ABSTRACT: Power-to-Gas enables the integration of renewable electricity and carbon into the chemical industry. The electricity is used to produce hydrogen, which is subsequently converted with CO₂ as the renewable carbon source. The resulting products can be used as feedstock for the chemical industry replacing current fossil-based feedstock. Because the integration of renewable electricity and carbon into the chemical industry is mainly environmentally motivated, we identify the conditions under which Power-to-Gas pathways are environmentally beneficial. The conditions are expressed as environmental threshold values for electricity supply. The threshold values are derived by a comparative life cycle assessment (LCA) of Power-to-Gas pathways to fossil-based processes. We analyze Power-to-Gas pathways to synthetic natural gas (Power-to-SNG) and to syngas (Power-to-Syngas). SNG is produced by the Sabatier reaction; syngas by reverse water gas shift (rWGS) and dry reforming of methane (DRM). The threshold values for electricity supply allow us to compare the environmental benefit of Power-to-SNG and Power-to-Syngas on an equal basis: how well they utilize the currently limited renewable electricity. Syngas production by the DRM process has the largest environmental potential. Both Power-to-Syngas pathways lead to larger environmental benefits than Power-to-SNG making syngas the more desirable product than methane as long as renewable electricity is limited.

KEYWORDS: LCA, CO₂ utilization, Reverse water gas shift, Dry reforming of methane, CO₂ methanation, Steam-methane-reforming, Power-to-Gas

INTRODUCTION

Renewable energies are the key to reconcile growing global energy demand with fossil resource depletion and climate change. Currently, the integration of renewable energy sources is most advanced in the power sector: Nearly 60% of the power generation capacity installed worldwide in 2014 was based on renewable energy sources.¹

Renewable electricity can also be integrated into further sectors such as the chemical industry through the Power-to-Gas concept. In the Power-to-Gas concept, renewable electricity is used in water electrolysis to produce hydrogen (Power-to-H₂).²,³ In a subsequent step, hydrogen reacts with CO₂ to yield carbon-containing gases such as methane.⁴⁻⁸ The carbon-containing gases can be used as renewable feedstock for the chemical industry replacing fossil feedstocks such as oil and natural gas. At the same time, Power-to-Gas provides energy storage due to the flexible operation of water electrolysis.⁹ Energy storage will be increasingly important because the majority of the newly installed renewable energy converters are wind and solar power, which are inherently intermittent and nondispatchable.¹⁰,¹¹ However, renewable electricity and, in particular, renewable electricity requiring storage will still be a limited resource in the foreseeable future. Thus, processes with highest environmental benefits per kWh renewable electricity must be identified.

Currently, the most advanced Power-to-Gas pathway based on hydrogen and CO₂ is the synthesis of a gas containing mainly methane, so-called synthetic natural gas (Power-to-SNG).⁴ Pilot SNG plants are already in operation.⁶ SNG can be fed directly into the existing natural gas grid and replace natural gas. Thus, all natural-gas-based processes could be switched directly to renewable feedstock without any downstream changes.

In the chemical industry, natural gas is used as chemical feedstock predominantly for steam-methane-reforming (SMR).¹² SMR yields a gas containing carbon monoxide (CO) and hydrogen, so-called syngas. The stoichiometric reaction has a molar H₂:CO ratio of 3:1. The syngas mixture can be directly processed to further chemicals such as methanol, or syngas can be separated into pure hydrogen and pure carbon monoxide. The pure products, hydrogen and CO, are used, e.g., to produce isocyanates which are important precursors for polyurethanes.¹³

Although syngas is currently produced from natural gas as outlined above, syngas can also be produced from renewable electricity (Power-to-Syngas). In this case, syngas is obtained from the reverse water gas shift (rWGS) reaction,¹⁴,¹⁵ or the
In the RWGS reaction, hydrogen and CO$_2$ react to CO. In the DRM reaction, natural gas and CO$_2$ react to CO and hydrogen. Both reactions require additional hydrogen to achieve the molar H$_2$:CO ratio of 3:1 of the established SMR. RWGS and DRM reactions have been intensively investigated in recent years.

The development of Power-to-SNG and Power-to-Syngas is motivated by their potential to reduce environmental impacts compared to the corresponding conventional processes. A suitable method to determine the environmental impacts is life cycle assessment (LCA). First LCA case studies have therefore been reported for Power-to-Gas pathways. Reiter and Lindorfer compared Power-to-SNG to conventional natural gas supply. van der Giesen et al. studied the RWGS process as intermediate step for the production of liquid hydrocarbon fuels (Power-to-Fuel) as alternative to diesel. In both case studies, Power-to-SNG reduced the nonrenewable energy demand compared to the fossil-based processes only if...
renewable electricity was used for electrolysis. Renewable electricity for electrolysis is also required in combination with CO₂ from biogenic sources or air capture to reduce global warming impacts compared to the conventional process as shown in both case studies. In our recent paper, we showed that Power-to-Syngas and Power-to-SNG could achieve lower global warming impacts than the fossil-based processes even if CO₂ was supplied by a coal-fired power plant. However, we studied the storage of surplus electricity which had no environmental impacts. Surplus electricity occurs when electricity generation exceeds demand to low full load hours. Thus, alternative electricity sources should be considered.

The first goal of this paper is to identify under which conditions the Power-to-SNG and both Power-to-Syngas (rWGS and DRM) pathways achieve lower environmental impacts than the corresponding conventional processes. Since previous case studies showed that electricity supply is the crucial factor for the environmental performance of Power-to-Gas pathways, we identify environmental threshold values for electricity supply. As Reiter and Lindorfer presented for Power-to-SNG, these threshold values for the environmental impact of electricity supply correspond to the break-even point of the environmental impacts of the Power-to-Gas pathways and the conventional processes. The second goal is to determine which Power-to-Gas pathway achieves the highest environmental benefit per kWh renewable electricity used.

We analyze the dependence of the environmental threshold values on technology choices within the supply chain, e.g., CO₂ supply and construction of hydrogen storage and electrolysis. In the main text, we focus on two environmental impact categories: global warming impact and fossil depletion impact. Twelve further environmental impact categories are presented in the Supporting Information.

The LCA method and the considered technologies for the assessment of the Power-to-Gas pathways are presented in the following section. In the subsequent section, the results on the environmental threshold values and options to achieve the environmental threshold values are presented. Finally, conclusions are drawn for the Power-to-Gas pathways.

### METHODOLOGY

Life cycle assessment (LCA) is a method to assess environmental impacts of products and processes. In the following subsection, the goal and scope of this case study is described.

**Goal and Scope Definition.** The goal of this paper is to identify under which conditions the considered Power-to-Gas pathways are environmentally beneficial compared to corresponding conventional processes. We express the conditions as environmental threshold values for electricity supply. Furthermore, we use the threshold values to compare the environmental benefit of the threec considered Power-to-Gas pathways. The considered Power-to-Gas pathways include two Power-to-Syngas processes (rWGS and DRM) and one Power-to-SNG process (Figure 1).

**System Boundaries and Functional Unit.** For the determination of the threshold values, the Power-to-Gas pathways are compared to the conventional production of syngas and SNG. Both considered Power-to-Syngas processes produce syngas with a molar H₂:CO ratio of 3:1. Conventionally, syngas is produced by steam-methane-reforming. Thus, we compare both Power-to-Syngas processes to steam-methane-reforming (SMR). We consider the separation of syngas into pure hydrogen and CO as required for subsequent chemical utilization, e.g., in polyurethane production.

Thus, the combined supply of the two pure products hydrogen and CO in a molar ratio of 3:1 is denoted syngas in this paper.

The Power-to-SNG process produces a gas containing mostly methane (>95 vol %) that is fed into the natural gas grid, so-called synthetic natural gas (SNG). The SNG is compared to conventional natural gas supply. Both products SNG and natural gas are considered equivalent based on the lower heating value.

In the comparison of Power-to-Gas pathways to conventional processes, the downstream processes are identical. Therefore, the use-phase and the end-of-life phase of syngas and SNG can be neglected for the present scope. We use the so-called cradle-to-gate approach considering only the upstream processes for syngas and SNG.

The Power-to-Gas pathways consist of the chemical conversion process, an electrolysis unit and a power plant (Figure 1). The power plant supplies the CO₂ for the chemical conversion process. Power plants are frequently discussed as CO₂ sources because they can supply large amounts of CO₂. All considered processes are explained in detail in the next section.

The considered Power-to-Gas pathways produce not only the desired products SNG and syngas but also electricity, oxygen, steam and purge gas. A sound comparison requires that all compared processes produce the same products. Therefore, an identical set of products has to be defined for all compared processes. This set of products is the functional unit in the LCA.

For the syngas processes, we choose the following functional unit:

i. **FU** _syngas_: Supply of 1 kg syngas and 1.11 kWh electricity.

Both products, syngas and electricity, are chosen because syngas is the main product of the chemical conversion process, and electricity the main product of the power plant. The amount of electricity results from the amount of CO₂ supplied by the power plant with CO₂ capture. The extension of the functional unit is called system expansion in LCA. System expansion is the preferred approach for allocation according ISO 14040. In our case, system expansion avoids the potential misassignment of all environmental benefits to the chemical conversion process only.

For the SNG process, we choose as the corresponding functional unit:

ii. **FU** _SNG_: Supply of 1 MJ SNG and 0.049 kWh electricity.

Here, SNG is considered equivalent to natural gas based on the lower heating value.

The other products (oxygen, steam and purge gas) are defined as byproducts according to the approach presented by Jung et al. The handling of the byproducts is explained in the next section.

The conventional production processes consist of the chemical conversion process and a power plant (Figure 1). Both conventional production processes for SNG and Syngas are extended by a coal-fired power plant without CO₂ capture to produce the electricity required for the selected functional unit.

**Environmental Impact Categories.** In the main text, the processes are compared based on their global warming (GW) and fossil depletion (FD) impacts. These impact categories are chosen because they reflect the major motivation for the installation of renewable energies. Further environmental impact categories are shown in the Supporting Information. The environmental impacts are determined according to ReCiPe 1.08 Midpoint (Hierarchist). The methodology ReCiPe 1.08 (Hierarchist) determines the global warming impact in CO₂-equivalents based on global warming potentials from the IPCC Fourth Assessment Report for a time frame of 100 years. The impact category fossil depletion aggregates the utilization of fossil resources such as oil and natural gas based on their energy content in kg oil-equivalents.

**Environmental Threshold Values for Electricity Supply.** In this paper, environmental threshold values for electricity supply are used to identify under which conditions Power-to-Gas pathways are environmentally beneficial and (ii) to compare the environmental benefit of the different Power-to-Gas pathways. Such a comparison is in particular important if limited renewable electricity is used.

**Definition.** The environmental threshold value for electricity supply (EVT <electricity supply>) is the specific environmental impact of electricity...
supply per kWh for which the environmental impacts of the Power-to-Gas pathways \((E_{\text{P2G}})\) and the corresponding conventional processes \((E_{\text{conventional}})\) are equal:

\[
E_{\text{P2G}}(E_{\text{electricity supply}}^{\text{TV}}) = E_{\text{conventional}}(E_{\text{electricity supply}}^{\text{TV}})
\]

(1)

**Application.** We use the environmental threshold value for electricity supply to

i. Identify electricity supply processes with environmental impacts \(E_{\text{electricity supply}}\) that reduce environmental impacts for the Power-to-Gas pathways compared to the conventional processes:

\[
E_{\text{electricity supply}}^{\text{TV}} \geq E_{\text{electricity supply}}
\]

(2)

The environmental threshold value for electricity supply can also be used to identify an environmentally beneficial combination of electricity supply processes. For example, the determination of the minimum required share \((X)\) of wind electricity \((E_{\text{wind electricity}})\) in combination with grid electricity \((E_{\text{grid electricity}})\):

\[
E_{\text{electricity supply}}^{\text{TV}} \geq E_{\text{wind electricity}} X + E_{\text{grid electricity}} (1-X)
\]

(3)

ii. Compute the actual environmental benefit of Power-to-Gas pathways for a given electricity supply process. Because the threshold value provides the maximum environmental impact for electricity supply allowable to still be beneficial, using electricity supply with less impacts will lead to an environmental benefit \((E_{\text{benefit}})\). The environmental benefit per kWh electricity is thus the difference of the threshold value and the environmental impact of the chosen electricity supply:

\[
E_{\text{benefit}} = E_{\text{electricity supply}}^{\text{TV}} - E_{\text{electricity supply}}
\]

(4)

**Considered Technologies for Syngas and SNG Production.** In this section, the considered technologies are briefly presented. The inputs and outputs of the chemical conversion processes are obtained from published data of process simulations (see Table S1 in the Supporting Information for all details).

**Reverse Water Gas Shift (rWGS).** Syngas can be produced from CO2 and hydrogen by the reverse water gas shift reaction \((\text{rWGS})\):

\[
\text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3\text{H}_2
\]

(7)

The rWGS reaction is favored at high temperatures and low pressures. To avoid the production of coke, temperatures over 1000 °C are required. At ambient pressure and 1000 °C, the equilibrium conversion of CO2 is about 95%. After the reaction, the raw gas contains CO, CO2, water, hydrogen and small amounts of methane. Both products hydrogen and CO can be separated by a similar raw gas treatment as for the rWGS reaction. The only difference is an additional pressure swing adsorption (PSA) in the last step to further purify the hydrogen-rich stream leaving the liquid methane scrubbing. For the DRM process, only CO2 is recycled to the reactor.

**Conventional Syngas Production.** The most used conventional production process for syngas is steam-methane-reforming (SMR):

\[
\text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3\text{H}_2
\]

(8)

The SMR reaction is typically carried out at 900 °C. After the reaction, the raw gas contains CO, CO2, water, hydrogen and methane. The raw gas treatment is the same as for the DRM process. However, only part of the separated CO2 is recycled to the reactor to achieve the desired molar H2:CO ratio of 3:1. The majority of separated CO2 is emitted into the atmosphere.

**Power-to-SNG.** Hydrogen produced by water electrolysis can react with CO2 to methane according to the Sabatier reaction:

\[
\text{4H}_2 + \text{CO}_2 \rightarrow \text{CH}_4 + 2\text{H}_2\text{O}
\]

(9)

The Sabatier reaction is favored at low temperatures. On the basis of experimental data, Müller et al. report that about 95% of CO2 is converted to methane at 250 °C. The product gas also contains water and hydrogen because not all CO2 is converted. After separation of water, the gas can be directly fed into the natural gas grid.

All considered chemical conversion processes are operated at steady-state conditions in 8000 h per year.

**Conventional Natural Gas Production.** For the conventional production of natural gas, we consider the German natural gas mix. The natural gas supplied to Germany originates from Russia (34%), Norway (28%), The Netherlands (24%) and Germany (14%). The environmental impacts are directly taken from the GaBi database. For the US, methane emissions have very high uncertainties. For the US, methane emission are reported for conventional natural gas supply between 5 and 39 g CH4 per kg natural gas. We therefore vary methane emissions in our sensitivity analysis presented in the Supporting Information. We chose 17 g CH4 per kg natural gas as maximum methane emissions because this value corresponds to the highest methane emissions for a European country (Slovakia) according to the GaBi database. This value is also in accordance to measured methane emissions for the Russian natural gas system in 2005. For PRODUCTION. Hydrogen is produced in a proton exchange membrane (PEM) electrolysis unit with an electricity demand of 50 kWh per kg H2. The main advantage of PEM electrolysis is that it is a very dynamic operation, which enables following intermittent electricity supply, e.g., wind electricity. In this paper, we consider both (I) steady-state and (II) part-load operation of electrolysis. Steady-state operation requires continuous electricity supply, e.g., through the utilization of grid electricity. Part-load operation is required if intermittent renewable electricity is directly fed into the electrolysis unit. For example, full load hours for solar and wind electricity in Germany are between 900–1100 and 1400–5000 h per year, respectively. For part-load operation, we consider 2500 full load hours in the main text. The influence of full load hours on results is analyzed in the Supporting Information. Furthermore, part-load operation of electrolysis requires hydrogen storage. In this case, we assume that hydrogen is stored in pressure tanks at 25 bar. In the main text, the hydrogen storage is sized to cover 10 days without intermittent renewable electricity. The influence of the size of the hydrogen storage is investigated in the Supporting Information.
Besides hydrogen, electrolysis produces also pure oxygen (8 kg O₂/ kg H₂).

CO₂ Supply (Power Plant). The CO₂ required for the Power-to-Gas pathways is supplied by a coal-fired power plant with CO₂ capture. The efficiency of the considered power plant is 27.25% (based on the higher heating value) and 90% of the CO₂ emissions are captured (DoE case 10).38 We also consider the production of monoethanolamine (MEA) as CO₂ capture solvent. Most of the MEA is recycled but about 0.0019 kg MEA/kg CO₂ captured is lost.39

The electricity in the functional units FUsyn gas and FUSNG is exactly the electricity produced in a power plant with CO₂ capture to provide the required amount of CO₂ for the rWGS process and SNG process, respectively.

For the conventional processes without CO₂ demand, electricity is produced in a coal-fired power plant without CO₂ capture. The efficiency of the power plant without CO₂ capture based on the higher heating value is 38.27% (DoE case 9).38 The DRM process requires only about half of the CO₂ of the rWGS process. To supply still the same electricity in the functional unit, the DRM process uses both the power plant with CO₂ capture and the power plant without CO₂ capture.

Utilities. The heat required for the chemical conversion process is supplied by natural gas boilers with an efficiency of 95%. For the electricity supply, we present most results as a function of the environmental impacts. However, for some results we require environmental impacts of grid electricity. In this case, we apply forecasted German grid electricity mixes (Scenario 2011 A).40 The forecasted grid electricity mixes are based on the climate targets of the German Federal Government. Import of electricity is not considered. The considered LCA data sets for the utilities are summarized in Table S3 in the Supporting Information.

Construction. For the Power-to-Gas pathways, we consider the environmental impacts for the construction of the units for electrolysis and hydrogen storage. The construction of chemical plants and power plants is neglected due to a lack of data. For fossil-based steady-state processes, the impact of construction is usually low compared to the impacts during operation.37 Furthermore, both Power-to-Gas and conventional processes employ similar chemical plants and power plants. Thus, for a comparative assessment, these impacts are of minor importance.

For the construction of the electrolysis unit, we use LCA data based on a PEM fuel cell. For the construction of the hydrogen storage pressure tanks, we only consider the steel demand following Mori et al.42 Details for the construction of electrolysis and hydrogen storage are summarized in Table S2 in the Supporting Information.

Byproducts. In the main text, we do not consider an environmental credit for byproducts, because utilization cannot always be achieved. The influence of environmental credits is investigated in the Supporting Information. The environmental credit for byproducts is then based on the avoided burden method (also called substitution method).43

■ RESULTS AND DISCUSSION

In this section, the threshold values for electricity supply are presented for global warming and fossil depletion. In the first subsection, we compare the impacts for global warming and fossil depletion of Power-to-Gas pathways to the corresponding conventional processes for steady-state operation of electrolysis (100% grid electricity). On the basis of the comparison, the environmental threshold values are derived for electricity supply. In the second subsection, we consider part-load operation of electrolysis (mix of renewable and grid electricity). In this subsection, we use the threshold values to determine minimum required shares of renewable electricity to achieve environmentally beneficial Power-to-Gas pathways and to compare the environmental benefit of the Power-to-Gas pathways (cf. eq 3).

Steady-State Operation of Electrolysis. Power-to-Syngas vs Conventional Syngas Production. Figure 2 shows the impacts for global warming and fossil depletion of Power-to-Syngas (rWGS and DRM) and conventional syngas production for the production of 1 kg syngas and 1.11 kWh electricity (FU syn gas) in Germany. Here, the forecasted German grid electricity mix for 2020 is applied for electricity supply.

The global warming impact of conventional syngas production is 2.5 kg CO₂-eq/FU syn gas. The functional unit FU syn gas includes electricity and syngas. About 45% of the global warming impact is due to the electricity production in the coal-fired power plant power and 55% due to the syngas production. The global warming impact of the syngas production stems from electricity supply (14%), heat supply from natural gas...
(18%) and purge gas (11%), CO₂ purge (2%) and supply of natural gas as feedstock (10%).

For the rWGS process, the global warming impact is about 5.8 kg CO₂-eq/FUₘₚₚ in 2020. About 85% of the global warming impact is due to the electricity supply for electrolysis. Hence, the rWGS process will strongly benefit from a future reduction of global warming impacts for electricity supply. The remaining 15% of the global warming impact is mainly due to the electricity supply for the chemical conversion process, emissions of the coal-fired power plant and the heat supply. The CO₂ emissions in the power plant are lower than in conventional syngas production because CO₂ is captured and used in the chemical conversion process. The global warming impact of the construction of the electrolysis unit is negligible if the process is operated continuously (<1%).

For the DRM process, the global warming impact is about 4.2 kg CO₂-eq/FUₘₚₚ in 2020. The global warming impact is lower than for the rWGS process, because the DRM process requires less hydrogen. Thus, less electricity is required for the electrolysis. About 65% of the global warming impact of the DRM process is due to electricity supply for electrolysis. About 35% of the global warming impact of the DRM process is due to electricity supply for the chemical conversion process, emissions of the power plant and the natural gas supply. The global warming impact of the power plant is higher compared to the rWGS process, because only half of the CO₂ is captured from the power plant compared to the rWGS process (see above).

The fossil depletion impact of the conventional syngas production is about 1.2 kg Oil-eq/FUₘₚₚ. About 20% of the fossil resources are required for electricity production and 80% for syngas production. For the rWGS and DRM processes, the fossil depletion impacts are 1.9 and 1.5 kg Oil-eq/FUₘₚₚ, respectively.

Thus, both Power-to-Syngas processes are expected to lead to higher GHG emissions and higher fossil resource depletion than conventional syngas production in 2020. This is mainly due to the impact of electricity supply. Since the environmental impacts from electricity supply should be reduced in the future, we study the impact from electricity supply on Power-to-Syngas in detail and derive environmental threshold values for electricity supply.

Figure 3 shows the global warming impact of conventional syngas production and both Power-to-Syngas processes as a function of the global warming impact of electricity supply. For both Power-to-Syngas processes, the global warming impact of electricity supply has a strong influence on the total global warming impact, because both processes require large amounts of electricity for electrolysis. For the DRM process, the influence of the global warming impact of electricity supply is slightly lower than for the rWGS process, because the DRM process requires less hydrogen. For electricity supply with reduced GHG emissions, Power-to-Syngas becomes beneficial over conventional production. This would be achieved after the year 2040 based on the forecast for the German electricity mix.

For conventional syngas production, the total global warming impact depends much less on the global warming impact of electricity supply, because the conventional syngas process requires electricity only for the compression and separation of gases.

The intersections of the rWGS and DRM process with the conventional syngas production are the threshold values for global warming impact of the electricity supply. The global warming threshold values for electricity supply are 0.14 and 0.19 kg CO₂-eq/kWh for rWGS and DRM process, respectively.

The fossil depletion threshold values can be determined in the same way as for the global warming threshold values. The fossil depletion threshold values for electricity supply are 0.056 and 0.073 kg Oil-eq/kWh for rWGS and DRM process, respectively.

In the Supporting Information, we investigate the dependence of the threshold values for global warming (Figure S1) and fossil depletion (Figure S2) on utilization of byproducts, CO₂ supply and efficiency of electrolysis. Both Power-to-Syngas processes require much cleaner electricity, i.e., have much lower global warming threshold values for electricity supply, if CO₂ supply does not avoid CO₂ emissions. CO₂ emissions are not avoided if, for example, CO₂ is used which otherwise would be stored underground. CO₂ emissions are also not avoided if the installation of a power plant with CO₂ capture delays installation of renewable electricity.

Our analysis of twelve further environmental impact categories shows that positive threshold values are only achieved for photochemical oxidant formation (rWGS and DRM). Only in impact categories with a positive threshold value, the Power-to-Syngas processes have the potential to reduce environmental impacts compared to the corresponding conventional processes. For Power-to-Syngas processes, the environmental hot spots of most impact categories are construction of electrolysis unit and coal supply. However, uncertainty is also higher in most other impact categories compared to global warming and fossil depletion impacts.

Power-to-SNG vs Conventional Natural Gas Production. Figure 4 shows the global warming and the fossil depletion impact of Power-to-SNG and conventional natural gas production for the production of 1 MJ SNG and 0.049 kWh electricity (FUₜₚₚ) in the year 2020.

The global warming impact of conventional natural gas production is 0.059 kg CO₂-eq/FUₜₚₚ. 85% of the emissions...
The fossil depletion impact of the conventional natural gas production is 0.037 kg Oil-eq/FUSNG. 30% of the fossil depletion impacts are due to the coal-fired power plant and 70% due to the natural gas supply. For Power-to-SNG, the fossil depletion impact is 0.072 kg Oil-eq/FUSNG. The fossil depletion threshold value for electricity supply is 0.040 kg Oil-eq/kWh.

For Power-to-SNG, we also present the sensitivity of threshold values depending on utilization of byproducts, CO₂ supply and efficiency of electrolysis in the Supporting Information. The results are similar to Power-to-Syngas. Power-to-SNG benefits most from an environmental credit for byproducts, because a large amount of heat is produced during the exothermic reaction. In contrast to Power-to-Syngas, the global warming threshold value becomes even negative if CO₂ is used which otherwise would be stored. In this case, Power-to-SNG would thus have higher global warming impacts than conventional natural gas supply even if global warming impacts for electricity supply were zero.

Part-Load Operation of Electrolysis. In this section, we investigate the influence of part-load operation of electrolysis. Part-load operation of electrolysis enables the direct utilization of intermittent renewable electricity. Through the direct utilization of renewable electricity such as wind or solar, the global warming and fossil depletion threshold values for electricity supply can be satisfied already today. However, if Power-to-Gas pathways use exclusively renewable electricity, the electrolysis unit has only low full load hours and huge hydrogen storage capacities are required. Through the combination of intermittent renewable and base-load grid electricity, the full load hours of electrolysis can be increased and the size of the hydrogen storage can be decreased. In the following, intermittent renewable electricity is only denoted renewable electricity and base-load grid electricity only grid electricity.

In Figure 5, the threshold values for global warming and fossil depletion are shown for renewable electricity as a function of the share of renewable electricity used for electrolysis. The threshold value for renewable electricity can be determined from eq 3 for a given share of renewable electricity and environmental impact of grid electricity (see eq S2 in the Supporting Information). For the environmental impact of grid electricity, two scenarios based on forecasted German grid electricity mixes are considered: 2020 and 2030.

Global Warming Impacts. For all Power-to-Gas pathways, the global warming threshold value for renewable electricity has its maximum if the electrolysis uses 100% renewable electricity. In 2020, the DRM process has a maximum global warming threshold value for renewable electricity of 0.13 kg CO₂-eq/kWh, followed by the rWGS and SNG processes with 0.10 and 0.048 kg CO₂-eq/kWh, respectively. The maximum threshold value differs from the value determined in the previous section due to lower full load hours for electrolysis unit, the additionally required hydrogen storage and the utilization of grid electricity for the chemical conversion process. For the utilization of 100% renewable electricity, utilizing wind and solar electricity can satisfy the threshold values for all Power-to-Gas pathways. Figure 5 also shows the minimum shares of renewable electricity that are required to be environmentally beneficial depending on the renewable electricity technology used for electrolysis. If wind electricity is used for electrolysis in 2020, the minimum required shares of wind electricity for DRM, rWGS, and SNG processes are 67%, 76%, and 90%, respectively.

The highest global warming benefit per kWh renewable electricity is achieved by the DRM process for all shares of renewable electricity and for all considered years. For example, if 100% wind electricity is used for electrolysis in 2020, the

![Figure 4](https://example.com/figure4.png) Global warming impact (top) and fossil depletion impact (bottom) of Power-to-SNG and conventional natural gas production. The functional unit (FUSNG) is the production of 1 MJ natural gas and 0.049 kWh electricity. The bars of the single processes show the breakdown of the impacts. The environmental impacts for grid electricity are based on forecasted German electricity mixes in 2020.
In 2030, the maximum global warming threshold values are almost the same as for 2020. However, the minimum required share of renewable electricity to be environmentally beneficial decreases compared to 2020.

**Fossil Depletion Impacts.** The results for the fossil depletion impact correspond qualitatively well to the global warming impact. However, quantitatively, much lower shares of renewable electricity are required compared to global warming impacts to be environmentally beneficial with a lowest value for 2020 of 34% for the DRM process (use of wind electricity). The maximum fossil depletion threshold values for DRM, rWGS, and SNG in 2020 are 0.069, 0.046, and 0.029 kg Oil-eq/kWh, respectively.

In the Supporting Information, a sensitivity analysis is presented for utilization of byproducts, CO2 supply, efficiency of electrolysis, and construction of electrolysis unit and hydrogen storage. Regarding the construction of electrolysis and hydrogen storage, the size of the hydrogen storage has the highest influence on global warming (Figure S3) and fossil depletion threshold values (Figure S4). However, the results for construction should only be considered as indicative because currently only very few LCA data sets are available for the construction of hydrogen storage and electrolysis unit.

## CONCLUSIONS

In this paper, we analyze the following three Power-to-Gas pathways: Power-to-Syngas (rWGS and DRM) and Power-to-SNG. Syngas and SNG are synthesized from conversion of CO2 and hydrogen. As the CO2 source, we consider a coal-fired power plant with CO2 capture. Hydrogen is supplied by water electrolysis. For operation of the electrolyzer, we consider both steady-state and part-load operation. The Power-to-Gas pathways are compared based on environmental threshold values for the electricity supply. These threshold values indicate the maximum environmental impact of electricity supply for which the environmental impact of the Power-to-Gas pathway is lower than for the corresponding conventional process.

For steady-state operation of electrolysis, the global warming threshold values for electricity supply are 0.19, 0.14, and 0.082 kg CO2-eq/kWh for DRM, rWGS, and SNG processes, respectively. For Germany, both Power-to-Syngas processes achieve lower global warming impacts than conventional syngas production if the forecasted electricity mix for 2050 is used (0.13 kg CO2-eq/kWh). For the Power-to-SNG process, not even the electricity mix of 2050 is sufficient to reduce global warming impacts compared to the conventional natural gas supply.

Through part-load operation of electrolysis, intermittent renewable electricity can be directly used in electrolysis. If the electrolysis unit uses only renewable electricity, the global warming threshold values for electricity supply are 0.13, 0.10, and 0.048 kg CO2-eq/kWh for DRM, rWGS, and SNG processes, respectively. Thus, all Power-to-Gas pathways have lower global warming impacts compared to corresponding conventional processes if 100% wind (0.012 kg CO2-eq/kWh) or solar (0.046 kg CO2-eq/kWh) electricity is used for electrolysis. Since the exclusive utilization of renewable electricity leads to low full load hours for electrolysis and requires huge storage capacities, we also investigate a hybrid strategy combining grid electricity and renewable electricity. If a mix of wind electricity and German grid mix in 2020 is used for electrolysis, DRM, rWGS and SNG processes require at least 67%, 76%, and 90% of wind electricity to reduce global warming impacts compared to the corresponding conventional process. If all Power-to-Gas pathways use the same amount of wind electricity, the DRM process achieves the highest environmental benefit.

Besides electricity supply, the CO2 supply is also crucial for the environmental performance of Power-to-Gas pathways. Power-to-Gas pathways can achieve lower global warming impacts than conventional processes if CO2 from a coal-fired power plant is captured, which would emit the CO2 otherwise. However, the global warming benefit of the Power-to-Gas pathways decreases if the utilized CO2 does not avoid CO2.
emissions (e.g., if CO$_2$ storage is avoided). In this case, the SNG process has even higher global warming impacts than conventional natural gas supply.

Overall, Power-to-Gas is a promising technology to integrate renewable electricity into the chemical industry and decrease dependence on fossil feedstocks. SNG has the advantage of the potential for integrating renewable energy into the chemical value chain.

**ASSOCIATED CONTENT**

[Supporting Information](#)

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acssuschemeng.6b00644.

LCA data, sensitivity analysis for global warming and fossil depletion threshold values, threshold values for further environmental impact categories, global warming of SNG production as a function of global warming impact of electricity, environmental threshold values for renewable electricity supply (PDF).

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**Notes**

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