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Life Cycle Assessment for full chain CCU demonstration in the ALIGN-CCUS project –dimethyl ether and polyoxymethylen dimethyl ethers production from CO_2 and its usages in the mobility and electricity sectors

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Abstract

As part of three years of research and development in the European ALIGN-CCUS project, a CCU demonstration plant was erected at the lignite-fired block K of the RWE power plant at Niederaussem, Germany. It was the aim to demonstrate the full chain of CCU - beginning with the capture of CO_2 from the power plant flue gas stream, via the one-step synthesis of dimethyl ether (DME) through to its usage for electricity generation in a peak power diesel engine generator. A secondary target was the synthesis of polyoxymethylen dimethyl ethers (OMEs) and their use in the mobility sector.
A thorough Life Cycle Inventory was collected based on real process data for the first time, supported by process modelling with AspenPlus. This includes the inventory for construction and operation of all elements of the demonstrator chain: the power plant with its monoethanolamine (MEA)-based post-combustion capture facility, the newly developed synthesis unit based on a one-step reactor using a bifunctional catalyst (up to 50 kg DME per day) and the reconversion of the produced DME into electricity by an adapted diesel power generator (60-80 litre per hour DME consumption, output 240 kW_{el}). Additionally to the DME-route, the synthesis and usage of OME_{3-5} as a fuel in an adapted 2 l diesel motor for mobility application were part of the investigation.
The extensive inventory enabled a Life Cycle Assessment (LCA) according to ISO standards showing results for both applications. Benchmarking technologies like diesel- and e-mobility and gas turbines for peak power supply enable a comparative analysis identifying advantages and disadvantages of the CCU route. A sensitivity analysis is used to identify weak points along the routes together with future development targets and potentials.
To investigate a broad array of results, several varying scenarios, especially regarding energy supply were considered. The use of renewable electricity sources for electrolytic hydrogen production, but also for other processes of the CCU chain was investigated. A range of different impact categories were considered: Global Warming Potential (GWP), Particulate Matter Formation Potential (PM), Fossil Depletion Potential (FDP), Photochemical Ozone Creation Potential (POCP) and Terrestrial Acidification Potential (AP).
The results show that the investigated CCU technology routes are highly emission-intensive when using the current German electricity mix: the reuse of CO_2 and the reduction of fossil fuel consumption that goes along with it, is outweighed by the energy-intensive nature of the synthesis processes. Using renewable energy sources can drastically reduce GHG emissions, especially to a point where the implementation of a mostly renewable energy supply for the process chain offers a viable mitigation option compared to the investigated benchmark technologies. Findings regarding sensitive LCI data, development targets and technological potentials are discussed in the results.

Keywords: CCU; CCUS; Power-to-fuel; Life Cycle Assessment; LCA; dimethyl ether; DME; polyoxymethylen ethers; OME; CCUS; CCU demonstration

1. Introduction

The energy systems are currently transforming into configurations with a significant share of renewable energies to reduce GHG emissions and to protect the environment. At the same time, power supply and consumption must be kept in balance to maintain security of supply and to improve economic efficiency. To reduce the CO_2 emissions according to the Paris climate protection goals

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by up to 95% until 2050 (base year 1990), however, it is far from sufficient to replace conventional fossil-fired power generation by renewable energies. Without a strong contribution from all economic sectors these goals cannot be achieved. At the same time the fluctuating nature of renewable energies demands reliable, efficient energy storage systems and back-up power. These requirements offer an opportunity for coupling of the energy and the transport sector and are unlocking the potential for carbon capture and utilization (CCU).

The ALIGN-CCUS project united 30 research institutes und industrial companies in the shared goal of supporting the quick and cost-effective deployment of carbon capture, utilization and storage (CCUS), enabling Europe's industrial and power sectors to contribute to a low-carbon future while remaining economically viable [1]. Proving the feasibility and viability of a demonstrated full CCU-chain and the utilization of the CCU-products in the power and transport sector was an important aim of the ALIGN-CCUS project.

For the first time a full CCU-chain was demonstrated as an element for large-scale energy storage together with CO_2 conversion and CCU-fuel usage as a promising blue print for sector coupling, pushing the technology towards a higher technology readiness level (TRL6) [2]. The demonstration comprises amine-based CO_2 post-combustion capture, liquefaction and storage processes, an alkaline water-electrolysis unit to split water into hydrogen and oxygen and an innovative one-step MeOH/DME synthesis unit. A diesel engine of a stationary power generator was adapted to enable use of dimethyl ether (DME) to demonstrate peak and back-up power production in the energy sector. Due to the lack of direct C-C bonds, one advantage of DME is the much lower tendency to build soot compared to diesel. Additionally, a passenger car was modified for the utilization of polyoxymethylen dimethyl ethers (OME_{3-5}), a diesel substitute for the transport sector that can be produced out of MeOH and DME.

Life Cycle Assessment (LCA) is a widely applied method to determine environmental impacts of different products, systems and services. It supports decision making processes during development of emerging technologies. By shedding light on potential environmental pitfalls or advantages of design decisions and benchmarking it with existing technologies, it supports development towards more sustainable products throughout their life cycle. While in recent years several LCA studies [3-8] of various CCUs have been published, none is based on real process data from a demonstrator unit.

2. Methodology

LCA is gaining more and more importance in the scientific community as it supports decision making processes towards more sustainable products. The evaluation of environmental impacts is done by investigating the complete life cycle of a product/technology – from its production including the supply of all raw materials needed, to the use/operation phase of the product/technology, including potential maintenance up to the end of life, which can include waste treatment or recycling. According to the ISO 14040 and ISO 14044 standards [9, 10], LCA is subdivided into four steps. During goal and scope definition the object, the system boundaries, and the functional unit (FU) of the analysis are described. In the life cycle inventory (LCI), information regarding material and energy flows and emissions to air, water or soil along the life cycle are collected. The collected data regarding material, energy and emissions is translated into potential impacts in different environmental effects, so called impact categories, during life cycle impact assessment (LCIA). This enables either a comparison to benchmarking products or the understanding of the magnitude of an impact in a bigger system. The results are summarized and conclusions are drawn to give recommendations for improvement in the final interpretation step.

2.1. Functional Unit

In the investigated system, the choice of the FU is not trivial. The production of the CCU fuels OME_{3-5} and DME are closely linked to the generation of electricity in the lignite-fired power plant. The implementation of the post combustion capture facility directly influences the processes, emissions, material flows and efficiencies in the power plant. Therefore the process needs to be treated as a two product system. The choice of a FU is therefore closely linked to the allocation method. Which product should the captured CO_2 be allocated to? What about the efficiency changes of the power plant processes? The method of LCA does not give one set way to handling these questions of allocation and has been topic of several controversial studies, which lies beyond the scope of this study [3] One approach to the problem of a two product system is the allocation of the arising impacts in a pre-defined way to one or both products. For the investigated system in the ALIGN-CCUS project, we define said allocation as follows: The FU is defined as *'1 kWh peak electricity'* or *'1 km distance driven'*. All impacts that arises due to the implementation of the CCU technology is allocated to the CCU product. This includes all additional emissions and material flows as well as avoided emissions and material flows. The advantages of this approach are an easy comparison of different scenarios and benchmarking routes. The CO_2 needed for the CCU route is shown as a negative impact in the results and is therefore added as a credit (labelled: allocation credit) to the FU. The overall sum of all emission shows the net impact of the FU.

2.2. Impact Categories

The investigated impact categories are determined according to the ReCiPe methodology for LCIA version 2016 v1.1 Midpoint (Hierarchist) [11]. The different process chains are compared based on the following six impact categories:

1. Climate change (GWP) [kg CO_2 eqv.],
2. Fossil depletion (FDP) [kg oil eqv.],
3. Fine Particulate Matter Formation (PM) [kg PM2.5 eqv.],
4. Terrestrial Acidification (AP) [kg SO_2 eqv.],
5. Photochemical Ozone Creation, Ecosystems (POCPe) [kg NOx eqv.],
6. Photochemical Ozone Creation, Human Health (POCPh) [kg NOx eqv.]

These categories are chosen because they reflect electricity and transportation driven impact categories and thus represent the motivation for CCU.

2.3. Data sources

The investigation at hand was carried out using the LCA software GaBi 6 by Sphera Solutions GmbH in the software version 9.5.2.49 [12]. The GaBi database version used was SP40. Most of the LCI data of the CCU production technologies (so called foreground data) are real process data from the demonstrator. However, delays and set-backs throughout the project made the acquisition of real process data in parts difficult or even impossible up to now. Therefore alternative data sources, such as process modelling by AspenPlus or alternative test data were included or development targets taken into account, while at the same time sticking closely to the developed system wherever possible. While the investigation of the demonstrator as-built and operated at Niederaussem was the first target, we also recognize the importance of taking into account potentials for future improvement to the overall concept. Further background LCI data are taken from the ecoinvent versions 3.4 and 3.6 as included in the GaBi software as well as the GaBi's own professional database.

3. System description

A full CCU-chain was demonstrated at the RWE Power Innovation Center in Niederaussem, Germany, including utilization of two CCU-products in the transport and power sector. Captured CO_2 and renewable hydrogen were used for the production of the synthetic fuels DME and OME_{3-5} and their application for transportation and for large-scale energy storage. The demonstrator system serves as a blueprint for Power-to-X technologies and provides input data for the LCI by the different project partners, specialists for the different steps along the CCU route. It is described briefly in the following. A more detailed description can be found in [2].

The system investigated includes elements from the CO_2 capture up to the use of the synthetic fuels either in an emergency generator for peak and back-up power generation or in a car for transport purposes (Figure 1). CO_2 is captured by a post-combustion carbon capture facility at the lignite-fired power plant from RWE Power including CO_2 liquefaction and intermediate storage. Hydrogen is produced in an alkaline water electrolyzer from Asahi Kasei and CO_2 and H_2 are subsequently fed into the synthesis process. This one-step DME synthesis unit developed by Mitsubishi Power Europe produces a MeOH/DME/H_2O mix. Separated DME is used to produce peak power in a stationary power generator equipped with a DEUTZ diesel engine which was adapted by VKA at RWTH Aachen University, FEV and Bosch to operate on liquid DME. Aside from the synthesis of DME in the demonstrator, the production of the fuel OME_{3-5} which could be produced from MeOH and DME is also part of the concept. Its usage as a fuel in an adapted passenger car is investigated.

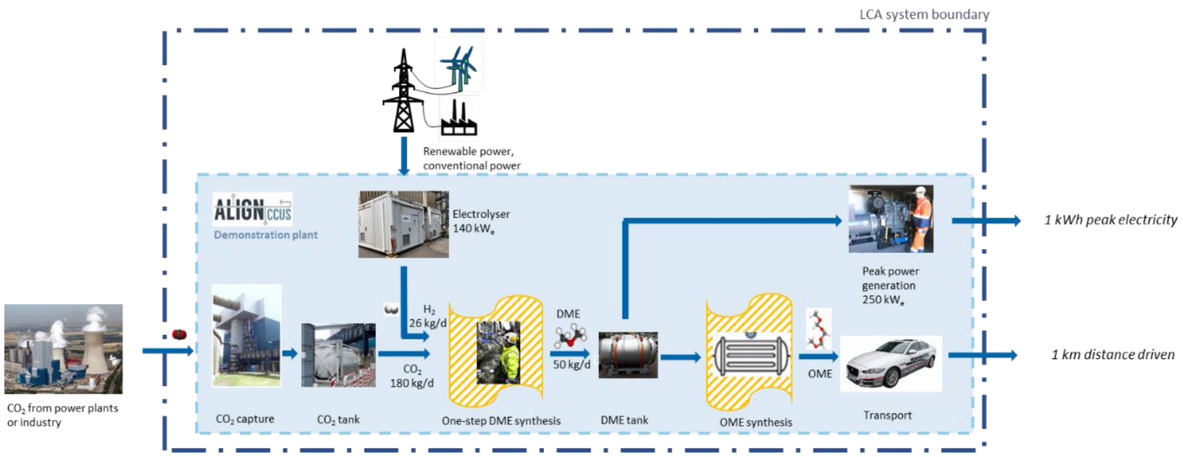

Fig. 1. LCA system boundaries of CCU demonstrator at Niederaussem.
light blue: demonstrator components installed at Niederaussem, yellow shaded: modelled components

3.1. Power Plant

Representative for an industrial CO_2 source the flue gas from RWE Power's 1 GW_{net} raw lignite-fired power plant at Niederaussem is the starting point for the process chain. LCI data for the construction of the power plant is based on previous works in [13] which in turn references and adapts the very detailed inventory from [14]. Regarding the operation phase as well as technical power plant parameters the project partner RWE Power supplied actual data of a power plant without carbon capture [15]. Ash disposal is included in the investigation. Denitrification is done by staged air combustion, selective catalytic reduction is not necessary.

When the CO_2 capture facility with a heat demand for regeneration of 3.5 GJ/t CO_2 is implemented, the resulting efficiency loss at the power plant is 11.8 %-points. This will in turn reduce the net power output while maintaining all other material and emission flows. The impact per kWh electricity produced therefore increases significantly by nearly 25%.

3.2. Carbon Capture Facility

The post-combustion carbon capture pilot plant operated more than 80,000 hours on site at Niederaussem since 2009. With a flue gas flow feed of 1,550 m_N^3/h 7.2 t of CO_2 can be captured per day (CO_2 content of flue gas at absorber inlet approximately 14.2% (v/v) dry; O_2 content in flue gas upstream of the capture plant 5.0% (v/v) dry). Besides the CO_2 scrubber also CO_2 compression and liquefaction units (pressure 16.5 bar (a) - 17.5 bar (a), temperature: -26.5 ° C) and a storage tank (CO_2 storage capacity approx. 18 t) are installed. The liquefied CO_2 has a process-related high purity (> 99.98% (v/v), O_2/Ar <0.016% (v/v), SO_2 <1 ppmv dry).

Only a small percentage of the captured CO_2 of Niederaussem is needed for the DME synthesis demonstrator. However, as data for a large scale MEA facility was available (carbon capture facility for 550 MW electricity generation) we decided to calculate the impact of a large-scale carbon capture system based on the size of the power plant to depict a future as realistic as possible.
For the scrubbing a 30% MEA solution was used. For the investigated optimized system a consumption of 300 g MEA/tCO_2 is assumed when the degradation process can be stabilized in the linear degradation regime. Data for MEA emission into air was gathered also from [16]. The range given is 1-5 mg MEA/m^3exhaust gas, for the investigation conservative 3 mg/m^3 were assumed.

3.3. Electrolyzer

Hydrogen is supplied on-demand with a mass flow of 0.1 – 1.1 kg/H_2 by an improved unit of the mature technology of alkaline water-electrolysis from Asahi Kasei Europe. The unit installed can produce up to 26 kg H_2 per day. Due to Asahi Kasei's novel material concept the efficiency was improved by 10%. The purity of H_2 is > 99.97%, the oxygen by-product is not used and emitted to the atmosphere. Power consumption is 4.3 kWh/Nm³ at a current density of 0.6 A/cm² and cell voltage of 1.8 V [17]. A detailed list of components describing weights and materials for the construction of the electrolyzer as well as regarding its performance was provided by the project partner [18].

3.4. Synthesis Unit

For the synthesis step two different concepts need to be investigated, one for DME and the other for $OME_{3\text{-}5}$ production. Both concepts include the CO_2 conditioning unit, where the CO_2 properties are adapted to the conditions required by the following synthesis unit (gaseous CO_2 at 11 bar(a) and 10°C). DME is produced in an innovative one-step MeOH/DME synthesis unit developed by Mitsubishi Power Europe. A novel bifunctional catalyst coated onto monoliths allows the production of methanol from CO_2 and H_2 and dehydration of methanol to yield DME in one reactor. Non-reacted educts and the by-product CO are recirculated to increase the yield. Flue gas thresholds (especially hydrocarbons and CO) are reached by the use of a catalytic afterburner provided by Forschungszentrum Juelich. For the construction of the unit Mitsubishi Power Europe has supplied a list of components with materials and weights for the demonstrator with a production capacity of up to 50 kg crude-DME per day. The demonstrator does not include the conditioning of the resulting MeOH/DME/H_2O mix to pure DME. As there was no data available regarding the different components of a conditioning unit this was omitted in the LCA in a first approach, therefore underestimating effects.

The synthesis step of $OME_{3\text{-}5}$ out of the MeOH/DME/H_2O mix is not demonstrated on site in the ALIGN-CCUS project. To not completely omit the construction phase for an $OME_{3\text{-}5}$ synthesis unit it is assumed that it is the same for $OME_{3\text{-}5}$ as it is for DME. This approach is a gross simplification as OME synthesis is an even more complex process, its significance regarding the results is discussed in chapter 5.

As no performance data for DME synthesis was available within the project time, the operation of both DME and $OME_{3\text{-}5}$ synthesis was simulated by Forschungszentrum Juelich using AspenPlus. A publication investigating different $OME_{3\text{-}5}$ synthesis routes, DME 1- and 2-step and methanol synthesis regarding their performance and LCA results will soon be available.

3.5. Peak engine

The diesel engine (DEUTZ-Motor TCD 2013 L6 4V, water cooled 6-cylinder in-line engine, Common-Rail-technology) in a stationary power generator of Henkelhausen was adapted by VKA at RWTH Aachen University, FEV Europe and Bosch to demonstrate peak and back-up power production by DME. As DME is gaseous under atmospheric conditions it must be liquefied first. The injection system is similar to a LPG-System. Due to a substantially lower kinematic viscosity of liquefied DME compared to diesel precautions for leakages have to be made. However, the changes to the engine itself – from a material input point of view – are minimal. Therefore the construction phase LCI is based on the data sheet of the original engine. Its weight is given as 870 kg [19]. The in GaBi included ecoinvent dataset *GLO: internal combustion engine production, passenger car* was then used, adapted to the given engine weight.

For the operation, the results of the test stand performance have been supplied by VKA at RWTH Aachen University [20]. As the investigated engine (year 2013 version) was not equipped with a state-of-the-art EGR (Exhaust Gas Recirculation) system, another set of emission data was also supplied presenting an educated estimate regarding emission changes given the application of an actual EGR.

Additionally, within the duration of the project, a novel concept for a multiple CO_2 recycling was considered but not implemented. In this concept the exhaust gas from the diesel engine will be fed into the inlet of the carbon capture unit and most of the CO_2 will again be recovered and used for the DME synthesis. Multiple CO_2 recycling will result in peak and backup power with very low GHG emissions. Potential improvements of impacts due to this recirculation process are currently under investigation.

3.6. Car

A mixture of $OME_{3\text{-}5}$ is a promising diesel substitute. Changes to a 4-cylinder passenger car diesel engine to apply $OME_{3\text{-}5}$ combustion are minimal. Therefore, for the construction of car and engine the data sets for diesel cars included in GaBi were used as LCI. Regarding the cars performance the diesel engine of a Jaguar XE was adapted according to the $OME_{3\text{-}5}$ specific flow-rate. Tests with diesel and $OME_{3\text{-}5}$ were performed by VKA at RWTH Aachen University, FEV Europe and Bosch. Project data of $OME_{3\text{-}5}$ mobility performance emissions are compared to available data from literature [21]. Hank et al. estimate the OME demand of a car based on the EU-wide transport model TREMOVE of the European Union [22].

3.7. Electricity supply

Existing LCAs of CCU have shown that the electricity source has a very high impact on the results. To show the range of the possible impacts two different options for electricity supply were considered. One representing the German electricity mix (2016) using the GaBi data set *DE: Electricity grid mix ts*, representing the upper bound of the spectrum. The other represents a renewable power source as lower bound. Here wind energy was considered by using the dataset *DE: Electricity from wind power ts*.

3.8. Benchmarking systems

To compare the environmental impacts to existing conventional but also to other emerging technologies a set of benchmarking systems for both use options are defined.

3.8.1. Benchmarking systems: Peak Power

As there is no unique definition of peak power, defining benchmark technologies is not trivial. We included LCA results for engine performance with diesel as well as electricity generation via gas turbine. Data regarding the diesel engine performance was supplied by VKA at RWTH Aachen University [20]. Regarding the gas turbine, the data set *DE: natural gas, burned in gas turbine*, for compressor station ecoinvent was used which is included in GaBi. This process includes the upstream of the gas supply, construction of the gas turbine and its emissions.

3.8.2. Benchmarking systems: Mobility

For the mobility scenarios several benchmarking technologies are chosen. These include conventional diesel and gasoline mobility but also e-mobility as one further option for emission mitigation in the transport sector. All car and engine construction elements use data from the GaBi and ecoinvent databases, in parts with adaptations. These include: *Passenger car (medium, gasoline, 1 piece)* for diesel and gasoline mobility; *GLO: passenger car production, electric, without battery ecoinvent 3.4* and *GLO: market for battery, Li-ion, rechargeable, prismatic ecoinvent 3.4* for e-mobility; *GLO: Car diesel, Euro 6 (from Sept 2019), engine size 1,4-2l ts <u-so>* and *Car petrol, Euro 6, engine size 1,4-2l* for performance of diesel and gasoline engines. For diesel mobility the emissions from the GaBi database was adapted by a set of measured data from VKA at RWTH Aachen University. The investigated engine performs with an under average fuel consumption. To enable a fair comparison with a similarly efficient gasoline engine, the GaBi dataset was adapted to the reduced consumption value. For e-mobility a battery was included in the vehicle construction and an electricity consumption of 20 kWh/100 km was assumed.

3.9. Scenario overview

Results for two scenarios (power generation and peak mobility) are generated. The scenario cases vary by choice of fuel (DME, $OME_{3\text{-}5}$ and benchmarking technology fuels), energy source for production (grid electricity or renewable to all or specific processes) and exhaust gas treatment options. This adds up to a total of 16 different scenario cases.

3.9.1. Peak power scenario overview

Over all, six different scenario cases using DME for peak power generation are studied, differing mainly in their electricity supply (for DME synthesis) and exhaust gas treatment at the motor. Included is electricity for DME synthesis from grid electricity (German electricity mix 2016) as well as from wind electricity. Furthermore the cases marked ALL WIND show results for a concept using renewable wind power for both carbon capture and electrolyzer/DME synthesis. As the tested engine is not fitted with an actual EGR system, additionally cases including an estimation of potential improvements and degradations when implementing the latest EGR technology are considered. Together with the two benchmarking technologies overall 8 different cases are investigated for the peak power application.

- DME Grid
- DME Wind
- DME Grid, EGR
- DME Wind, EGR
- DME ALL WIND
- DME ALL WIND, EGR
- Diesel
- Gas Turbine

3.9.2. Mobility scenario overview

For $OME_{3\text{-}5}$ scenario cases we look at two different data sets – a dataset received from motor testing performed by our ALIGN partners and one exclusively modelled taken from literature marked as 'Hank' [21]. Both data sets were analysed using either grid or wind energy for the elctrolyzer. For the benchmarking technologies adapted diesel and gasoline data sets were used. Additionally we include e-mobility as benchmarking scenarios, one using grid electricity, and the other wind energy. Overall 8 different scenarios are investigated for the mobility application.

- OME Grid (Hank)
- OME Grid (ALIGN)
- OME Wind (Hank)
- OME Wind (ALIGN)
- Diesel
- Gasoline
- E-Mobility Grid
- E-Mobility Wind

4. Results

The following results are split into the two scenarios '*CCU Peak Power*' and '*CCU Mobility*'. First, the results for GWP are discussed in detail, as the reduction of CO_2 is the main driver for CCU. The comparison of the results from the demonstrator to the benchmarking technologies shows to which extent this goal can be reached. Results for other impact categories can only be given in a condensed form, but a more thorough publication regarding these impacts is currently in the works. The GWP results are displayed in two different figures. One shows the contribution of the different steps along the process chain to the overall impact. This helps to identify hotspots along the CCU chain. This form of depiction includes the credit for the use of CO_2. The second figure aggregates the net GWP of the entire CCU chain. This facilitates the comparison of the different scenario cases.

4.1 CCU Peak Power

4.1.1 Global Warming Potential

The list of emissions impacting the Global Warming Potential (GWP) is long, the most frequent emissions are CO_2, methane and hydrocarbons, though. The results for the peak power scenario show the immense impact on GWP of the electricity source of the electrolyzer (Figure 2). The second highest contribution to GWP results from the exhaust gas of the peak power engine. GWP emissions at the engine equally occur in all DME cases. The introduction of the EGR system, however, does not have an impact on the GWP results.

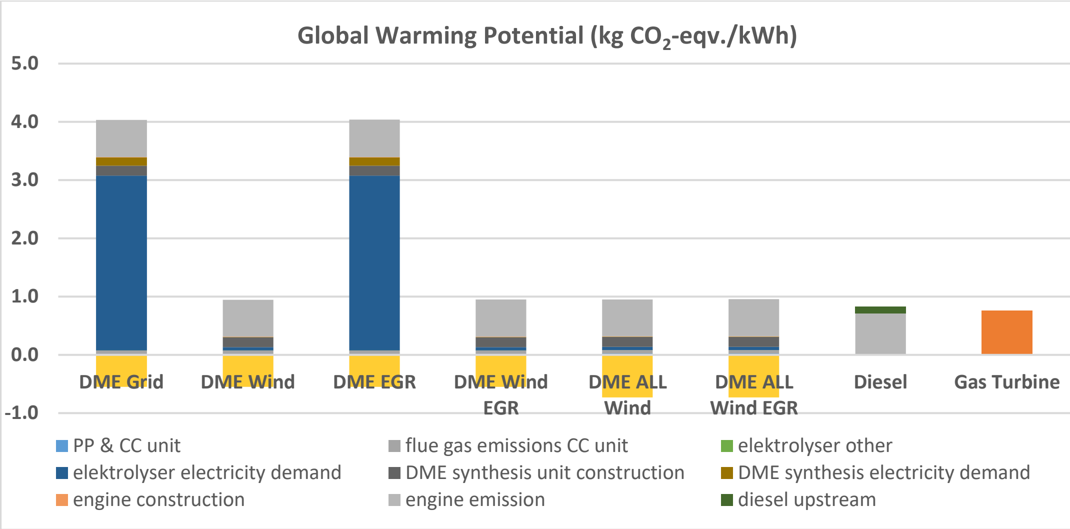

Fig. 2: Global Warming Potential (GWP, g CO_2-eqv./kWh) results for peak power scenario, divided into sources along the process chain. Grid = electricity supply for fuel synthesis with todays' grid electricity mix in Germany (2016), Wind = synthesis using wind energy only, ALL Wind = synthesis and carbon capture using wind energy only.

When using grid electricity, the net GWP (Figure 3) is as high as 3.48 kg CO_2-eqv./kWh (DME Grid & DME Grid, EGR). Using wind power for the electrolyzer and DME synthesis this can be reduced by nearly one order of magnitude to 0.396 kg CO_2-eqv./kWh (DME Wind & DME Wind, EGR). If the use of wind energy is also included for the carbon capture process, further reductions down to 0.226 kg CO_2-eqv./kWh (DME ALL Wind & DME ALL Wind EGR) are possible.

In comparison, benchmarking technologies achieve net GWP values of 0.832 kg CO_2-eqv./kWh for the diesel powered peak engine and 0.761 kg CO_2-eqv./kWh using a gas turbine. Overall, DME produced using wind power shows always better performance concerning GWP compared to today's technology.

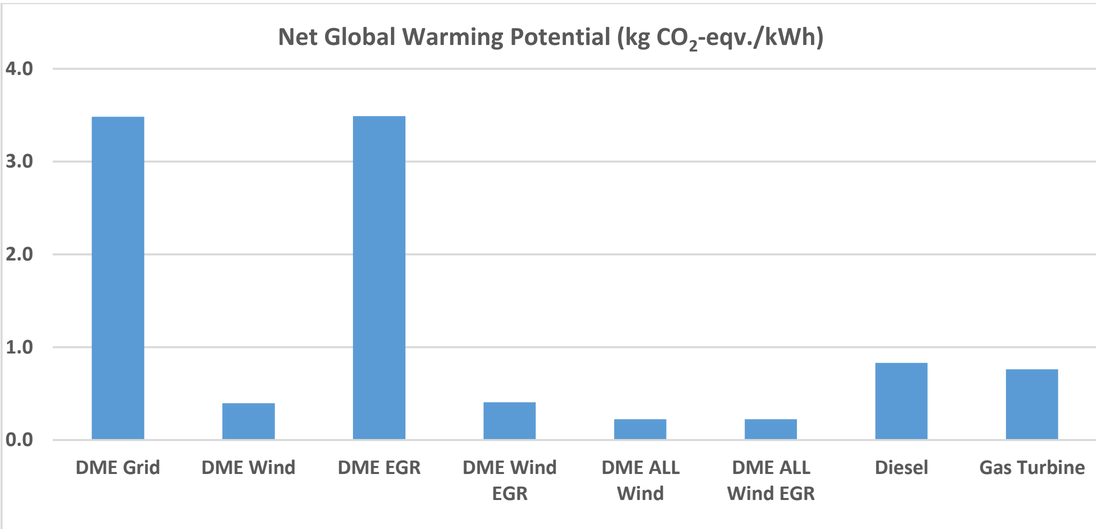

Fig. 3: Aggregated Global Warming Potential (Net GWP, g CO_2-eqv./kWh) results for peak power scenario. Grid = electricity supply for fuel synthesis with todays' grid electricity mix in Germany (2016), Wind = synthesis using wind energy only, ALL Wind = synthesis and carbon capture using wind energy only.

4.1.2 Results in further Impact Categories

For the impact category Fine Particulate Matter Formation (PM) the most influential emissions are particulates, ammonia, nitrate, NOx and SOx. Three effects can be highlighted for this category. First, grid electricity leads to a higher PM impact than wind energy. Second, the construction of the synthesis unit has a significant impact in all cases using DME. This is due to the large amounts of steel per unit of DME produced at demonstrator scale for DME synthesis. Third, high emissions on the engine side are identified. This sums up to 2.46 g PM-eqv./kWh for the DME Grid case. Benchmark technology results for diesel performance are 0.68 g PM-eqv./kWh, for a gas turbine application 0.38 g PM-eqv./kWh. The impact reduces significantly when actual EGR is applied. As EGR leads to a significant NOx emission reduction the category of PM improves greatly compared to the engine performance without actual EGR (1.47 g PM-eqv./kWh DME EGR; 0.60 g PM-eqv./kWh DME Wind EGR; 0.58 g PM-eqv./kWh DME ALL Wind EGR). An implementation of actual EGR for diesel performance was not included in the investigation. It is likely, that this would have a similar effect for the diesel case.

The category Fossil Depletion (FDP) sums up the consumption of fossil resources. The use of grid electricity for DME generation leads to very high impacts of 1,145 g oil eqv./kWh in this category. It is strongly linked to the usage of fossil fuels for electricity generation, so it makes sense that the impact in this category reduces significantly once electricity supply for DME generation is sourced from wind power. All cases using wind power lead to an overall reduction of FDP (254 g oil eqv./kWh). Diesel and gas turbine peak electricity impacts sum up to 247 g oil eqv./kWh and 324 g oil eqv./kWh respectively.

The main emissions for both categories of Photochemical Ozone Creation, ecosystems (POCPe) & Photochemical Ozone Creation, human health (POCPh) are NOx, though a long list of other, rarer emissions can also have an impact. POCPe is measured in NOx-equivalents, which already indicates the significance of NOx emissions for this category. Therefore the impact of EGR implementation on the results also in this category is self-evident. The NOx emissions at the engine together with grid electricity for the elctrolyzer have the highest overall impact on the result. All this leads to a very high impact for the DME Grid case (13.70 g NOx-eqv./kWh), a slightly reduced impact when using wind power (DME Wind & DME ALL Wind: 10.49 g NOx-eqv./kWh), much lower emissions for DME Grid with EGR (4.64 g NOx-eqv./kWh) and very low impact for DME using wind power and EGR (Wind & ALL Wind: 1.47 g NOx-eqv./kWh). Both benchmarking technologies have higher impacts (diesel: 5.42 g NOx-eqv./kWh; gas turbine: 2.48 g NOx-eqv./kWh) than DME Wind and EGR, though the lack of EGR implementation also for the diesel case needs to be considered, which is not considered here.

The results for POCPh are also measured in NOx-equivalents and show a very similar distribution to POCPe: DME Grid case 13.60 g NOx-eqv./kWh, DME Wind 10.11 g NOx-eqv./kWh; DME ALL Wind 10.00 g NOx-eqv./kWh, DME Grid with EGR 4.25 g NOx-eqv./kWh; DME Wind EGR & DME ALL Wind EGR 1.47 g NOx-eqv./kWh. Both benchmarking technologies have higher impact (diesel: 5.41 g NOx-eqv./kWh; gas turbine: 2.45 g NOx-eqv./kWh).

Terrestrial Acidification (AP) is caused by emission of SO_2, ammonia, ammonium compounds, nitrate, NOx, SOx and sulphuric acid. It shows a similar distribution as POCPe & POCPh. Again, both engine emission without EGR and fossil electricity for DME generation have the biggest overall impact. Even though SO_2 emissions were not measured during testing, the emissions of NOx that also affect AP generate a sizable impact. The construction of the DME synthesis unit also has a notable effect on AP. This is due to the large amount of steel used per unit of DME generated due to the demonstration size. Overall the highest impact was found for the DME Grid case (7.31 g SO_2-eqv./kWh), followed by DME Wind & DME ALL Wind (4.52 g SO_2-eqv./kWh) . DME EGR (4.07 g SO_2-eqv./kWh) is still relatively high but for DME Wind EGR & DME ALL Wind EGR (1.27 g SO_2-eqv./kWh) the impact is slightly lower than the benchmarking technology diesel (2.20 g SO_2-eqv./kWh) and the same as the gas turbine (1.23 g SO_2-eqv./kWh). Again, the lack of EGR implementation for the diesel case needs to be considered.

4.2 CCU Mobility

4.2.1 Global Warming Potential

For $OME_{3\text{-}5}$ mobility using grid electricity, the highest source of impact along the process chain is the electricity consumption of the fuel synthesis (see Figure 4). Specifically the hydrogen production by means of electrolysis has the highest share of impact. For $OME_{3\text{-}5}$ cases using grid electricity impacts are as high as 1,120 g CO_2-eqv./km (OME Grid (Hank)) and 700 g CO_2-eqv./km (OME Grid (ALIGN)). These impacts alone surpass all total impacts of the investigated benchmark technologies.

When applying wind energy the results change significantly, though for OME mobility it is still significant with values as high as 308 g CO_2-eqv./km (OME Wind (Hank)) and 226 g CO_2-eqv./km (OME Wind (ALIGN)). This is due to a considerable demand of non-renewable process steam for the synthesis steps, which does not change with the application of renewable electricity. However, the impact of the electricity demand decreases significantly. The difference regarding emissions at the vehicle between the $OME_{3\text{-}5}$ scenario cases OME (Hank) and OME (ALIGN) are the result of a higher fuel consumption values given by the publication [21]. This leads not only to higher emission values at the car, but also all impacts due to the OME production increase, as their share per km driven distance is higher.

For the aggregated GWP (Figure 5) $OME_{3\text{-}5}$ mobility has still higher impacts than its benchmark technology diesel mobility (180 g CO_2-eqv./km Diesel). The lowest GWP can be reached using e-mobility with renewable wind electricity (107 g CO_2-eqv./km).

Throughout the work in the ALIGN-CCUS project, DME synthesis showed more promising results. The application of DME in a car engine was not investigated in the ALIGN-CCUS project, therefore no engine testing data was available. Nevertheless a detailed investigation is in progress showing promising results for DME mobility using wind, which further reduction potential to approx. 130 g CO_2-eqv./km. First results can be found in [2].

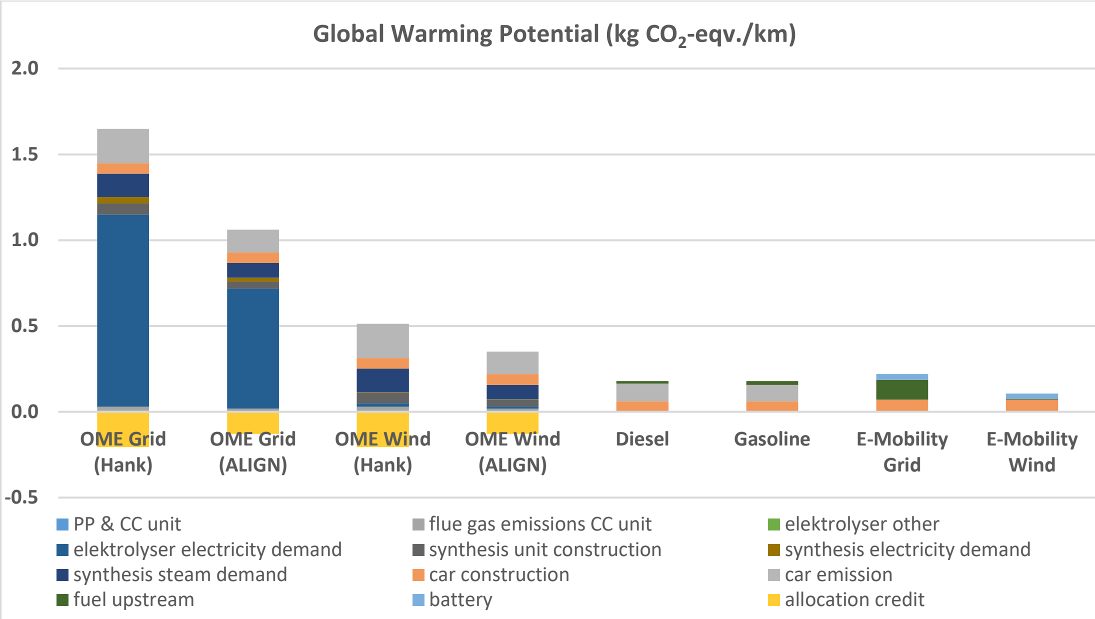

Fig. 4: Global Warming Potential (GWP, kg CO_2-eqv./km) results for mobility scenario, divided into sources along the process chain. Grid = electricity supply for fuel synthesis with todays' grid electricity mix in Germany (2016), Wind = synthesis & e-mobility using wind energy only.

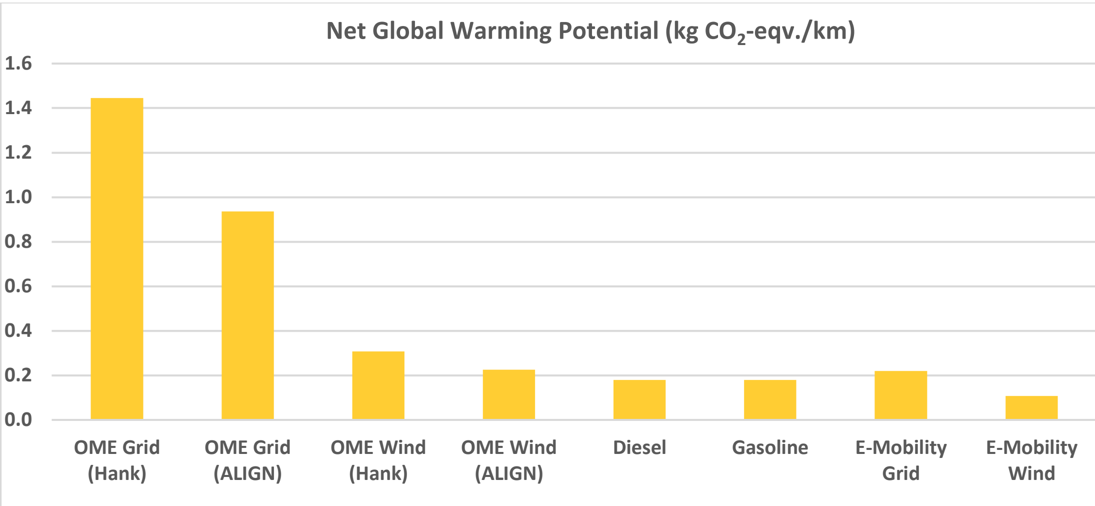

Fig. 5: Aggregated Global Warming Potential (Net GWP, kg CO_2-eqv./km) results for mobility scenario. Grid = electricity supply for fuel synthesis with today's grid electricity mix in Germany (2016), Wind = synthesis & e-mobility using wind energy only.

4.2.2 Results in further Impact Categories

For all cases several effects for the category PM were identified. First, when comparing $OME_{3\text{-}5}$ cases with each other, the cases using car emission and fuel consumption data from Hank [21] have a significantly higher impact (OME Grid (Hank): 0.79 g PM 2.5-eqv./km; OME Grid (ALIGN): 0.50 g PM 2.5-eqv./km). The difference is mainly due to Hank [21] giving both a higher fuel consumption and much higher NOx emissions of the engine than measured in the ALIGN-CCUS project. The higher fuel consumption has again the effect that also other elements of the process chain, like the construction phase of the synthesis unit, increase per unit distance driven. Therefore the high NOx emissions lead to increased impact levels for PM. Using wind electricity reduces the upstream emissions significantly (OME Wind (Hank): 0.47 g PM 2.5-eqv./km; OME Wind (ALIGN): 0.30 g PM 2.5-eqv./km). The car construction plays a significant role in all cases though it does not change the relative difference between cases. In the e-mobility cases (E-Mobility Grid: 0.33 g PM 2.5-eqv./km; E-Mobility Grid: 0.30 g PM 2.5-eqv./km) it was found out that battery construction has a major impact on PM results.

As FDP is closely linked to energy consumption of non-renewable sources the grid cases have the highest impact. $OME_{3\text{-}5}$ case impacts (496 g oil-eqv./km OME Grid (Hank); 317 g oil-eqv./km OME Grid (ALIGN); 167 g oil-eqv./km OME Wind (Hank); 111 g oil-eqv./km OME Wind (ALIGN)) are up to 9 times higher than classic diesel or gasoline mobility (55 g oil-eqv./km Diesel; 55 g oil-eqv./km Gasoline). The introduction of renewable energy leads to significantly lover FDP values.

The results of the cases are for both POCPe und POCPh categories very similar and show a comparable relative distribution as the GWP results. NOx emission is closely linked to fossil energy generation, therefore the energy intensive mobility with OME_{3-5} using fossil-based steam and grid electricity has a very high impact. When looking at the cases using the car emission data from Hank [21], the assumed NOx emissions in the car exhaust gas are very high compared to all other cases. Once wind energy is applied to the synthesis process, a significant reduction of POCPe and POCPh emissions occurs. In contrast to the GWP results, no emissions fall below benchmarking technology levels.

Looking at OME_{3-5} cases a link between fossil based energy consumption and impacts in the AP category are detected. The high grid electricity demand for both OME_{3-5} Grid cases leads to very high impacts for AP. There is a significantly higher impact from flue gas emissions in the cases using data from Hank [21] (2.49 g SO_2-eqv./km OME Grid (Hank) and 1.19 g SO_2-eqv./km OME Grid (ALIGN)). This is due to the fact that SOx emissions were not measured in the ALIGN-CCUS project but are estimated in Hank. Furthermore the higher fuel consumption of Hank has again the effect that also other elements of the process chain like the construction phase of the synthesis unit increase per unit distance driven. Additionally for AP another significant impact from the battery needed for e-mobility was identified. This causes a higher AP for e-mobility compared to today's conventional technology (0.68 g SO_2-eqv./km E-Mobility Wind; 0.33 g SO_2-eqv./km Diesel; 0.38 g SO_2-eqv./km Gasoline). OME Wind (ALIGN) also shows lower values than e-mobility (0.54 g SO_2-eqv./km), but still higher than the conventional options.

5. Discussion and Conclusions

The chance to use real process data of the demonstrator for an entire CCU chain enables a better understanding of dependencies and interrelationships. Hereby, it contributes strongly to close the knowledge gap for the discussion of environmental benefits of CCU concepts. The broad spectrum of cases for the two different CCU applications 'peak power' and 'mobility' give an overview over some of the most impactful routes and route changes that can be taken. However, at this level of development there are still some major parameters which cannot be determined appropriately. Therefore this chapter discusses important parameters and their sensitivities on the results obtained so far.

5.1. Important parameters

5.1.1. Electricity source

The electricity demand for the synthesis of OME_{3-5} and DME is high. For the synthesis of 1kg DME nearly 16.5 kWh of electricity are needed. For 1 kg OME_{3-5} – which has a 33% lower LHV – it is nearly the same with 16.1 kWh, but in addition 4.1 kWh high pressure steam are needed. For the different CCU applications this translates into an input of approx. 5.5 kWh of electricity (grid or wind) to generate 1 kWh of peak electricity. For mobility using OME_{3-5} it is approx. 1.26 kWh per km distance driven plus 0.32 kWh high pressure steam demand. First investigations regarding DME mobility show a consumption of around 1 kWh per km driven. This is 5-6 times as high as the electricity demand of e-mobility. With regard to the energy demand, direct electrification of the private transport sector is favorable. This is not different for heavy duty applications, but currently there are no suitable batteries for long-haul transportation available and direct electrification could only be achieved by overhead contact lines (as it is done in railway transportation). However, transportation with exclusively renewable power requires 100% availability and a match between renewable power generation and traffic consumption. Synthetic fuels have the capability to store renewable energy in large quantities and thereby solve an inherent weakness of fluctuating renewable power: the mismatch between supply and demand. This crucial advantage is not addressed in the environmental assessments so far. LCA methodology is mainly used to analyze static situations and a dynamic approach would be necessary to include the time aspect.

Therefore it is not a surprise that the source of electricity - especially for hydrogen production - has the biggest impact on most impact categories or rather the highest potential for improvement. To understand how much impact it has, we investigated not only hydrogen production and other synthesis steps using grid electricity but also cases using wind energy. For the important category of GWP we can see an improvement by over 80 % when using wind energy compared to their respective cases. In fact, results show that CCU using grid electricity is no suitable option for both fuels OME_{3-5} and DME and for both CCU applications if GWP reduction is the overall target. All cases combining CCU and grid electricity have much higher GWP impact results than their respective benchmarking technologies. For all CCU cases for the GWP and FDP category the choice of a renewable electricity source has the highest potential for improvement. For POCPe, POCPh and AP it also has one of the highest potentials, only rivalled by exhaust gas treatment options at the engines.

The electricity grid mix of 2020/2021 is already quite different than the applied mix of 2016 and will change towards even higher percentages of renewable energy in the near future. This will bring down most impact categories for the grid cases significantly. However, only a renewable energy share of nearly 100% will be able to bring the GWP results to a lower level than those of the benchmark technologies. When data gaps as for the conditioning of the water/DME/methanol mix or construction of OME plant are filled by additional data the break-even point of renewable energy supply can be estimated.

Electrification of the carbon capture process and therefore the complete CCU fuel synthesis chain can lead to a significant additional reduction. Per 1 kg DME generated this adds another nearly 1.4 kWh of electricity demand for synthesis, bringing it up to 17.9 kWh per kg DME and 6 kWh per kWh peak electricity generated. When using wind energy for capture and synthesis steps a GWP impact of 0.226 kg/kWh peak electricity can be achieved. For mobility, OME_{3-5} leads to higher GWP emissions than the benchmark technologies, even if using renewable electricity sources. Investigations regarding DME mobility are under investigation, first results suggest that it can compete with the benchmark technologies.

5.1.2. Exhaust gas treatment

The impact of exhaust gas treatment can be grave. There are several instances in the process chain of the investigated CCU processes that include exhaust gas emissions into air. The difference between the inclusion and non-inclusion of EGR illustrates how significant a change on flue gas treatment can be. In this case it is particularly true for impact categories that include emissions like NOx and SOx, though many other emissions need to be considered as well. However, it was not possible to measure all emissions and apply or even optimize EGR in time of the project. Moving forward it is important to keep track on changes of exhaust gas emission composition with regard to the LCA to gain a more thorough understanding as to which chances and limitations there are. Soot and NOx reduction is often mentioned as an argument pro DME/OME application, which gives further investigations even more significance. An interesting technological option for the peak power production from DME is to recirculate the exhaust gas of the engine back upstream to the capture plant. It holds the potential to reduce the GWP of exhaust gas emissions by re-using the carbon several times. Here more detailed investigations are ongoing.

5.1.3. Construction phases

The LCI for the construction phases for the different elements of the CCU process chain have different levels of depth, completeness and accuracy. The construction of the MEA-wash facility is an educated estimation from developers, including the complete set up. The electrolyzer construction has the most level of detail and is based on the actual, implemented electrolyzer. Construction of the DME synthesis unit is incomplete as it does not include processing of the water/DME/methanol mix. A purification of the mix into DME needs a considerable amount of further equipment. This could not be included yet, due to lack of data. For OME_{3-5} synthesis, construction data were not available at all, therefore the estimation was made that it is the same as DME synthesis. This is of course a fairly inaccurate approach and needs to be updated once actual data for OME_{3-5} synthesis exist. The same goes for the conditioning of the water/DME/methanol mix synthesis which could not be included in the investigation at hand.

Another point that needs to be considered when reviewing the results is, that the investigated system is at demonstrator size with the purpose of research and development. There is very large room for improvement regarding the most efficient relation between material usage and DME output.

Still, the impact of the DME construction phase is relatively low for all impact categories. If assuming a doubling of material usage due to the processing of the water/DME/methanol mix it will become more significant, especially in the categories GWP, PM and AP. At the same time, a full scale application will lead to a significant reduction of material usage per unit DME output and therefore to impact reduction. Further investigations will definitely be needed, and it is fair to say that no final conclusion can be drawn at this point.

5.1.4. Research and development targets

Along the process chain, the efficiency of CO_2-capture, H_2 generation, DME/OME_{3-5} synthesis and the engines (car and peak power) all have a significant influence on the LCA results. It is difficult to foresee potential improvements for the future. Asahi Kasei hopes to improve efficiencies by 8-18 % in the long term. DME/OME_{3-5} is still in an early stage of development, so room for improvement can be expected even though it is difficult to quantify. Regarding the engines, researchers are hopeful to reach efficiencies that are comparable with their respective diesel alternatives.

Transport of intermediates and products was not investigated in this study. Depending on the necessary additional infrastructure for the implementation of different climate protection measures it can have sizable impact. The synthetic fuels produced from CO_2 and renewable hydrogen may offer the potential to use more existing infrastructure (e.g. filling stations, cars) in comparison with the benchmarking technologies.

5.1.5. Carbon dioxide source

CO_2 is a by-product of many production processes, mainly as part of a flue gas. Besides CO_2 from lignite-fired power plants as demonstrated in the ALIGN-CCUS project there are several other sources that can be considered to be used in an overall process chain of CCU. As upstream processes for CO_2 capture have a significant impact on the overall environmental impact of the investigated process chain, it makes sense to compare these different CO_2 sources. Depending on the CO_2 concentration in the flue gas, the amount and condition of flue gas, the required purity of the CO_2 product and the availability of waste heat the supply of CO_2 needs different additional expenditure. As these additional expenditures are allocated to the CCU product, the best point source needs to be chosen. CCU product, CCU chain location, network, transport availabilities, CCU product sink can all have an impact on the overall cradle-to-grave result. The conclusion is, that only a LCA for every specific CO_2 source with its respective boundary conditions can determine if a CO_2 source is a good choice for the combination with CCU in terms of environmental impact.

5.2. Conclusions

The LCA of the CCU demonstrator that was developed in the ALIGN-CCUS project generated extensive results in six different impact categories for two different scenarios and a total of 16 cases. Both peak power generation and mobility using CCU fuel DME or OME_{3-5} were investigated. The discussion of important parameter settings made clear, that there are many factors that determine the overall result for the functional units *1 kWh peak electricity* and *1 km driven distance*. Only some of these were investigated in the form of scenario cases like the influence of the electricity source on the overall impact or the implementation of different EGR rates. Others were not investigated, mainly due to lack of data. Especially the impact of construction phases for DME and OME_{3-5} synthesis deserve a closer look in the future once more data is available. Also the composition of the engine exhaust gas needs further investigations once more detailed and holistic inventory data is available. However, the availability of first hand data from an existing demonstrator improves the understanding of environmental impacts of a real system. It helps to classify previous studies on exclusively modelled data.

From the results gained in the LCA at hand, several conclusions can be drawn. Firstly, OME_{3-5} as a CCU fuel does not perform well at this point from a life cycle perspective. The very energy intensive synthesis of OME_{3-5} leads to very high impacts in the GWP category, even when using renewable electricity for hydrogen generation and other synthesis processes.

The results regarding DME draw a different picture. Implementing CCU with DME into today's energy mix and using grid electricity for the synthesis processes leads to very high impacts for the CCU products. However, the higher the share of renewable electricity sources in the grid, the lower those impacts will be. If using only renewable energy for synthesis processes (in this investigation wind electricity), the CCU product can reach impact values that are lower than its benchmark technologies. The downside is the very large input of electricity. It takes 5.5 kWh renewable electricity to generate 1 kWh of peak electricity. Likewise driving 1 km with DME uses 1-1.08 kWh renewable electricity, which is 5-6 times as much as e-mobility. In a scenario of electric power solely from renewables and at the same time a constant demand for e.g. fuels for transport, a reliable and efficient storage solution must inevitably be established, and CCU fuels like DME can present an overall solution due to the sector coupling co-benefits for energy and transportation.

To prove operability with the demonstrator is a first step to use real process data for further discussion. The identification of hot spots and improvement potentials within the CCU route, but also the discussion of improvement potentials supports the development of CCU technologies.

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