EU Framework Programme for Research and Innovation
H2020-Competitive Low-Carbon Energy
Call topic 11-2014

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Photofuel - Biocatalytic solar fuels for sustainable mobility in Europe

Deliverable D6.3

LCA on cultivation and fuel production and options for improving the LCA

This project has received funding from the European Union’s Horizon 2020 research and innovation programme under grant agreement No 640720

Editorial	
Deliverable Nº:	D6.3
Title	LCA on Photofuel processes
Workpackage:	WP6
Responsible beneficiary:	KIT, IFPEN
Authors:	Christine Rösch, Daniela Villacreses, Kirsten Moore, Franziska Ketzer, Pierre Collet
Contributors:	KIT, IFPEN
Version:	
Due date of deliverable:	
Version date:	
Contact:	christine.roesch@kit.edu
Dissemination level:	PU-Public
Nature:	Report
Review status	WP-leader accepted
	GA accepted
	Coordinator submitted

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1. Introduction

In the Photofuel project biologists at ICL, UU and UniBi have investigated different production processes with microalgae or cyanobacteria in order to find out the best performing strains and processes with regard to fuel yields. Not all of these processes investigated have been transferred to pilot scale mainly due to the lack of high productivities. This has been the case for bisabolene, n-butanol and 1-octanol. The most productive system is 1-butanol, however 1-octanol productivity increased at lab scale towards the end of the project. Consequently, the work on LCA, which will be reported here, is focusing on the production of 1-butanol by *Synechocystis PCC6803* since this strain developed at UU showed very high productivities not only at lab scale but also on pilot scale at A4F.

In WP 6 a comprehensive questionnaire was developed to collect relevant and detailed information on the process design and the technology and equipment used to model and calculate the LCA from well-to-wheel. Based on this questionnaire, there was a lively and ongoing exchange with the project partners in Florence (UniFi) and Lisbon (A4F) with personal communication, virtual meetings and email correspondence. By this, we collected as much information and original data for the LCA inventory as possible. Based on this inventory and supplemented by data from the literature and the Ecoinvent database we first provided an environmental impact assessment for the pilot scale. The insights we drew from the pilot scale results were used to design and model a large-scale production plant (20 ha) by upscaling the process in order to increase efficiencies, optimise the production system and gain environmental benefits from economies of scale. In a further upscaling scenario we have modelled and implemented the process of hydrothermal liquefaction (HTL), which is a thermal biomass valorisation process used to convert wet residual algal biomass into crude-like oil under moderate temperature and high pressure and to recycle main nutrients such as nitrogen and phosphate. Moreover, we developed and calculated an upscaled scenario with a renewable electricity mix used for the Photofuel production.

As reference to the 1-butanol process, we designed, modelled and performed an LCA on the production of total fatty acids (TFA) with the natural algae strain *Nannochloropsis oceanica*, which was not genetically modified. Based on this reference, conclusions can be drawn about the ecological excellence of the Photofuel process, designed to run in a chemostat operation allowing for a "milking" of the genetically modified cyanobacteria compared to photobioreactor harvesting for extraction of biofuel from cells of non-genetic modified algae.

The following paragraphs describe the production system and process for the 1-butanol and TFA production in detail including the system boundaries and the functional unit as well as the results for the pilot scale, three scenarios for the upscaled process and the total well-to-wheel LCA.

2. Methodology

For all LCA studies the open source software OpenLCA is applied to model the production chain in a well-to-wheel analysis.
For quantifying the life cycle impacts, the recommendations of the ILCD Handbook 2011 have been followed. This impact assessment method, developed and promoted by the JRC European commission (2011) is already implemented in the OpenLCA software. In addition, data from the pilot plant operation at UniFi and A4F as well as from literature and the database Ecoinvent 3.2 [1], which provides complex datasets to derive a proper LCI, have been used. The impact assessment is carried out by calculating recommended impact categories according to the ILCD handbook [2]. An overview of the 16 midpoints impacts categories considered in the ILCD are given in Table 1.

Table 1: Impacts on midpoint level based on the ILCD Handbook

Midpoint applications			
Classification	*Impact category*	*deflaut LCIA method*	*Indicator*
I	Climate Change	ILCD 2011: Baseline model of 100 years of IPCC	Radiative forcing as Global Warming Potential (GWP 100)
I	Ozone Depletion	ILCD:2011: Steady-state ODPS 1999 as in WMO assessment	Ozone depletion Potential (ODP)
I	Particulate matter/Respiratory inorganics	RiskPoll model (Rabl and Spadaro, 2004) and Greco et al. 2007	Intake fraction for fine praticles (kg $PM_{2.5}$-eq/kg)
II	Ionising radiation, human health	Human health effect model as developed by Dreicer et al. 1995 (Frischknecht et al. 2000)	Human exposure efficiency relative to U^{235}
II	Photochemical ozone formation	LOTUS-EUROS (Van Zelm et al., 2008) as applied in ReCiPe	Topospheric ozone concentraion increase
II	Acidification	Accumulated Exceedance (Seppälä et.al., 2006, Posch et al., 2008)	Accumulated Exceedance (AE)
II	Eutrophication, terrestrial	Accumulated Exceedance (Seppälä et.al., 2006, Posch et al., 2008)	Accumulated Exceedance (AE)
II	Eutrophication, aquatic	EUTREND model (Struijts et al., 2009b) as implemented in ReCiPE	Fraction of nutrients reaching freshwater end compartment (P) or marine end compartment (N)
II	Resource depletion, mineral, fossil and renewable (Depletion of renewable resources is included in the analysis but none of the analysed methods is mature for recommendation)	CML 2002 (Guinée et al. 2002)	Scarcity
II/III	Ecotoxicity (freshwater)	USEtox model (Rosenbaum et al., 2008)	Comparative Toxic Unit for humans (CTUe)
II/III	Human toxicity, cancer effects	USEtox model (Rosenbaum et al., 2008)	Comparative Toxic Unit for humans (CTUh)
II/III	Human toxicity, non-cancer effects	USEtox model (Rosenbaum et al., 2008)	Comparative Toxic Unit for humans (CTUh)
III	Land use	Model based on Soil Organic Mater (SOM) (Milà i Canals et al., 2007b)	Soil Organic Matter
III	Resource depletion, water	Model for water consumption as in Swiss Ecoscarcity (Frischknecht et al., 2008)	Water use realted to local scarcity of water
Interim	Ionising radiation, ecosystems	No methods recommended	
	Ecotoxicity (terrestrial and marine)	No methods recommended	

It was stated in Deliverable 6.1 that only the impact categories classified as “I” will be assessed; however the “III”-classified categories of water resource depletion and land use are used based on relevance and communication with the consortium during the duration of the project. The ILCD handbook [2] explains the classification levels as recommended and satisfactory (“I”), recommended but in need of some improvements (“II”), and recommended, but to be applied with caution (“III”).

The database for the processes comprises raw material extraction as well as the assembly to pre-products e.g. borosilicate tubes. However, transport, storage and end of life options are excluded because they are outside the system boundaries of the investigated processes. In addition to that, the delivery of inputs and raw materials to the production site as well as machine abrasion and disposal are defined to be outside the system boundaries of this study.

Several pathways of (bio) fuels production and utilization are assessed. Each one is described in detail in the following sections. Environmental impacts are evaluated on the basis of two driving cycles:

- The New European Driving Cycle (NEDC), last updated in 1997, is designed to assess the emission levels of car engine. It was the test protocol in the European Union on which the CO_2 targets were based until September 2017. It has received criticism regarding its effectiveness to represent real-life driving and to reduce CO_2 emissions in real world operating conditions.
- The worldwide harmonized light vehicles test procedure (WLTP), which is supposed to address issues relative to the NEDC, consists in a more realistic test procedure and is the current test protocol in the European Union since 2017. Main differences between NEDC and WLTP are: test cycle and gear-shifting sequence, vehicle mass definition, road load determination, chassis dynamometer preconditioning and temperature [3] (ref doc JRC)

For the well-to-wheel (wtw) LCA impacts figures represent the different impacts generated in the process of delivering 1 km of vehicle motion on the different driven cycles assessed.

Data used to calculate the environmental impacts of the different fuels by IFPEN come from KIT for the impacts linked to the production of the precursors (1-butanol and TFA). Only the upscaled scenario results are considered in the wtw calculations, as lab scale results are several orders of magnitude higher than the prospective industrial scale and fossil references. Photofuel partners Fiat, Volkswagen and Volvo provided the combustion emissions of the different fuels (respectively for the gasoline, diesel light duty and diesel heavy duty fuels). Combustion emissions are summarized in the Annex 1. Ecoinvent database 3.4 [1], with the cut-off assumption for end of life management for the other required data was applied by IFPEN.

3. Photofuel process: 1-Butanol production

The LCA analysis is based on 1-butanol produced by *Synechocystis PCC6803*. Productivities for 1-butanol were extrapolated from lab scale batch experiments at UU and continuous cultivation experiments at ICL regarding both, scale as well as cultivation time. A year-round production of 360 days was considered with an average butanol productivity of 600 mg/l/d.

As the project is dealing with genetically modified cyanobacteria, a stable, closed system has to be used to keep the risk of leakage low and prevent an exposure and cross-breeding. The system boundaries of the pilot scale are therefore based on the installation of the plant of A4F in Lisbon. After analysing different reactor types, the unilayer horizontal tubular (UHT) PBR was chosen since this reactor has the same areal productivity as the multilayer horizontal tubular (MHT-PBR) reactor but needs less material and pumping energy.

The pilot plants at UniFi and A4F are designed to carry out experiments with different microalgae and settings rather for research than for commercial algae fuel production. Thus it was not surprising that the LCA results of 1-butanol production at pilot scale did not meet the expectation compared with already established fully mature technologies operating at large scale. In order to assess the environmental impacts in a prospective way and to improve comparability, scenarios with upscaled industrial production systems for algae-based 1-butanol of 20 ha were modelled and calculated. It is expected that economies of scale will reduce energy and material demand compared to the pilot plant at A4F, which runs with 5 m^3 volume (LCA results reported in deliverable 6.2).

3.1 System boundaries and process description

Three scenarios were considered and calculated:
(1) industrial UHT-PBR (20 ha)
(2) industrial UHT-PBR (20 ha) with biomass valorisation and nutrient recycling by HTL
(3) industrial UHT-PBR (20 ha) with HTL process and renewable (Norwegian) energy mix

The upscaled system is based on the concept and design of the well-established pilot scale cultivation unit at A4F with a continuous process and daily product (ethanol) harvesting and on confidential information on the upscaling of the UHT-PBR system for ethanol provided by A4F. The land occupied by the reactor was assumed to be 20 years in use and classified as industrial area. The considered system is overall based on theoretical estimates. For a final cultivation scale, three consecutive production volumes have to be achieved. Precisely, the underlying system is composed of three production steps: Pre Inoculation UHT-PBR, Inoculation UHT-PBR and Production UHT-PBR. The occupied area as well as the associated volume and number of units is listed in Table 1. However, butanol is continuously excreted from the cells and separated from the culture ('milking') so that the inoculum production is of limited importance for the overall impacts. During the cultivation period, no maintenance and breakdowns were considered

Table 1: UHT-PBR setup for the 20 ha cultivation system for 1-butanol production

	Amount	Volume [m³]	Area [m²]
Pre inoc. UHT-PBR	12	1	16
Inoc. UHT-PBR	4	21	590
UHT PBR	72	98	2,769
Total		7,115	201,924

Production and preparation steps providing less than 1 m^3 of culture were neglected and considered as lab work outside the system boundaries. Main materials of the production system were taken into account without assembling, forming and construction processes. Whenever we used catalogue data on electric devices like the blower, we assumed 70% of the total mass being stainless steel, only. The main materials used within the cultivation phase are listed in Table 2.

Table 2: Main components and specifications for the 1-butnaol production system (Inventory)

Component	Material	UHT PBR [kg material/a]	Inoc UHT PBR [kg material/a]	Pre-inoc. UHT PBR [kg material/a]	Assumption
Tubing	PMMA	15,577	541	122	A4F
Circulation pump 1	Stainless steel			5	Lowara CO(M) 350/05
Circulation pump 2	Stainless steel		9		Lowara ESHS 50-160/75
Circulation pump 3	Stainless steel	2,036			Lowara FHS-FHF 125-200/450 45 KW
Harvesting pump	Stainless steel	113			Lowara FHS-FHF 125-200/450 45 KW
Nutrient pump	Stainless steel	0.4			Lowara CO (M) 350/03
Blowers	Stainless steel	6,143	73	11	MANVAC 220V Single Phase Blower LD 015 H21 R15
U-Bends	Borosilicate glass	2,808	33	5	A4F
Support	Cast iron	61,776	732	106	A4F
Fittings	Rubber	3,510	42	6	A4F
Manifolds	Stainless steel	9,477	112	16	A4F
Tank	Stainless steel	111,618	1,323	191	A4F
Thermal regulation coil	Stainless steel	24,921	295	43	A4F
Tube connectors	PP	2,106	25	4	A4F
CO_2 circuit	Polyurethane	228	3	0.4	A4F

The system and equipment lifetime was considered to be 20 years. Pump work and culture bubbling was implied. Optimal pumps were selected using the flow rates (30 m^3/h, 83 m^3/h, 500 m^3/h) per reactor size given by A4F to ensure a culture speed of 0.5 m/s. No power was used for thermoregulation, only water was assumed to cool down the culture medium. Therefore, the energy consumption for the 360 days production can be depicted from Table 3. Sensors and controlling equipment were neglected in the LCA as well as connecting pipes between the different productions steps.

Table 3: Baseline assumptions for pumping energy inputs

Item	Amount	Capacity [kW]	Operating scheme [h/d]	Yearly energy consumption [kWh/a]	Assumption
Blower	1	1.5	24	18,442,909	scaled by volume, A4F
Circulation pump 1	12	0.55	24	57,024	Lowara CO(M) 350/05 0.55 KW
Circulation pump 2	4	7.5	24	259,200	Lowara ESHS 50-160/75 7.5KW
Circulation pump 3	72	45	24	27,993,600	Lowara FHS-FHF 125-200/450 45 KW
Harvesting pump	4	45	1	64,800	Lowara FHS-FHF 125-200/450 45 KW
Nutrient pump	1	0.37	1	133.2	scaled by nutrient flow

Numbers on operational materials like fertilizer as well fresh water for cleaning purposes, thermoregulation or fresh culture supply were calculated based on information provided by A4F. Like this, we considered 107 g N/kg DM biomass as $NaNO_3$ and 15 g P/kg DM biomass as P_2O_5. 62.5% of nitrogen and 90% of phosphate could be recycled within the HTL process, significantly reducing their demand from primary sources, which are either limited (phosphate rock) or linked to a high-energy demand (production of N). The LCA comprises 3 kg of CO_2 per kg of algae biomass produced which are released while burning the butanol in a vehicle engine or producing electricity from the bio crude oil.

As it is assumed that the system is running continuously, only one cleaning per year takes place, flushing the tubes with twice the water volume of the reactors and a solution with chlorine (7.020 kg) and thiosulfate (5.616 kg). In order to keep fresh water demand low, recycling of the culture medium is assumed to reduce the water consumption, which summarises including the water demand for the cooling system through evaporation up to 433,145 m^3 per year. As 90% of the culture broth harvested can be recycled and fed back to the PBR system total fresh water volume per year can be reduced to 82,625 m^3 per year.

The yearly 1-butanol production was considered to be about 2,000 t and 1,080 t of biomass as co-product since replacement of around 30 % of the culture every day was assumed. Differences in densities of biomass, butanol and culture medium were not considered for technical configuration and processing.

After the cultivation process in the UHT-PBR the product flow was separated through filtration into two different phases according to the information from ICL and A4F: a 1-butanol rich medium (ca. 65 vol.%) that was sent to the pervaporation unit and the biomass slurry (ca. 35

vol.%), which was harvested and concentrated using a polypropylene-microfilter (5 years lifetime). To harvest and separate 1-butanol, the pervaporation process was implemented based on data provided by ICL (see Deliverable 6.2). In order to match with the industrial scale, the data for the equipment and their energy consumption were linearly up-scaled. The culture regime was assumed to be continuous. Main equipment used for the pervaporation process is listed in Table 4.

Table 4: Main equipment for the pervaporation process to harvest 1-butanol from the product flow

Component	Material	Amount [kg material/a]	Assumption
Pre-Heater	Stainless steel	338	Tranter HE
Cooler	Stainless steel	338	Tranter HE
Heater 1	Stainless steel	338	Tranter HE
Condenser 1	Stainless steel	338	Tranter HE
Heater 2	Stainless steel	338	Tranter HE
Condenser 2	Stainless steel	338	Tranter HE
Pervaporator Filtration 1	Polysulfone	115	ICL, own estimation
Pervaporator Filtration 2	Polysulfone	115	ICL, own estimation
Pervaporator pump 1	Stainless steel	13,234	Lowara FHS-FHF 125-200/450 45 KW
Pervaporator pump 2	Stainless steel	713	Lowara FHS-FHF 125-200/450 45 KW
Distillation Boiler	Stainless steel	338	Tranter HE
Distillation Condenser	Stainless steel	338	Tranter HE

The frequency of the partial harvests was determined to be daily and the harvested amount remained 30% of volume. For the pervaporation, the following energy inputs were considered in the LCA model (see Table 5).

Table 5: Energy inputs for the pervaporation process to harvest 1-butanol from the product flow

Item	Yearly energy consumption [kWh/a]	Assumption
Net of pre Phase (heating + condensing)	24,840	Upscale of ICL-Data
Net of pervaporation 1 (heating + condensing)	16,560	Upscale of ICL-Data
Net of pervaporation 2 (heating + condensing)	8,280	Upscale of ICL-Data
Pervaporator pump 1	2,111,400	Upscale of ICL-Data
Pervaporator pump 2	107,640	Upscale of ICL-Data
Net Distillation	8,280	Upscale of ICL-Data

According to the data provided by the Photofuel partners, the butanol concentration was 2.65 g/l. Parallel to the harvest, medium including fresh fertilizer was added to maintain constant conditions.

The butanol-rich flow in the system undergoes a combination of two pervaporation steps (Figure 1). Each pervaporation unit required a heater, a condenser, and a vacuum pump to reduce the outlet pressure. However, the vacuum pumps were not integrated in the model. All aqueous streams are being recycled to minimize the amount of required fresh water and to recover all residual butanol.

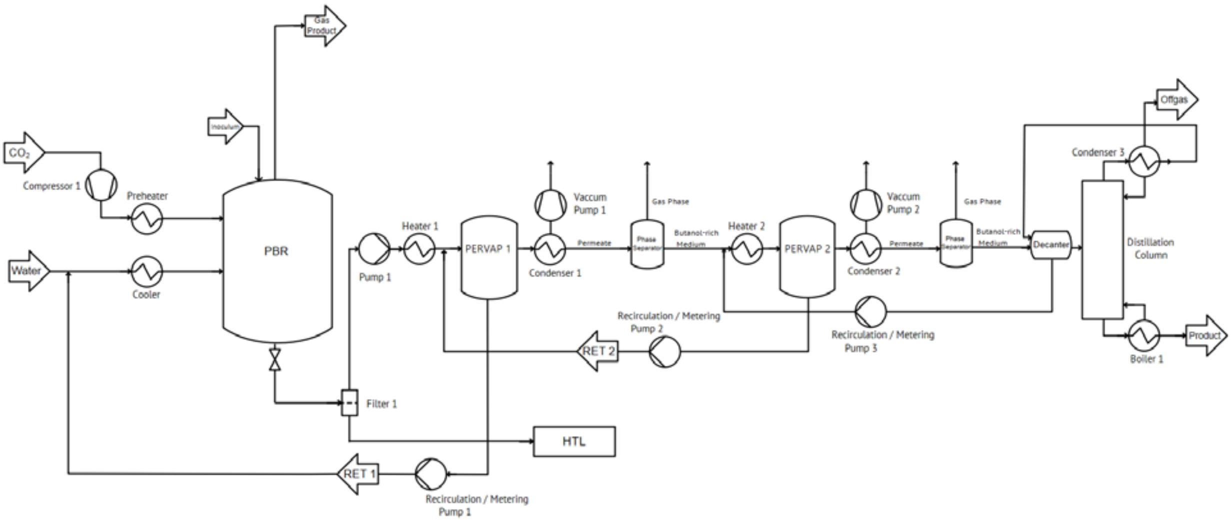

Figure 1: Flow diagram of the 1-butanol production process including the HTL process

Pre-studies showed, that biomass valorisation should be included to improve the overall Photofuel process e.g. by recycling of nutrients as well as increasing the energetic output. We considered the hydrothermal liquefaction (HTL) process as the most suitable technology to convert the residual algae biomass since HTL in general is appropriate for the conversion of wet feedstocks [3]. Besides, with HTL a liquid energy carrier (so called biocrude oil) is produced, which can also be upgraded and used as fuel. Although the majority of HTL research has been performed using small batch reactors, typically a few hundred milliliters in volume, the present LCA study comprises an up-scaled HTL-Process, which was designed according to literature. As a result, residual algal slurries with 20% dry matter (DM) content were processed and converted trough high temperature (350°C) and pressure (210 bar) reaction processes into four streams. A generic organic co-solvent (1,1 dimethylcyclopentan) is also included in order to support the separation of the bio-oil from the other products. The solvent flowrate was set to 10% of the total flowrate entering the HTL process (ICL). Figure 2 shows the HTL process as used in our model, the product yields and the nutrients amount in each of them according to data from a case model by [3]

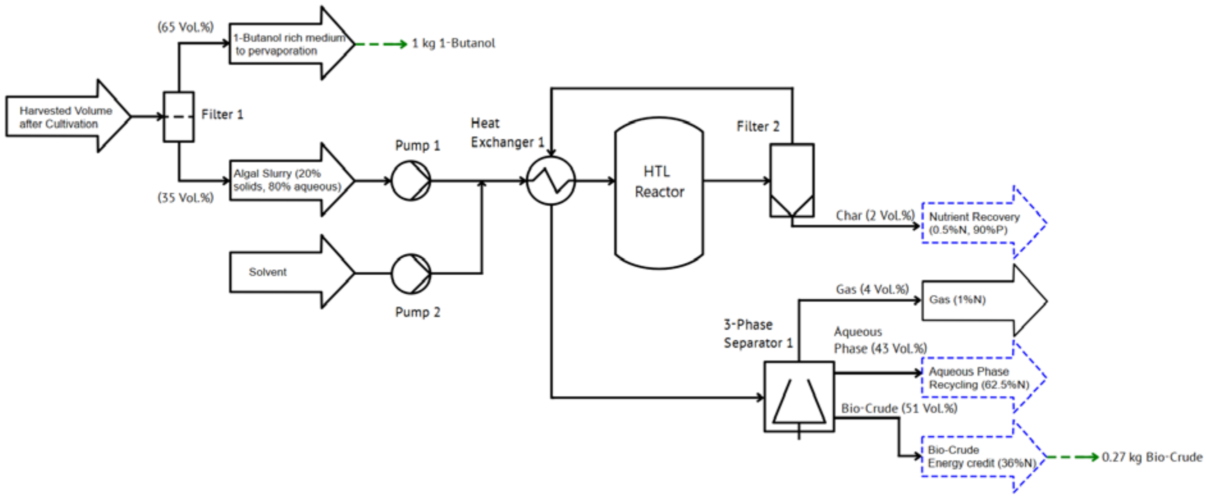

Figure 2: Flow diagram of the HTL process as implemented in the LCA model (recycled flows in colors)

A list of the equipment used in the HTL phase is given below (see Table 6). All equipment inputs were scaled according to the flows that have to pass the system. After the HTL process, the product flow is separated using a ceramic filter as a first step and a 3-phase separator subsequently.

Table 6: Components for the hydrothermal liquefaction (HTL) process

Component	Material	[kg material/a]	Assumption
Reactor	Stainless steel	3.85	ICL, own estimation
Prinicipal pump	Stainless steel	10.91	ICL Dosing pump LDE3_LEWA
Solvent pump	Stainless steel	11.6	ICL, Syringe pump ISCO 1820
Solvent tank	Stainless steel	32.5	own estimation
Heat exchanger	Stainless steel	1.47	Tranter HE
3-Phase Separator	Stainless steel	13.65	Flottweg Separator
Ceramic filter	Ceramic Al_2O_3	0.00002	own estimation

The HTL product contains 51 vol.% bio-crude, 43 Vol.% of aqueous phase, 4 Vol.% of product gas and 2 Vol.% of solids. Figure 2 shows the nitrogen balance in the product streams as estimated from experimental results according to the literature [3]. The nitrogen and phosphorus bound in the solid and aqueous phases are internally recycled and used as a credit in the model reducing the external nutrient demand during cultivation. The HTL solids would normally require a conversion step (such as acid digestion) to make it bio-available before re-use [3]. However, any further processing of nutrients to enhance bioavailability were considered outside the system boundaries.

For the HTL the following energy inputs are considered (see Table 7).

Table 7: Energy inputs for HTL process

Item	Yearly energy consumption [kWh/a]	Assumption
Prinicpal pump	47,520	own estimation
Solvent pump	864	own estimation
Heat exchanger	1,598,903	own estimation
3-Phase Separator	47,520	own estimation

The bio-crude oil produced (0.27 kg per 1 kg 1-Butanol produced), with a High Heating Value (HHV) of 39 MJ/kg (ICL), is implemented in the model as an energy credit along the butanol production. In the LCA model it is assumed that this bio-crude oil is being used to produce electricity with a conversion efficiency of 40%. The produced gas is not recycled or used as its amount is negligible (4 vol.%).

3.2 LCA results for the production of 1-butanol

The results of the LCA for the production of 1-butanol are depicted and described in the following. The results in the following paragraphs are displayed in the same way, always referring to the production of 1 kg of 1-butanol. The most relevant ILCD impact categories were selected. Main processes were considered as described above. Especially electricity, operational materials e.g. fertilizers and the embedded burdens of the materials used contribute to the impact assessment results. Processes contributing to less than 2% to each impact category were summarized to "others".

As can be derived from Figure 3 the highest share within climate change and fresh water eutrophication is resulting from the energy consumption especially for cultivation (66% and 85% respectively). As expected, the land use category as well as the mineral depletion category are mainly related to the infrastructure (83% and 71% respectively). Operational materials contribute to the biggest share in water depletion (72%), as here fresh water inputs for cooling as well as for medium preparation are considered. Electricity and infrastructure equally drive the particulate matter formation. Additionally after the main contributors (electricity and infrastructure), fertilizer inputs represent a major share in the overall impacts.

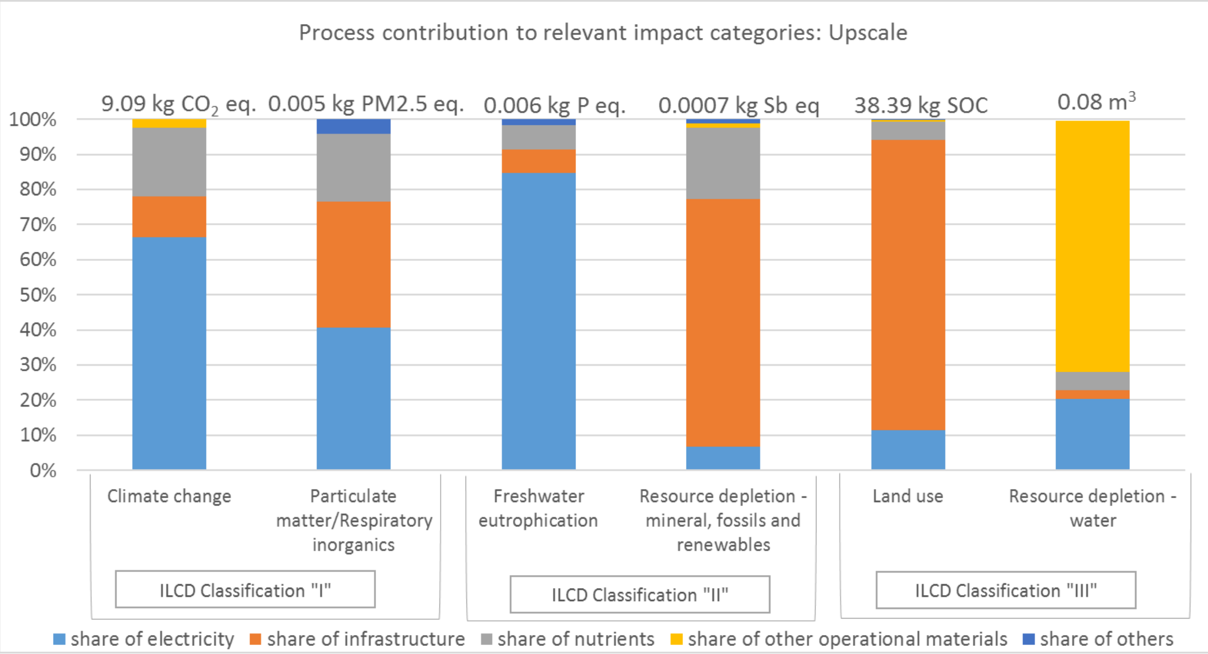

Figure 3: LCA results for 1-butanol production with *Synechocystis PCC6803* in UHT-PBR (Scenario 1).

The second scenario (see Figure 4) includes with the HTL process a further downstream step to valorize the biomass as well as recycle nutrients. Climate change is now slightly better than in scenario 1, while the kg of SOC in "land use" is increasing. However, the pattern of main contributors to the impacts remains the same. The expected improvements with the HTL can be seen at the important reduction of the absolute results especially related to the significant reduction of nutrients due to high recycling rates.

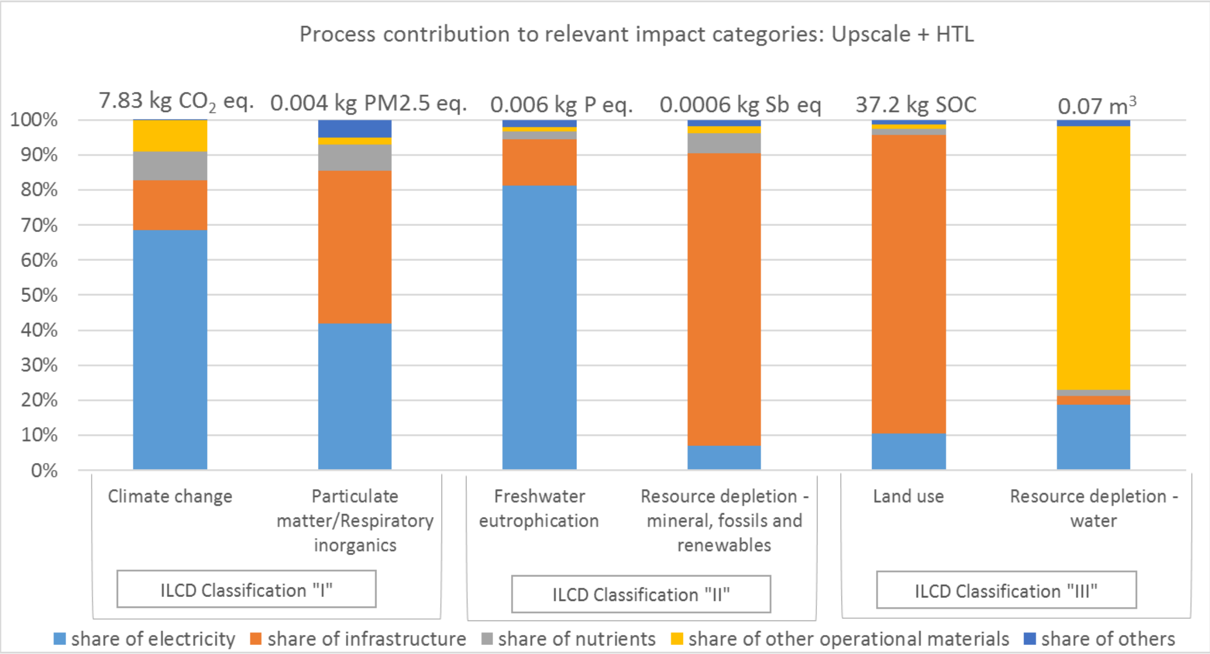

Figure 4: LCA results for 1-butanol production with *Synechocystis PCC6803* in UHT-PBR (Scenario 2)

In the third scenario (see Figure 5), the standard European electricity mix has been substituted by a renewable energy mix, based on the conditions of Norway, where hydropower is dominating the market (98%). In this way, the overall results improve for all categories except water depletion. Here, the methodology of including water used in turbines, might be misleading and could be further discussed. If the electricity was set to a Norwegian scenario, the impacts of infrastructure showed up and dominated the results.

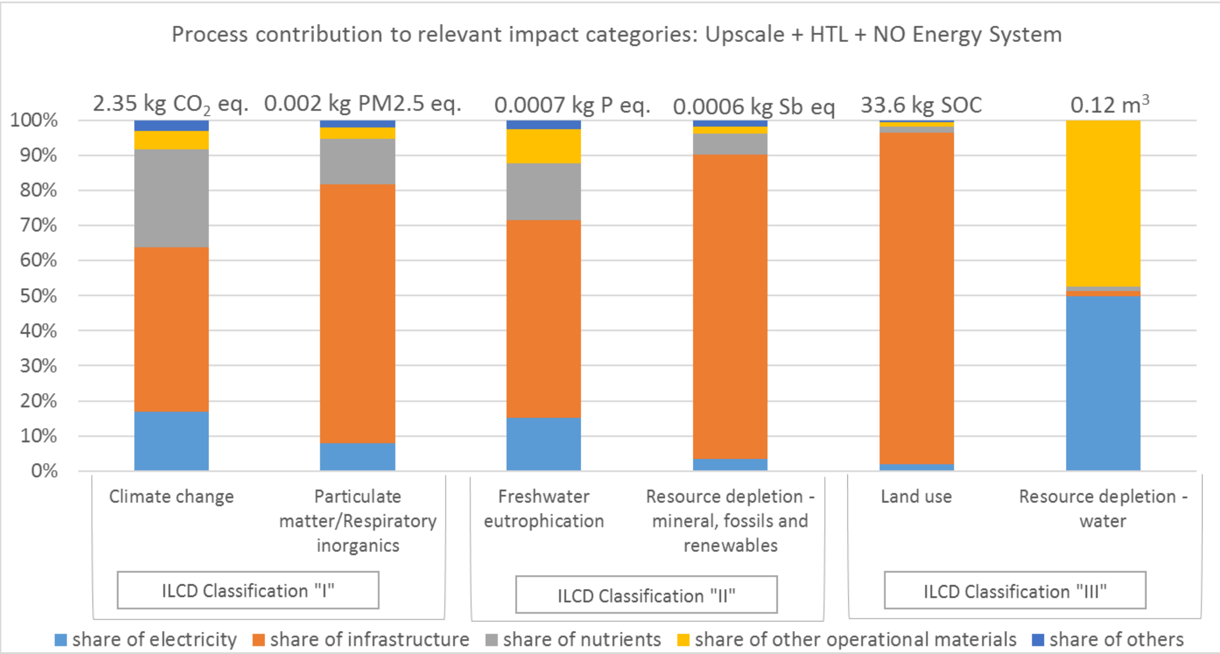

Figure 5: LCA results for 1-butanol production with *Synechocystis PCC6803* in UHT-PBR (Scenario 3)

3.2.1 Comparison of scenarios and fossil reference

The importance of this future-oriented approach, analyzing different scenarios with respect to technology development and design (lab and pilot scale), becomes visible by the comparison of the scenarios (Figure 6). The normalized results of the scenarios in relation to the results of the fossil reference are presented in Figure 6. The black bars indicate the fossil reference system (1 kg of 1-butanol from hydroformylation). The LCA results indicate that the Norwegian scenario can environmentally compete with the fossil reference in terms of environmental burdens for climate change (Upscale + HTL + NO: 2.35 kg CO_2 eq/kg biobutanol versus fossil reference: 2.45 kg CO_2 eq./kg 1-Butanol) and freshwater eutrophication (Upscale + HTL + NO: 0.0007 kg P eq./kg biobutanol versus 0.0008 kg P eq./kg 1-butanol). The result show, that there is a perspective for further optimizations of the biocatalytic production of biobutanol by cyanobacteria.

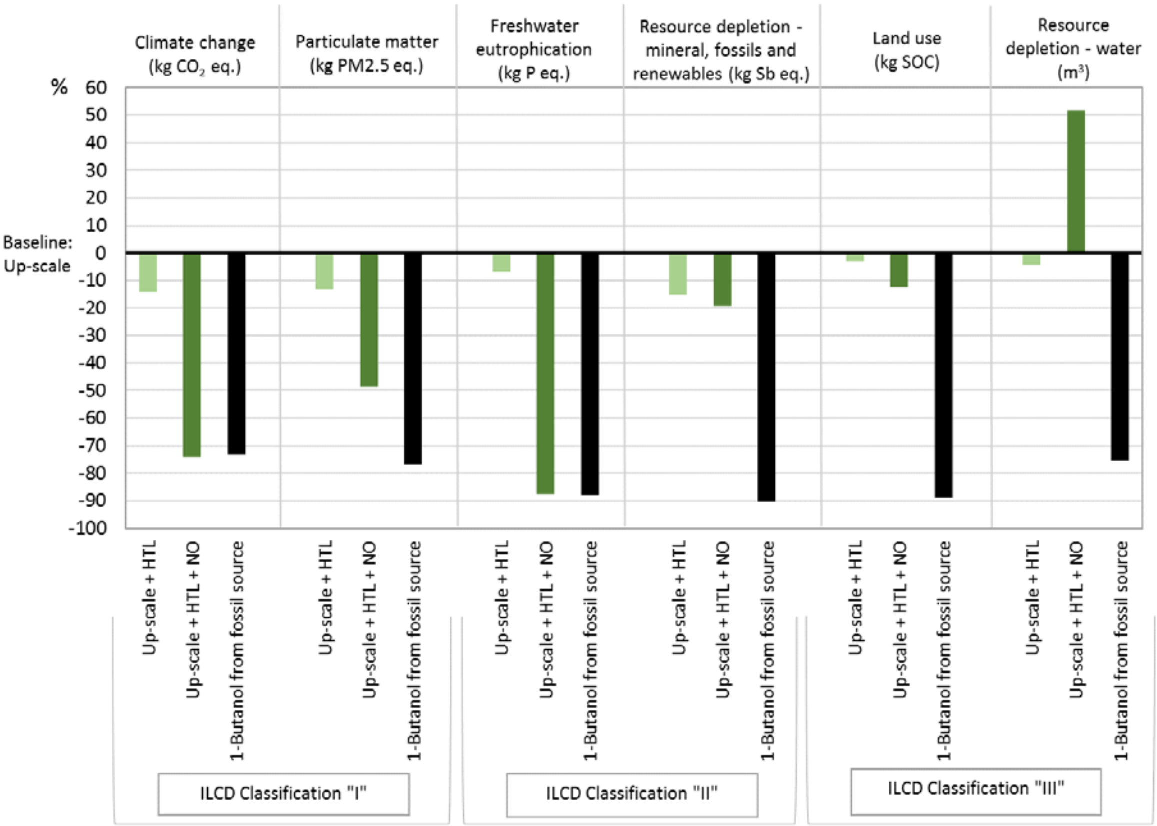

Figure 6: Comparison of absolute results by scenario and fossil reference, normalized to the upscaled scenario

Another result of the LCAs is the identification of the process hotspots, which should be addressed for future improvements. These are mainly related to electricity demand, infrastructure and fertilizer inputs during the cultivation of the cyanobacteria.

During data collection, we were facing challenges, either because some parameters were not measured and available or data were not applicable for upscaling to fit an industrial scale of 20 ha size.

Another main factor, which should be addressed, is the modular structure of the LCA model. Some production steps are on different technology readiness levels and developed for other application. Therefore, it is a methodological challenge to integrate the individual process steps into the LCA as this is only possible with assumptions and high uncertainties about the quality of data. Besides, the scale of the considered systems can hardly be matched to one ideal system and possible savings and improvements might still occur when a complete process chain is built up in practice.

Nevertheless, LCA approaches for immature technologies are important to show possible directions for technology development and system design as well as the systematic and coherent identification of environmental hotspots and drivers.

Future investigations should improve the approach and focus on complete process pathways with scaled equipment. It can be concluded that the LCA results contribute to the discussion on the environmental feasibility of an innovative technology to produce a liquid energy carrier based on a biocatalytivc process.

The upscaling approaches and scenario analysis indicate that the biobutanol production could become competitive to the fossil reference under the described conditions. Future genetic modification of cyanobacteria could further increase productivities of biobutanol and in combination with improvements of the technical settings and design of the production process, cyanobacteria could deliver a puzzle stone in meeting the EU targets for biofuels within the energy sector.

3.3 LCA-results of using 1-butanol in cars

The gasoline test fuel 1 (But45) is composed of 45%vol butanol and 55%vol gasoline. Gasoline used is low-sulphur petrol. The gasoline test fuel 2 (But55) is composed of 55%vol butanol and 45%vol gasoline, with the same inputs for gasoline and butanol. Main characteristics of But45 and But55 fuels are presented in Table 8.

Table 8: But45 and But55 fuel properties from own calculations

		Butanol	But45	But55
LHV (MJ/kg)		33	38.4	37.4
Density (kg/m^3)		0.810	0.775	0.782
Volume proportion (%)	Gasoline	0	55	45
	Butanol	100	45	55
Mass proportion (%)	Gasoline	0	47	57
	Butanol	100	53	43
Fossil carbon content (% total carbon)		0	60	50

Two reference fuels are assessed: E10 (90%vol gasoline and 10%vol ethanol) and E25 (75%vol gasoline and 25%vol ethanol). In both cases, gasoline used is low-sulphur petrol. Data used is: “Petrol, low-sulfur {Europe without Switzerland}| market for | Cut-off, U”. Petrol is one of the many coproducts of the refinery process in Ecoinvent, which delivers these other coproducts: bitumen, diesel, light fuel oil, heavy fuel oil, kerosene, naphtha, propane/ butane, refinery gas, secondary sulphur and electricity.

Ethanol is produced from sugar beet. Economic allocation is done between the three coproducts *ethanol*, *beets chips* and *vinasse* of the multi output process ‘sugar beets, to fermentation‘. A corrective term to compensate the carbon balance is also included, as the carbon content of the coproduct is not proportional to the economic values. Data used is: “Ethanol, without water, in 95% solution state, from fermentation {CH}| ethanol production from sugar beet | Cut-off, U”.

Main characteristics of gasoline, ethanol, and E10 and E25 fuels are presented in Table 9.

Table 9: Gasoline, ethanol, and E10 and E25 fuel properties, from (ref) and own calculations

		Gasoline	Ethanol	E10	E25
LHV (MJ/kg)		43.2	26.8	41.4	38.9
Density (kg/m^3)		0.745	0.794	0.75	0.758
Volume proportion (%)	Gasoline	100	0	90	75
	Ethanol	0	100	10	25
Mass proportion (%)	Gasoline	100	0	89.4	73.7
	Ethanol	0	100	10.6	26.2
Fossil carbon content (% total carbon)		100	0	93	82

In line with the new Renewable Energy Directive proposition [6] for the period 2020-2030, no Land Use Change (LUC) emission factor is included in the calculation of climate change impact of first generation biofuels, even if the Directive (EU) 2015/1513 [7] recognizes that indirect land- use change is capable of negating some or all greenhouse gas emissions savings of some biofuels. However, research has shown that the scale of the effect depends on a variety of factors (type of feedstock used for fuel production, the level of additional demand for feedstock triggered by the use of biofuels…), and so single emission factors are not directly included in the greenhouse gas emission calculation methodology. However, LUC emission factors are proposed in this directive, to prepare for the transition towards advanced biofuels and minimize the overall direct and indirect land-use change impacts. Therefore, sensitivity analyses are performed in section 3.4 to include the effect of LUC on climate change for bioethanol production.

3.4 Comprehensive LCA-results (from well to wheel)

Impacts generated in the transportation on 1 km with the different fuels assessed are shown in Figure 7. For each fuel the two driven cycles (WLTP and NEDC) are assessed. The ILCD method [2] has been used to assess potential environmental impacts. The selected impacts are: climate change / freshwater eutrophication / land use / particulate matter / mineral, fossil and renewable resource depletion / water depletion. For the references fuels (E10 and E25), the impacts increase with the proportion of ethanol in the fuel for freshwater eutrophication, land use, mineral, fossil and renewable resource depletion and water depletion. On the contrary, the impacts decrease for climate change (due to the climate neutral effect of biogenic carbon) and for particulate matter.

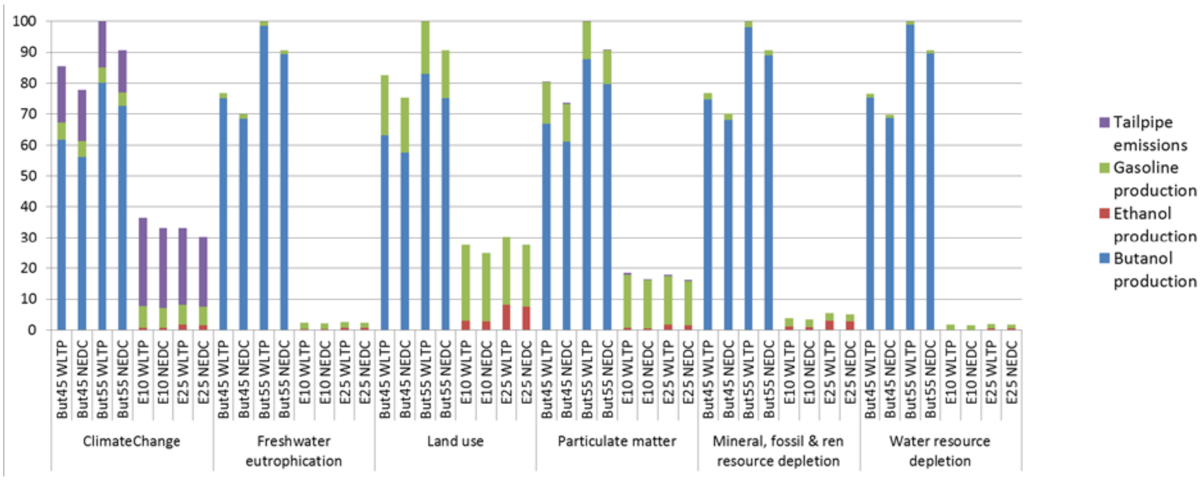

Figure 7: Comparison of impacts generated by 1 km transportation with the different assessed fuels

Figure 7 presents the contribution of each process to the various environmental impacts with a contribution analysis based on the step in the Life Cycle (butanol / ethanol production, gasoline production and tailpipe emissions). Fuels from microalgae (But45 and But55) are characterized by significantly higher impacts than other biofuels (here E10 and E25) in all the assessed impact categories. Butanol production step is the highest contributor in the But45 and But45 fuels, for all the assessed impacts. This step is even higher than the overall contributions of the other fuels for all the impacts. The contribution analysis of the butanol production step is extensively described in section 3.2. The impacts increase with the proportion of butanol in the fuel. Another general pattern is the higher impact of WLTC compared to NEDC. This is in line with results from European Commission on CO_2 emissions of light-duty vehicles [3]. This is due to both higher emissions and higher fuel consumption per km (see Annex 1).

The comparison of technologies in different stages of development (here butanol from algae vs. first generation ethanol and gasoline), can end up in incomplete analysis. In fact, most reference technologies are mature and have been optimized over decades, while, low TRL processes usually have higher impacts. However, a comparison between a low TRL CCU technology and a high TRL reference technology still reveal valuable insights to guide research. Furthermore, modelling approaches could be used to represent future systems when data is not readily available [8]. Industry cost-curves [9], dynamic interaction simulations with agent-based models [10] or models of innovation diffusion at macro and micro-level [11] are some examples of approaches that have recently been explored. Most of current research policies recommend the use of the LCA methodology at the early stages of development to decrease the environmental impacts of future technologies. Therefore integrating specific aspects related to emerging technologies (like data availability or rapid technology changes), could be adapted to complete classic sensitivity analyses.

3.5 Focus on climate change

Climate change impact for the different fuels ranges from 139 g CO_2eq/km (E25 NEDC) to 409 g CO_2eq /km (But55 WLTP) and are presented on Figure 8:

- For microalgae fuels, values are between 338 g CO_2eq /km (But45 NEDC) and 432 g CO_2eq /km (But55 WLTP).
- Reference fuels (E10 and E25) range from 139 g CO_2eq /km (E25 NEDC) to 169 g CO_2eq /km (E10 WLTP),
- 100% fossil references [3] are 168 g CO_2eq /km (NEDC) and 194 g CO_2eq /km (WLTP).

As in the global impact picture, microalgae fuel are hampered by the high share of climate change impact due to the butanol production. However, due to the higher share of biofuel in the microalgae fuels (45% and 55% compared to 10% or 25%), contribution of tailpipe emissions are the lowest of the assessed fuels. Therefore improvements of the butanol production could lead to important reduction of GHG emissions.

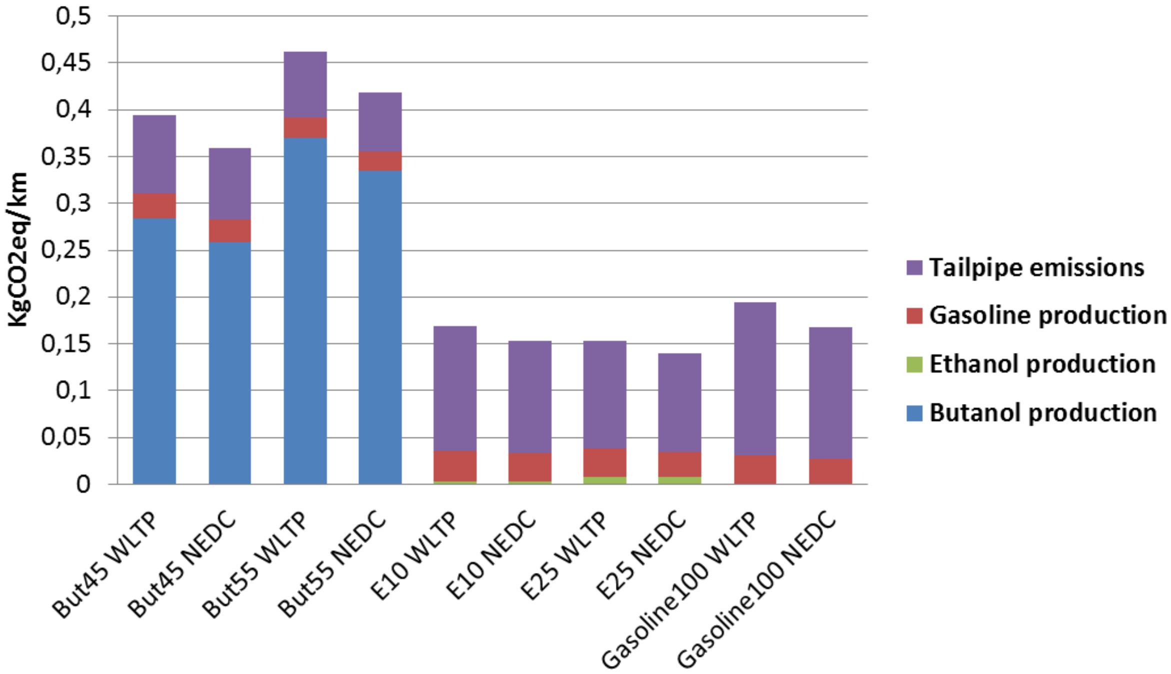

Figure 8: Climate change impact of the assessed fuels

3.5.1 Sensitivity analysis

Sensitivity analysis are performed on both reference fuels inputs and microalgae fuels inputs:

- For reference fuels ('E10 and E25), two sensitivity analysis are done: the inclusion of LUC emissions factors and the use of corn instead of sugar beet as feedstock for ethanol production.
- For microalgae fuels (But45 and But55), the electricity mix was changed from European grid mix to the highly renewable Norwegian grid mix

As the ranking of the fuels are the same for NEDC and WLTP driven cycles (Figure 8), these sensitivity analysis are only presented for the WLTP driving cycle for the sake of clarity and simplicity. Results on climate change with the inclusion of electricity mix from Norway, corn feedstock for ethanol production and with the LUC emissions factors are presented on Figure 9.

- LUC emission factors

As already pointed out, this study does not account for indirect land use changes; however opposite to other biodiesel feedstock, it is very likely that algae production facilities will not be installed on arable lands and hence will not create significant indirect land use changes. On the contrary, considering indirect land use changes would probably increase climate change impact [12] and biodiversity damages [13] of other first generation biodiesels. LUC emissions factors values are taken from the REDII [6]: 13 g CO_2eq /MJ for ethanol (feedstock group sugar beet and corn here). Results are presented on Figure 8. The inclusion on LUC emissions factors has no significant influence on the GHG emissions: only + 1.6% in E25 from sugar beet and + 2.8% in the E25 from corn.

- Ethanol production

In the reference case studies, sugar beet is used as feedstock for the ethanol production. In this section, ethanol is produced from corn, using data from the Ecoinvent 3.4 database [1]. Results are presented on Figure 9. The use of corn instead of sugar beet has no significant influence on the GHG emissions: only + 2% for E10 and + 6% for E25.

- Electricity mix

Electricity mix has an important potential on the reduction of the upstream impacts of butanol production, as it is presented in Figure 5. When focusing on climate change, the use of the Norwegian electricity mix (around 30 g CO_2eq / kWh) for the butanol production instead of the European one (around 500 g CO_2eq / kWh) leads to very strong reduction of GHG emissions: But45 are reduced by almost 50 %, and But55 by more than 55%. Consequently, with a very low carbon electricity source, But45 and But55 assessed fuels compete with the E10 and E25 reference fuels.

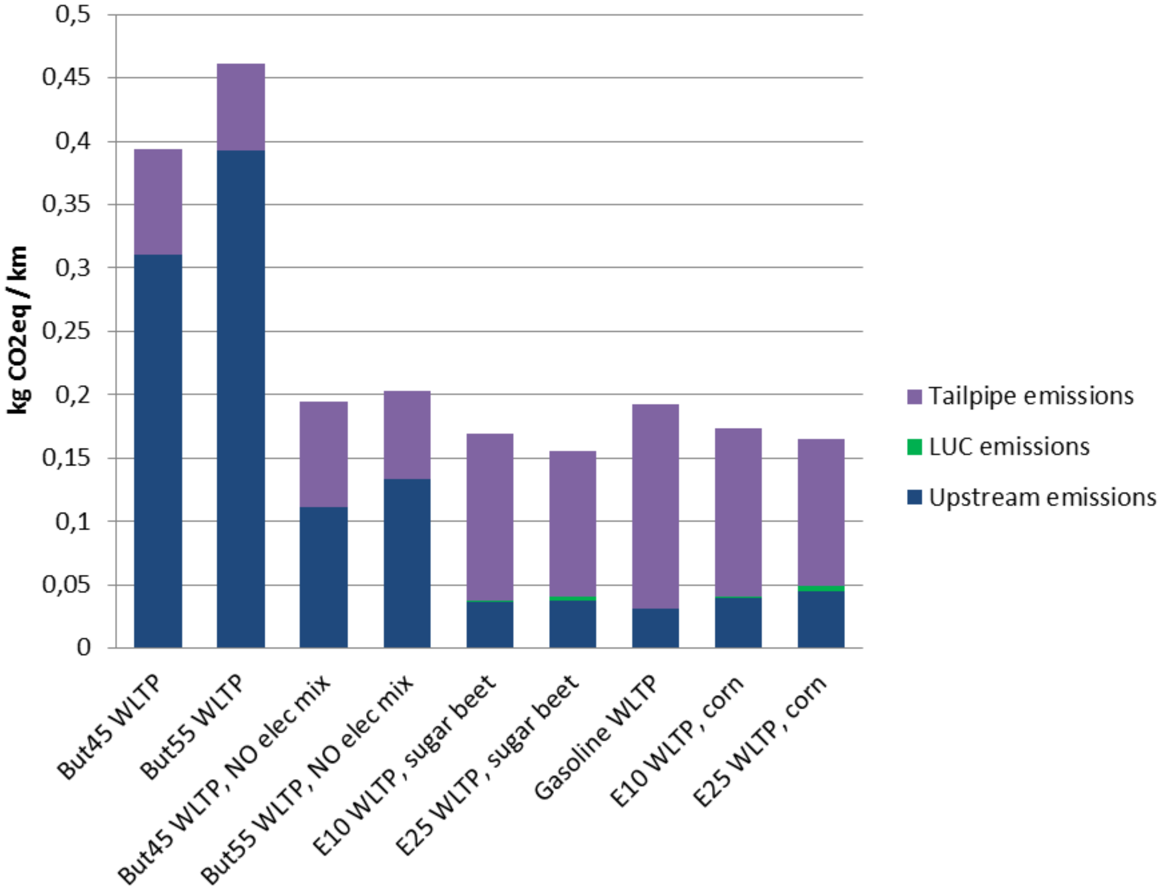

Figure 9: Sensitivity analysis on ethanol resource, land use change and electricity mix

4. Reference process: TFA production with Nannochloropsis

The reference process to 1-butanol production with genetically modified cyanobacteria is the production of fatty acids with the oil-accumulating and naturally-occurring microalgae species *Nannochloropsis oceanica*. Information and data on the process design and LCA model, as well as numbers for productivities at pilot scale, were received from UniFi based on two experimental runs (summer/winter) in a 1.4 m³ Green Wall Panel (GWP) pilot facility. In the large-scale setting modelled for the LCA scenario, the cultivation unit was assumed to hold 6,304 m³ (8 independent modules per ha). The concentration of TFA in the reactor by the end of the cultivation cycle is 1 g/l culture and the ratio of biomass to TFA is 2:1. The average productivity is 38 mg TFA/l culture/day, or 300 tons of total fatty acids (TFA) and 600 tons of biomass per year.

As with the three scenarios for 1-butanol production, data given for the pilot scale were extrapolated to a 20 ha large-scale system with 360 d production. However, the process is quite different to the continuous production of butanol. Two phases can be distinguished: The green phase (7 days), in which the biomass production is produced with nitrogen supply; followed by a nitrogen starvation phase to promote lipid accumulation in the biomass, also known as the yellow phase (16.5 days). Hence, an average cycle was assumed to be 23.5 days allowing for 15 full cycles per year.

4.1 System boundaries and process design

The system boundaries of the *Nannochloropsis* LCA begin with cultivation within the Green Wall Panel PBR and end at with the separated final product of TFA. The processes required after cultivation to obtain the final TFA product include culture centrifugation, cell disruption via homogenization, TFA extraction using solvent, and another centrifugation process using a three-phase separator. These processes are explained in detail in the following paragraphs. Additionally, a schematic overview below serves to visualize the processes. The functional unit is 1kg TFA.

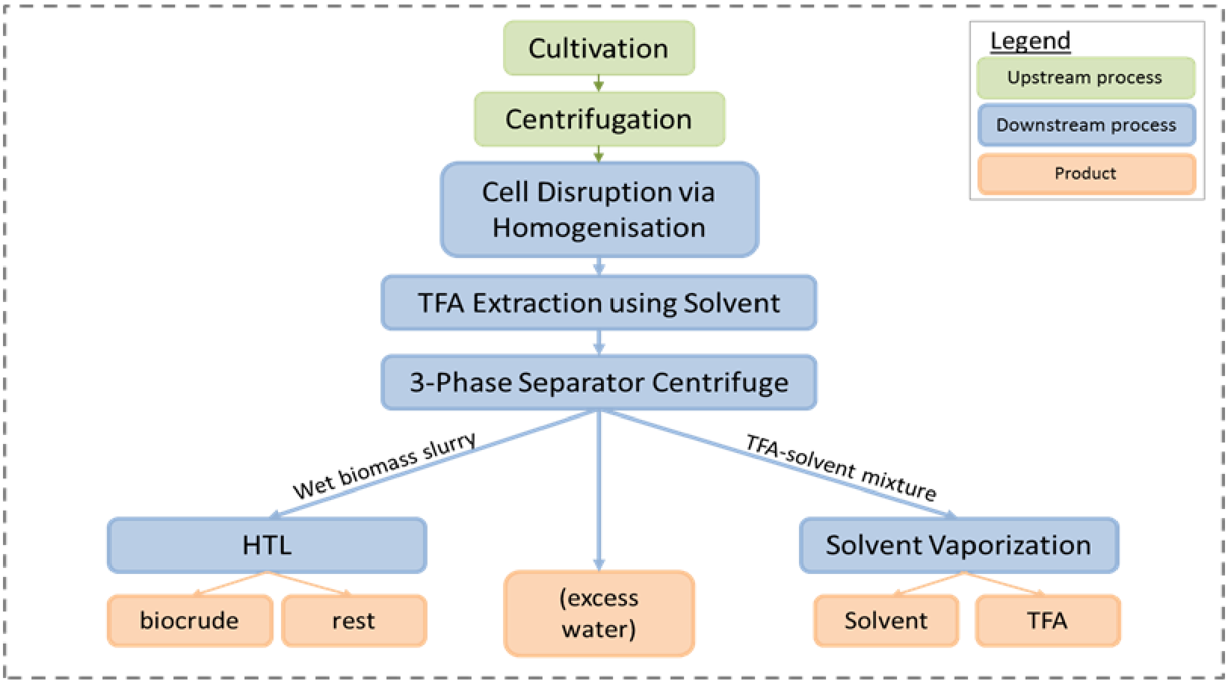

Figure 10: Schematic overview of TFA production with *Nannochloropsis oceanica*

The detailed LCI of the equipment and energy used for the production could be obtained by [4], which describes a 1 ha pilot facility. To reach a scale of 20 ha, material inputs were linearly upscaled. For electricity inputs, savings towards the large scale of 15 % were considered. A list of electricity inputs for cultivation is given below (see Table 10).

Table 10: Electricity inputs for cultivation

Item	Yearly Energy Consumption (kWh/a)	Assumption
Blowers	3,283,992	Tredici et al. 2016, UNIFI estimation (15% saving)
Submersible Pumps	642,600	Tredici et al. 2016, UNIFI estimation (15% saving)
Open Impeller Pumps	893,520	Tredici et al. 2016, UNIFI estimation (15% saving)
Centrifugal Separators	581,400	Tredici et al. 2016, UNIFI estimation (15% saving)

During cultivation, seawater (8,081 m^3/a) is pumped (37,800 kwh/ha/a) to cool down the culture medium. Optimal culture process conditions include the input of nutrients: 2.96 kg sodium nitrate/kg TFA and 0.28 kg sodium phosphate/kg TFA, plus 15.7 kg CO_2/kg TFA were fed to the culture.

After the two-phase cultivation of the *Nannochloropsis oceanica* in the GWP reactor, the cells are disrupted with a homogeniser. The materials and electricity inputs for cell disruption are depicted in Table 11 and Table 12, respectively.

Table 11: Material inputs for homogenization of *Nannochloropsis oceanica*

Component	Material	(kg material/a)	Assumption
Homogeniser	Stainless steel	18.025	GEA Ariete NS2006
	Lubricating oil	1.5	GEA Ariete NS2006
	Lubricating water	1500000	GEA Ariete NS2006

Table 12: Electricity inputs for homogenisation of *Nannochloropsis oceanica*

Item	Yearly energy consumption (kWh/a)	Assumption
Homogeniser	416,666.67	GEA Ariete NS2006

The culture containing disrupted cells is then ready for TFA extraction. TFA are extracted using Methyl tert-butyl ether as organic solvent - in total 1,656.12 t/a. The slurry of TFA, biomass and solvent is then divided into excess water, a TFA-solvent mixture, and a watery biomass slurry using a three-phase separator centrifuge. The three-phase separator (Flottweg Separator: 37.975 kg stainless steel/a) uses 6,660 kWh/a. The LCA model assumes that 99% of the solvent can be recycled by vaporization, which consumes 666,666 kWh/a. Similar to the butanol model, a valorization of biomass and recycling of nutrients via HTL is assumed. While the equipment used is identical to the materials used within the butanol pathway, the electricity inputs changed due to a difference in flow volume and frequency of use (15 times/a). Thus the biomass treatment consumed 23,009 kWh/a.

4.2 LCA results of TFA production

The impact category results from the LCA for the three TFA scenarios are depicted as aggregated bar graphs below. The following table describes the contributing flows to each process category, which are seen in the aggregated bar graphs.

Table 13: Process categories and the contributing flows- for the aggregated bar graphs

Process Category	LCA Phase	Flow
electricity flows:	cultivation:	blower energy
		open impeller pump energy
		submersible pump energy
		centrifuge energy
	HTL:	heat exchanger energy
		principal pump energy
		3 phase separator energy
		solvent pump energy
		(-) biocrude credit
	downstream:	solvent evaporisation energy
		cell homogenisation energy
		TFA extraction energy
infrastructure flows:	cultivation:	Green Wall Panel
		Industrial land use
	HTL:	solvent tank
		reactor
		filter
	downstream:	cell homogeniser
		3 phase separator
nutrient flows:	cultivation:	sodium phosphate
		sodium nitrate
		ferric chloride
other operational material flows:	cultivation:	sodium hypochlorite for cleaning
		tap water for cleaning
	downstream:	cell homogenisation operational oil
		cell homogenisation operational water
		TFA extraction solvent

In the 20ha Upscale scenario (scenario 1), the largest contributing process to climate change, freshwater eutrophication, and resource depletion- water categories is electricity. For the cultivation phase, 21 kwh/kg/a are required, which is 85% of the total electricity demand of the TFA LCA. The other 15% are from the downstream processes and HTL. The share of infrastructure also contributes to climate change, particulate matter, and water resource depletion categories; this is largely due to the short lifespan and high demand for polyethylene bags, which are part of the Green Wall Panel process flow.

A large share of the land use impacts is related to “operational materials”: 68% of this impact is dedicated to the CO_2 fertilization to promote algae growth. As mentioned previously, the production of *Nannochloropsis* requires 15.7 kg CO_2/ kg TFA. The remaining 32% is from the cleaning materials (sodium hypochlorite, hydrochloric acid) required for GWP maintenance. These values do not change between scenarios because these flows do not change between scenarios, unlike nutrient input and electricity impact, which do vary between scenarios.

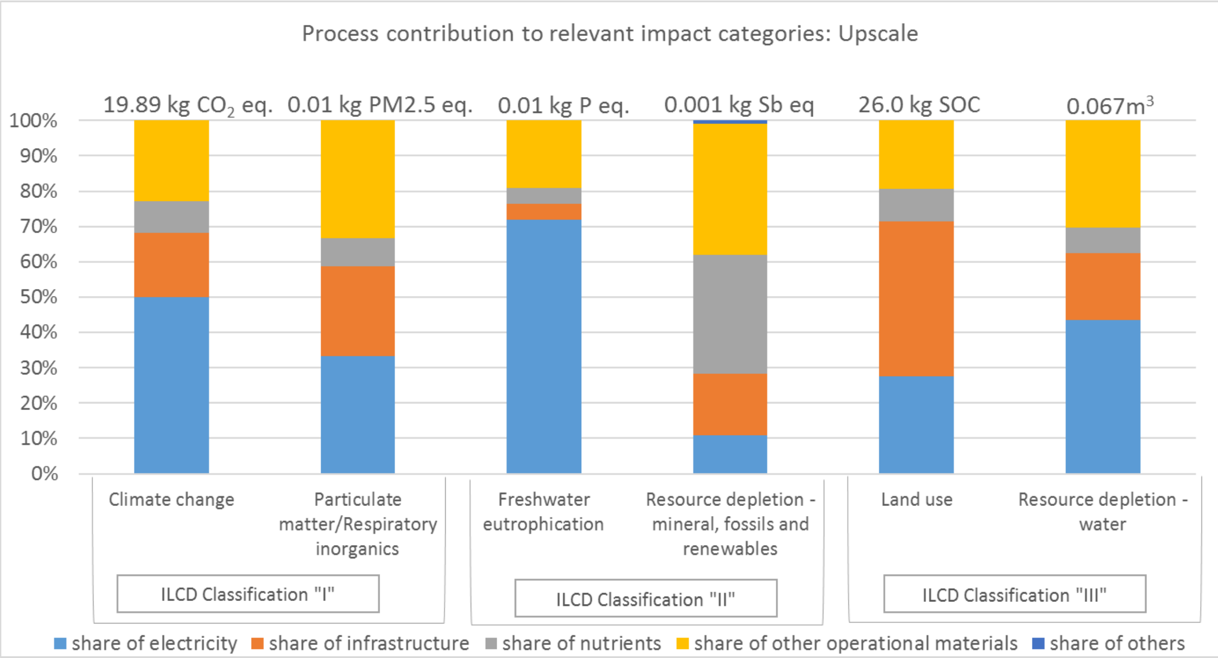

Figure 11: LCA results for TFA production with *Nannochloropsis oceanica* in a GWP-PBR (Scenario 1)

For the Upscale + HTL scenario (scenario 2, Figure 11), the share of nutrients is reduced from the previous scenario due to the recycling of nutrients from the HTL process. Although HTL does require some energy (0.076698061 kwh/kg TFA/a), it is a very small amount in relation to the energy credit gained from the biocrude via the HTL (2.17 kWh/kg TFA/a). Therefore, the overall climate change impact is reduced from the previous scenario but the shares stay relatively similar.

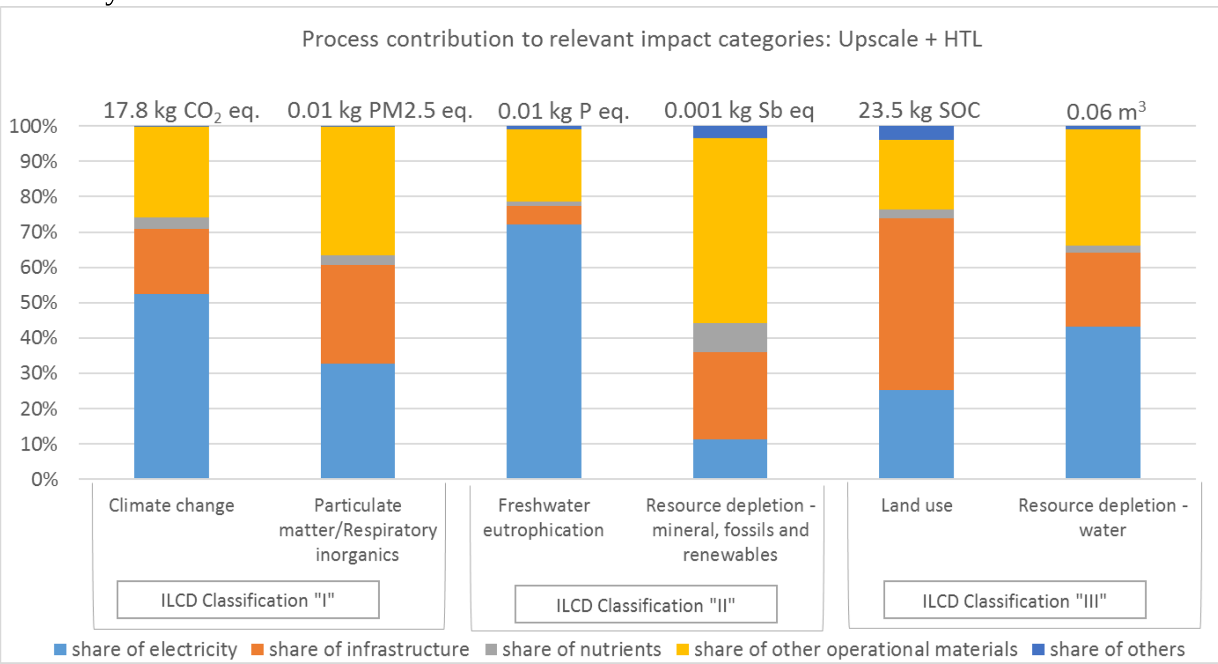

Figure 12: LCA results for TFA production with *Nannochloropsis oceanica* in a GWP-PBR and biomass use for HTL (Scenario 2)

For scenario 3, the share of electricity dramatically reduces (and therefore the total impact) in each impact category except for water resource depletion. This is due to the use of water as renewable energy in the Norwegian energy mix as already described in the butanol pathway (see section 3.2)

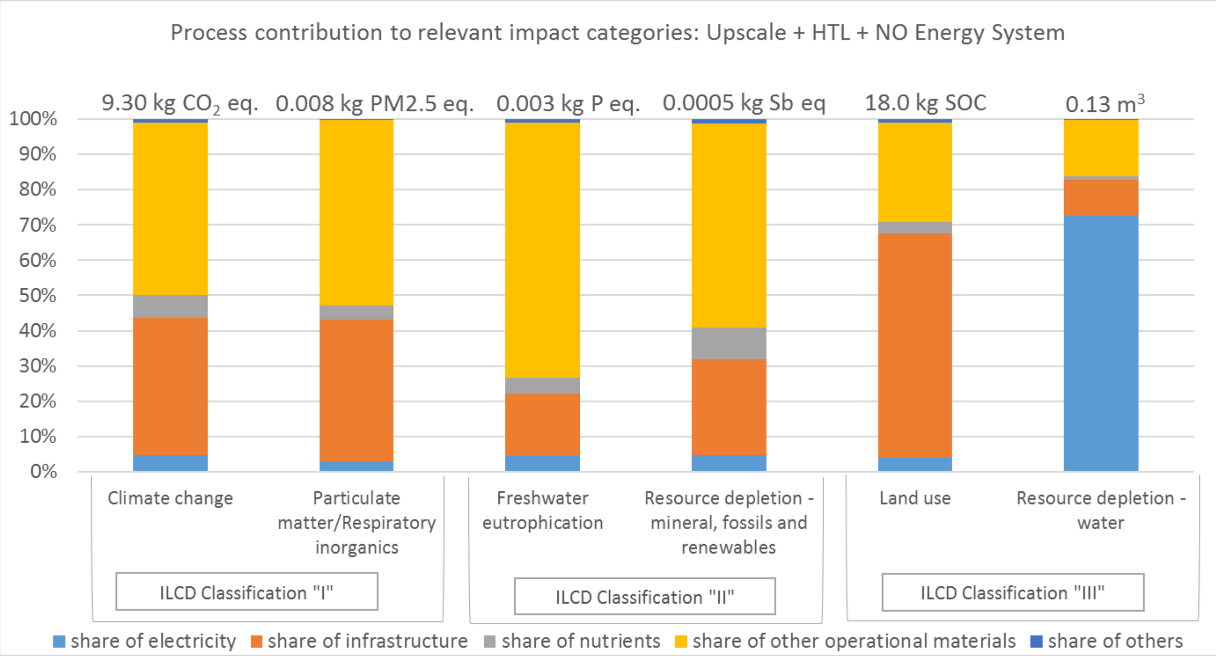

Figure 13: LCA results for TFA production with *Nannochloropsis oceanica* in a GWP-PBR including HTL and Norwegian power grid mix (Scenario 3)

5. LCA comparison of 1-butanol and TFA production

The following paragraph shows the comparison of results and critically discusses the way forward. The results are listed in Table 14 for1 kg of butanol and 1 kg of TFA produced in three similar scenarios. This is only indicative as the systems are hardly comparable because of different processes and equipment use, different operation schemes but also assumptions and level of details.

Table 14: Comparison of LCA results for the production of 1-butanol and TFA in different scenarios

ILCD Classification	Impact Category	Scenario	per 1 kg Butanol	per 1kg TFA
"I"	**Climate change (kg CO_2eq.)**	Up-scale	9.1	19.9
		Up-scale + HTL	7.8	17.8
		Up-scale + HTL + NO energy system	2.4	9.3
	Particulate matter (kg PM2.5 eq.)	Up-scale	0.0052	0.0123
		Up-scale + HTL	0.0045	0.0112
		Up-scale + HTL + NO energy system	0.0027	0.0077
"II"	**Freshwater eutrophication (kg P eq.)**	Up-scale	0.0061	0.0121
		Up-scale + HTL	0.0057	0.0108
		Up-scale + HTL + NO energy system	0.0008	0.0032
	Resource depletion - mineral, fossils and renewables (kg Sb eq.)	Up-scale	0.0007	0.0008
		Up-scale + HTL	0.0006	0.0005
		Up-scale + HTL + NO energy system	0.0006	0.0005
"III"	**Land use (kg SOC)**	Up-scale	38.4	26.0
		Up-scale + HTL	37.2	23.5
		Up-scale + HTL + NO energy system	33.6	18.0
	Resource depletion - water (m3)	Up-scale	0.0777	0.0676
		Up-scale + HTL	0.0745	0.0612
		Up-scale + HTL + NO energy system	0.1180	0.1250

Due to different processes and equipment use, different operation schemes, assumptions and level of details, the systems cannot be directly compared. However, we can see that the production of 1-butanol performs better in the climate change, freshwater eutrophication, particulate matter formation and resource depletion (mineral) impact categories. Butanol is worse in impacts concerning land use. This is because different PBRs are used for each algae and therefore require different materials, which vary in land intensity. The materials required for the precursor separation phases of butanol (pervaporation and distillation) are 13x greater than the precursor separation phases for TFA (cell homogenization and TFA extraction via solvent additional and centrifugation), even when considering higher areal productivity of butanol. This is due to the low efficiency of the pervaporation and distillation phases.

Compared to the results described in deliverable 6.2, process upscaling proved to be deliver high energy efficiency gains and leads to an important reduction of the impacts (177.8 vs. 7.06 kg of CO_2-eq in the small- and large-scale 1-Butanol scenarios). The LCA results for scenario 3 with the Norwegian scenario shows the impact of a change in the energy mix and how crucial it is to further improve the processes, not only by changing to a renewable energy mix but also to reduce energy inputs. Nonetheless, this Norwegian energy mix scenario still does not result in an upstream algae fuel precursor production with the expected superior environmental impacts compared to existing biofuel or even fossil fuel production.

6. Literature

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Annex 1: Summary of combustion emissions

Fuel type		Gasoline								Diesel								
Vehicule		Light duty								Light duty						Heavy duty		
Source		CRF (Delivrable 5.2)								Volkswagen delivrable 5.1						Volvo		
Blending		E10		E25		But 45		But 55		Reference		70/20/10		70/30		Reference	70/20/10	70/30
Cycles		NEDC	WLTP	NEDC	WLTP	NEDC	WLTP	NEDC	WLTP	NEDC	WLTP	NEDC	WLTP	NEDC	WLTP	VECTO	VECTO	VECTO
Emissions	HC (mg/km)	34	31	30	31	27	20	32	29	17	4	20	5	11	4	21	21	21
	CO (mg/km)	361	544	300	520	232	497	252	542	151	273	166	167	77	213	522	519	520
	NOx (mg/km)	11	30	9	27	7	23	10	32	77	55	72	41	67	37	584	582	583
	CO_2 (g/km)	129	142	128	141	126	139	125	138	85	89	84	88	84	88	762	758	755
	PM (mg/km)	0,5	0,7	0,6	0,5	0,2	0,3	0,1	0,3	0,09	0,04	0,01	0,1	0	0	1,3	0,8	1,1
	CH4 (mg/km)									3	2	5	2	3	2	0	0	0
	SO_2 (mg/km)									0,32	0,34	0,23	0,24	0,24	0,25	2,9	2,1	2,1
Consumption (L/100 km)		5,6	6,2	6,1	6,7	7,3	8	7,8	8,6	3,3	3,4	3,4	3,5	3,3	3,5	29	30	29