Life cycle assessment of a small-scale methanol production system: A Power-to-Fuel strategy for biogas plants

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Abstract

Power-to-Fuel (PtF) systems use carbon dioxide and hydrogen as feedstock together for renewable fuel production and can hence contribute to climate change mitigation. This study assesses the environmental performance, from cradle to gate, of an innovative PtF system for synthetic methanol production, which integrates a biogas plant based on manure and feed residues and a combined heat and power unit. Under this concept, the residual carbon dioxide from biogas production is used for the synthesis of methanol, whereas hydrogen is obtained via wind-based electrolysis. A life cycle assessment (LCA) is carried out here for 1 kg of methanol produced with the integrated system proposed, operated on a small scale. In view of the multi-functionality of the process, the uncertainty in LCA outcomes is assessed by considering different assumptions on co-product credits for both the electricity from cogeneration and the digestate from the anaerobic digestion of organic raw materials. Additionally, a sensitivity analysis is performed to examine the influence of variability in life cycle inventory data on the results. All the analysed scenarios show significant improvements compared with conventional methanol production from fossil resources (with only a few exceptions for acidification and eutrophication). The sensitivity analysis shows that parameters determining the overall energy

requirements as well as methane losses from anaerobic digestion in the PtF system greatly influence its environmental performance, and should be carefully considered in process design and upscaling. In spite of the uncertainty inherent in LCA, the system is presented as an interesting option to produce renewable methanol while contributing towards a circular economy, provided that the economic performance is also beneficial relative to the fossil alternative.

Abbreviations

AD	Anaerobic digestion
AP	Acidification potential
BGP	Biogas plant
CC	Climate Change
CH_4	Methane
CHP	Combined heat and power
CO	Carbon monoxide
CO_2	Carbon dioxide
EP	Eutrophication potential
EPD	Environmental Product Declaration
EU	European Union
FD	Fossil depletion
GHG	Greenhouse gases
H_2	Hydrogen
HT	Human toxicity
LCA	Life cycle assessment
LCI	Life cycle inventory
LCIA	Life cycle impact assessment
LHV	Lower heating value
MOP	Muriate of potash or potassium chloride
NG	Natural gas
ODP	Stratospheric ozone depletion
PEM	Polymer electrolyte membrane
POF	Photochemical ozone formation
PSA	Pressure swing adsorption
PtF	Power-to-Fuel
RED	Renewable Energy Directive
SSP	Single superphosphate

Keywords
advanced fuel, biofuel, circular economy, environmental impact, renewable energy, uncertainty

1. Introduction

The revised Renewable Energy Directive (REDII) (EC, 2018) requires the European Union (EU) to produce 32% of its energy consumption and at least 14% of its total fuel consumption with renewable energy sources by 2030. This implies that further efforts must be made across Member States in order to meet their respective national targets. In Germany, the share of renewables in primary energy consumption was 13.7% as of 2018, while it was 7.8% in transportation energy use (BMWI, 2019). To meet their national targets, Member States can promote the use of non-fossil-based feedstock, as a substitute for conventional energy sources. However, alternative raw materials must allow for feasible and sustainable fuel production pathways in both technical and environmental terms. Power-to-Fuel (PtF) technologies constitute an opportunity to make progress towards the REDII targets, as they employ carbon dioxide (CO_2) –together with hydrogen (H_2)– as feedstock for renewable fuel production and can hence contribute to climate change mitigation (Dietrich et al., 2018). PtF systems can be for instance used to produce methanol, which is consumed as a commercial fuel and represents an interesting non-fossil alternative for both the transport and energy sectors (Peters et al., 2020). As such, methanol already accounts for a large market share in industrialised countries, e.g. more than 20% in China (Yang and Jackson, 2012). Conventional methanol production is mainly based on a chemical synthesis process that uses H_2 and carbon monoxide (CO). The latter is produced via steam methane reforming of natural gas (NG), a process which is well established on an industrial scale (Pontzen et al., 2011). However, the production of methanol from H_2 and CO_2 has recently gained more attention in the scientific literature, where it has been the focus of model-based process analyses (Peters et al., 2020) as well as life cycle assessments (LCAs) (González-Garay et al., 2019; Matzen and Demirel, 2016).

Biogas plants (BGP) and biogas upgrading plants in Germany are a promising carbon source for large-scale renewable CO_2 provision. Germany had a biogas production capacity of 10.4 million tonnes in 2016, plus 1.5 million tonnes from biogas upgrading plants; only the latter could cover the entire methanol production of the country (VCI, 2018). In a BGP, raw biogas is burned in a combined heat and power (CHP) unit in order to generate electricity. The exhaust heat is captured by a heat exchanger and used for heating in the same installation. Biogas upgrading plants have a similar setup but include an additional step to upgrade the raw biogas from anaerobic digestion (AD) into biomethane, which has the quality of natural gas (NG) and can be injected into the NG grid. Biogas upgrading allows increasing the share of methane (CH_4) in the product gas by segregating CO_2 and other substances. Possible upgrading technologies include amine scrubbing, cryogenic membranes and pressure swing adsorption (PSA); the latter is the most common technology in Germany, together with classic membrane processes (Viebahn et al., 2018). To date, CO_2 from most plants is usually released into the atmosphere and not made available for other applications (Billig et al., 2019). There is, however, the possibility of using flue gas from gas separation in PtF concepts, since it can provide a high CO_2 concentration with few trace gases (Viebahn et al., 2018). Billig et al. (2019) highlight the technical potential of CO_2 capture and utilization for developing sustainable technologies of alternative fuel production.

Employing CO_2 from BGPs as feedstock also comes with disadvantages as it is often available at the local level, posing logistic challenges especially in rural areas with only a few BGPs. Given that central processing plants can only be profitable if there is enough CO_2 in close proximity (Viebahn et al., 2018), a possible solution could be to implement a small-scale CO_2 processing plant on farms where manure is readily available (Lee et al., 2016). BGPs increasingly employ organic residues such as liquid manure from pigs and cattle, which can easily be pumped, transported and stored due to its low dry matter content (FNR, 2013). Moreover, collecting manure from nearby farms can also substantially reduce the impacts from manure transport (Esteves et al., 2019). The production of fuels from animal manure ultimately yields "advanced biofuels" as defined by the REDII, while contributing towards a circular economy by reusing by-products and minimizing waste generation (Meng and McKechnie, 2019). In Germany, small-scale manure-based BGPs are abundant and are known as *Güllekleinanlagen*, with an average biogas production capacity of 75 kW and a minimum requirement of manure of 80% (Daniel-Gromke et al., 2018). The number of plants increased from 120 in 2012 to 582 in 2016 (Scholwin et al., 2019), which could provide 126 Mt of CO_2 annually. Nevertheless, only one third of the manure produced annually in Germany is used in BGPs, while the remaining amount is stored without processing and spread onto fields, hence generating GHG emissions (Scholwin et al., 2019). These BGPs are mostly concentrated in the Northwestern region of Germany, where a high number of livestock farms can be found (Daniel-Gromke et al., 2017). The Northwestern coastline also represents some excellent wind locations, which could provide low-carbon energy for H_2 production (Decker et al., 2019; Welder et al., 2018). This motivates the choice of this region as the case study where to establish a PtF system integrated with a manure-based BGP in Germany.

The environmental performance of alternative technologies or production strategies is commonly analysed by means of LCA, since it makes the production options comparable (Cherubini et al., 2009). In the context of biogas production, LCAs have been applied, for instance, to compare the environmental impacts from manure- and crop-based BGPs to produce electricity (Fuchsz and Kohlheb, 2015); or from different agricultural substrates to produce biogas in integrated CHP plants (Lansche and Müller, 2012). Other authors use LCA to quantify impacts of classic biogas upgrading from different raw materials (Buratti et al., 2013), as well as from more innovative production of biosynthetic methane from H_2 and CO_2 (Castellani et al., 2018). In the context of transport fuels, LCAs also tackle the utilization of methanol as a fuel (Verhelst et al., 2019), while others assess the production of a variety of synthetic fuels from biogas through AD, e.g., compressed and liquefied biogas, methanol, or Fischer-Tropsch Diesel and dimethyl ether (DME) (Moghaddam et al., 2015; 2016). Lee et al. (2016) evaluate the production of DME from landfill gas or manure-based biogas. However, all these systems use the CH_4 from biogas for steam reforming to syngas. To the best of the authors' knowledge, there are no LCAs addressing an integrated PtF system as the one presented in this case study, including on-site biogas production and utilization, for both energy generation and CO_2 provision and using CO_2 in biogas with wind-based H_2 for renewable fuel production.

Integrated fuel production systems with cogeneration usually have more than one function and hence pose the so-called 'multi-functionality problem' in LCA (Escobar et al. 2015). According to the ISO 14040/44:2006 (International Organization for Standardization, 2006a, b), this can be solved by either partitioning (often referred to as 'allocation') or system expansion. ISO 14044 (International Organization for Standardization, 2006b) recommends system expansion over allocation for attributional LCAs when the system delivers more than one product or function. This entails assuming that co-products replace other products in the market, generating co-product credits under system expansion approaches. The need for assumptions produces uncertainty due to modelling choices, in addition to parameter and model uncertainty (Huijbregts et al., 2001). The products to be replaced normally depend on the relative prices, amongst other market factors, which in turn depend on the geographical and temporal scope of the LCA. In attributional LCA, co-product credits are normally estimated by considering those co-products to be most likely replaced in the market, i.e. from average suppliers (Finnveden et al., 2009). On the contrary, consequential LCA considers suppliers of marginal technologies by incorporating economic reasoning (Ekvall and Weidema, 2004). Thus, the influence of such assumptions on results from both attributional and consequential LCA can be critical, especially when comparing systems against each other, and must be conveniently assessed through scenario analysis (Escobar et al., 2014).

The goal of this study is to assess the environmental performance of an innovative PtF system to produce methanol by using CO_2 from manure-based biogas combined with H_2 produced from wind energy. The concept was specifically developed for this LCA case study, as implemented on a pilot scale in the Northwestern region of Germany, where there are many BGPs and an excess of manure. In order to quantify the environmental benefits brought about by the proposed system, environmental impacts along the life cycle are compared to a reference system for fossil-based methanol production, also considering results uncertainty due to modelling choices in LCA and parameter variability. The economic performance of the same system will be subsequently evaluated in order to understand trade-offs among sustainability dimensions.

2. Materials and methods

We carry out an attributional LCA by following ISO 14040/44:2006 (International Organization for Standardization, 2006a, b), which consists of the following phases.

2.1 Goal and scope

The LCA aims at quantifying the environmental impacts from a novel PtF system specifically developed at the Research Centre 'Forschungszentrum Jülich' (North-Rhine Westphalia, Germany), from cradle to gate. This implies that the system boundaries include processes up to the stage at which the main product, i.e., methanol, is delivered at the plant gate, hence excluding further processing, use and disposal. The functional unit (FU) is thus defined as 1 kg of methanol produced by a PtF plant with a capacity of 216 tonnes of methanol per year (135 kW as lower heating value –LHV–), designed as proposed by Decker et al. (2018). It is a site-specific integrated system, which includes the following components: a) a biogas production plant; b) a CO_2 recovery unit, which upgrades biogas to biomethane using PSA and a post-combustion unit with a CO_2 tank; c) a CHP unit; d) a wind turbine and a polymer electrolyte membrane (PEM) electrolyzer for H_2 supply and e) a methanol synthesis plant with a H_2 storage facility for buffering. Most components meet the technology readiness level (TRL) of 9 and are readily available to be used in an operational environment. However, methanol production is not yet available on such a small scale. The PSA upgrading technique is applied by Hygear (2015) on a similar scale, though it is not widely available for small-manure plants. The TRL of the PEM electrolyzer is assumed to be 8 as defined by Saba et al. (2018). PEM systems can be purchased from several manufacturers, although their current market penetration is limited in Germany.

The main processes included in the PtF system assessed are described below (Figure 1), while major technical characteristics and associated process parameters are included in Table 1:

a) Biogas is produced in a fermenter through AD of a mix of manure and grain residues from feed processing. Both types of feedstock are considered as residues, hence their respective upstream production processes up to the PtF facility gate are not included within the system boundaries. The manure is directly transported from the stable to the facility through an automatic manure scraper into the preliminary storage tank, which is located underground and covered with concrete. The manure is pre-stored for a short period of time until it is pumped into the fermenter. The digestate obtained as a co-product from AD is then openly stored on-site.
b) A PSA upgrading process generates biomethane and separates the CO_2 from the biogas. A flue gas stream with 98.25% of CO_2 and 1.75% CH_4 is considered, assuming that biomethane from upgrading has a purity of 95% (Lohse, 2019). Post-combustion of biomethane through recuperative afterburning is carried out to achieve a pure stream of CO_2. This process requires a minimum CH_4 of 0.3vol.% (Graf and Bajohr, 2013).
c) The biomethane stream is subsequently burnt in the CHP unit. In industrial scale installations, the biomethane is injected into the gas grid in Germany. Nevertheless, there is not always a connection to the grid next to small-manure plants, which are often located in remote areas. This is why it was assumed that biomethane is used on site.
d) A wind turbine is also implemented to produce H_2 from surplus wind energy via electrolysis. A H_2 tank provides a buffer facility whereby H_2 can be stored after production. It is assumed that the wind turbine has a capacity of around 1.2 MW, producing electricity for water separation with a 960 kW electrolyzer. It is also assumed that the H_2 is produced in close proximity to the farm, hence no transport is involved.
e) Methanol is finally produced on a pilot scale in a plant with a nameplate capacity of 216 tonnes of methanol per year, assuming 8500 full load hours (FLH), corresponding to 135 $kW_{th,LHV}$. It should be noted that the methanol synthesis runs between 80-100 bar and 250 °C.

Table 1: Main process parameters characterising the innovative Power-to-Fuel system and associated data sources.

Electricity CHP (kW)	75[a]
Heat CHP (kW)	98[a]
Engine output CHP (kW)	205[a]
Plant electricity demand	0.08[a]
Plant heat demand	0.35[a]
Composition of biogas	
CH_4 (%)	53[a]
CO_2 (%)	46[a]
O_2 (%)	1[a]
Number of cows (providing manure for a 75 kW biogas plant)	126[b]
CH_4 losses during AD (%)	1.40[a]
Methane slip during PSA (%)	1.50[c]
Share of CO_2 gain from post-combustion (%)	1.83[d]
Electrolyzer capacity (kW)	960[d]
Wind turbine capacity (kW)	1200[d]
Capacity of methanol plant ($kW_{th,LHV}$[e])	135[d]

[a] Rau (2019), [b] Rutzmoser et al. (2014), [c] Lohse (2019), [d] own calculation, [e] Lower heating value

Several co-products are generated across the life cycle such as fertilizer, heat and electricity. In order to subtract additional functions delivered by the co-products, besides methanol production (i.e. the FU), the 'system expansion' approach is applied according to ISO 14044 (ISO 2006b). It is thus assumed that co-products generate environmental credits by substituting for average products available in the market. The system boundaries are shown in Figure 1, including the so-called avoided processes that generate those co-product credits.

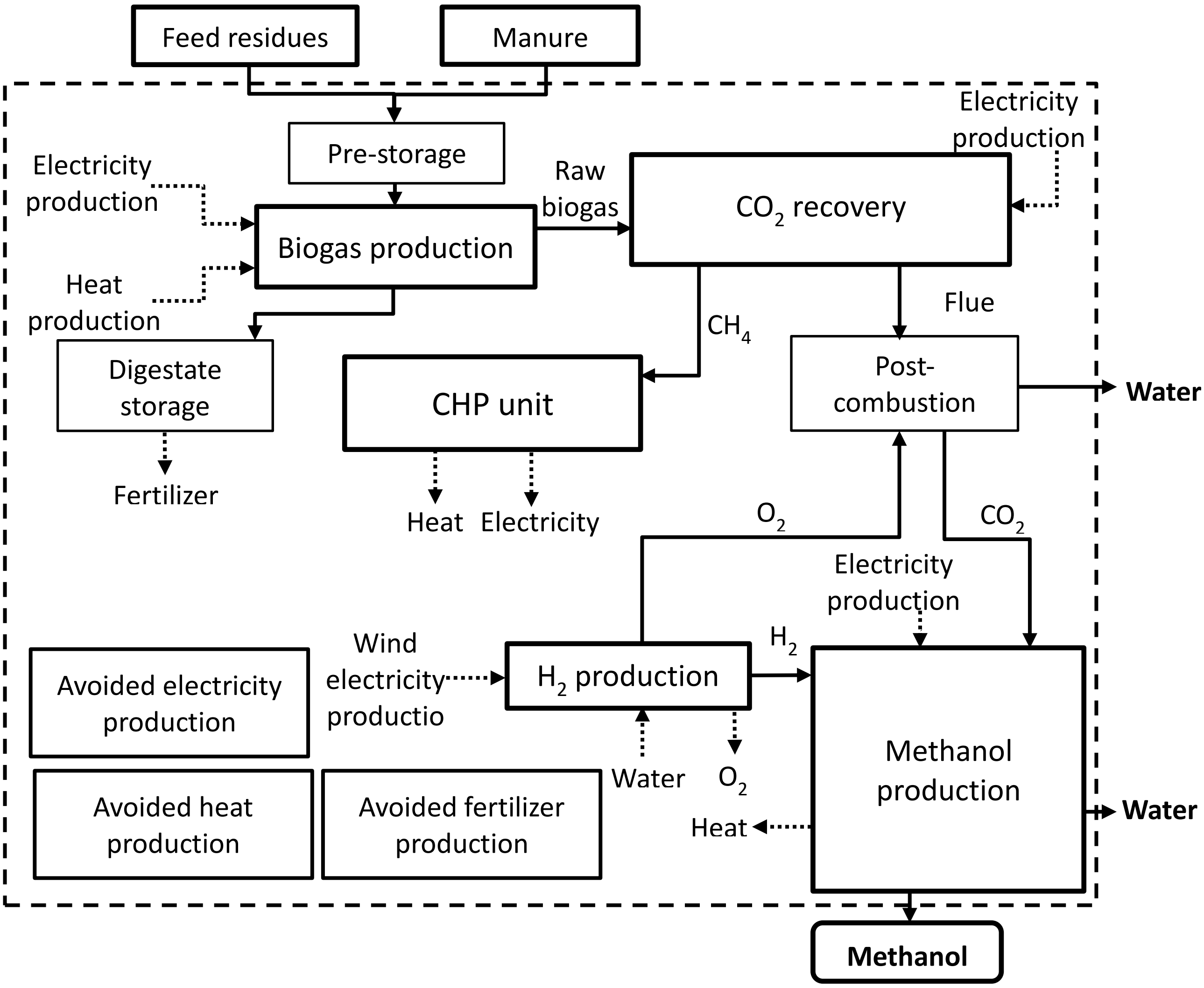


Figure 1. Flow diagram of the Power-to-Fuel system proposed, from cradle to gate, by applying system expansion to subtract impacts from co-product generation.

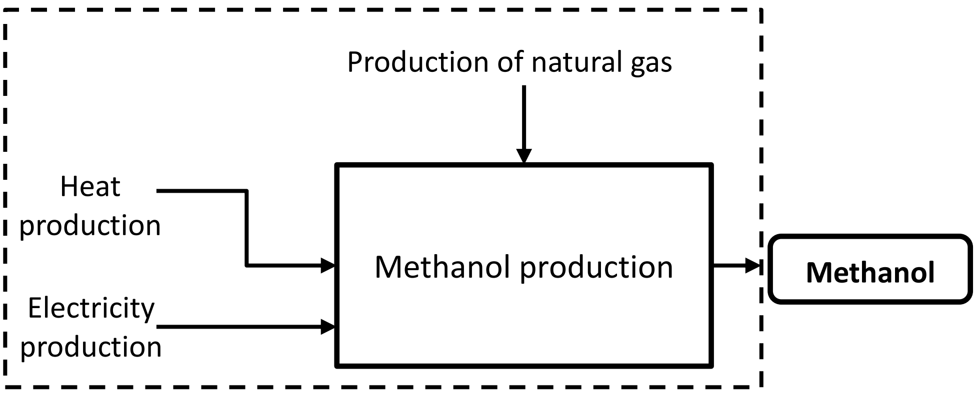


Figure 2. Flow diagram of the reference process for fossil-based methanol production, from cradle to gate.

Several scenario formulations were defined to tackle uncertainty in LCA results due to assumptions on avoided processes, as shown in Table 2. It is firstly assumed that the digestate can be used as a fertilizer since it contains 3.4 wt% elemental nitrogen (N), as well as 5.3 wt% potassium (as K_2O) and 2.4 wt% phosphorus (as P_2O_5). Hence, in the first scenario formulation, i.e. A1, the substitution is based on the N content, assuming that digestate replaces urea as a major organic fertilizer in the market, with an average N content of 46%. In scenario A2, we assume that K_2O in digestate replaces potassium chloride –also known as muriate of potash (MOP)–, with a K_2O content of 60% (EC, 2019). In scenario A3, using the digestate as fertilizer based on its P_2O_5 content avoids producing single superphosphate (SSP) with a P_2O_5 content of 20% (IPNI, n.a.). Overall, these fertilizers were chosen due to their relevance as the main commercial fertilizers in the EU (EC, 2019). In addition to the digestate, the assessed PtF system generates electricity as a co-product from the CHP, which can be sold to the grid. In order to capture uncertainty in the source of electricity that is most likely to be replaced, we assumed that it substitutes for electricity from the average German electricity mix (A). As alternative scenario formulations, electricity from the CHP replaces average off-shore wind electricity (B) or electricity from coal (C), as best-case and worst-case scenarios in the context of Germany, respectively, from the environmental point of view. Finally, excess heat from the CHP and methanol production replaces heat from NG at industrial furnaces in the EU, which is assumed to be the major heat source in Germany. In order to calculate environmental benefits brought about by the proposed system, fossil-based methanol production was considered as the reference process (shown in Figure 2), which does not deliver additional co-products according to the Ecoinvent 3.5 database (Wernet et al., 2016).

Table 2. Scenario formulations of the PtF system assessed with choices on avoided processes under the system expansion approach.

	N content in digestate replaces urea production (as N)	K_2O content in digestate replaces potassium chloride (MOP) production (as K_2O)	P_2O_5 replaces single superphosphate (SSP) production (as P_2O_5)
CHP-electricity replaces average electricity from the German mix	A1	A2	A3
CHP-electricity replaces wind-based electricity produced in Germany	B1	B2	B3
CHP-electricity replaces from a coal-based electricity produced in Germany	C1	C2	C3

2.2 Life cycle inventory (LCI)

LCI data for the main processed at the foreground level was collected from our own measurements, as combined with technical process parameters for the biogas and methanol plants. The energy efficiency was calculated at 35.58 % based on the individual energy efficiency rates shown in Table 3. The process of CO_2 recovery is the most intensive in the use of electricity, followed by the electricity demand of the BGP and the methanol production. Yet, only 19% of the electricity production of the CHP and 35% of the produced heat is used within the system, given that H_2 production is wind-based. Primary data was combined with secondary data for the energy demand of the PSA and methanol synthesis, as well as certain emission factors of the BGP. It must be noted that the production of capital goods for the main processes considered in the foreground system are not included within the system boundaries due to data limitations at this stage. Manure application and transport are also not included since it is assumed that the integrated system is located on-farm.

Table 3. Energy efficiency of the main sub-processes included in the system boundaries to produce 1 kg of methanol by means of the Power-to-Fuel system proposed.

	Biogas production plant (incl. CHP)	Polymer electrolyte membrane electrolysis	CO_2 recovery plant	Methanol synthesis plant
$\eta_{sub-system}$	0.65[a]	0.70[b]	0.92[c]	0.85[d]

[a] Rau (2019), [b] Schiebahn et al. (2015), [c] Sun et al. (2015), [d] Schemme et al. (2020)

The main assumptions made and data sources employed through the LCI, are described as follows, while the LCI is shown in Table 4:

a) Biogas production: The LCI was taken from a BGP located in Eastern Germany, via personal communication with the Technical University Bergakademie Freiberg (Rau, 2019). The plant has two large CHPs with 344 kW and 180 kW biogas production capacity connected to three fermenters (1180 m³ each), respectively, as well as another 75 kW CHP with a fermenter of 1250 m³. The feedstock used consists of 80% of liquid cattle manure and 20% of grain shreds from feed production. Both are considered waste from livestock farming in a nearby dairy and entail zero emissions according to the REDII. We calculated CH_4 emissions from pre-storage of manure by following the IPCC (2006a) guidelines, and employing reference values for Germany (Haenel et al., 2020). Specifically, we considered manure generated by 126 cows, which is the number of cows on-site corresponding to 75kW. According to Haenel et al. (2020), NH_3 emissions from pre-storage of manure are zero, when the facility has a roof made out of concrete, as is the case. Moreover, manure is only kept inside the pre-storage tank temporarily; hence the short retention time avoids the formation of floating covers that enable nitrification (Wulf et al., 2019). N_2O emissions from AD were also neglected according to (IPCC, 2006b) as data is scarce, while the process releases negligible quantities of H_2, H_2O and other trace gases. Air is induced into the fermenter to capture some of the hydrogen sulphide (H_2S), measured at 200 ppm. We further considered CH_4 losses from biogas production arising both from the fermenter and the storage of digestate. These are estimated at 1 kg/MWh equivalent to 1.4%, mainly coming from the storage facility (Rau, 2019). The leakage of NH_3 from the fermenter is lower than 0.05% of the N content in the digestate and hence excluded (EMEP/EEA, 2016). NH_3 emissions of 2.66% of the N in the digestate, occurring during storage, were included in the inventory based on a tier 2 approach from EMEP/EEA (2016). The values for N content in digestate arise from on-site measurements from the larger fermenter at the pilot plant, which are fed with the same feedstock and have the same retention time of 150 days.
b) CO_2 recovery: PSA delivers high purity of CO_2 in the flue gas stream (i.e. about 87-99.9%) (Viebahn et al., 2018). The technique includes upstream desulfurization, which guarantees that the biogas no longer contains sulphur when entering the adsorption (FNR, 2014). Desulphurization is also part of the process technology described by Viebahn et al. (2018).

Hence, the LCI includes the demand of electricity for desulfurization as part of the CO_2 separation process (Viebahn et al., 2018).

c) CHP: Emissions were measured for the 75 kW CHP. These, however, arise from the combustion of raw biogas, and hence the emission values serve as an approximation. When calculating the CH_4 losses due to biogas production and upgrading, we assumed that an additional 3% of biogas is required to guarantee a full load drive of the CHP. This translates into additional feedstock requirements in input that can be easily operated with the existing fermenter. Emissions data for the CHP Otto gas engine without a catalyst was measured at the plant in Eastern Germany in November of 2018.

d) H_2 production: Production data for the wind turbine and H_2 electrolysis as well as the methanol production were obtained from own simulations performed by the Institute of Electrochemical Process Engineering within the Institute of Energy and Climate Research at the Forschungszentrum Jülich. Assuming 2000 FLH of the wind turbine, a factor of 4.25 was considered for 8500 FLH for methanol synthesis and the AD process.

e) Methanol production: The methanol production process was carried out under 250°C at 80 bar inside an isothermal reactor which uses 1.37 kg of CO_2 per kg methanol as also described by Billig et al. (2019). The thermal discharge from the methanol production process can be used in the system. The 135 $kW_{th,LHV}$ methanol synthesis plant uses 34.3 kg of CO_2 per hour, and 4.7 kg of H_2.

For the reference process, we considered that conventional methanol production in Germany is carried out via the steam reforming of NG. Associated LCI data was taken from the process in Ecoinvent 3.5 (Wernet et al., 2016) by considering energy consumption only and neglecting capital goods production, same as in the proposed system.

Table 4. Life cycle inventory of all inputs and outputs associated with the production of 1 kg of methanol by means of the Power-to-Fuel system proposed.

INPUTS		**OUTPUTS**	
Methanol synthesis		**Methanol synthesis**	
CO_2 (kg)	1.37	Methanol (kg)	1.00
H_2 (kg)	0.19	Water (kg)	0.56
Electricity (MJ)	0.56	Thermal discharge (MJ)	1.75
CO_2 recovery (PSA)		**CO_2 recovery (PSA)**	
Biogas (m^3)	1.58	CO_2 in flue gas (kg)	1.37
Electricity (MJ)	1.03	H_2O (m^3)	1.96E-05
		Biomethane 95vol.% (kg)	0.66
Biogas production		**Biogas production**	
Electricity (MJ)	0.85	Biogas (m^3)	1.56
Heat (MJ)	4.86	Urea as N (kg)	0.78
		Potassium chloride as K_2O (kg)	0.96
		Single superphosphate as P_2O_5 (kg)	1.28
		NH_3 emissions from digestate storage (kg)	1.54E-03
		CH_4 emission from pre-storage of manure (kg)	1.40E-02
		CH_4 losses from AD and digestate storage (kg)	8.07E-03
CHP		**CHP**	
CH_4 (kg)	0.66	Electricity (MJ)	10.63
		Heat (MJ)	13.89
		Emissions	
		SO_2 (kg)	7.76E-05
		NOx (kg)	1.13E-03
		CO (kg)	6.03E-04
		NMVOC (kg)	1.30E-05
		CH_4 (kg)	1.82E-03
H_2 Production (PEM)		**H_2 Production (PEM)**	
Electricity (MJ)	32.27	Oxygen (kg)	1.33
Water (kg)	1.69E-03	H_2 (kg)	0.19

2.3 Life cycle impact assessment (LCIA)

The characterization method 'ReCiPe 2016' (Huijbregts et al., 2016) was chosen for the calculation of environmental impacts at the midpoint level, as implemented in GaBi Life Cycle Engineering Suite (Kupfer et al., 2019). This proves to be a comprehensive method for comparative impact assessments of energy systems and transport fuels (Cavalett et al., 2013; Treyer et al., 2014). The "hierarchist perspective" was taken, as a neutral scenario for the analysis of future socio-economic developments (Huijbregts et al., 2016). Moreover, it estimates the climate change potential from GHG emissions over a 100-year horizon, in line with the temporal scope for developing low-carbon economies, according to the REDII. We considered the following impact categories due to their importance in the environmental performance of alternative fuels (Morales et al., 2015; Rocha et al., 2014): climate change (excluding biogenic carbon) (CC) (CO_2-eq.); freshwater and marine eutrophication potential (EP) (kg P-eq.); terrestrial acidification potential (AP) (kg SO_2-eq.); fossil depletion (FD) (kg oil-eq.); photochemical ozone formation (POF) (kg NO_x-eq.); human toxicity (HT) (kg 1,4-DB-eq.); and stratospheric ozone depletion (ODP) (kg CFC-11-eq.).

2.4 Sensitivity analysis

In addition to the scenario analysis described above (section 2.1), a sensitivity analysis was also carried out in order to assess the influence of parameter uncertainty on the results. In particular, we considered a range of variability for those parameters that showed the highest variability in the PtF implementation on a pilot scale, namely: i) the CH_4 emission intensity of the BGP associated with the fermenter and the digestate storage; ii) the energy efficiency of the PEM electrolysis; iii) the electricity input for methanol production; iv) the energy input to the fermenter; and v) the heat input to the fermenter. The sensitivity analysis was carried out by means of the GaBi Analyst Tool. We assumed lower and upper bounds for each of the aforementioned parameters based on the literature, as is shown in Table 5.

Table 5. Parameters of the sensitivity analysis.

Parameter	Base value	Range of variability (±CV[1])	Reference
i) CH_4 losses from anaerobic digestion and digestate storage (kg/h)	0.21	±75%	Graf and Bajohr (2013), Scheutz and Fredenslund (2019)
ii) Electricity demand of electrolysis (MJ/kg H_2)	171.63	±17%	Buttler and Spliethoff (2018), Brynolf et al. (2017)
iii) Electricity demand for methanol synthesis (MJ/kg methanol)	0.56	±40%	Fasihi and Breyer (2017)
iv) Electricity demand of the fermenter (MJ/h)	21.60	±25%	Stinner et al. (2015), Scholwin et al. (2019)
v) Heat demand of the fermenter (MJ/h)	123.48	±30%	Zielbauer et al. (2007), Daniel-Gromke et al. (2017)

[1]CV: coefficient of variation relative to the base value

3. Results

3.1 Impact assessment results

Results from the LCIA are shown in Table 6. All scenario formulations for the proposed PtF system (see Table 2) yield negative values due to co-product credits, which offset net impacts from the integrated system itself. This translates into negative impact values per FU. Only scenario B2 generates positive values for the CC, both the marine and freshwater EP and the POF and AP; while positive values are also generated by B1 in the freshwater EP. When comparing the CC results to the other scenarios, the lowest impact values in absolute terms are achieved in scenarios C1 and C3 (-5.48 and -4.92 kg CO_2-eq., respectively). In both cases, electricity replaces electricity from coal as the most CO_2-intensive electricity mix. CC outcomes are quantified at -3.83 and -3.27 kg CO_2-eq. in scenarios A1 and A3, respectively, which consider the average electricity mix as an avoided process; by contrast, CC is relatively smaller in scenarios B1 and B3, in which wind-based electricity is replaced. When comparing scenario formulations that differ in terms of the fertilisers being replaced, scenario B2 generates the greatest impact for CC (0.22 kg CO_2-eq.), while the lowest value is found for scenario C1 with avoided urea production (-5.48 kg CO_2-eq.).

In general, those scenarios that include replaced coal electricity generate the largest reductions across impact categories. The worst scenario across all formulations is B2, which shows greater impacts across categories, with positive values for CC, EP, POF and AP. The lowest impact values for FD are found in scenarios C1 and C3 (-2.26 and -2.29 kg oil-eq.). The three scenarios, including replacement of the MOP fertiliser (A2, B2 and C2), yield the highest values for FD. This is due to the relatively higher content of K2O (60%) in MOP, which translates into smaller environmental credits than those from replacing urea (46%) and SSP (20%). Fertiliser credits in all categories - except for CC - are the greatest when SSP is replaced, followed by urea and then MOP. The lowest impact values for AP are achieved in scenario A3 (-2.99E-02 kg SO_2-eq.), in which electricity from the grid is replaced, implying that the German grid mix causes more kg SO_2-eq. emissions than coal-based electricity. The reason for this can be found in the composition of the average electricity mix in Germany, which includes a large share of NG according to Ecoinvent 3.5 (Wernet et al., 2016). As a result, electricity from the average German mix generates acidifying emissions (as SO_2-eq.) in larger amounts per MJ compared to coal-based electricity. The same is observed for the ODP category.

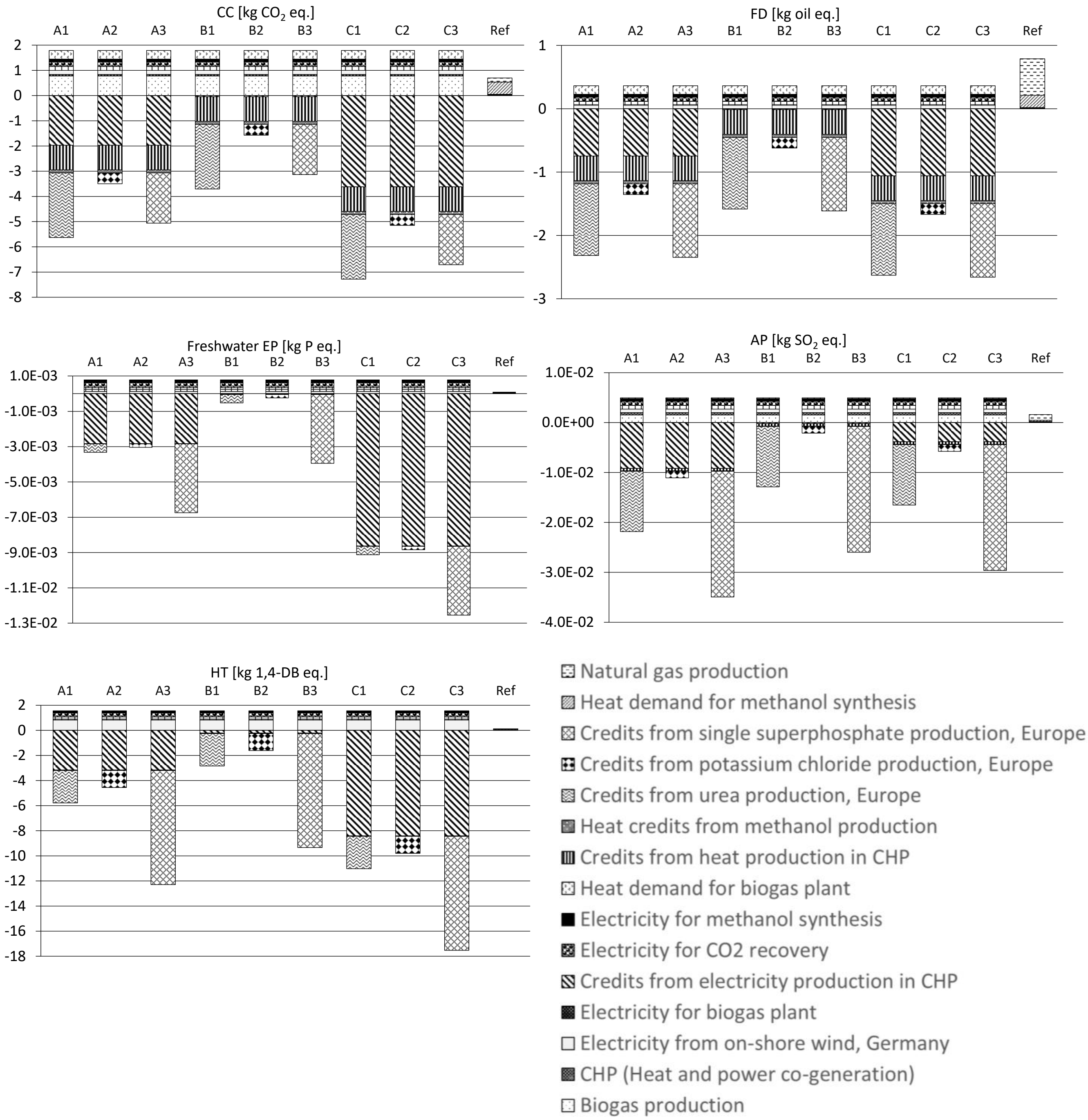
When comparing the LCIA results from the proposed scenarios with those obtained from the reference scenario, all perform better in every impact category, except for scenario B1 in terms of freshwater EP and scenario B2 in EP (both marine and freshwater) and AP. The greatest savings are observed for the impact categories of EP, AP and HT, which show values that are between 131 and 137 times lower than those from fossil-based methanol production. Savings in the CC category are still notable, associated with impact values around 9 times lower relative to the reference scenario. In general, the worst-case scenarios for co-product credits in the PtF system generate the greatest savings relative to conventional methanol production, and the other way around. Scenario B2 generates net emissions in the categories of CC, AP and POF, entailing between 1 and 5 times greater impacts than the reference scenario. Scenario B1 generates impact values in EP freshwater that are twice as high as those in the reference scenario.

Table 6. Results from the Life Cycle Impact Assessment of the different scenarios assessed for producing 1 kg of methanol by means of the Power-to-Fuel system proposed, as compared with conventional fossil-based methanol production.

	CC [kg CO_2-eq.]	FD [kg oil-eq.]	EP freshwater [kg P-eq.]	EP marine [kg N-eq.]	HT [kg 1,4-DB-eq.]	POF [kg NO_x-eq.]	ODP [kg CFC-11-eq.]	AP [kg SO_2-eq.]
A1	-3.83	-1.95	-2.55E-03	-2.62E-04	-4.22	-7.12E-03	-2.01E-06	-1.69E-02
A2	-1.75	-0.99	-2.26E-03	-1.47E-04	-3.00	-2.93E-03	-1.32E-06	-6.09E-03
A3	-3.27	-1.98	-5.97E-03	-2.97E-04	-10.73	-1.42E-02	-2.11E-06	-2.99E-02
B1	-2.35	-1.38	-3.88E-04	-1.20E-04	-1.95	-4.13E-03	-1.11E-06	-7.92E-03
B2	-0.22	-0.42	-1.04E-04	-5.29E-06	-0.73	6.78E-05	-4.11E-07	2.87E-03
B3	-1.79	-1.42	-3.81E-03	-1.55E-04	-8.46	-1.12E-02	-1.20E-06	-2.10E-02
C1	-5.11	-2.19	-7.02E-03	-5.40E-04	-8.27	-7.87E-03	-1.82E-06	-1.16E-02
C2	-2.98	-1.23	-6.73E-03	-4.25E-04	-7.04	-3.68E-03	-1.13E-06	-7.80E-04
C3	-4.54	-2.22	-1.04E-02	-5.74E-04	-14.72	-1.49E-02	-1.92E-06	-2.46E-02
Reference scenario	0.70	0.79	8.65E-05	6.15E-06	0.12	1.37E-03	1.48E-07	1.62E-03

The contribution of each sub-process to the overall impacts from the proposed PtF scenarios is shown in Figure 3 for those categories that deliver greater savings as compared to the reference scenario, namely AP, EP and HT; CC and FD are also included, since these are the two impact categories to be potentially improved by a renewable fuel production system. The remaining categories are included in Figure S1 in the electronic supplementary material (ESM).

Figure 3. Contribution analysis of the processes included in the expanded system for the impact categories climate change (CC), fossil depletion (FD), acidification (AP), eutrophication (EP) and human toxicity (HT); the underlying data can be found in the LCI (see Table 4).



In the PtF system, electricity credits associated with CHP production account for 27-38% of the overall impacts in the CC in scenarios A1-A3, and for shares of 40-52% in scenarios C1-C3. The share of electricity credits is, however, much smaller in scenarios B1-B3, i.e., 1-2%. In the latter scenarios, the replaced heat and fertiliser production (especially urea and SSP) make the greatest contribution to the most impact categories, i.e., 48% of FD in B2 and 83% of freshwater EP in B3, respectively. Electricity credits from the CHP account for a larger share of the absolute impacts when the electricity from the average mix (A) and especially that from coal (C) are considered. In scenarios in which wind electricity is replaced (B), environmental credits of the heat from CHP make the greatest contribution to CC and FD in scenario B2. Credits associated with the digestate account for the larger share of impacts in scenarios B1 and B3. In terms of HT, EP and AP, the effect of the heat credit is negligible. Biogas production makes the greatest positive contribution to AP (22%) in B2, offsetting co-product credits and generating net SO_2-eq. emissions. P-eq. savings from replaced electricity account for 75-90% of the impact (in scenarios A and C), while fertiliser credits account for 37-83% across scenario B. In scenarios A and C, electricity credits offset the environmental burdens from the use of on-shore wind electricity in H_2 production in HT, EP and AP. The greatest impact reductions in scenarios B are achieved by replacing SSP, followed by urea except for CC, where it is the other way around. As indicated, the environmental credits from MOP are comparatively smaller, but still represent a significant share of the absolute impacts, e.g., 43% of HT in B2. The CHP unit is the largest source of NO_x-eq. emissions (accounting for between 9% and 35% of POF across the scenarios), followed by the consumption of electricity. In the reference system for fossil-based methanol production, electricity production accounts for the largest share of the impacts of EP and HT. While heat production makes the greatest contribution to CC and ODP, NG production accounts for the largest shares of FD, POF and AP (see Table S1 in the ESM).

3.2 Results from the sensitivity analysis

Results from the sensitivity analysis are shown in Table 7, including only those scenarios and impact categories for which the difference in the impact value relative to the reference scenarios is higher than ±10%. Note that the variability in outcomes greatly depends on the extent to which co-products credits are affected, which vary asymmetrically with the +10% and -10% change in the parameter, respectively.

Variability in CH_4 losses from both the AD and subsequent storage of the digestate are critical for the GHG performance of scenario B2, inducing a change of around ±100% in the CC value. This is due to the relatively significant contribution of biogas production to the overall impact in B2 (23%). CH_4 losses also prove influential in scenarios A2, B2 and B3. Another decisive parameter is the electricity demand in the electrolyser, which generates the greatest variability in the categories CC, EP, HT and POF in scenario B2; as well as for HT in B1 and AP in C2. These correspond to those categories to which credits from replacing wind-based electricity make a substantial contribution (i.e., ≥20%). Electricity demand for both methanol synthesis and for the BGP also causes a change in HT greater than ±100% of the base value in scenario B2 due to the relatively smaller credits from electricity substitution compared to the other scenarios, which increases the contribution of the other sub-stages to the overall impact. Similarly, variability in electricity demand for methanol synthesis and the BGP also yields significant changes (larger than ±15% of the base value) in CC and OPD for scenario B2, in freshwater EP for scenario B1 and in AP for scenario C2. Variability in the heat demand of the BGP is only relevant in scenario B2, and especially in CC, causing a change of around ±45% of the base value. Parameters determining the overall energy requirements across sub-processes thus greatly influence the environmental performance of the integrated PtF system and must be carefully considered in process design and upscaling. CH_4 losses from AD and subsequent storage of the digestate are also decisive for the GHG performance. It must be noted, however, that CH_4 emissions from BGPs are close to zero in recently built plants (FNR, 2016), which translates into significant improvements in the GHG performance.

Table 7. Change in results from the life cycle impact assessment relative to the reference values, when changing the parameters indicated by ±10% through sensitivity analysis. Only changes over ±10% relative to the reference values are shown.

Parameter	CC [CO_2 -eq.]	FD [oil -eq.]	Freshwater EP [P-eq.]	HT [1,4-DB-eq.]	Marine EP [N-eq.]	POF [NO_x -eq.]	ODP [CFC-11-eq.]	AP [SO_2 -eq.]
CH_4 losses of the biogas plants								
A2	12.87% -12.87%							
B1	11.05% -12.11%							
B2	98.16% -104.36%							
B3	16.30% -16.30%							
Electricity demand of electrolyser								
B1				10.97% -10.98%				
B2	12.44% -12.44%			250.74% - 257.78%		17.14% -17.66%		
C2								14.74% -15.38%
Electricity demand for methanol synthesis								
B1			23.43% -23.53%					
B2	19.35% -19.35%		11.15% -11.15%	122.96% - 124.07%	10.84% -10.84%		18.31% -17.61%	
C2								24.62% -25.13%
Electricity demand of the fermenter								
B1			23.53% -23.53%					
B2	18.43% -17.97%		10.59% -10.59%	116.67% - 116.48%	10.30% -10.30%		16.90% -16.90%	
C2								23.33% -23.59%
Heat demand of the fermenter								
B2	45.16% -44.70%	15.18% - 15.18%				13.06% -12.96%	14.79% -14.79%	

CC: climate change, FD: fossil depletion, EP: eutrophication potential, HT: human toxicity, POF: photochemical ozone formation, ODP: stratospheric ozone depletion, AP: acidification potential

4. Discussion

The scenario analysis of the multi-functional PtF system assessed shows that environmental impacts are largely dependent on the choices regarding co-product credits when performing system expansion in attributional LCA. Specifically, credits from the electricity generated by the CHP are the main contributor to the impact savings estimated for the PtF system across impact categories, as also found by Lansche and Müller (2012). In this way, onsite emissions released by the CHP itself are only relevant for POF, although these account for a relatively small share compared with other sub-processes, as also found by De Vries et al. (2012). These findings support the idea that electricity produced from manure-based biogas in CHP units is more environmentally-friendly than that from NG, in terms of CC, AP, POF and ODP (Esteves et al., 2019). Environmental credits associated with the use of the digestate as a fertiliser account for a significant share of the absolute impacts across all categories, especially when SSP is assumed to be replaced in the market. This is in line with the results from FNR (2013), as for CC only. It must be noted that using digestate as fertiliser can deliver further GHG benefits in agricultural production, relative to applying untreated manure (Esteves et al., 2019). Other commercial fertilisers could have been considered instead of urea, e.g. ammonium nitrate, which is the most commonly used inorganic N fertiliser in the EU and globally (Fertilizers Europe, 2019). Since the digestate originates from organic residues, i.e. manure and straw residues, it was considered that it rather substitutes for organic fertilisers. Such an integrated PtF system also valorises manure following circular economy principles (EC, 2020) to supply renewable energy in a flexible way. An alternative system design was taken into account, in which renewable energy is not produced in sufficient amounts within the system and electricity from the German grid is needed to generate H_2. This scenario causes even greater impacts than fossil-based methanol (see Table S2 in the ESM), highlighting the need to generate renewable energy for self-consumption. As a limitation, this study does not include upstream impacts associated with the production of capital goods for the PtF system production. However, these are expected to be comparatively low in contrast to impacts from operational stages (Esteves et al., 2019).

This attributional LCA applies the system expansion approach to include those processes that would potentially be replaced in the German market by the several co-products generated (i.e. digestate, heat and electricity), according to ISO (2006b). A scenario analysis was carried out by assuming environmental credits associated with both average and also marginal technologies (e.g. wind electricity), in order to assess the variability in the results due to such modelling choices. Attributional LCA is usually based on average LCI data, while consequential LCA uses marginal data to estimate impacts from a change in demand the FU (Tillman, 2000). However, consequential LCA requires additional economic modelling to simulate how changes in the life cycle affect the whole economic system (Earles and Halog, 2011; Weidema et al., 2018). Another alternative could have been to apply partitioning to solve the multi-functionality problem and allocate environmental impacts among co-products up to the stage in which these are generated. As an accounting exercise, GHG savings brought about by the proposed system compared to conventional methanol production were estimated based on the relative energy content of co-products, as the REDII recommends. The following values were considered: 19.9 MJ/kg for methanol (McAllister et al., 2011), 17.4 MJ/kg (as dry matter) for the digestate (Gardoni and Guarino, 2016) and 22.3 MJ/m^3 for biogas (FNR, 2016); besides, the net energy output in MJ of electricity and heat. The REDII assumes CH_4 savings from manure management, which were included based on EC (2018). Methanol from the PtF system delivers GHG savings of 55.5% relative to fossil-based methanol (Wernet et al. 2016), while savings increase to 83.6% if wind electricity is used across sub-processes, i.e. biogas production, CO_2 recovery and methanol synthesis. This means that methanol production in a PtF system is energy intensive and would only meet the REDII's sustainability requirements for transport fuels after January 2026 (GHG savings >65%) if the wind-based electricity is readily available in the installation. The REDII provides that advanced biofuels based on non-food and waste feedstock (e.g. animal manure or sewage sludge) should account for at least 3.5% of the transport fuel market by 2030.

The way to deal with multi-functionality has been broadly discussed in the LCA literature. For instance, Pelletier et al. (2015) suggest that system expansion should not be prioritized in attributional LCA, but the choice depends on the rationale of the analysis. Meng and McKechnie (2019) emphasize that system expansion is a suitable method for understanding the system's overall impact when evaluating a novel technology. In this study, the system expansion approach was chosen to consider the effects of the multiple products delivered to the market by the integrated PtF system, as compared to conventional methanol production, in order to highlight benefits from 'closing loops' in fuel production towards a circular economy. Applying partitioning may constitute a simplification when analysing integrated systems in which each sub-process delivers multiple co-products, some of which are used as inputs in other units. Indeed, most LCAs of integrated or circular production processes apply system expansion to deal with the multi-functionality issue (Escobar et al., 2015; Lansche and Müller, 2012); although this can hinder comparative sustainability assessments with other systems. The REDII identifies challenges when applying energy allocation if CHP is used in the processing of biofuels, bioliquids and biomass fuels, as is the case here; while the system expansion approach is accepted for the purposes of policy analysis. Similarly, the International Organization for Standardization (2006b) even recommends avoiding partitioning in both closed-loop and open-looped product systems.

In LCA, impacts are not proportional to the FU but specific to the scale of production. When performing system expansion, the scale of production also determines the quantity of co-products generated and hence the products to be potentially replaced in the market. For instance, it can be expected that if digestate production increases with the biogas production capacity, not all of it could be employed as fertilizer, depending on the demand for it by the agricultural sector and associated market prices. This shows the importance of performing prospective analyses on co-product credits with consequential approaches in order to capture the current and future socio-political conditions affecting market behaviour (Zamagni et al., 2012). Furthermore, additional waste treatment scenarios could be explored from cradle-to-grave by applying a consequential LCA perspective (Ahlgren et al., 2015; Lund et al., 2010). The system has been conceptualized as a methanol production system (with a product-based FU), but could also be assessed as a waste treatment system (with an input-based FU). In this case, the Environmental Product Declaration (EPD) could contribute to the further harmonization of the cut-off criteria (Borghi et al., 2007). In any case, the system is aligned to the EU's circular economy strategy (EC, 2020) which aims at reducing waste generation, by enhancing reuse and recycling and the establishment of a market for secondary products. This should not, however, encourage the increased production of either feed residues or manure from industrial livestock farming, which highlights the need for adequate regulations in feedstock markets, by taking into account market responses from a supply chain perspective.

5. Conclusions

This study consists of an LCA of an integrated PtF system that produces methanol based on biogas from manure and H_2 generation in combination with a CHP unit in the context of Germany. In view of the multi-functionality of the process, nine scenarios in total were assessed to understand the uncertainty in the modelling choices of co-product credits when applying system expansion. Most of them entail impact reductions relative to the conventional methanol production, generating negative impact values, with only few exceptions (i.e., 8.3% of the impact values). The scenario in which the digestate from AD replaces SSP as a fertiliser delivers greater environmental benefits, regardless of the energy carrier to be replaced in the market by the electricity from the CHP. CC and FD are the only exceptions, as urea production generates more credits than SSP. Assumptions on co-product credits thus play an important role when assessing an integrated system, such as the one proposed here, under the system expansion approach. The choice of avoided processes should consider the market conditions in which co-production takes place, as well as the scale of production of the process itself, which determines subsequent market responses from co-product generation. Applying a consequential LCA perspective could provide further insights on the price-mediated effects triggered by marginal changes in supply and demand of the main product, considering co-product substitution across sectors, although this requires further economic modelling. LCA outcomes are also subject to parameter variability (e.g., in measurements), which should be assessed by means of a sensitivity analysis, especially in the case of emerging technologies to be implemented on an industrial scale. Variability in parameters determining the energy needs and CH4 losses of the small-scale PtF system assessed proves to be critical for the overall environmental performance. A more comprehensive sensitivity analysis based on on-site measurements combined with uncertainty analysis would be necessary to better inform decision-making in other technical, geographical and socio-political contexts.

In spite of uncertainty, the proposed system shows potential to outperform conventional methanol production provided that raw materials are readily available and co-products are generated at competitive prices. In this sense, adopting the PtF technology on a large scale could help meeting the EU's REDII goals as for consumption of advanced fuels, although actual GHG savings are conditional on the kind of electricity used within the system. Besides mitigating CC and FD by substituting for fossil fuels, the PtF system proposed reduces the quantity of manure waste to be treated and disposed of safely, hence contributing towards the goal of a circular economy. Technology adoption is, however, largely dependent on the economic performance of the system as compared to available alternatives. This is why comprehensive life cycle costing of the proposed PtF technology will be carried out at a later stage, in order to estimate trade-offs amongst sustainability dimensions, while providing technical assistance on planning and upscaling.

Declarations of interest

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Supplementary material

Figure S1. Contribution analysis of the processes to the impact categories of marine eutrophication (EP marine), photochemical ozone formation (POF), and stratospheric ozone depletion (ODP).

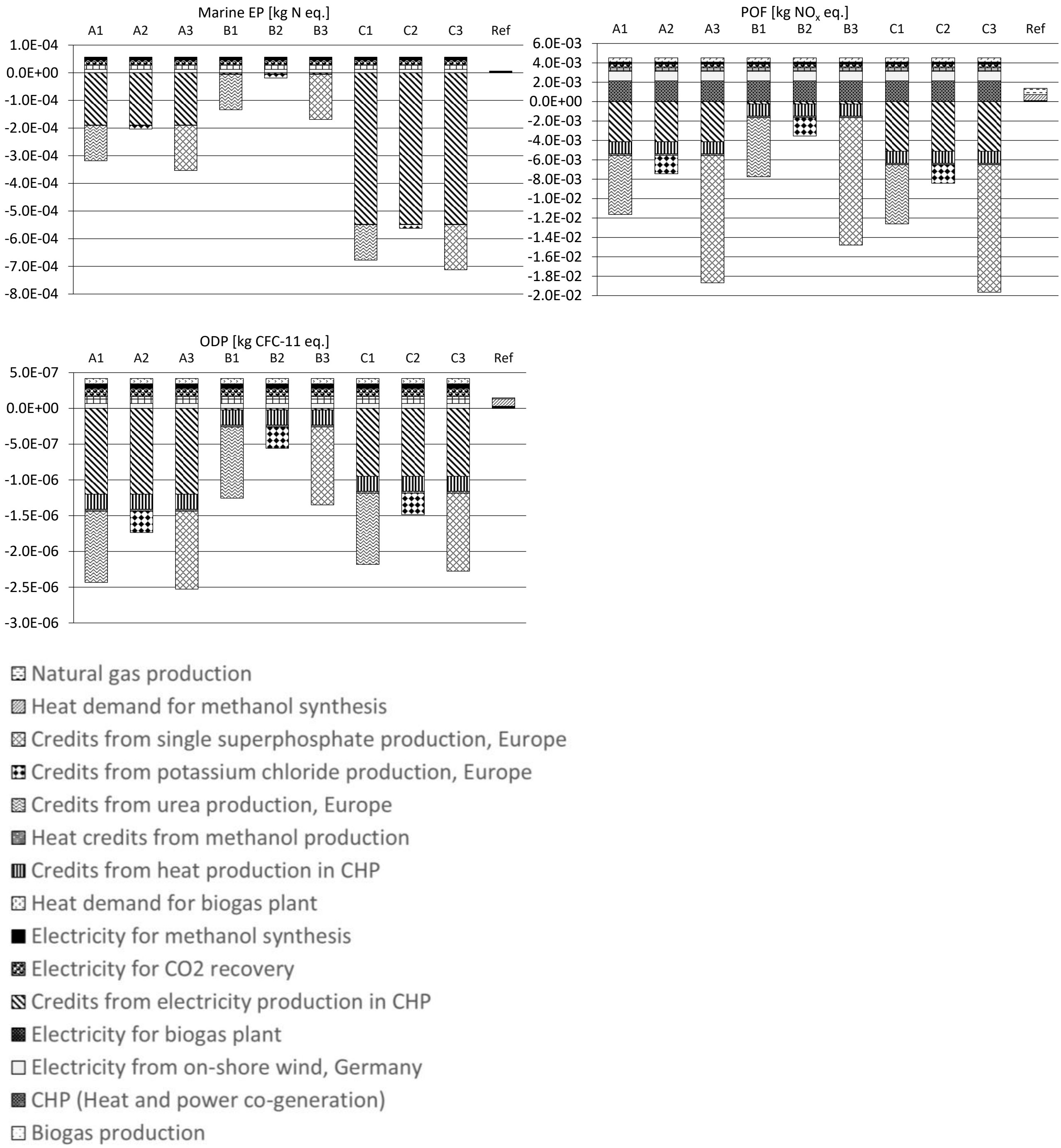


Table S2. Values of the contribution analysis shown in Figure S1.

	Biogas production	CHP (Heat and power co-generation)	Electricity from onshore wind, DE[1]	Electricity for biogas plant	Credits from CHP electricity production	Electricity for CO_2 recovery	Electricity for metha-nol synthesis	Heat demand for biogas plant	Credits from CHP heat production	Heat credits from methanol production	Credits from urea production, EU[2]	Credits from potassium chloride production, EU[2]	Credits from single superphosphate production, EU[2]	Heat demand for methanol synthesis	Natural gas produc-tion
Climate Change (CC)															
A1	0.78	0.06	0.16	0.16	-1.97	0.19	0.10	0.34	-0.98	-0.12	-2.56	0.00	0.00	0.00	0.00
A2	0.78	0.06	0.16	0.16	-1.97	0.19	0.10	0.34	-0.98	-0.12	0.00	-0.43	0.00	0.00	0.00
A3	0.78	0.06	0.16	0.16	-1.97	0.19	0.10	0.34	-0.98	-0.12	0.00	0.00	-1.99	0.00	0.00
B1	0.78	0.06	0.16	0.00	-0.04	0.00	0.00	0.34	-0.98	-0.12	-2.56	0.00	0.00	0.00	0.00
B2	0.78	0.06	0.16	0.00	-0.04	0.00	0.00	0.34	-0.98	-0.12	0.00	-0.43	0.00	0.00	0.00
B3	0.78	0.06	0.16	0.00	-0.04	0.00	0.00	0.34	-0.98	-0.12	0.00	0.00	-1.99	0.00	0.00
C1	0.78	0.06	0.16	0.29	-3.62	0.35	0.19	0.34	-0.98	-0.12	-2.56	0.00	0.00	0.00	0.00
C2	0.78	0.06	0.16	0.29	-3.62	0.35	0.19	0.34	-0.98	-0.12	0.00	-0.43	0.00	0.00	0.00
C3	0.78	0.06	0.16	0.29	-3.62	0.35	0.19	0.34	-0.98	-0.12	0.00	0.00	-1.99	0.00	0.00
Ref	0.00	0.00	0.00	0.00	0.00	0.00	0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.49	0.16
Fossil depletion (FD)															
A1	0.00	0.00	0.06	0.06	-0.75	0.07	0.04	0.14	-0.39	-0.05	-1.13	0.00	0.00	0.00	0.00
A2	0.00	0.00	0.06	0.06	-0.75	0.07	0.04	0.14	-0.39	-0.05	0.00	-0.17	0.00	0.00	0.00
A3	0.00	0.00	0.06	0.06	-0.75	0.07	0.04	0.14	-0.39	-0.05	0.00	0.00	-1.16	0.00	0.00
B1	0.00	0.00	0.06	0.00	-0.01	0.00	0.00	0.14	-0.39	-0.05	-1.13	0.00	0.00	0.00	0.00
B2	0.00	0.00	0.06	0.00	-0.01	0.00	0.00	0.14	-0.39	-0.05	0.00	-0.17	0.00	0.00	0.00
B3	0.00	0.00	0.06	0.00	-0.01	0.00	0.00	0.14	-0.39	-0.05	0.00	0.00	-1.16	0.00	0.00
C1	0.00	0.00	0.06	0.08	-1.06	0.10	0.06	0.14	-0.39	-0.05	-1.13	0.00	0.00	0.00	0.00
C2	0.00	0.00	0.06	0.08	-1.06	0.10	0.06	0.14	-0.39	-0.05	0.00	-0.17	0.00	0.00	0.00
C3	0.00	0.00	0.06	0.08	-1.06	0.10	0.06	0.14	-0.39	-0.05	0.00	0.00	-1.16	0.00	0.00
Ref	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.19	0.58

	Biogas production	CHP (Heat and power co-generation)	Electricity from onshore wind, DE[1]	Electricity for biogas plant	Credits from CHP electricity production	Electricity for CO_2 recovery	Electricity for methanol synthesis	Heat demand for biogas plant	Credits from CHP heat production	Heat credits from methanol production	Credits from urea production, EU[2]	Credits from potassium chloride production, EU[2]	Credits from single superphosphate production, EU[2]	Heat demand for methanol synthesis	Natural gas production
Acidification potential (AP)															
A1	1.54E-03	4.54E-04	7.00E-04	7.33E-04	-9.17E-03	8.87E-04	4.79E-04	1.79E-04	-5.11E-04	-6.44E-05	-1.21E-02	0.00	0.00	0.00	0.00
A2	1.54E-03	4.54E-04	7.00E-04	7.33E-04	-9.17E-03	8.87E-04	4.79E-04	1.79E-04	-5.11E-04	-6.44E-05	0.00	-1.33E-03	0.00	0.00	0.00
A3	1.54E-03	4.54E-04	7.00E-04	7.33E-04	-9.17E-03	8.87E-04	4.79E-04	1.79E-04	-5.11E-04	-6.44E-05	0.00	0.00	-2.52E-02	0.00	0.00
B1	1.54E-03	4.54E-04	7.00E-04	7.33E-04	-2.00E-04	8.87E-04	4.79E-04	1.79E-04	-5.11E-04	-6.44E-05	-1.21E-02	0.00	0.00	0.00	0.00
B2	1.54E-03	4.54E-04	7.00E-04	7.33E-04	-2.00E-04	8.87E-04	4.79E-04	1.79E-04	-5.11E-04	-6.44E-05	0.00	-1.33E-03	0.00	0.00	0.00
B3	1.54E-03	4.54E-04	7.00E-04	7.33E-04	-2.00E-04	8.87E-04	4.79E-04	1.79E-04	-5.11E-04	-6.44E-05	0.00	0.00	-2.52E-02	0.00	0.00
C1	1.54E-03	4.54E-04	7.00E-04	7.33E-04	-3.85E-03	8.87E-04	4.79E-04	1.79E-04	-5.11E-04	-6.44E-05	-1.21E-02	0.00	0.00	0.00	0.00
C2	1.54E-03	4.54E-04	7.00E-04	7.33E-04	-3.85E-03	8.87E-04	4.79E-04	1.79E-04	-5.11E-04	-6.44E-05	0.00	-1.33E-03	0.00	0.00	0.00
C3	1.54E-03	4.54E-04	7.00E-04	7.33E-04	-3.85E-03	8.87E-04	4.79E-04	1.79E-04	-5.11E-04	-6.44E-05	0.00	0.00	-2.52E-02	0.00	0.00
Ref	0.00	0.00	0.00	0.00	0.00	0.00	2.29E-04	0.00	0.00	0.00	0.00	0.00	0.00	2.55E-04	1.13E-03

	Biogas production	CHP (Heat and power co-generation)	Electricity from onshore wind, DE[1]	Electricity for biogas plant	Credits from CHP electricity production	Electricity for CO_2 recovery	Electricity for methanol synthesis	Heat demand for biogas plant	Credits from CHP heat production	Heat credits from methanol production	Credits from urea production, EU[2]	Credits from potassium chloride production, EU[2]	Credits from single superphosphate production, EU[2]	Heat demand for methanol synthesis	Natural gas produc-tion
Freshwater eutrophication potential (EP)															
A1	0.00	0.00	1.25E-04	2.27E-04	-2.83E-03	2.74E-04	1.48E-04	4.97E-06	-1.42E-05	0.00	-4.78E-04	0.00	0.00	0.00	0.00
A2	0.00	0.00	1.25E-04	2.27E-04	-2.83E-03	2.74E-04	1.48E-04	4.97E-06	-1.42E-05	0.00	0.00	-1.94E-04	0.00	0.00	0.00
A3	0.00	0.00	1.25E-04	2.27E-04	-2.83E-03	2.74E-04	1.48E-04	4.97E-06	-1.42E-05	0.00	0.00	0.00	-3.90E-03	0.00	0.00
B1	0.00	0.00	1.25E-04	2.47E-06	-3.09E-05	3.00E-06	1.62E-06	4.97E-06	-1.42E-05	0.00	-4.78E-04	0.00	0.00	0.00	0.00
B2	0.00	0.00	1.25E-04	2.47E-06	-3.09E-05	3.00E-06	1.62E-06	4.97E-06	-1.42E-05	0.00	0.00	-1.94E-04	0.00	0.00	0.00
B3	0.00	0.00	1.25E-04	2.47E-06	-3.09E-05	3.00E-06	1.62E-06	4.97E-06	-1.42E-05	0.00	0.00	0.00	-3.90E-03	0.00	0.00
C1	0.00	0.00	1.25E-04	6.90E-04	-8.63E-03	8.35E-04	4.51E-04	4.97E-06	-1.42E-05	0.00	-4.78E-04	0.00	0.00	0.00	0.00
C2	0.00	0.00	1.25E-04	6.90E-04	-8.63E-03	8.35E-04	4.51E-04	4.97E-06	-1.42E-05	0.00	0.00	-1.94E-04	0.00	0.00	0.00
C3	0.00	0.00	1.25E-04	6.90E-04	-8.63E-03	8.35E-04	4.51E-04	4.97E-06	-1.42E-05	0.00	0.00	0.00	-3.90E-03	0.00	0.00
Ref	0.00	0.00	0.00	0.00	0.00	0.00	7.09E-05	0.00	0.00	0.00	0.00	0.00	0.00	7.09E-06	8.48E-06

	Biogas production	CHP (Heat and power co-generation)	Electricity from onshore wind, DE[1]	Electricity for biogas plant	Credits from CHP electricity production	Electricity for CO_2 recovery	Electricity for methanol synthesis	Heat demand for biogas plant	Credits from CHP heat production	Heat credits from methanol production	Credits from urea production, EU[2]	Credits from potassium chloride production, EU[2]	Credits from single superphosphate production, EU[2]	Heat demand for methanol synthesis	Natural gas production
Marine eutrophication potential (EP marine)															
A1	0.00	0.00	1.24E-05	1.51E-05	-1.89E-04	1.83E-05	9.88E-06	4.05E-07	-1.16E-06	-1.46E-07	-1.28E-04	0.00	0.00	0.00	0.00
A2	0.00	0.00	1.24E-05	1.51E-05	-1.89E-04	1.83E-05	9.88E-06	4.05E-07	-1.16E-06	-1.46E-07	0.00	-1.32E-05	0.00	0.00	0.00
A3	0.00	0.00	1.24E-05	1.51E-05	-1.89E-04	1.83E-05	9.88E-06	4.05E-07	-1.16E-06	-1.46E-07	0.00	0.00	-1.63E-04	0.00	0.00
B1	0.00	0.00	1.24E-05	3.64E-07	-4.56E-06	4.41E-07	2.38E-07	4.05E-07	-1.16E-06	-1.46E-07	-1.28E-04	0.00	0.00	0.00	0.00
B2	0.00	0.00	1.24E-05	3.64E-07	-4.56E-06	4.41E-07	2.38E-07	4.05E-07	-1.16E-06	-1.46E-07	0.00	-1.32E-05	0.00	0.00	0.00
B3	0.00	0.00	1.24E-05	3.64E-07	-4.56E-06	4.41E-07	2.38E-07	4.05E-07	-1.16E-06	-1.46E-07	0.00	0.00	-1.63E-04	0.00	0.00
C1	0.00	0.00	1.24E-05	4.39E-05	-5.48E-04	5.31E-05	2.87E-05	4.05E-07	-1.16E-06	-1.46E-07	-1.28E-04	0.00	0.00	0.00	0.00
C2	0.00	0.00	1.24E-05	4.39E-05	-5.48E-04	5.31E-05	2.87E-05	4.05E-07	-1.16E-06	-1.46E-07	0.00	-1.32E-05	0.00	0.00	0.00
C3	0.00	0.00	1.24E-05	4.39E-05	-5.48E-04	5.31E-05	2.87E-05	4.05E-07	-1.16E-06	-1.46E-07	0.00	0.00	-1.63E-04	0.00	0.00
Ref	0.00	0.00	0.00	0.00	0.00	0.00	4.73E-06	0.00	0.00	0.00	0.00	0.00	0.00	5.77E-07	8.48E-07

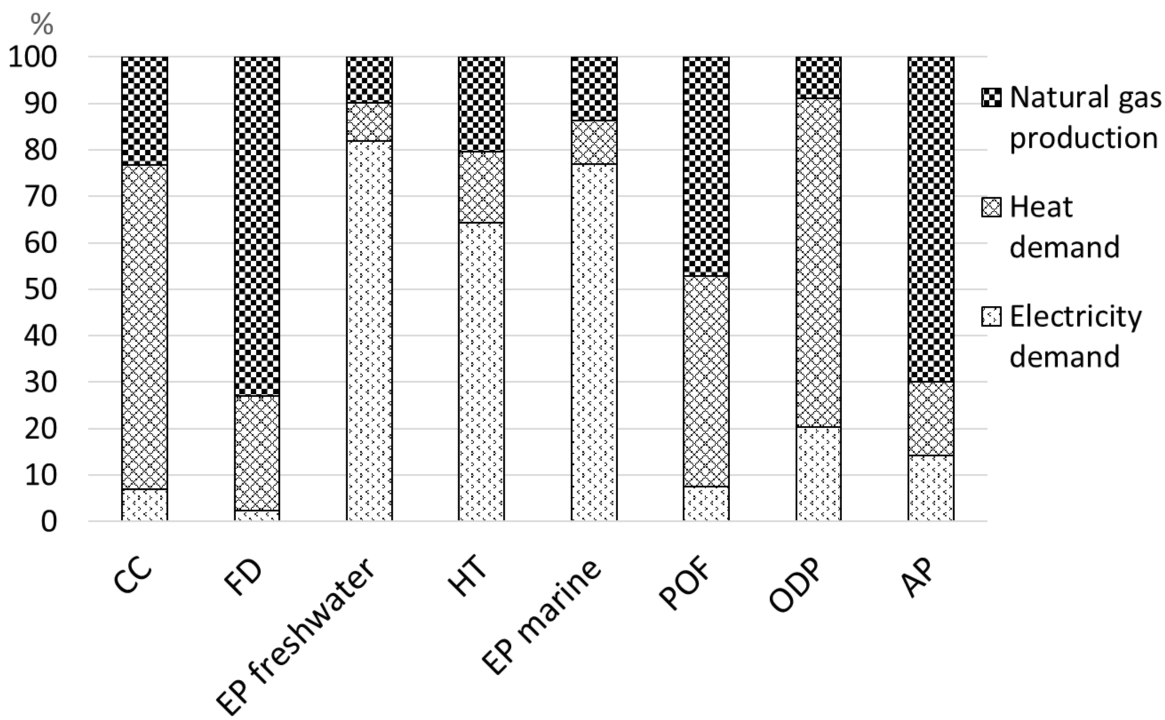
	Biogas production	CHP (Heat and power co-generation)	Electricity from on-shore wind, DE[1]	Electricity for biogas plant	Credits from CHP electricity production	Electricity for CO_2 recovery	Electricity for methanol synthesis	Heat demand for biogas plant	Credits from CHP heat production	Heat credits from methanol production	Credits from urea production, EU[2]	Credits from potassium chloride production, EU[2]	Credits from single superphosphate production, EU[2]	Heat demand for methanol synthesis	Natural gas production
Human toxicity (HT)															
A1	0.00	0.00	8.21E-01	2.52E-01	-3.16	3.06E-01	1.65E-01	1.33E-02	-3.78E-02	-4.76E-03	-2.58	0.00	0.00	0.00	0.00
A2	0.00	0.00	8.21E-01	2.52E-01	-3.16	3.06E-01	1.65E-01	1.33E-02	-3.78E-02	-4.76E-03	0.00	-1.35	0.00	0.00	0.00
A3	0.00	0.00	8.21E-01	2.52E-01	-3.16	3.06E-01	1.65E-01	1.33E-02	-3.78E-02	-4.76E-03	0.00	0.00	-9.09	0.00	0.00
B1	0.00	0.00	8.21E-01	1.71E-02	-2.14E-01	2.08E-02	1.12E-02	1.33E-02	-3.78E-02	-4.76E-03	-2.58	0.00	0.00	0.00	0.00
B2	0.00	0.00	8.21E-01	1.71E-02	-2.14E-01	2.08E-02	1.12E-02	1.33E-02	-3.78E-02	-4.76E-03	0.00	-1.35	0.00	0.00	0.00
B3	0.00	0.00	8.21E-01	1.71E-02	-2.14E-01	2.08E-02	1.12E-02	1.33E-02	-3.78E-02	-4.76E-03	0.00	0.00	-9.09	0.00	0.00
C1	0.00	0.00	8.21E-01	6.72E-01	-8.40	8.14E-01	4.40E-01	1.33E-02	-3.78E-02	-4.76E-03	-2.58	0.00	0.00	0.00	0.00
C2	0.00	0.00	8.21E-01	6.72E-01	-8.40	8.14E-01	4.40E-01	1.33E-02	-3.78E-02	-4.76E-03	0.00	-1.35	0.00	0.00	0.00
C3	0.00	0.00	8.21E-01	6.72E-01	-8.40	8.14E-01	4.40E-01	1.33E-02	-3.78E-02	-4.76E-03	0.00	0.00	-9.09	0.00	0.00
Ref	0.00	0.00	0.00	0.00	0.00	0.00	7.90E-02	0.00	0.00	0.00	0.00	0.00	0.00	1.89E-02	2.50E-02

	Biogas production	CHP (Heat and power co-generation)	Electricity from onshore wind, DE[1]	Electricity for biogas plant	Credits from CHP electricity production	Electricity for CO_2 recovery	Electricity for methanol synthesis	Heat demand for biogas plant	Credits from CHP heat production	Heat credits from methanol production	Credits from urea production, EU[2]	Credits from potassium chloride production, EU[2]	Credits from single superphosphate production, EU[2]	Heat demand for methanol synthesis	Natural gas production
Photochemical ozone formation (POF)															
A1	0.00	2.14E-03	9.96E-04	3.31E-04	-4.14E-03	4.00E-04	2.17E-04	4.35E-04	-1.24E-03	-1.57E-04	-6.10E-03	0.00	0.00	0.00	0.00
A2	0.00	2.14E-03	9.96E-04	3.31E-04	-4.14E-03	4.00E-04	2.17E-04	4.35E-04	-1.24E-03	-1.57E-04	0.00	-1.90E-03	0.00	0.00	0.00
A3	0.00	2.14E-03	9.96E-04	3.31E-04	-4.14E-03	4.00E-04	2.17E-04	4.35E-04	-1.24E-03	-1.57E-04	0.00	0.00	-1.31E-02	0.00	0.00
B1	0.00	2.14E-03	9.96E-04	2.00E-05	-2.49E-04	2.41E-05	1.31E-05	4.35E-04	-1.24E-03	-1.57E-04	-6.10E-03	0.00	0.00	0.00	0.00
B2	0.00	2.14E-03	9.96E-04	2.00E-05	-2.49E-04	2.41E-05	1.31E-05	4.35E-04	-1.24E-03	-1.57E-04	0.00	-1.90E-03	0.00	0.00	0.00
B3	0.00	2.14E-03	9.96E-04	2.00E-05	-2.49E-04	2.41E-05	1.31E-05	4.35E-04	-1.24E-03	-1.57E-04	0.00	0.00	-1.31E-02	0.00	0.00
C1	0.00	2.14E-03	9.96E-04	4.09E-04	-5.11E-03	4.95E-04	2.68E-04	4.35E-04	-1.24E-03	-1.57E-04	-6.10E-03	0.00	0.00	0.00	0.00
C2	0.00	2.14E-03	9.96E-04	4.09E-04	-5.11E-03	4.95E-04	2.68E-04	4.35E-04	-1.24E-03	-1.57E-04	0.00	-1.90E-03	0.00	0.00	0.00
C3	0.00	2.14E-03	9.96E-04	4.09E-04	-5.11E-03	4.95E-04	2.68E-04	4.35E-04	-1.24E-03	-1.57E-04	0.00		-1.31E-02	0.00	0.00
Ref	0.00	0.00	0.00	0.00	0.00	0.00	1.04E-04	0.00	0.00	0.00	0.00	0.00	0.00	6.20E-04	6.44E-04

	Biogas production	CHP (Heat and power co-generation)	Electricity from onshore wind, DE[1]	Electricity for biogas plant	Credits from CHP electricity production	Electricity for CO_2 recovery	Electricity for methanol synthesis	Heat demand for biogas plant	Credits from CHP heat production	Heat credits from methanol production	Credits from urea production, EU[2]	Credits from potassium chloride production, EU[2]	Credits from single superphosphate production, EU[2]	Heat demand for methanol synthesis	Natural gas production
Stratospheric ozone depletion (ODP)															
A1	0.00	0.00	6.93E-08	9.58E-08	-1.20E-06	1.16E-07	6.27E-08	7.39E-08	-2.11E-07	-2.66E-08	-9.95E-07	0.00	0.00	0.00	0.00
A2	0.00	0.00	6.93E-08	9.58E-08	-1.20E-06	1.16E-07	6.27E-08	7.39E-08	-2.11E-07	-2.66E-08	0.00	-2.99E-07	0.00	0.00	0.00
A3	0.00	0.00	6.93E-08	9.58E-08	-1.20E-06	1.16E-07	6.27E-08	7.39E-08	-2.11E-07	-2.66E-08	0.00	0.00	-1.09E-06	0.00	0.00
B1	0.00	0.00	6.93E-08	1.76E-09	-2.20E-08	2.13E-09	1.15E-09	7.39E-08	-2.11E-07	-2.66E-08	-9.95E-07	0.00	0.00	0.00	0.00
B2	0.00	0.00	6.93E-08	1.76E-09	-2.20E-08	2.13E-09	1.15E-09	7.39E-08	-2.11E-07	-2.66E-08	0.00	-2.99E-07	0.00	0.00	0.00
B3	0.00	0.00	6.93E-08	1.76E-09	-2.20E-08	2.13E-09	1.15E-09	7.39E-08	-2.11E-07	-2.66E-08	0.00	0.00	-1.09E-06	0.00	0.00
C1	0.00	0.00	6.93E-08	7.59E-08	-9.49E-07	9.19E-08	4.96E-08	7.39E-08	-2.11E-07	-2.66E-08	-9.95E-07	0.00	0.00	0.00	0.00
C2	0.00	0.00	6.93E-08	7.59E-08	-9.49E-07	9.19E-08	4.96E-08	7.39E-08	-2.11E-07	-2.66E-08	0.00	-2.99E-07	0.00	0.00	0.00
C3	0.00	0.00	6.93E-08	7.59E-08	-9.49E-07	9.19E-08	4.96E-08	7.39E-08	-2.11E-07	-2.66E-08	0.00	0.00	-1.09E-06	0.00	0.00
Ref	0.00	0.00	0.00	0.00	0.00	0.00	3.00E-08	0.00	0.00	0.00	0.00	0.00	0.00	1.05E-07	1.32E-08

[1]DE: Germany, [2]EU: Europe

Figure S2. Contribution analysis of each sub-process to the impacts from conventional methanol production.



CC: climate change, FD: fossil depletion, EP: eutrophication potential, HT: human toxicity, POF: photochemical ozone formation, ODP: stratospheric ozone depletion, AP: acidification potential

Table S2. Results for a system, which utilizes German grid-mix electricity for hydrogen production instead of wind-based electricity.

	Climate change [kg CO_2 eq.]	Fossil depletion [kg oil eq.]	EP freshwater [kg P eq.]	Human toxicity [kg 1,4-DB eq.]	EP marine [kg N eq.]	POF [kg NO_x eq.]	ODP [kg CFC-11 eq.]	Acidification [kg SO_2 eq.]
Total	3.69	0.37	6.24E-03	4.861	3.18E-04	4.70E-03	1.70E-06	1.26E-02
Biogas production	0.79	0	0	0	0	0	0	3.06E-03
CHP (Heat and power co-generation)	0.05	0	0	0	0	1.90E-03	0	4.03E-04
Electricity for methanol synthesis	0.16	0.06	2.31E-04	0.26	1.54E-05	3.37E-04	9.75E-08	7.46E-04
Credits from CHP electricity production	-1.75	-0.66	-2.52E-03	-2.81	-1.68E-04	-3.68E-03	-1.06E-06	-8.15E-03
Electricity for CO_2 recovery	0.19	0.07	2.79E-04	0.31	1.86E-05	4.08E-04	1.18E-07	9.03E-04
Electricity for biogas plant	0.10	0.04	1.48E-04	0.17	9.88E-06	2.17E-04	6.27E-08	4.79E-04
Electricity for H_2 production	5.98	2.27	8.60E-03	9.59	5.73E-04	1.26E-02	3.64E-06	2.78E-02
Heat demand for biogas plant	0.35	0.14	5.06E-06	0.01	4.12E-07	4.43E-04	7.52E-08	1.82E-04
Credits from CHP heat production	-0.87	-0.35	-1.26E-05	-0.03	-1.03E-06	-1.11E-03	-1.88E-07	-4.55E-04
Heat credits from methanol production	-0.12	-0.05	-1.79E-06	0	-1.46E-07	-1.57E-04	-2.66E-08	-6.44E-05
Credits from urea production, Europe	-1.19	-1.15	-4.86E-04	-2.62	-1.31E-04	-6.21E-03	-1.01E-06	-1.24E-02

EP: Eutrophication potential, POF: Photochemical ozone formation, ODP: Stratospheric ozone depletion