₂ -eq/ kg-enzyme	MJ/ kg-enzyme	MJ/ kg-enzyme	kg PO ₄ -eq/ kg-enzyme	kg SO ₂ -eq/ kg-enzyme	kg-C ₂ H ₄ -eq/ kg-enzyme
Gilpin & Andrae 2017	A cornstarch glucose	10600	81*	*	0.044	0.049	0.0016
	B sugar cane molasses	9100	62*	*	0.025	0.055	0.0049
	C softwood	7900	52*	*	0.0087	0.032	0.0020
	A-C average	9200	65*	*	0.026	0.045	0.0028
ecoinvent 3.4	potato starch	6670	82	76	0.0312	0.0428	0,0013
G&A 2017 av./ecoinvent 3.4		1.4	0.8	---	0.8	1.1	2.1

* In Gilpin & Andrae 2017 only a value for “CED” is given. Based on the comparison with ecoinvent, especially concerning the GWP, it is assumed that “CED” represents CED n.r. .

Source: Öko-Institut e.V. based on Gilpin & Andrae (2017) and ecoinvent 3.4 (APOS, CML 2016)

The accordance of the two datasets is very good. The higher GWP found by Gilpin & Andrae (2017) may be explained by direct emissions in the fermentation phase for cellulase production. These might be due to emissions from nitrogen-based nutrients but this is, however, not mentioned explicitly. For consistency reasons the ecoinvent dataset was used for this LCA.

The catalyst requirement for the conversion of 2,3-butanediol to butanone (protonated form of ZSM-5) has not yet been simulated and is neglected for the present assessment.

With respect to the combustion of side streams the following assumptions are made:

- Ø The energy content of the **solid residue from hydrolysis** is calculated based on the composition given by Viell et al. 2013 [SI]. The energy required to evaporate the residual water content is subtracted.
- Ø The energy content of the **cell mass** from 2,3-butanediol fermentation is calculated assuming a carbon content of 43 % and a lower heating value of 18 MJ/kg. The energy required to evaporate the residual water content is subtracted based on assumptions concerning water in and outside the cells (70 % and 50 % referring to dry cell mass, respectively). Side components make up a small share.
- Ø **by-products from furfural synthesis**, see MF concept Chapter 2.3.2.
- Ø **non-xylose components in the XRS**, see MF concept Chapter 2.3.2.

In order to test the effect of the assumed LHV-correction factor of 0.5 a sensitivity analysis was carried out where it was set 1. The effect is discussed in the results sections (Chapters 2.4.3 and 2.4.4).

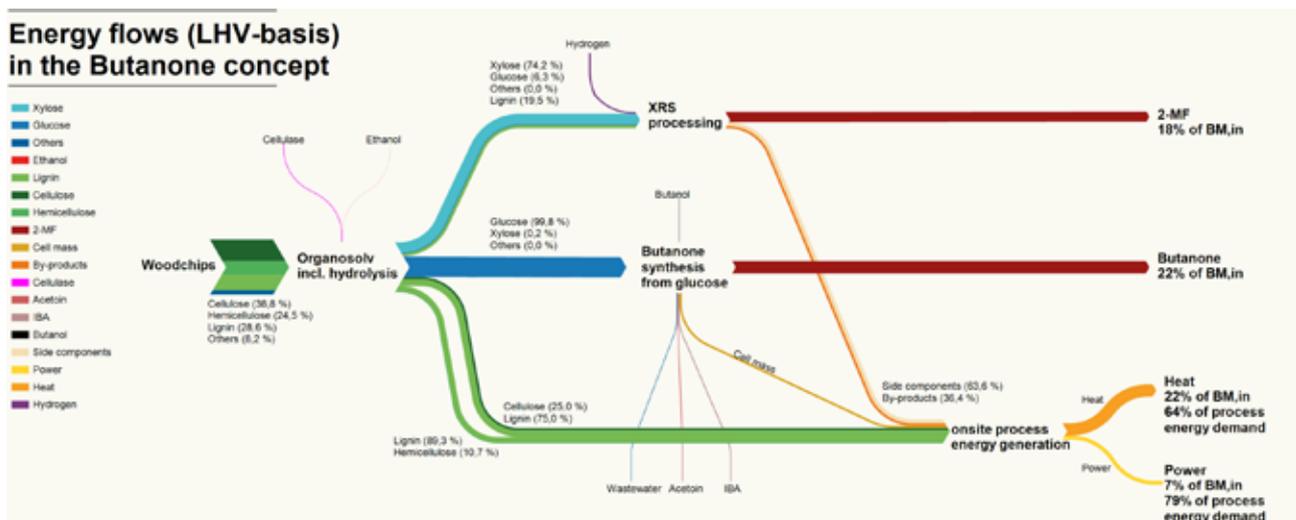
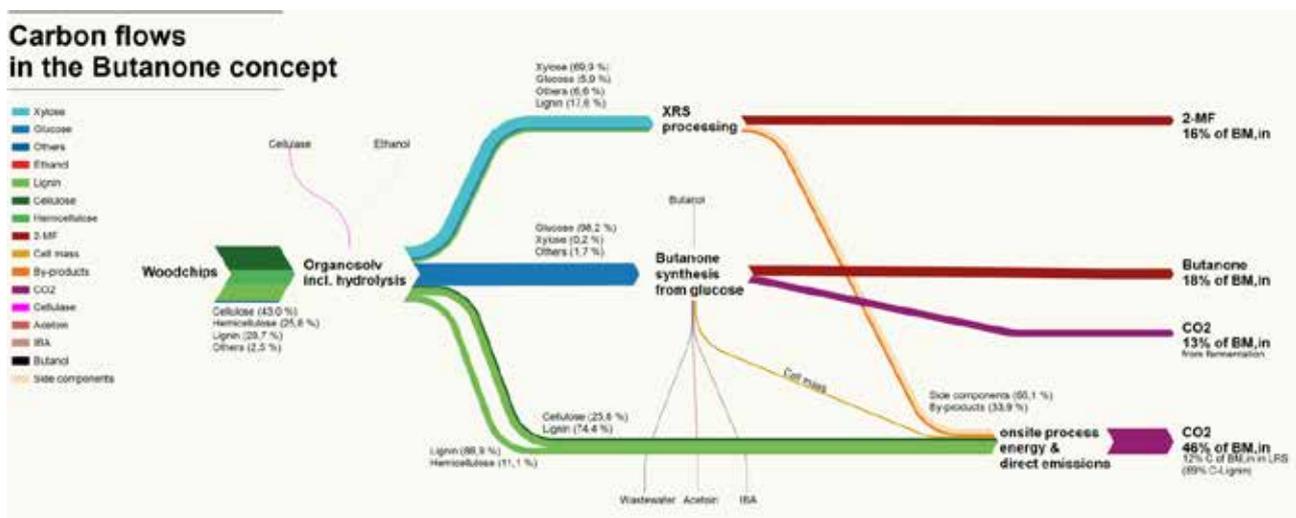
Butanone synthesis actually produces two more side streams (acetoin and IBA) which are pure according to the current simulation and might be marketed. These streams, representing 4 %

($MJ_{Acetoin}/MJ_{Butanone+MF}$) and 7 %⁶ ($MJ_{IBA}/MJ_{Butanone+MF}$), are not yet considered within this assessment in order to leave the focus to the desired large-quantity outputs of the process (butanone and MF fuels). Future more detailed work may take their potential benefits into account.

2.4.3. Carbon and energy flow analyses

Based on the process set up and material and energy flow modelling described in Chapters 2.4.1 and 2.4.2 the carbon and energy flows through the Butanone concept are calculated. In Figure 2-13 they are shown and discussed for Scenario 3-2. For Scenario 4-2 very similar results are obtained because the main difference between the scenarios is the external energy demand (see Table 2-16). The energy flows in Figure 2-13 are indicated based on their lower heating value with respect to dry mass (for onsite process energy generation water content is subtracted).

Figure 2-13: Carbon and LHV-based energy flows in the butanone concept



Note: in „Energy flows“ only biomass based energy flows and hydrogen input are shown. The butanone concept moreover requires an external input of heat and electricity (see Table 2-16).

Source: Öko-Institut e.V.

⁶ 6 % in Scenario 4-2. For acetoin the value is 4% in both scenarios.

About one third of the carbon is converted to fuel, two thirds are converted to CO₂ (nearly 50 % in CHP generation or other not specified direct emissions). The CRS represents the biggest fraction from the Organosolv step resulting in a transfer of 18 % of the carbon to butanone. In butanediol fermentation 13 % of the overall carbon input is converted to CO₂. A smaller amount of carbon is transferred to the cell mass and the by-products acetoin and IBA. The XRS is smaller, but carbon efficiency to furfural is higher so that nearly the same amount of carbon is transferred to MF (16 %). It has to be kept in mind that this assessment assumes a direct conversion of furfural to MF based on stoichiometric hydrogenation. However, real pathways would probably follow an indirect route via furfuryl alcohol (see Chapter 2.3.1).

The energy efficiency for butanone and MF with respect to biomass input is 40 % equalling a biomass input of 2.47 MJ/MJ_{fuel}. The hydrogen input in this case is rather moderate so that even if it is taken into consideration an overall energy efficiency of 39 % is obtained.

Approx. 80 % of the electricity requirement and two thirds of the heat requirement of Scenario 3-2 can be satisfied by the combustion of the side streams. The remaining external energy requirement is shown in Table 2-16. Moreover, the external energy requirement resulting for Scenario 4-2 is shown. It represents a lower bound for process energy requirement assuming ideal water separation (see Chapter 2.4.1).

Table 2-16: External energy demand of the butanone concept

	MJ/MJ _{fuel}	MJ/MJ _{fuel}	Comment
	Sc. 3-2	Sc. 4-2	Sc. 4-2 theoretical lower bound for energy requirement
Electricity	0.043	0.047	
Heat	0.30	0.18	distribution losses neglected
Hydrogen	0.08	0.08	0.12 MJ _{el} /MJ _{fuel} for electrolytic H ₂

Source: Öko-Institut e.V.

If the correction factor for the energy content of the XRS side components and by-products (see Chapter 2.4.2) is set “1”, the fraction of the process energy generated onsite increases to 90 % and 70 %, for electricity and heat, respectively, in Scenario 3-2.

Approx. 12 % of the carbon and LHV in the biomass are incorporated into the LRS, about 90 % of it lignin. A slightly larger share of the lignin in the biomass is still transferred into the CRS and separated during hydrolysis. Also the XRS contains some residual lignin. In the base case all residual streams are assumed to be combusted for onsite energy generation. In a sensitivity analysis the utilization of the LRS as a phenol substitute is regarded (see Chapter 2.7).

The carbon and LHV content of auxiliary materials is comparably negligible.

The carbon and energy efficiencies are summarized in Table 2-17.

Table 2-17: Carbon and energy efficiencies of the butanone concept

	Carbon	Energy	Comment
in CRS fuel	18%	22%	
in XRS fuel	16%	18%	
overall	35%	40%	2.47 MJ _{biomass} /MJ _{fuel}
overall ext. H ₂		39%	external hydrogen demand included as energy input

Source: Öko-Institut e.V.

2.4.4. LCA results

Following the quantification of the material and energy flows for the entire production process and the corresponding auxiliary material and energy demand these are evaluated concerning their lifecycle environmental impacts. The results are shown in Figure 2-15 and Figure 2-16. The corresponding values are listed in Table 2-18.

2.4.4.1. Main observations for the base case Base 2030

For the base case Figure 2-14 additionally shows the relative contribution of the biomass upstream impact and the impacts related to the TMFB conversion processes differentiating their origin by heat, electricity and auxiliary material input.

While with respect to the gross values energy requirement is responsible for over 50 % of the GWP, with respect to the net values energy, auxiliary material requirement and biomass upstream (incl. soil carbon debt, see also Chapters 2.1.3 and 0) each represent approximately one third. At a transport distance of 200 km, harvesting and transport both contribute similarly to GWP, and together account for approx. 10 % of the net impact. For CED n.r. still 50 % are due to energy requirements dominated by heat. The renewable CED r. is vastly dominated by the energy contained in the biomass: 2.47 MJ/MJ_{fuel} in this case. However, also the impact of cellulase production, the reason for CED r. of auxiliary materials, becomes visible. Also EP and AP are dominated by auxiliary material impact, the main driver being cellulase consumption. For POCP about 60 % of the net impact is caused in the biomass upstream (mainly harvesting) while the remaining is dominated by auxiliary material consumption for the woodchip conversion processes (half of it due to cellulase).

The net GWP is lower than that of fossil gasoline (incl. combustion assuming the same efficiency). The impact of the conversion processes only is about half of its value. When also considering the upstream impact (incl. the soil carbon debt according to Globiom 2015) the reduction is close to one forth. CED n.r. is reduced by one third. The total CED amounts to 3.5 MJ/MJ_{fuel} (see Figure 2-15a-d)).

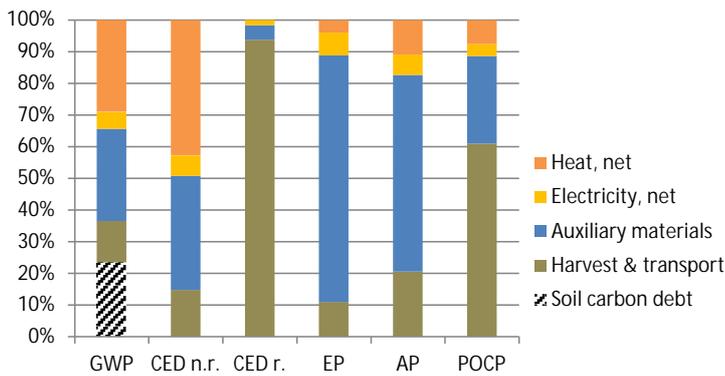
With respect to the conversion processes the relevance of OS fractionation is increased compared to the other concepts because hydrolysis included. Although it does not affect the external energy requirement (see Table 2-3) in the LCA it becomes visible through the impact of cellulase production. The main contributor to the impact of OS fractionation remains electricity (42 %) followed by heat demand and cellulase input. Heat demand is the main driver for the impact of CRS processing and XRS processing (in both cases about 80 %). The absolute contribution of XRS processing to

the overall impact is considerably higher. Beside the steam demand for furfural synthesis, hydrogen requirement contributes.

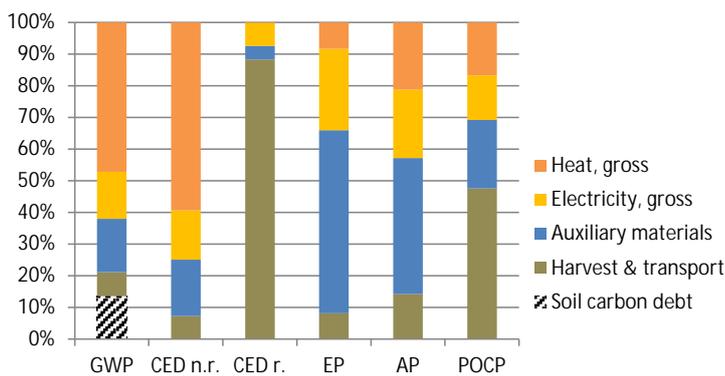
As mentioned in Chapters 2.4.1 and 2.4.2 the energy available for CHP generation from the CRS and XRS side streams may be underestimated by the possibly conservative LHV correction factor of 50 %. However, the influence is not very high in this case. When it is set 1 the overall net GWP is reduced by 10 %. A bigger reduction is obtained in the case of Scenario 4-2 (cf. Chapter 2.4.1, Figure 2-12, ideal effort-less separation of water): the overall net GWP is 17 % lower, the net GWP of the conversion processes is reduced by one forth, indicating the potential of effective water separation concepts.

AP and especially EP are dominated by the Organosolv step with cellulase consumption being responsible for 56 % and 67 %, respectively. The remaining is mainly due to electricity requirement. Heat plays again an important role for CRS and XRS processing followed by auxiliary materials (butanol and sulfuric acid, respectively). The POCP of the conversion processes is mainly caused by heat demand and auxiliary material requirement – cellulase, butanol and sulfuric acid/hydrogen for the OS step, CRS processing and XRS processing, respectively.

Figure 2-14: Contribution analysis for the different impact categories, Base 2030



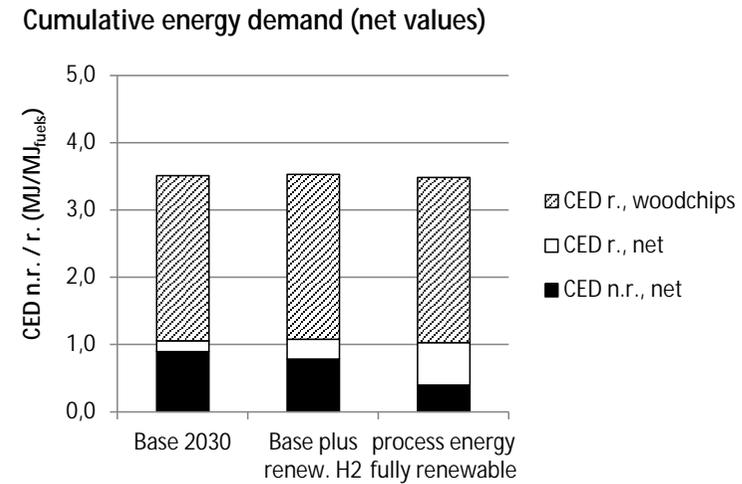
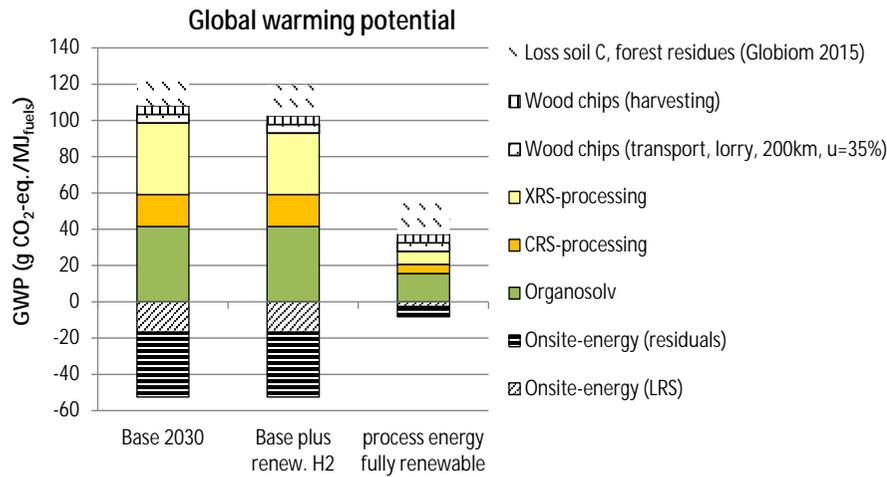
a) Contribution with respect to the **net impact**, i.e. considering external process energy demand only (incl. energy recovery by CHP)



b) Contribution with respect to the **gross impact**, referring to total process energy demand (excl. energy recovery by CHP)

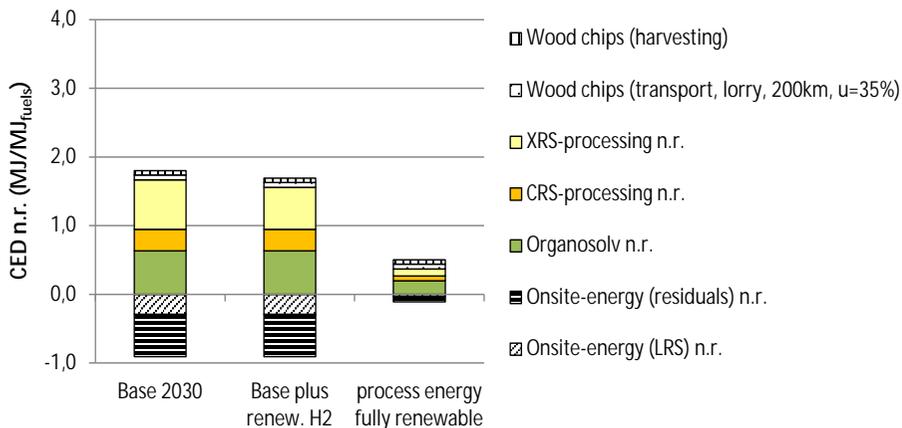
Source: Öko-Institut e.V.

Figure 2-15: Global warming potential (GWP) and cumulative energy demand (CED) for the butanone concept



a) Global warming potential*

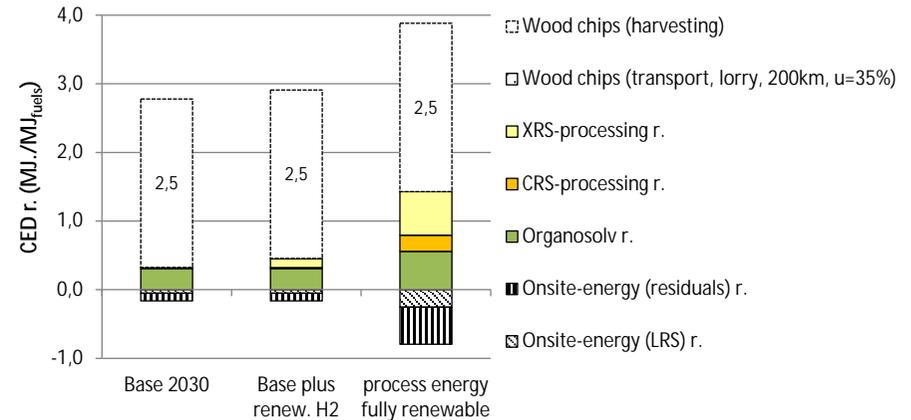
Cumulative energy demand, non-renewable



c) Non-renewable cumulative energy demand

b) Total cumulative energy demand, net values

Cumulative energy demand, renewable

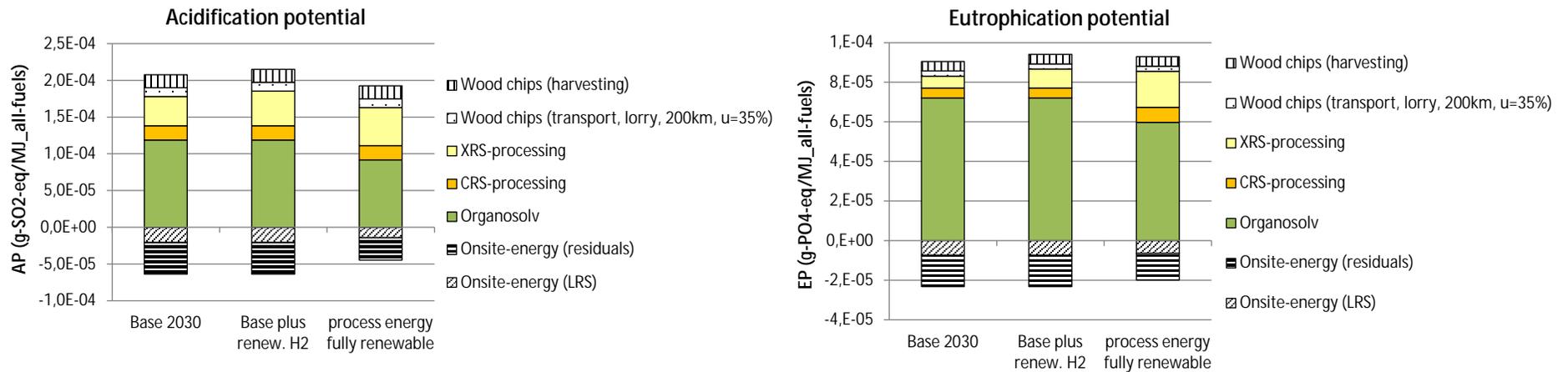


d) Renewable cumulative energy demand

* GWP including the CO₂ emissions resulting upon combustion.

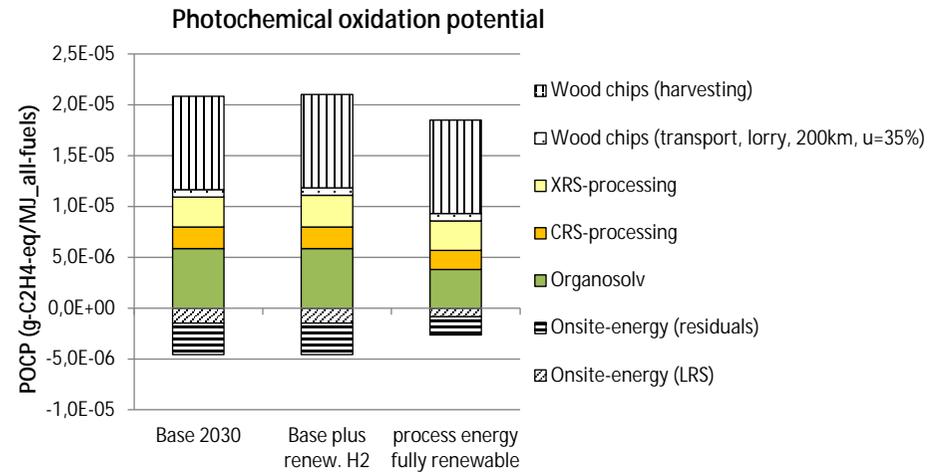
Source: Öko-Institut e.V.

Figure 2-16: Acidification, eutrophication and photochemical oxidation potentials for the Butanone concept



e) Acidification potential

f) Eutrophication potential



g) Photochemical oxidation potential

Source: Öko-Institut e.V.

Table 2-18: Lifecycle impacts of the butanone concept

Butanone concept	Base	Base plus re- new. H2	process energy fully renewable
GWP forest residues, soil-C	17	17	17
GWP biomass harvest & transport	9	9	9
GWP TMFB processes, net	46 (39)*	40	19
GWP overall**	72 (65)*	67	46
CED n.r. biomass harvest & transport	0,1	0,1	0,1
CED n.r. TMFB processes, net	0,8	0,6	0,3
CED n.r. overall	0,9	0,8	0,4
CED r. biomass harvest & transport	2,5	2,5	2,5
CED r. TMFB processes, net	0,2	0,3	0,6
CED r. overall	2,6	2,7	3,1
EP biomass harvest & transport	7,4E-06	7,4E-06	7,4E-06
EP TMFB processes, net	6,0E-05	6,3E-05	6,5E-05
EP overall	6,7E-05	7,1E-05	7,3E-05
AP biomass harvest & transport	3,0E-05	3,0E-05	3,0E-05
AP TMFB processes, net	1,1E-04	1,2E-04	1,2E-04
AP overall	1,4E-04	1,5E-04	1,5E-04
POCP biomass harvest & transport	9,9E-06	9,9E-06	9,9E-06
POCP TMFB processes, net	6,3E-06	6,5E-06	5,9E-06
POCP overall	1,6E-05	1,6E-05	1,6E-05

* The values in parentheses represent a sensitivity analysis concerning a potentially higher energy content of the side components and by-products of the XRS stream sent to the CHP unit (see Chapter 2.4.2 and 2.4.3). Given that it reduces external energy requirements, it affects all impact categories but for sake of clarity is shown ownly indicatively for GWP.

** GWP including the CO₂ emissions resulting upon combustion

Source: Öko-Institut e.V.

2.4.4.2. Changes for renewable hydrogen and a completely renewable process energy supply

[The upstream impact of the biomass supply remains unchanged in all cases.]

As the renewable energy cases (see Table 2-2) are the same for all concepts the effects go in the same direction in all three concepts (see also discussion for the MTHF concept, Chapter 2.2.4.2). The degree varies with the specific characteristics of each concept.

In general, while utilizing renewable electricity to substitute the Base-2030 mix leads to reduced impacts in all categories except CED r., its utilization to substitute natural gas-based hydrogen production and heat decreases the impacts of GWP and CED n.r., but increases the impacts for EP and AP (by factors of 9 and 3, and 3 and 1.2, respectively; see Chapter 2.2.4.2, Table 2-8). The impact of POCP stays similar.

In the Butanone concept the change in the “Base 2030 plus renewable H₂”-case reflects only in XRS processing. The overall net GWP is reduced by 8 %, with respect to the conversion processes only the reduction is of 12 %. CED r. increases slightly more than CED n.r. decreases, but as the quantity of hydrogen is rather small, the overall CED remains essentially constant.

The overall net EP and AP increase by only 5 % (with respect to the conversion processes only by around 6 %) due to the fact that cellulase production is an additional important driver in these categories in the Butanone concept. POCP stays approximately constant.

For a completely renewable process energy supply the net GWP and CED n.r. of the woodchip conversion processes fall in the same order of magnitude as the upstream impact of the biomass (incl. soil carbon debt). While the contribution of energy consumption is decreased, the upstream impact of auxiliary materials remains unchanged. Under these assumptions the impact of cellulase consumption then amounts to 50 % of the net GWP of conversion processes.

Also for the Butanone concept CED r. concomitantly increases so that the total CED remains essentially constant. This is remarkable because in general renewable electricity generation is linked to a smaller total CED due to decreased conversion losses (and system boundary definition starting after solar/wind energy is first captured). However, in the present LCA not only direct electricity is substituted by renewable generation but also heat and hydrogen where the total CED remains approx. constant (heat) or is even increased (hydrogen) based on the datasets used.

For EP and AP cellulase consumption and heat (via PtH) remain the main contributors to the impact of OS fractionation and CRS-/XRS processing, respectively. The overall net EP remains higher than in the base case. The POCP is largely dominated by the biomass upstream impact. For the woodchip conversion processes it varies by less than +/- 10 %.

2.4.4.3. Discussion of upstream impact of renewable energy generation

For considerations on the upstream impact of renewable energy generation please refer to Chapter 2.2.4.3.

2.5. Synthesis and conclusions

The lifecycle assessment illustrates the challenges related to the real implementation of tailor-made fuel production in an integrated complex biorefinery concept. Starting with Organosolv fractionation various conversion and purification steps are included with their concomitant auxiliary material and especially energy requirements. For the three proposed candidates following OS fractionation two synthesis steps are necessary in all cases (see Table 2-1).

The total cumulative energy demand for production varies between 3 to 4 MJ/MJ_{fuel}, with biomass requirement varying between 2.4 and 2.5 MJ/MJ_{fuel}.

The carbon and energy efficiencies of the three concepts are summarized in Table 2-19.

Table 2-19: Carbon and energy efficiencies of the three concepts

	MTHF	MF	Butanone
Carbon (kg-C _{fuel} /kg-C _{biomass})	32%	38%	35%
Energy, LHV-basis (MJ _{fuel} /MJ _{biomass})	40%	42%	40%
Energy, LHV-basis (MJ _{fuel} /MJ _{biomass+H₂})	36%	40%	39%

Source: Öko-Institut e.V.

The highest carbon and energy efficiency was found for the MF concept. Through a CO₂-recycle in this concept it is assured that only 1 mol-C/mol-fuel is lost from cellulose processing. MTHF and Butanone concept perform equally well with respect to biomass requirement. However, the MTHF concept relies on additional hydrogen input in a much stronger way. Thus, the total energy efficiency with respect to biomass and hydrogen input is lower. If in the MF concept the pure CO₂ stream was reduced with hydrogen, the carbon efficiency would increase to 42 %, the energy efficiency to 43 % (MJ_{fuel}/MJ_{biomass+H₂}) or 41 % (MJ_{fuel}/MJ_{biomass+electricity}). The required hydrogen input would still be smaller than that of the MTHF concept. By TMFB research potential biofuel blends with energy efficiencies > 50 % (referring to biomass and hydrogen input) were identified by model-based product and pathway design (Dahmen & Marquardt 2017). The resulting blends vary concerning hydrogen requirement and complexity. A lifecycle assessment of the most promising candidates will be helpful in order to evaluate the trade-off between process complexity (concomitant process energy requirement) and performance in terms of carbon efficiency and biomass requirement.

The lifecycle impacts differ considerably between the concepts. In general, the MF concept has the lowest values mostly due to the very low energy demand estimated for the CRS processing. However, toxicity concerns were identified for this candidate which need to be substantiated in order to decide on its suitability as non-hazardous biofuel. In case MF production can be pursued the concept for 2-MF from C5-sugars via furfural needs to be revised. This preliminary LCA already revealed a relevant impact of steam requirement for furfural production even based on the relatively low value reported for the advanced Suprayield process.

For the butanone concept a relevant contribution of cellulase consumption was found. More research is needed here and other hydrolysis methods should be evaluated. Also alternative water separation concepts could reduce the impact further. Additionally, depending on achievable yields, a combined fermentation of C6 and C5 sugars could be looked at.

In the MTHF concept steam requirement for levulinic acid synthesis is an important driver. It might be investigated whether it can be reduced or better covered when omitting a previous fractionation step. The effort and losses in 3-MTHF production are comparably high. The lower heating value of itaconic acid is rather low so that hydrogen requirement is high. This suggests that in the case of itaconic acid other uses than fuel may be more promising.

Finally, it might be seen whether the OS fractionation, which in all concepts contributes to the impact, can be simplified if no material use of the lignin fraction is envisaged or which conditions justify the effort of fractionation (e.g. higher-value material use of products). The results of a sensitivity analysis for the utilization of lignin as phenol substitute are presented in Chapter 2.7.

According to the present modelling a relevant fraction of the biomass is consumed on-site for the generation of process energy. In order to substantiate the LCA results more research is needed to determine the gross energy demand of the processes under real conditions (using real instead of pure substrate as feed), as well as their net impact based on a more precise estimate of the energy actually recoverable on-site. If the only product is fuel from the carbohydrate fractions, it appears that viable concepts need to be at least energetically self-sustaining at the biomass quantities required in the analysed concepts (see also Chapter 3).

The use of renewable electricity for hydrogen production and especially directly for process energy reduces the global warming potential. The total cumulative energy demand (CED) remains essentially constant. While also for the eutrophication and acidification potentials the impact of the fully renewable electricity mix is considerably reduced compared to the base mix in 2030, the substitution of natural-gas based hydrogen and heat leads to increased impacts in these categories. For further analyses the question of availability of renewable electricity and hydrogen (also including possible storage losses or transport requirements) should be considered in detail. In order to increase the share of direct renewable electricity the development of flexible production concepts able to respond to fluctuating renewable generation is of interest.

The upstream impact of the biomass becomes relevant when the net impacts of the processes are low, especially in case self-sustaining concepts are reached. At a transport distance of 200 km by lorry, the GWP of transport is similar to that of harvesting and chipping, together amounting to about 10 % of the fossil comparator value (incl. combustion). Depending on the biomass used, there may be additional climate-relevant effects which further increase the upstream GWP. In the case of forest residues soil carbon loss may occur upon removal (Globiom 2015). Future assessments should consider this issue in more detail and also evaluate the use of other feedstock, like short rotation coppice or straw.

Moreover, the LCA needs to be extended to include the combustion of the fuels.

Overall, it has to be taken into account that the processes assessed in this study are in an early stage of development. The focus was on the conversion processes and data for the assessment are mostly based on simulations. The results should hence be taken as indicative and need to be substantiated under real conditions, especially based on the utilisation of real substrate.

2.6. Impact of biomass input

A lignocellulosic feedstock is explicitly chosen as the basis for all activities of the TMFB cluster in order to avoid direct competition with food. However, also the utilization of lignocellulosic material is linked to questions concerning availability and sustainability which are dependent on the concrete source of biomass being processed. As forests are already under high harvesting pressure the potential for additional round-wood harvesting from forests on a sustainable basis is rather limited. Also industrial residues of wood processing are already mostly dedicated to other uses (see e.g. Brosowski et al. 2015). In order to also avoid the implications of dedicated agricultural energy crop production (land use, fertilizer use) in this study forest residues were chosen as feedstock for biofuel production.

As indicated in the LCA results in Chapters 2.2.4, 2.3.4 and 2.4.4 also their utilization may, however, be linked to climate-relevant CO₂ emissions. According to the Globiom model a global warming potential of 17 g CO₂-eq/MJ_{fuel} was calculated due to the loss of soil carbon in forests upon residue extraction (Globiom 2015). An additional, regionally varying, aspect which may add to the GWP is that if forest residues are taken out of the wood and converted into a product like fuel which is burnt shortly afterwards CO₂ is released into the atmosphere which would otherwise have stayed in the forest for a longer period (especially in Nordic countries slow-decaying residues like stumps may remain for several decades).

It is interesting to note that the total amount of available forest residues underlying the modelling in the Globiom study is rather low because the study considers only sustainably harvested residues with no current uses to be extracted for biofuel production. In areas that are not ecologically vulnerable 60-70 % of the available residues are removed, depending on the harvesting method (Globiom 2015). Stumps are explicitly excluded. This results in an estimated residue availability of about 7.2 Mt (dry mass) in the EU 28, equalling about 130 PJ per year. The Globiom study moreover states that it cannot determine if the emission level per quantity of residue would be lower or higher for a more limited extraction rate because it is unclear whether the soil carbon response is linear with the extraction rate.

A recent project carried out by Oeko-Institut on the role of bioenergy in the electricity and heat markets up to 2050 (Oeko 2018) considered the technical potential of biogenic residues for Germany based on a DBFZ study (Brosowski et al. 2015). Based thereon the solid biogenic residue potential amounts to 880 PJ of which 20 % or 30 % are allocated to the transport sector in 2030, i.e. 176 PJ and 264 PJ, respectively. According to Brosowski et al. 2015 the currently unused technical potential of forest residues in Germany is 218 PJ of which around 90 PJ are hardwood.

It has to be stressed that this project based itself on the technical potential so that these numbers rather have to be considered as upper bounds. With respect to soil carbon balances, nutrient balances and biodiversity requirements further restrictions are necessary. While a restriction to the utilisation of fast-decaying residues is recommendable from a carbon balance perspective (see e.g. Forest Research 2018) it may adversely effect the nutrient balance because most nutrients are contained in fast decaying residues. Slow decaying residues, especially stumps, should be left in the forests for biodiversity reasons because biodiversity highly depends on the availability of large dimensioned deadwood (Reise et al. 2017). One approach for safeguarding would be to limit forest residue utilisation to current levels. This also implies the diversion from current inefficient uses, like e.g. heating with split logs, in order to liberate potentials for other demands, e.g. biofuel production, an approach which is taken in another current project. According to Brosowski et al. 2015 the current use of hardwood forest residues in Germany is 60 PJ. In the project "Nature protection and

advanced biofuels” the effects of advanced biofuels on nature protection are being evaluated.⁷ Current criticism concerning the update of the renewable energy directive (RED II 2018) and nature protection is summarized in Hennenberg et al. 2018.⁸

With respect to the sustainable sourcing of biomass for biofuel production also the quality requirements represent a crucial parameter. Processes which can utilize lower quality input, e.g. rotting wood, or wood containing a higher fraction of bark, as it is the case for smaller residues, or feedstock of varying composition are less likely to compete with other uses. This aspect should hence be taken into account in process development. The plant capacity is limited by sustainable biomass availability, transport distances and economic considerations. The originally intended output of the TMFB plant of 100 000 t/a biofuel from the cellulose-rich stream only at current efficiency levels corresponds to a woodchip input of 0.6-0.8 Mt/a, equalling around 13 PJ/a. Based on the above considerations only a single plant would thus consume a large fraction of the available potential.

In general, limitations of biomass potentials have been recognized in recent years so that short-term likely scenarios for national transport limit biofuel amounts to the currently used volume of around 120 PJ/a in Germany (renewability.de). In long-term sustainable scenarios liquid fuels are rather allocated to transport modes where the potential of direct electrification is restricted (esp. shipping and aviation).

2.7. Material use of lignin

In the project “Lignocellulose-Bioraffinerie” options for the material use of lignin were studied (Fh IGB 2013/14). It was demonstrated that a substitution of phenol in phenol-formaldehyde resins up to 70 % was possible directly with Organosolv lignin (Leschinsky 2018a). The companies involved further aim at substituting up to 100 % of the phenol.

Based on this a sensitivity analysis for the three concepts (Chapters 2.2-2.4) was carried out to evaluate the impact of a direct material utilization of the lignin on the LCA results.

The following assumptions are made:

- i. The LRS can be used directly to substitute phenol, no further work-up is required.
- ii. The substitution factor is 0.7.
- iii. The LCA impact is allocated between the LRS and the fuels output based on the respective carbon content.
- iv. For the specific impact of the phenol-substitute only the lignin-share of the LRS (capable of substituting phenol) is considered.

In the present assessment the lignin-rich stream contains 84 % lignin (by mass), while the lignin obtained from the project “Lignocellulose-Bioraffinerie” had a purity of 98 % (Leschinsky 2018b). It is hence likely that at present a phenol substitution with the LRS from the concepts of this study is not possible in the same way as with the higher purity lignin. Still, the outcome of the sensitivity analysis provides an indication of the potential involved and to some extent the purity is considered by point iv). The purity is an optimization parameter subject to a trade-off between raw material, purification effort and purity (Viell 2018b).

⁷<https://www.natur-und-erneuerbare.de/projektdatenbank/projekte/fortschrittliche-biokraftstoffe/>

⁸https://www.nature.com/articles/s41559-018-0659-3.epdf?author_access_token=2D2LTJWegLedxucH6-GpvdRgN0jAjWel9jnR3ZoTv0PLJOrWO9J9BR444f7uYpyOxmgAxnmR3f_qvXypELwO417nkKiwC87ccBgqhJPesRXoKuTRjlkspocx2NXaxaFY4pzPLVUobxvF5VYKEqsA%3D%3D

The result of the sensitivity analysis is shown in Table 2-20. The impact of fossil phenol (ecoinvent 3.4 dataset: “market for phenol”) is divided by the carbon-allocated LCA result for the lignin in the LRS. It can be seen that in almost all cases the impact of the phenol-substitute is lower than that of fossil phenol. The main exception is of course the renewable CED which for the phenol-substitute is higher by a factor of 50-60. In the MTHF concept also the GWP is higher than for fossil phenol.

Table 2-20: Ratio between impact of fossil phenol and lignin as phenol-substitute

	MTHF concept	MF concept	Butanone concept	
GWP*	0,8	1,5	1,0	
CED n.r.	1,4	3,3	2,0	
CED r.	0,0	0,0	0,0	
EP	2,8	5,5	1,6	yellow: < 10% deviation
AP	2,3	3,3	2,0	green: LRS as phenol substitute > 10% lower
POCP	3,5	4,8	3,7	red: LRS as phenol substitute > 10% higher

* incl. biomass upstream impact incl. soil carbon loss-value for forest residues as modelled by Globiom 2015.

Source: Öko-Institut e.V.

The carbon-allocated LCA results for the fuels remain similar. In most cases they are even reduced by about 20 % (see Table 2-21).

Table 2-21: Impact of fuels - Ratio between results for energetic utilisation of LRS (no allocation) and carbon-based allocation between fuels and LRS

	MTHF concept	MF concept	Butanone concept	
GWP	1,2	1,0	1,1	
CED n.r.	1,1	0,8	1,0	
CED r.	1,3	1,3	1,3	
EP	1,1	0,9	1,2	yellow: < 10% deviation
AP	1,2	1,1	1,2	green: C-allocated value > 10% lower
POCP	1,3	1,2	1,2	red: C-allocated value > 10% higher

Source: Öko-Institut e.V.

3. Considerations on alternative biofuels and drives

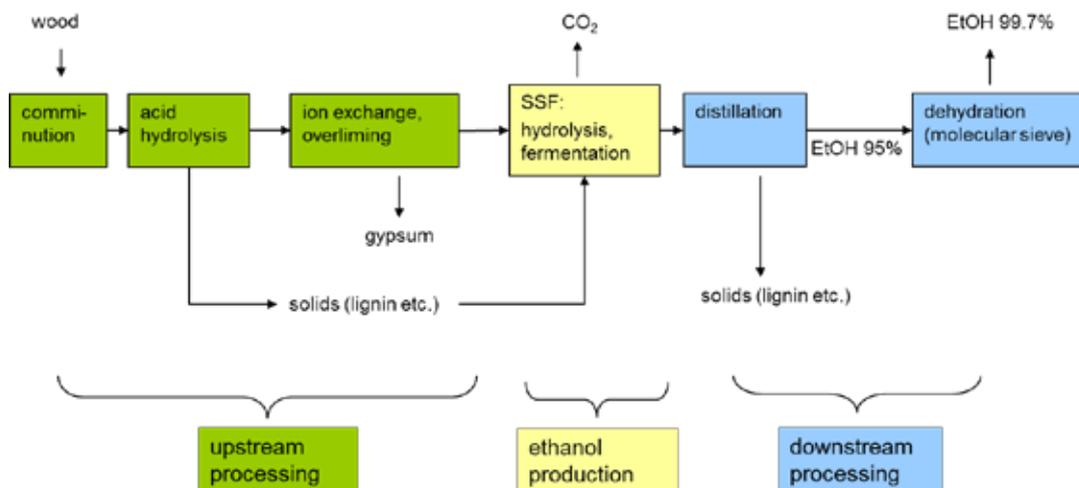
3.1. Ethanol

Ethanol is the most important biofuel that is available on the European fuel market. In the following the dataset for ethanol from lignocellulose fromecoinvent database is analysed as benchmark. A description of the modelled process has been published in Jungbluth et al. 2007.

Process chain

The production of ethanol from lignocellulosic biomass consists of three stages: biomass pretreatment, hydrolysis and fermentation. Chemical and physical pretreatment break down plant cell structures and separate the lignin from cellulose and hemicelluloses and thereby facilitates their hydrolysis. Acid or enzymatic hydrolysis converts the cellulose and hemicelluloses into fermentable sugars. The sugars are fermented to ethanol, which is then purified and dehydrated. The lignin residue can be used for electricity and heat generation. The single process steps are shown in Figure 3-1.

Figure 3-1: Process chain of the ethanol production



Source: Öko-Institut e. V.

In a pretreatment step the wood chips are held 10 minutes at about 160-190°C with 0.5-1.0% dilute sulphuric acid to hydrolyze the hemicelluloses. By a sudden drop in pressure the temperature is rapidly lowered and the reaction is stopped. The liquid and the solid fractions are separated. Most of the cellulose and lignin is contained in the solid fraction. The liquid is pumped to an ion exchange operation to remove a portion of the acetic acid and virtually all of the sulphuric acid. By addition of lime the pH of the liquid is increased to about 10 to neutralize the remaining acetic acid and to remove other substances toxic to downstream fermentation (overliming). Then the liquid is mixed back with the solid fraction and fed into the fermentation step. A small portion of the solids and the treated liquid is used to produce the enzyme cellulase by the fungus *Trichoderma reesei* for the SSF process.

In the simultaneous saccharification and fermentation process (SSF) cellulases and an organism that ferments the sugars from hemicelluloses plus the glucose released from cellulose to ethanol are added, e. g. yeasts. Integration of the two process steps in one reactor avoids product inhibition of the enzymatic hydrolysis as the resulting sugars are directly fermented to the product. The fermented beer contains about 5% (vol.) ethanol. It is fed into a distillation step where the ethanol

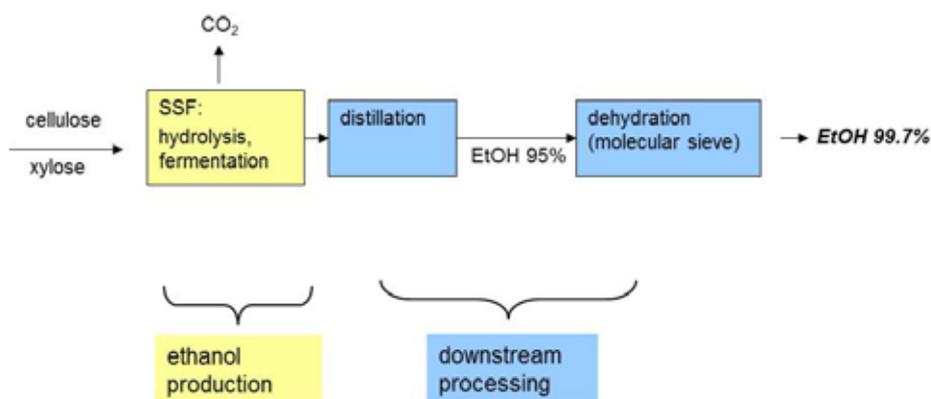
is concentrated to approximately 95% in the overhead. With a molecular sieve the hydrated ethanol 95% is dehydrated to anhydrous ethanol 99.7%.

Water is treated by anaerobic digestion, and the resulting biogas is combusted for steam generation. The residual solids, containing mostly lignin and insolubles from distillation are concentrated and combusted to generate steam that can provide the entire heat and electricity for the process with some excess electricity left to be exported. For the described ethanol production process from wood chips Jungbluth et al. (2007) give values for GWP and CED, non-renewable of 0.016 kg-CO₂-eq/MJ ethanol and 0.2 MJ/MJ ethanol, respectively. The values exclude the burden related to the supply of wood chips.

Process steps

A simplified process chain for the ethanol route is shown in Figure 3-2.

Figure 3-2: Simplified process chain of the ethanol production route



Source: Öko-Institut e. V.

Ethanol can be purified by rectification and dehydrated with a molecular sieve.

Equations of reactions

The overall equations for the fermentation of glucose and xylose to ethanol are given here:

- (1) glucose: $C_6H_{12}O_6 \rightarrow 2 C_2H_5OH + 2 CO_2$
- (2) xylose: $3 C_5H_{10}O_5 \rightarrow 5 C_2H_5OH + 5 CO_2$

Calorific values

Table 3-1 shows the lower heating values of the main raw materials and products of the ethanol route.

Table 3-1: Properties of raw materials and fuels

	Formula	LHV [MJ/kg]
glucose	C ₆ H ₁₂ O ₆	14.4
xylose	C ₅ H ₁₀ O ₅	14.4
ethanol	C ₂ H ₅ OH	26.7

Source: ecoinvent 3.5 (2018)

Fermentation of sugars to ethanol increases the calorific value from 14.4 MJ/kg to 26.7 MJ/kg.

Yields

Yields that have been realized in pilot scale ethanol production plants are given Table 3-2.

Table 3-2: Realized yields for ethanol fermentation

	Fermentation of glucose	Fermentation of xylose
yield based on mass	47 %	43 %
yield based on C-content	61 %	57 %
yield based on energy content (LHV)	87 %	81 %

Source: ecoinvent 3.5 (2018)

In Table 3-3 the overall yields are shown for production of ethanol 99.7 % from sugars (Wooley et al. 1999, Jungbluth et al. 2007).

Table 3-3: Overall yields for ethanol 99.7% production

Production route	sugars to ethanol 99.7%
yield based on mass	45 %
yield based on C-content	59 %
yield based on energy content (LHV)	84 %

Source: ecoinvent 3.5 (2018)

In the IA-route the final energy content of the product is increased by the additional hydrogen input. When this contribution is neglected in the calculation of the energetic O/I ratio, the overall energetic yield of the IA-route (94 %) is superior to the ethanol route (84 %). However, when the energy input with the hydrogen is included, the overall energetic yield of the IA-route decreases to 67 %.

It is very remarkable that if the complete process energy demand of the ethanol route is included in the calculation of the overall energetic efficiency of ethanol production a value of 66 % results.

$$\text{Overall energetic efficiency, ethanol production} = \frac{1 \frac{\text{MJ}_{\text{Ethanol}}}{\text{MJ}_{\text{Ethanol}}}}{1.2 \frac{\text{MJ}_{\text{Sugars}}}{\text{MJ}_{\text{Ethanol}}} + 0.32 \frac{\text{MJ}_{\text{Process energy}}}{\text{MJ}_{\text{Ethanol}}}} = 66\%$$

Auxiliary materials consumption & process energy

As the pre-treatment, the enzyme production, and the wastewater treatment are excluded from this comparison no auxiliaries are needed in the considered ethanol production steps.

Energy inputs for the ethanol route are shown in Table 3-4.

Table 3-4: Ethanol production: consumption of electricity and heat

energy consumption	electricity [MJ/MJ ethanol]	heat [MJ/MJ ethanol]
SSF	0.02	
Rectification	0.003	0.26
Dehydration	0.001	0.04
Total	0.02	0.3

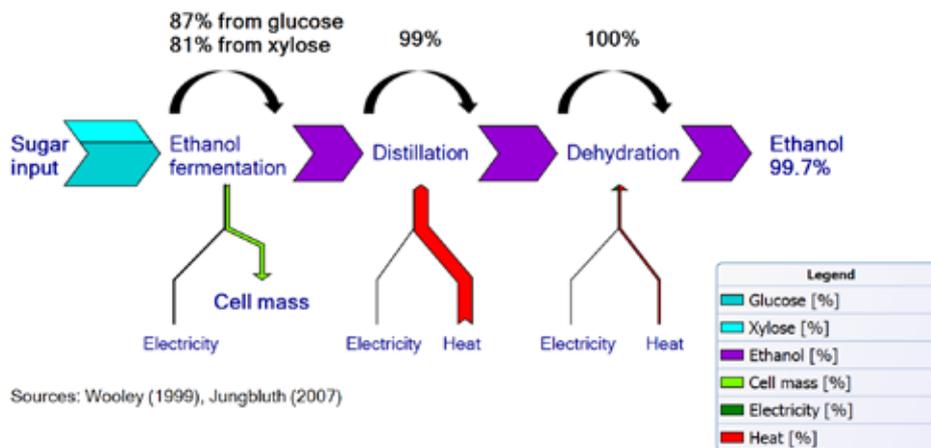
Source: ecoinvent 3.5 (2018)

Energy flows

Figure 3-3 shows the energy flows through ethanol fermentation and purification. The energy related to the material flows through the process is based on LHV.

Figure 3-3: Material related energy flows (LHV based) and process energy input through ethanol route

Energy flows (LHV-based) through ethanol fermentation and purification and process energy inputs



Sources: Wooley (1999), Jungbluth (2007)

Source: Öko-Institut e. V.

Table 3-5 shows impact assessment results for ethanol production 99.7 % from wood, RoW (ecoinvent 3.5, 2018)

Table 3-5: Impact assessment results for 1 kg ethanol from wood, RoW, 99.7%

Impact method	Impact category	Value	Unit	Comment
CML 2016	acidification potential	3.98E-03	kg SO ₂ -eq	generic
	climate change	0,55	kg CO ₂ -eq	GWP 100a
	eutrophication potential	1.1E-03	kg PO ₄ -eq	generic
	land use	3.78	m ₂ a	competition
	photochemical oxidation (summer smog)	1.59E-04	kg ethylene-eq	high NOx POCP
	photochemical oxidation (summer smog)	8,73E-05	kg ethylene-eq	low NOx POCP
	resources	3.18E-04	kg antimony-eq	depletion of abiotic resources
	cumulative energy demand	renewable energy resources, biomass	17,05	MJ-eq
renewable energy resources, geothermal, converted		1.9E-03	MJ-eq	
renewable energy resources, solar, converted		2.96E-04	MJ-eq	
renewable energy resources, potential (in barrage water), converted		0,11	MJ-eq	
renewable energy resources, kinetic (in wind), converted		1.62E-02	MJ-eq	
non-renewable energy resources, fossil		6,52	MJ-eq	
non-renewable energy resources, nuclear		0,21	MJ-eq	
non-renewable energy resources, primary forest		3.8E-03	MJ-eq	
total renewable		17,18	MJ-eq	
total non-renewable		6,73	MJ-eq	
total		23,9	MJ-eq	
IPCC 2013	climate change	0,50	kg CO ₂ -Eq	GTP 100a
USEtox	ecotoxicity	2.03	CTU	total
	human toxicity	1,79E-08	CTU	carcinogenic
	human toxicity	5,92E-07	CTU	non-carcinogenic
	human toxicity	6,10E-07	CTU	total

Source: ecoinvent 3.5 (2018)

Many other publications on LCA of ethanol production from lignocellulosic biomass have been analysed.⁹ As there is a huge variation in specific system boundaries, allocation rules, credit for electricity offtake, calculation of iLUC and dLUC, the results for global warming potential given in these publications are hardly comparable. The yields of ethanol based on wood input (mainly forest resi-

⁹ Borrion et al. 2012a, Borrion, McManus, Hammond, 2012, Brodeur-Campbell 2012, Budsberg et al. 2012, Cherubini et al. 2010, Daystar et al. 2012, Gonzalez et al. 2012, Gonzalez-Garcia et al. 2012, Guo et al. 2014, Hipolito 2011, Jeswani et al. 2015, Karlsson 2014, Karlsson et al. 2014, Mu et al. 2010, Prasad et al 2015, Reed et al. 2012, Roy et al 2012, Roy 2014, Scott et al. 2013, Shen et al 2015, Steele et al. 2012, Swana et al. 2011, Terlouw 2013, Tsalidis et al. 2014, Wiloso et al. 2012

dues, willow, and poplar) lie between 0.14 and 0.28 kg ethanol per kg wood. The excess electricity lies between 0.38 and 0.65 kWh/kg ethanol. While the ethanol yield in theecoinvent dataset (0.26 kg ethanol per kg wood chips) is in the range of other LCAs the value for excess electricity is significantly lower (0.045 kWh / kg ethanol).

3.2. Biomass-to-liquid

Another means to convert wood into liquid fuels is by gasification and subsequent Fischer-Tropsch or methanol synthesis. With respect to biomass requirement a simulation-based analysis of the entrained-flow gasification concept (as was used for the Choren plant) reported that the BtL process can be operated in an energetically self-sufficient way at an of $2.4 \text{ MJ}_{\text{biomass}}/\text{MJ}_{\text{diesel}}$ or $\approx 2 \text{ MJ}_{\text{biomass}}/\text{MJ}_{\text{all-liquids}}$, equalling an energetic efficiency of $\approx 50\%$ for LHV-based allocation between the outputs (Renew 2008 WP 5.4.2 “Starting point scenario” without additional hydrogen input). Also the Globiom study considers about $2 \text{ MJ}_{\text{biomass}}/\text{MJ}_{\text{fuel}}$ for FT-Diesel production (Globiom 2015).

At the time Choren failed with the commercialization of BtL fuels it was stated that the reasons were not insurmountable technological barriers but rather related to economic constraints (Rapier 2011). Today DBFZ indicates a technological readiness level of TRL 5 (Hobohm et al. 2018). They assume a plant with 98 MW (biomass input 182 000 t/a) delivering 24 000 t/a of FT-fuels and 102 000 MWh/a electricity. If 8000 operating hours per year are assumed this equals a higher biomass requirement of $\approx 2.8 \text{ MJ}/\text{MJ}_{\text{fuel}}$ (neglecting allocation to electricity output; naphtha quantities not mentioned).

BtL fuels especially represent a benchmark with respect to their potential for the utilization of external hydrogen to increase the carbon efficiency. If hydrogen can be added to the syngas in the required quantities practically a full conversion of all carbon can be achieved with the same technology. According to DBFZ this can be reached by the utilization of $0.7 \text{ MJ}_{\text{biomass}}/\text{MJ}_{\text{fuel}}$ and $1.3 \text{ MJ}_{\text{el}}/\text{MJ}_{\text{fuel}}$ ($\approx 0.9 \text{ MJ}_{\text{H}_2}/\text{MJ}_{\text{fuel}}$), equalling a total energetic efficiency of around 60% ($\text{MJ}_{\text{fuel}}/\text{MJ}_{\text{biomass}+\text{H}_2}$) (Hobohm et al. 2018). Only dimensioning of the FT-synthesis unit has to be adapted.

3.3. Battery electric mobility

Especially when considering passenger cars battery electric vehicles (BEV) constitute a relevant alternative. Their lifecycle impact compared to internal combustion engine vehicles (ICEV) is a subject of intense debate. While the impact of vehicle production is higher for BEV, especially due to the battery, they have the potential for locally emission-free driving and lower impacts in the use phase especially when powered by renewable electricity. Current LCAs of BEV including all lifecycle stages suggest a GWP of $\approx 200 \text{ g.CO}_2\text{eq/vkm}$ based on the current German grid mix (Ifeu 2016, ICCT 2018). Own analyses for a VW e-Golf point into the same direction (Oeko 2017). This means that concerning GWP BEV are today in the break-even region or even already advantageous for the German grid mix (Ifeu 2016, ICCT 2018). Concerning the total cumulative energy demand a value in the region of 3-4 MJ/vkm is found which is comparable to fossil-based ICEV (Oeko 2017, ifeu 2016). Given that the total CED of biofuel production is higher than that of fossil fuel production (in the range of 2-4 MJ/MJ_{fuel} instead of 1.4 MJ/MJ_{fuel}) the total CED of ICEV running on biofuels will be higher. For proceeding decarbonisation of the electricity generation the impact of the BEV use phase in these categories will be lowered progressively. Concerning the production phase, the LCA of battery production is still related to important data uncertainties and subject to very dynamic development, e.g. concerning energy density, size, cell chemistries, and optimisation of production processes when entering larger scales.

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Annex 1 LCA datasets

Material	Dataset	Source		GWP, 100a	CEI, n.r.	CEI, r.	EP, generic	AP, generic	POCP, high NOx
				g CO2-eq.	MJ	MJ	kg PO4-eq.	kg SO2-eq.	kg C2H4-eq.
Wood chips	hardwood forestry, beech, sustainable forest management wood chips	ecoinvent 3.4	per kg	35	0,46	18,3	3,6E-05	1,3E-04	6,8E-05
Transport lorry	transport, freight, lorry, all sizes, EURO 6 to generic market for transport, freight, lorry, unspecified	ecoinvent 3.4	per tkm	132	1,92	0,03	7,2E-05	3,3E-04	2,1E-05
Ethanol	ethanol, from ethylene, RER combustion of C2H6O	ecoinvent 3.4	per kg	1,2E+03	4,6E+01	6,1E-01	1,8E-03	3,8E-03	1,4E-03
		calculated	per kg	1,9E+03					
Butanol	market for 1-butanol, GLO combustion of C4H10O	ecoinvent 3.4	per kg	2,9E+03	8,3E+01	2,4E+00	3,5E-03	1,3E-02	2,3E-03
		calculated	per kg	2,4E+03					
Water	market for tap water, Europe without CH	ecoinvent 3.4	per kg	3,8E-01	6,4E-03	7,8E-04	9,4E-07	1,8E-06	1,1E-07
Hydrogen	renewable hydrogen	own estimate, 70%	per kg	2,4E+03	3,0E+01	1,9E+02	5,7E-03	1,5E-02	9,4E-04
	hydrogen, steam reforming NG	Gabi 6	per kg	1,1E+04	1,9E+02	3,0E-01	5,9E-04	4,5E-03	6,9E-04
Catalyst	platinum group metal (50% Pd, 50% Pt)	ecoinvent 3.4 - mix	per kg	1,8E+07	2,4E+05	8,3E+03	9,4E+01	1,8E+03	8,6E+01
Toluene sul-fonic acid	toluene sulfonic acid	own estimate based on naphthalene sulfonic acid (ETH Zürich, Sutter, J.)	per kg	1,7E+03	4,8E+01	8,2E-01	2,2E-03	9,1E-03	7,5E-04
NH4PF6	market for lithium hexafluorophosphate	ecoinvent 3.4	per kg	2,2E+04	3,0E+02	2,2E+01	4,4E-02	1,6E-01	9,9E-03
aIL	1-butyl-3-methylimidazolium chloride	own estimate based on Righi et al. 2011	per kg	8,1E+03	2,0E+02	6,4E+00	1,2E-02	3,9E-02	4,5E-03
triphos	market for organophosphorus-compounds, GLO	ecoinvent 3.4	per kg	8,8E+03	1,5E+02	9,1E+00	5,7E-02	9,2E-02	6,1E-03
Sulphuric acid	market for sulfuric acid, GLO	ecoinvent 3.4	per kg	1,6E+02	6,7E+00	2,0E-01	7,6E-04	6,4E-03	2,9E-04
Sodium hydroxide	market for sodium hydroxide, 50% in H2O, GLO	ecoinvent 3.4	per kg	1,4E+03	1,8E+01	1,8E+00	2,8E-03	6,6E-03	2,8E-04
Cellulase	Enzymes (Alpha-amylase, Glucoamylase, Cellulase)	ecoinvent 3.4 (excl. bio-C)	per kg	6,7E+03	8,2E+01	7,6E+01	3,1E-02	4,3E-02	1,3E-03
Electricity	electricity 2030, medium voltage	own modelling based on OEKO/ISI 2015 and ecoinvent 3.4 datasets	per MJ	8,1E+01	1,2E+00	8,9E-01	1,0E-04	2,0E-04	1,3E-05
	electricity production, photovoltaic, 570kWp open ground installation, multi-Si, DE	ecoinvent 3.4	per MJ	2,9E+01	3,8E-01	1,1E+00	6,8E-05	1,6E-04	8,7E-06
	electricity production, wind, 1-3MW turbine, onshore, DE	ecoinvent 3.4	per MJ	5,5E+00	7,1E-02	1,1E+00	1,4E-05	2,9E-05	1,9E-06
	Renewables mix 75 % wind, 25 % PV		per MJ	1,1E+01	1,5E-01	1,1E+00	2,7E-05	6,1E-05	3,6E-06
Heat	heat production, natural gas, at industrial furnace > 100kW, Europe without CH	ecoinvent 3.4	per MJ	6,9E+01	1,3E+00	3,0E-03	8,9E-06	5,2E-05	4,1E-06
	PtH, based on renewables mix	own estimate	per MJ	1,1E+01	1,5E-01	1,1E+00	2,7E-05	6,1E-05	3,6E-06

Annex 2 Energy generation structure in 2030 and 2050 according to OEKO/ISI 2015, 95 % reduction scenario

OEKO/ISI 2015	2030 KS 95		2050 KS 95	
	TWh	Shares	TWh	Shares
Nuclear	0	0%	0	0%
Lignite	5	1%	0	0%
Hard coal	30,4	7%	0	0%
Natural gas	93,3	20%	13,1	2%
Other	12,6	3%	4,2	1%
Oil	0,5	0%	0	0%
Top gas	4,7	1%	0	0%
Coke oven gas	1,2	0%	0	0%
Litter	5,3	1%	3,6	0%
Other	1	0%	0,7	0%
Renewables	322,6	69%	733,9	98%
Hydropower	24,1	5%	24,7	3%
Wind	204,9	44%	569,8	76%
onshore	153,9	33%	389,8	52%
offshore	51	11%	180	24%
PV	66,1	14%	123,4	16%
Biogas	12,6	3%	0,6	0%
Biomass	11,6	2%	3,2	0%
Geothermal	4,2	1%	12,3	2%
Pumped storage	3,7	1%	8	1%
Back-up	0,2	0%	4,6	1%
Total	464,9	100%	751,4	100%