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LIFE CYCLE ASSESSMENT OF A NOVEL POWER-TO-FUEL SYSTEM FOR METHANOL PRODUCTION USING CO_2 FROM BIOGAS

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ABSTRACT

Power-to-Fuel technologies are important for reducing fossil fuel consumption in the transport sector. Carbon dioxide (CO_2) from biogas and hydrogen (H_2) production from wind electrolysis provide convenient components for the synthesis of methanol. This study evaluates the environmental performance of a novel Power-to-Fuel (PTF) system, in which methanol is produced at a biogas production plant. A Life Cycle Assessment is carried out considering five feasible process routes in order to identify the one that delivers greater environmental benefits. The proposed system is also compared to a system of conventional methanol production. Results show improvements in most impact categories, which make it interesting from the environmental point of view.

Keywords: Power-to-Liquid, biogas from manure, small-scale methanol production, environmental impacts, scenario analysis

NOMENCLATURE

Abbreviations	
AP	Acidification potential
CHP	Combined heat and power
EP	Eutrophication potential
FC	Freshwater consumption
FD	Fossil depletion
FLH	Full load hours
GHG	Greenhouse gas
HT	Human toxicity
LCA	Life cycle assessment
LCI	Life cycle inventory
LCIA	Life cycle impact assessment
LHV	Lower heating value
ODP	Stratospheric ozone depletion
PEM	Polymer electrolyte membrane
POF	Photochemical ozone formation
PtF	Power-to-Fuel
Symbols	
η	Energy efficiency

1. INTRODUCTION

The European Union's (EU) goal of reducing its greenhouse gas (GHG) emissions by 2050 by 80-95% relative to 1990 levels requires innovative concepts to achieve the transformation to a low-carbon economy [1]. One possible way to do it can be replacing fossil-based fuels and chemicals, such as methanol, by the same materials produced with renewable resources. The often named Power-to-Fuel (PtF) technologies (also known as Power-to-Liquid) or e-fuels combine H_2 from renewable energies via electrolysis with recycled CO_2 from industry or agriculture to produce liquid fuels [2]. Such concepts have a high potential to support the German energy transition by using less fossil resources [3].

Methanol, e.g., is conventionally produced from natural gas, based on synthetic gas from steam reforming of natural gas (NG) and H_2. In the light of climate change, the CO_2-based direct methanol synthesis has increasingly gained scientific interest throughout recent years [4, 5]. Biogas from anaerobic digestion can constitute a source of CO_2; being underused nowadays as it is mostly released into the atmosphere. The potential of CO_2 captured by biogas upgrading plants in Germany is estimated at 1.5 Mt [5]. This could yield 1.1 Mt of methanol, assuming that there are no constraints in H_2 availability. This roughly corresponds to the overall methanol production in Germany in 2016 [6].

Life cycle assessment (LCA) is a widely used method for the analysis of the environmental performance of complex production systems. It allows comparing technological alternatives and identifying those processes to be improved further [7]. There are various LCA case studies on synthetic fuel production [8-11] and biogas upgrading [12-15]. The latter quantify GHG savings due to reduced emissions and the substitution of chemical fertilizers [16]. Although CO_2 capture and utilization plays a role in some LCAs [13, 17], it does so in different contexts, e.g., for the production of H_2 or methane (CH_4). Besides, none of the studies consider CO_2 from anaerobic digestion for the production of synthetic fuels such as methanol.

In order to fill this gap, this study assesses small-scale methanol production which includes biogas upgrading on a farm site and uses wind power for H_2 production. The ultimate objective of the study is to assess the energy and environmental performances of the proposed system; while evaluating the influence of model assumptions on co-product environmental credits. Finally, the system is compared to the average fossil-based methanol production process.

2. MATERIAL AND METHODS

The methodology applied in this study is explained in detail in Eggemann et al. [18] and, thus, will only be summarized in the remainder of this chapter. It is carried out in accordance with the ISO 14040 and 14044 [19] and made up of the following phases:

2.1 Goal and scope

The objective is to measure environmental impacts from six different scenarios of a PtF system from cradle to gate. The system boundaries enclose all production steps towards 1 kg of methanol, which is the final product, and also defined as the functional unit (FU).

2.2 System description

The system described has been designed as proposed by Decker et al. [20] and is based on Eggemann et al. [18]. It is composed of the following sub-systems: *a)* a dairy cow farm with biogas production corresponding to the small manure plants widely found in Germany which use manure in large amounts, *b)* a CO_2 capturing unit using biogas upgrading technology and a CO_2 storage for buffering, *c)* a combined heat and power (CHP) unit for the energetic utilization of the CH_4, *d)* a wind turbine and electrolyzer (PEM) for H_2 supply and *e)* a methanol production plant with an H_2 storage. The system boundary is set at the farm gate, assuming close proximity of the wind turbine for simplification (Fig 1).

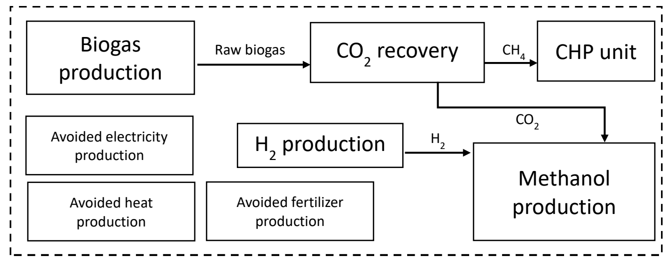

Fig 1 System boundary

The system, designed for the pilot scale, has a potential annual methanol production of 212.5 t assuming 8500 full load hours (FLH) and corresponding to 137.5 $kW_{th,LHV}$ (lower heating value). It has an energy efficiency of 36% based on the individual energy efficiency rates shown in Tab 1. Due to data availability constraints, capital goods are not included.

Tab 1 Energy efficiency of the multiple production steps as in Eggemann et al. [18]

	Biogas plant (incl. CHP)	Electro-lyzer	CO_2 re-covery plant	Methanol synthesis plant
η sub-system	0.65 [21]	0.70 [22]	0.92 [23]	0.85 [24]

Using the ‘system expansion’ approach, co-product credit is included in the system which generates ‘avoided processes’. In this case, power and heat from the CHP unit and the methanol production and nitrogen (N) fertilizer from digestate are obtained together with the FU. Further details can be found in Eggemann et al. [18].

The assessment considers six different scenarios which mostly have the same system boundary and FU as the first one. However, they vary in terms of the choice of the avoided processes, based on potential use options for the co-products. The last scenario refers to conventional methanol production where the system boundaries enclose the methanol production process and heat, electricity and NG production. The FU with 1 kg of methanol remains the same.

The scenarios are as follows:

1) *Base* – as described in this section above; N from digestate is used as substitute for average ammonium nitrate production in the EU; surplus thermal energy replaces heat production from the average mix in the EU and surplus electricity is sold to the grid; hence replacing electricity from the average German production mix.
2) *Urea* – includes the credit for replacing urea production instead of ammonium nitrate to be applied as fertilizer.
3) *Grid mix* – H_2 produced with electricity from German grid mix instead of wind power.
4) *Wind* – electricity production from the CHP substitutes offshore wind power for simulating a future electricity mix with renewables.
5) *Coal* – CHP electricity replaces coal-based (lignite) electricity for conventional simulation.
6) *Conventional* – reference process of conventional methanol synthesis via steam reforming of NG

2.3 Life cycle inventory (LCI)

In this chapter, all emissions and material flows per FU are collected with respect to the different scenarios described in section 2.2. The GaBi ts software was used to model the processes mostly using data from the Ecoinvent 3.5 database [25]. Other data sources, as well as the main assumptions are based on Eggemann et al. [18] and only presented in a nutshell.

The farm corresponds to a small-manure plant with a capacity of 75 kW. It uses feedstock which is made up of residues from dairy farming, namely 80% manure and 20% grain shreds from feed production. The composition of biogas is measured at 53% CH_4, 46% CO_2 and 1% oxygen. For reasons of simplicity, other components were neglected such as N_2O, H_2, H_2O and NH_3 which can leak during the digestion process. However, due to partly open digestate storage at the farm, NH_3 emissions according to EMEP/EEA [26] and methane emissions (1 kg/MWh) were included in the inventory. An N content of ammonium nitrate of 34% and of urea of 46% were considered [27, 28] which are substituted by the content of elemental N in the digestate. This means that for 1 kg of N in digestate, 2.94 kg of ammonium nitrate or 2.17 kg of urea must be produced.

No losses are assumed during the PSA process, simulating an ideal process. Furthermore, it is not accounted for the trace gases and other components of the flue gases. The total CO_2 is used in the methanol production process while the O_2 emits into the air. It should be noted that the biogas plant, as it is, produces a slight excess in the system of about 1 kg of CO_2, thus, a storage facility should be considered to guarantee a constant methanol production.

The methanol synthesis occurs in an iso-thermal reactor with 1.37 kg CO_2 per kg methanol according to Billig et al. [5]. The thermal discharge from this process is used to substitute heat production. A 133 $kW_{th,LHV}$ methanol synthesis plant demands 34.25 kg/h CO_2 and 4.7 kg/h H_2. The H_2 amount corresponds to 158 $kW_{th,LHV}$. A wind turbine capacity of 1.2 MW and that of an electrolyzer of 960 kW are assumed for the H_2 production. Further assumptions are explained in Eggemann et al. [18]. Conventional methanol synthesis excludes the production of some inputs, e.g. aluminum and copper oxide, because only very small amounts are used, hence, generating small impacts. Furthermore, the scenario assumes imported NG from Russia.

2.4 Life cycle inventory assessment (LCIA)

The environmental impacts, arising from the LCI, are calculated using the ReCiPe 2016 method. Results refer to the midpoint level from the *hierarchist perspective*. The chosen impact categories are as follows: "Climate change (excl. biogenic carbon) with a 100-year time horizon (GHG), […] eutrophication potential (freshwater and marine) (EP), acidification potential (terrestrial) (AP), fossil depletion (FD), photochemical ozone formation (POF), human toxicity (HT) and stratospheric ozone depletion (ODP)", p.7 in [18].

3. RESULTS AND DISCUSSION

Results for the selected impact categories and scenarios are summarized in Tab 2. All of the scenarios, but the *Grid mix* and *Conventional* scenarios, deliver mainly negative emissions, i.e. savings; which means that co-product environmental credits offset overall emissions. The other two only show positive emissions in all categories, i.e. environmental burdens, except for ODP for *Grid mix*.

CO_2 eq. savings of -5.41 kg are achieved for *Coal*, closely followed by *Base* with -4.22 kg. *Urea* notes savings of -3.64 kg and *Wind* of -2.83 kg. *Conventional* and *Grid mix*, on the other hand, note GHG emissions of 0.74 kg CO_2 eq. and 1.6 kg CO_2 eq., respectively, accounting for increases by 114.8% and 127.5% with respect to the *Base* scenario. The overall emissions in *Conventional* appear to be reasonable compared to Otto [29], who mentions emissions for the conventional methanol production process from NG of 0.51 kg CO_2.

Base performs better than *Urea*, which indicates that the process of urea production causes more emissions than the production of ammonium nitrate. Especially, when considering that conventionally produced urea even contains a higher share of N than ammonium nitrate, thus, requiring a lower amount of urea for the same amount of N.

Tab 2 Results of the LCIA for the different scenarios as mentioned in section 2.2

	Base	Urea	Grid mix	Wind	Coal	Conven -tional
GHG [kg CO_2 eq.]	-4.220	-3.640	1.600	-2.830	-5.410	0.735
FD [kg oil eq.]	-0.841	-0.820	1.370	-0.312	-1.060	0.789
EP freshwater [kg P eq.]	-2.02E-03	-1.97E-03	6.46E-03	2.73E-06	-6.20E-03	9.66E-05
EP marine [kg N eq.]	-1.58E-04	-1.36E-04	4.03E-04	-2.50E-05	-4.17E-04	6.16E-06
AP [kg SO_2 eq.]	8.33E-03	1.01E-02	3.55E-02	1.48E-02	1.22E-02	1.68E-03
HT [kg 1,4-DB eq.]	-2.044	-1.619	6.724	0.078	-5.825	0.123
POF [kg NO_x eq.]	0.177	0.180	0.189	0.181	0.177	0.002
ODP [kg CFC-11 eq.]	-2.06E-05	-1.01E-06	-1.70E-05	-1.97E-05	-2.04E-05	1.49E-07

The contribution that each sub-stage makes to the total climate change impact is shown in Fig 2. The results show that assumptions on the credits from thermal energy are not relevant when comparing the scenarios. The largest contributions for all scenarios are made by the biogas production and the heat and power co-generation of the CHP unit, e.g., for *Base* with 42.2%

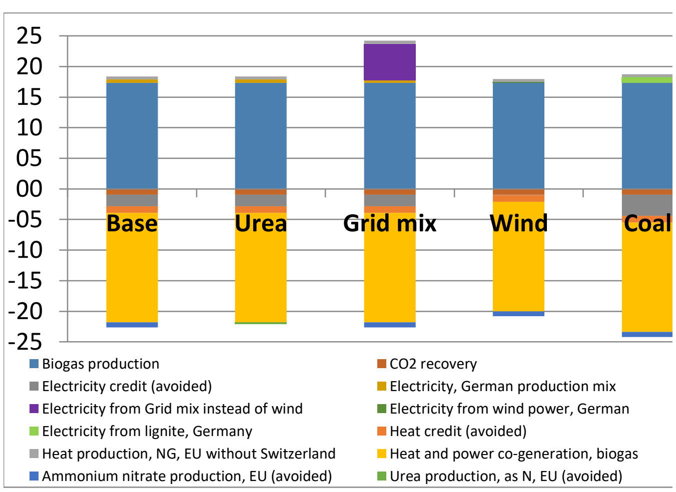

Fig 2 Climate change impact for the first five scenarios [the x-axis describes the different scenarios; the y-axis is in units of kg CO_2 eq.]

and 43.7%, respectively. Fertilizer savings with approx. 2% for the scenarios *Base*, *Grid mix*, *Wind* and *Coal* and only 0.6% for *Urea* are marginal but make a small difference according to the chosen avoided process. The use of the electricity from the German mix instead of wind power accounts for 12.8% of GHG emissions in the *Grid mix* scenario. This shows the importance of obtaining H_2 from renewable sources. Therefore, wind energy was chosen for H_2 production for the other scenarios.

When comparing *Base* with *Wind* and *Coal*, *Wind* performs worse and *Coal* better than *Base*. *Coal* notes the greatest GHG emission savings, as the electricity, which is replaced by power from the CHP, is coal-based and therefore has higher emissions. *Wind* still notes savings, although they are rather small. The electricity credit plays a part in the outcome, contributing 4.5% to the system in *Base*, 0.1% in *Wind* and 8% in *Coal*. The *Conventional* scenario is not included in Fig 2, as the processes are different from the rest. Nevertheless, the sub-stage, which accounts for the highest emissions, is heat production with 66% due to the high heat demand of the methanol production process.

Savings in EP, FD can be achieved for all scenarios but Grid mix and Conventional. The only burdens by all scenarios occur for the impact categories of AP and POF, and HT for some. The impacts for the proposed system range for AP from 7.85E-03 to 8.79E-03 kg SO_2 eq. and for POF from 1.77E-01 to 1.79E-01 kg NO_x eq. The main contribution to the HT savings of -2.04 kg 1,4-DB eq. in *Base* is made by the replaced electricity with 56%. In *Coal*, these savings are even greater, where replaced electricity, as the main contributor, accounts for 69% of the savings.

In general, the avoided processes have an impact on the entire system. Without them, the system does not account for savings. The proposed system shows relatively low impacts for all sub-systems but the biogas production and the CHP unit. In particular, the generated electricity achieves the highest share in the contribution to most categories. If electricity from wind is replaced by electricity from the grid mix, the system performs poorly. Another benefit by the system, apart from its flexibility, is that the carbon in the system is biogenic, coming from animal feces and vegetable resources, and not from fossil sources. Therefore, if compared to conventional methanol production, the system achieves high emission savings, using internal heat and electricity production instead of the offered German mix.

4. CONCLUSIONS

The study assessed a novel PtF system for a small-scale, local biogas plant with respect to different scenarios. The scenarios of the comprehensive LCA show that the system can achieve environmental benefits, especially, when compared to the conventional system. However, the environmental performance of such an integrated system with multiple co-products is subject to the modeling choices in the application of the LCA methodology. These should try to represent the actual situation in the geographical and temporal context in which the system is going to be applied. The application of a consequential LCA perspective is recommended for including the economy-wide environmental implications of successive market substitution effects due to multiple co-product generation. Similarly, the scale of production may alter the results from the comparative assessment, since impacts do not vary in a linear fashion; hence the importance of assessing upscaling.

In conclusion, the system under study, with the assumption of using renewable energies, can make a contribution to a low-carbon economy, especially when considering the fact that conventional methanol production could be replaced by it. The aspect that the CO_2 originates from biogenic and not from fossil sources is another advantage.

The novel system can further be modified or expanded by applying different electrolyzer technologies or CO_2 recovery methods and including the production of capital goods. The conventional system can be expanded including more inputs and losses from NG transport, causing increased relative savings by the proposed system. The economic performance of the system is equally important in determining its overall sustainability and driving investment decisions. This will be investigated by means of a techno-economic analysis.

REFERENCES

[1] European Commission. EU climate action. Available from: https://ec.europa.eu/clima/citizens/eu_en. 25.04.2019.

[2] Dietrich RU, Albrecht F, Pregger T. Production of Alternative Liquid Fuels in the Future Energy System. Chemie Ingenieur Technik. 2018;90:179-92.

[3] Varone A, Ferrari M. Power to liquid and power to gas: An option for the German Energiewende. Renewable and Sustainable Energy Reviews. 2015;45:207-18.

[4] Pontzen F, Liebner W, Gronemann V, Rothaemel M, Ahlers B. CO2-based methanol and DME – Efficient technologies for industrial scale production. Catalysis Today. 2011;171:242–50.

[5] Billig E, Decker M, Benzinger W, Ketelsen F, Pfeifer P, Peters R, et al. Non-fossil CO2 recycling—The technical potential for the present and future utilization for fuels in Germany. Journal of CO2 Utilization. 2019;30:130-41.

[6] VCI, Verband der Chemischen Industrie e.V. Chemiewirtschaft in Zahlen 2018. Frankfurt am Main2018.

[7] Baumann H, Tillman A-M. The hitch hiker's guide to LCA. 2004.

[8] Moghaddam EA, Ahlgren S, Hulteberg C, Nordberg Å. Energy balance and global warming potential of biogas-based fuels from a life cycle perspective. Fuel Processing Technology. 2015;132:74-82.

[9] Moghaddam EA, Ahlgren S, Nordberg Å. Assessment of novel routes of Biomethane Utilization in a life cycle Perspective. Frontiers in bioengineering and biotechnology. 2016;4:89.

[10] Lee U, Han J, Wang M, Ward J, Hicks E, Goodwin D, et al. Well-to-wheels emissions of Greenhouse gases and air pollutants of dimethyl ether from natural gas and renewable feedstocks in comparison with petroleum gasoline and diesel in the United States and Europe. SAE International Journal of Fuels and Lubricants. 2016;9:546-57.

[11] Deutz S, Bongartz D, Heuser B, Kätelhön A, Langenhorst LS, Omari A, et al. Cleaner production of cleaner fuels: wind-to-wheel–environmental assessment of CO 2-based oxymethylene ether as a drop-in fuel. Energy & Environmental Science. 2018.

[12] Starr K, Gabarrell X, Villalba G, Talens L, Lombardi L. Life cycle assessment of biogas upgrading technologies. Waste Management. 2012;32:991-9.

[13] Castellani B, Rinaldi S, Bonamente E, Nicolini A, Rossi F, Cotana F. Carbon and energy footprint of the hydrate-based biogas upgrading process integrated with CO2 valorization. Science of The Total Environment. 2018;615:404-11.

[14] Buratti C, Barbanera M, Fantozzi F. Assessment of GHG emissions of biomethane from energy cereal crops in Umbria, Italy. Applied Energy. 2013;108:128-36.

[15] Collet P, Flottes E, Favre A, Raynal L, Pierre H, Capela S, et al. Techno-economic and Life Cycle Assessment of methane production via biogas upgrading and power to gas technology. Applied Energy. 2017;192:282-95.

[16] Huttunen S, Manninen K, Leskinen P. Combining biogas LCA reviews with stakeholder interviews to analyse life cycle impacts at a practical level. Journal of Cleaner Production. 2014;80:5-16.
[17] Susmozas A, Iribarren D, Zapp P, Linβen J, Dufour J. Life-cycle performance of hydrogen production via indirect biomass gasification with CO2 capture. International Journal of Hydrogen Energy. 2016;41:19484-91.
[18] Eggemann L, Escobar N, Peters R, Burauel P, Stolten D. A Power-to-Fuel Strategy for Biogas Plants – Life cycle assessment of a Small-Scale Fuel Production System. 14th Conference on Sustainable Development of Energy, Water and Environment Systems. Dubrovnik, Croatia 2019.
[19] International Organization for Standardization. Environmental Management: Life Cycle Assessment; Principles and Framework: ISO; 2006.
[20] Decker M, Schorn F, Samsun RC, Peters R, Stolten D. Techno-economic analysis of a stand-alone power-to-liquid concept. UFZ EnergyDays – Energy landscapes of today and tomorrow Leipzig2018.
[21] Rau F. Personal Communication. 2019.
[22] Schiebahn S, Grube T, Robinius M, Tietze V, Kumar B, Stolten D. Power to gas: Technological overview, systems analysis and economic assessment for a case study in Germany. International journal of hydrogen energy. 2015;40:4285-94.
[23] Sun Q, Li H, Yan J, Liu L, Yu Z, Yu X. Selection of appropriate biogas upgrading technology-a review of biogas cleaning, upgrading and utilisation. Renewable and Sustainable Energy Reviews. 2015;51:521-32.
[24] IEK-3. Internal assumption for process at 100 bar 2019.
[25] Wernet G, Bauer C, Steubing B, Reinhard J, Moreno-Ruiz E, Weidema B. The ecoinvent database version 3 (part I): overview and methodology. The International Journal of Life Cycle Assessment. 2016;21:1218-30.
[26] EMEP/EEA. Biological treatment of waste - Anaerobic digestion at biogas facilities. EEA air pollutant emission inventory guidebook 2016 (EEA Report No 21/2016). Luxembourg2016.
[27] University of Minnesota. Fertilizer urea. Available from: https://extension.umn.edu/nitrogen/fertilizer-urea#advantages-755161. 14.05.2019.
[28] Mitchell CC. Nutrient Content of Fertilizer Materials. In: Universities AAMaA, editor.1999.
[29] Otto A. Chemische, verfahrenstechnische und ökonomische Bewertung von Kohlendioxid als Rohstoff in der chemischen Industrie: Universitätsbibliothek der RWTH Aachen; 2015.