Is hydrogen production through natural gas pyrolysis compatible with ambitious climate targets in the United States? A location-specific, time-resolved analysis

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Abstract
Pyrolysis of natural gas to produce \( \text{H}_2 \) and solid carbon through methane cracking can be characterized as a high-\( \text{CH}_4 \), low-\( \text{CO}_2 \) process. It results in low \( \text{CO}_2 \) emissions because no direct \( \text{CO}_2 \) is generated at the point of \( \text{H}_2 \) generation if solid carbon is not combusted further. However, it results in high \( \text{CH}_4 \) emissions because of its higher natural gas consumption compared to the direct use of natural gas and, thus, higher \( \text{CH}_4 \) losses along the natural gas supply chain. Here, I analyzed whether this process can provide climate benefit in comparison to the direct, unabated utilization of natural gas and also in comparison with \( \text{H}_2 \) produced from water electrolysis with grid electricity. To this end, Monte Carlo simulations of time-resolved and US state-specific emission profiles and their impact on mid-century global warming under different \( \text{CH}_4 \) mitigation scenarios were conducted. It was found that the climate benefit of natural gas pyrolysis is highly dependent on plant location and the speed at which \( \text{CH}_4 \) emissions can be abated. New York, Pennsylvania, and Ohio emerged as the most promising locations. This is because of their projected long reliance on natural gas for power generation, which renders electrolysis using grid electricity less attractive, as well as the relatively low estimate of current \( \text{CH}_4 \) emissions from the natural gas supply chain. However, without fast action on \( \text{CH}_4 \) emission mitigation, the climate benefit of natural gas pyrolysis is small or non-existent, irrespective of the plant location. Overall, the uncertainty in the relative climate benefit of natural gas pyrolysis was found to be large; however, this study developed an easy-to-adapt MS Excel/visual basic for applications (VBA) tool that can be updated as soon as more accurate data on \( \text{CH}_4 \) emissions becomes available. Policymakers, businesspeople, and scholars can use this tool to estimate the climate impact within their own scenarios and locations.

1. Introduction
Pyrolysis of natural gas, which consists predominantly of \( \text{CH}_4 \), to produce \( \text{H}_2 \) and solid carbon is being actively researched as a means of reducing greenhouse gas emissions from fossil fuel combustion. The \( \text{H}_2 \) gas produced in this manner is often referred to as 'turquoise hydrogen' and, together with 'blue hydrogen' produced from natural gas reforming with carbon capture and storage, it is frequently considered as a bridge technology toward a fully renewable energy system (Sánchez-Bastardo et al 2021).

\[
\text{CH}_4 \rightarrow \text{C} + 2\text{H}_2 \Delta H_R = +75 \text{ kJ.} 
\] (1)

Several processes have been used or suggested for the practical implementation of natural gas pyrolysis, such as catalytic cracking on metal or carbon substrates, thermal cracking, plasma conversion, and molten metal pyrolysis (Abbas and Wan Daud 2010, Amin et al 2011, Upham et al 2017). In natural gas pyrolysis, one mole of \( \text{CH}_4 \) with 891 kJ of higher heating value (HHV) yields a maximum of 2 moles of \( \text{H}_2 \) with an HHV of 572 kJ. The energy incorporated in the solid carbon (394 kJ) must remain unused to prevent \( \text{CO}_2 \) release from its oxidation. Therefore, on an HHV basis, natural gas consumption during pyrolysis increases by at least a factor of 1.56 compared to the direct use of natural gas.
The benefits of low-CO₂ natural gas technologies, such as pyrolysis, have recently come under scrutiny (Timmerberg et al 2020, Howarth and Jacobson 2021, Bauer et al 2022). To assess the potential of H₂ production from pyrolysis for the mitigation of global warming, it is useful to compare it to the climate benefit of H₂ produced from electric power through electrolysis (i.e. the splitting of water into H₂ and O₂). Furthermore, it should also be compared to the climate impact of direct utilization of natural gas on an HHV basis, as many potential applications of H₂ are those where H₂ is thought to substitute natural gas, such as space heating, power generation in gas turbines or solid oxide fuel cells, as process heat in industry, or in direct reduction of iron ore for steel production.

Natural gas pyrolysis can be considered emission-free at the point of hydrogen production. However, greenhouse gases (GHG) are emitted during production, processing, and transmission of natural gas. CH₄ emissions are of particular importance due to a much higher global warming potential (GWP) of CH₄ compared to CO₂. Because H₂ from pyrolysis consumes at least 1.56 times of natural gas per unit of energy delivered, it can be characterized as a high-CH₄/low-CO₂ technology. Because CH₄ is emitted via leaks or venting and emissions are not linearly caused by the consumption or production of natural gas, it is difficult to calculate an accurate estimate (Burns and Grubert 2021a). CH₄ emission factors are also basin-specific and vary considerably throughout the US (Omara et al 2018). Nevertheless, Grubert et al recently compiled consumption-based, state-specific first-order estimates of production-stage CH₄ emissions (Burns and Grubert 2021a). To assess the impact of CH₄ emissions from natural gas use on global warming, it is also important to consider CH₄ mitigation efforts and their timelines (Ocko et al 2021).

In contrast to H₂ from natural gas pyrolysis, emissions from H₂ production via water electrolysis are dominated by CO₂ emissions from fossil-fuel combustion for electric power production. Achieving ambitious climate goals will require rapid decarbonization of electric power production, and thus a phase-out of fossil fuels such as coal and natural gas. Specific emissions from power production, and thus, by extension, from water electrolysis, are projected to change drastically in the coming decades. It should be noted that electrolysis could also be powered by dedicated renewable energy production, however, electrolyzers are expensive and benefit greatly from continuous operation, which incentivizes their connection to an existing grid in many cases.

A previous work assessed the climate benefit of natural gas pyrolysis based on a fixed CH₄ emission factor in comparison to water electrolysis coupled with a specific power generation plant (either renewable or natural-gas fired) (Timmerberg et al 2020). However, this approach does not address whether natural gas pyrolysis yields a climate benefit compared to electrolysis against the backdrop of decreasing power generation CO₂ emission timelines that are compatible with ambitious climate targets.

In this paper, for the first time, I provide an analysis of the climate benefit of natural gas pyrolysis based on time-resolved emission profiles that result from net-zero-by-2050 grid power generation decarbonization trajectories and CH₄ mitigation scenarios. I further provide a state-specific analysis to account for the significant spatial differences in power decarbonization trajectories and CH₄ emissions. Furthermore, the uncertainty in climate benefits through Monte Carlo simulation is assessed.

Thus, I ask the question: Can natural gas pyrolysis provide climate benefits toward the Paris Agreement’s mid-century temperature goal in comparison with water electrolysis from grid electricity as well as in comparison with continued and unabated natural gas use?

2. Method

2.1. Technological pathways and scope of emissions included in calculation

Three technological pathways were considered in this study. In the first, business-as-usual pathway, the direct utilization of natural gas continues until 2050 (‘direct natural gas use’). In the second pathway, the direct utilization of natural gas is replaced by natural gas pyrolysis starting, at the earliest, from 2030 (‘natural gas pyrolysis’). 2030 was considered as the earliest possible deployment year because CH₄ pyrolysis with a focus on hydrogen production is currently at a low technological readiness level; thus, further R&D and upscaling is required until commercial deployment is possible (Schneider et al 2020). In the third pathway, the direct utilization of natural gas is replaced with hydrogen produced from water electrolysis as soon as the specific CO₂-equivalent emissions of hydrogen produced through electrolysis from grid electricity are lower than those from direct natural gas use (‘water electrolysis’). For water electrolysis, 2025 was considered the earliest possible deployment year as considerable upscaling of electrolyzer production is currently underway. These three pathways are shown in figure 1(a).

The scope of the emissions included in this analysis is shown in figure 1(b). Excluded are comparatively small emissions, such as coal supply chain CO₂ emissions, N₂O emissions from fuel combustion, and methane slip from natural gas combustion or utilization. Also excluded are the lifecycle emissions of any non-coal, non-natural gas power generation, such as wind, solar power, and nuclear power, as these...
are also comparatively small and expected to decline further with the progression of industrial decarbonization. Further, the effect of hydrogen emissions on global warming was excluded because the magnitude of emissions from hydrogen production, storage and transport, as well as the GWP of hydrogen are not yet fully understood. Recent research indicates that the GWP of hydrogen might be higher than previously thought, and hydrogen emissions could be a useful addition to future iterations of the model developed herein (Ocko and Hamburg 2022).

2.2. Metric for CO₂ equivalency of CH₄ emissions

To facilitate the comparison of technological pathways that emit highly varying amounts of CO₂ and a short-lived climate pollutant (SLCP) such as CH₄, a metric that reflects their relative impact on anthropogenic climate change needs to be chosen.

An overview of climate metrics that establish the CO₂ equivalency of SLCPs has recently been provided by Balcombe et al (2018). The most commonly used metric is the GWP, which assesses the cumulative radiative forcing of an emission pulse over a specified time horizon, such as a 100 year period (GWP₁₀₀) or a 20 year period (GWP₂₀). Instead, here I selected a temperature-based metric to assess the impact of emissions on mean surface temperature analogously to the global temperature potential (GTP) metric, albeit with a fixed endpoint instead of a rolling 20-, 50-, or 100 year time horizon. While a temperature-based equivalence introduces additional uncertainty, it allows the comparison of different technology pathways based on global mean surface temperature goals, as stipulated in the Paris Agreement. As an endpoint, the year 2050 was selected because the ambitious 1.5 °C/representative concentration pathway (RCP) 1.9 scenarios commonly exhibit stabilization or a mild overshoot/peak of global mean temperatures around the mid-century (Riahi et al 2017, Rogelj et al 2018, 2019). It is noted that some researchers argue that, due to current emission trends, a high mid-century overshoot over the 1.5 °C target should be expected, and mitigation of methane emissions should thus be focused later in the century for reasons of cost-effectiveness (Tanaka et al 2021). Nevertheless, 2050 was selected as an endpoint, in order to explicitly investigate the compatibility of methane pyrolysis with ambitious, net-zero by 2050 mitigation scenarios that avoid a high mid-century temperature overshoot. A fixed endpoint metric, such as the one used in this study may thus not account for climate impacts beyond that fixed endpoint, i.e. 2050, but can be considered a good compromise between simplicity and comprehensiveness.

The absolute global temperature change potential AGTP_CO₂,2050 of CO₂ emitted in year t toward 2050 was obtained through the procedure described in IPCC AR5

\[
AGTP_{CO_2,2050}(t) = A_{CO_2} \sum_{j=1}^{2} \left\{ a_0 c_j \left( 1 - \exp \left( -\frac{2050 - t}{d_j} \right) \right) \right. \\
+ \sum_{k=1}^{3} a_k \tau_j c_j \left( \exp \left( -\frac{2050 - t}{\tau_k} \right) - \exp \left( -\frac{2050 - t}{d_j} \right) \right) \} [K kg^{-1}] .
\]

The absolute global temperature change potential AGTP_CH₄,2050 of CH₄ emitted in year t toward 2050 was also obtained using the Intergovernmental Panel on Climate Change (IPCC) AR5 procedure, including the effects of ozone and stratospheric H₂O

\[
AGTP_{CH_4,2050}(t) = 1.65 A_{CH_4} \sum_{j=1}^{2} \frac{\tau_j c_j}{\tau - d_j} \\
\times \left( \exp \left( -\frac{2050 - t}{\tau} \right) - \exp \left( -\frac{2050 - t}{d_j} \right) \right) [K kg^{-1}] .
\]

Both AGTP functions are shown in figure 2(a). Note that the AGTP with a fixed endpoint in 2050 shows the contribution of 1 kg of gas emitted in the year t towards global temperature rise in 2050. The AGTP curves thus approach zero towards 2050, because one kg of gas emitted in 2050 will have no effect on the temperature in the year 2050 (because it
is emitted too late). The resulting GTP_{2050} of methane as a function of the emission year is shown in figure 2(b) with comparison to static metrics.

For each technological pathway i, the specific CO_{2}-equivalent emissions $\dot{e}_{\text{CO}_2}^{\text{eq}}$ in year t were obtained

$$
\dot{e}_{\text{CO}_2}^{\text{eq}}(t) = \dot{e}_{\text{CO}_2}^{\text{eq}} - GTP_{2050}(t) \times \dot{e}_{\text{CH}_4}^{\text{eq}}(t)
$$

(4)

The specific CO_{2}-equivalent emissions were then used to decide the deployment year of CH_{4} pyrolysis and water electrolysis, that is, they were deployed as soon as the specific emissions in year t became lower than those in the ‘direct natural gas use’ scenario (see figure 1(a)).

From equations (4) and (5), the temperature change in 2050 resulting from pathway i could then be approximated by summation of emissions from the years 2025–2050

$$
\Delta T_{t=2025}^{2050} = \sum_{t=2025}^{2050} \frac{AGTP_{\text{CO}_2,2050}(t) \times \dot{e}_{\text{CO}_2}^{\text{eq}}(t) \Delta t}{K(MJ_{\text{HHV}}Y^{-1})}
$$

(5)

2.3. Power generation emissions

The present-day specific emissions from coal- and gas-fired power generation were obtained from EIA data (U.S. Energy Information Administration 2020).

Progress toward decarbonization of power generation can be described in terms of the fraction of coal- and gas-generated power of the total power generated. Projections of these fractions were obtained from Williams et al, who studied eight different carbon-neutral pathways for the US to reach net-zero emissions by 2050 (2021). For each grid region and year, the fractions of coal and gas power generation were obtained from all eight pathways and aggregated into a Box-Cox transformed normal distribution. State-specific values were then obtained by assigning each state to a grid region. Coal supply chain CH_{4} emissions were added according to EPA data (United States Environmental Protection Agency 2021a). In addition, natural gas supply chain emissions were added as detailed in the following subsection.

2.4. Natural gas supply chain emissions

State-specific, consumption-normalized, production-stage CH_{4} emission factors have been estimated by Burns and Grubert (Burns and Grubert 2021a). Because they did not provide upper and lower bounds for their estimation, the upper and lower bounds determined by Omara for national production were applied instead (Omara et al 2018). For non-production CH_{4} emissions, state-specific estimates are not available, instead 0.5% of CH_{4} emissions are added in all states (Burns and Grubert 2021b).

End use emissions of CH_{4} were not included in this analysis because an end use of the natural gas and hydrogen was not specified. It should be noted that inclusion of end use emissions would likely render the ‘direct natural gas use’ pathway less attractive because of the higher global warming impact of CH_{4} compared to H_{2}. For supply chain CO_{2} emissions, an estimate of 7.5% of combustion emissions was used (Howarth and Jacobson 2021). The obtained specific CO_{2} emissions of power generation are compiled in table S1.

2.5. Methane mitigation scenarios

Three scenarios for methane emission mitigation along the natural gas supply chain were considered.

In the first, most conservative scenario ‘current regulations’, only the effect of state regulations (Canada), where an estimate of or target for the regulations’ effect on methane emissions has been provided (CA, CO, NM, PA, Canada, see table 1) is considered. In this scenario, year-by-year basin-specific emission factor decreases as a consequence of state regulation are calculated. Then, the effect of basin-specific emission factors on state-specific, consumption-normalized emission factors are traced.
Table 1. Summary of CH$_4$ emission factor reductions in the three CH$_4$ mitigation scenarios.

| Location      | Description                        | References                                      |
|---------------|------------------------------------|------------------------------------------------|
| 'Current regulations' scenario          |                                   |                                                  |
| CA            | 40% reduction until 2025, 45% until 2030 | California Air Resources Board (2020)          |
| NM            | 50% reduction until 2026           | New Mexico Environment Department (2022)       |
| CO            | 33% reduction until 2025, 50% until 2030 | Colorado Energy Office (2021)                  |
| PA            | 5% reduction until 2023            | Pennsylvania Department of Environmental Protection (2021) |
| Canada        | 40% reduction until 2025           | Government of Canada (2018)                    |
| All other states | No reduction                      | —                                               |
| 'EPA rule' scenario                |                                   |                                                  |
| US            | Yearly reductions according to RIA, 50% reduction until 2026 | United States Environmental Protection Agency (EPA) (2021b) |
| Canada        | 40% reduction until 2025           | Government of Canada (2018)                    |
| 'Rapid action' scenario          |                                   |                                                  |
| US + Canada   | 69% reduction until 2030           | International Energy Agency (IEA) (2021b)      |

Note: linear reduction in emission factor from 2023 to target year assumed in all cases.

Based on the natural gas flow analysis provided by Burns and Grubert (Burns and Grubert 2021a).

In the second scenario, 'EPA rule', the effect of a proposed federal EPA rule (RIN: 2060-AV16) on CH$_4$ emissions nationwide based on the EPA Regulatory Impact Assessment (incl. Canadian regulation) is considered.

In the third, most ambitious scenario 'rapid action', it is assumed that, starting from 2023, the CH$_4$ emission factors are linearly reduced until 2030 to 31% of the current emission factors and then remain constant until 2050. This corresponds to the abatement potential of 69% that has been identified by the IEA for US natural gas production (IEA 2021b). The target date for mitigation until 2030 is in agreement with the Global Methane Pledge. Other researchers have used more ambitious goals of 77%-83% global abatement, which include company commitments based on Oil and Gas Climate Initiative (OGCI) goals; however, OGCI members produce only a small fraction of US natural gas (Ocko et al 2021).

For methane emissions from coal mining, a 45% linear reduction in the methane intensity of coal supply from 2023 to 2030 is applied across all three scenarios (IEA 2021a). It should be noted that methane emissions from coal mining contribute little to temperature changes in 2050 in these calculations because of the rapid phase-out of coal in the net-zero-by-2050 scenarios.

2.6. Process efficiencies

Methane pyrolysis consumes both methane and power. The minimum CH$_4$ consumption for H$_2$ production from CH$_4$ pyrolysis is $E_{\text{CH}_4, \text{pyr,min}} = 221$ GJ$_{\text{HHV}}$ t$_{\text{H}_2}^{-1}$. Considering the endothermicity of the pyrolysis reaction, the minimum energy consumption becomes $E_{\text{pyr,min}} = 239$ GJ$_{\text{HHV}}$ t$_{\text{H}_2}^{-1}$. This additional required heat can be provided either from the combustion of CH$_4$, H$_2$, or through electrical heating. With the recent progress toward decarbonization of power production, electrical heating typically results in the lowest GHG emissions, which is why only electrically heated processes were considered in this study. Here, the two parameters efficiency (equation (6)), and share of power of total energy consumption (equation (7)) are used to fully describe CH$_4$ and power consumption and, thus, associated GHG emissions. Of note, it is assumed that neither CH$_4$ nor CO$_2$ is released during the methane pyrolysis process itself

$$\text{eff.} = \frac{E_{\text{pyr,min}}}{E_{\text{CH}_4} + E_{\text{power}}}$$

$$\text{power share} = \frac{E_{\text{power}}}{E_{\text{CH}_4} + E_{\text{power}}}.$$ 

The results of a literature survey and the generated cumulative density function (CDF) for pyrolysis efficiency and power share are shown in figure A1.

The efficiency of water electrolysis was obtained from predictions for AEC and PEMEC water electrolysis and SOEC steam electrolysis in 2030. For SOEC steam electrolysis, an energy penalty of 0.06 kWh Nm$^{-3}$ is applied for compression of H$_2$ from 10 to 30 bar, as well as a penalty of 0.26 kWh Nm$^{-3}$ to compensate for loss in power production when the required steam is extracted from a thermal power plant steam cycle (Tenhumberg and Büker 2020). The year 2030 was chosen because, in most of the calculations, electrolysis is deployed
Table 2. Summary of model input parameters.

| State-specific | Distribution shape | Value | Unit | References |
|----------------|--------------------|-------|------|------------|
| Natural gas supply chain |                   |       |      |            |
| CH₄ emission factor—production | Yes | Normal | μ: state-specific, US average: μ = 1.6 σ = 0.194μ | % | Burns and Grubert (2021a), Omara et al (2018) |
| CH₄ emission factor—processing, transmission, and storage | No | Deterministic | 0.5 | % | Burns and Grubert (2021b) |
| CO₂ emission factor | No | Deterministic | 7.5% of combustion CO₂ emissions | % | Howarth and Jacobson (2021) |
| Coal supply chain |                   |       |      |            |
| CH₄ emissions | No | Skew normal | ξ = 1.51, ω = 23.9, α = 0.23 | kgCH₄ MWhₑ,coal⁻¹ | US Environmental Protection Agency (2021) |
| Power generation |                   |       |      |            |
| Spec. CO₂ emissions coal | Yes | Deterministic | State-specific | kgCO₂ MWhₑ,coal⁻¹ | U.S. Energy Information Administration (2020) |
| Spec. CO₂ emissions gas | Yes | Deterministic | State-specific | kgCO₂ MWhₑ,gas⁻¹ | U.S. Energy Information Administration (2020) |
| Fraction of coal power gen. | Yes | Normal, Box-Cox transformed | Grid-region-specific, year-specific | MWhₑ,coal | Williams et al (2021) |
| Fraction of gas power gen. | Yes | Normal, Box-Cox transformed | Grid-region-specific, year-specific | MWhₑ,gas | Williams et al (2021) |
| CH₄ pyrolysis |                   |       |      |            |
| Efficiency | No | Truncated normal | μ = 88.3, σ = 7.56, truncated at 100 | % | See figure A1(a) |
| Power consumption | No | Laplace | μ = 16.6, b = 5.41 | % | See figure A1(b) |
| Water electrolysis |                   |       |      |            |
| Efficiency | No | Skew normal | ξ = 61.3, ω = 6.32, α = 10.4 | % LHV | See figure A2 |

from that year onward. The CDF for electrolysis efficiency was obtained from a literature survey, with a focus on expert elicitation studies, and is shown in figure A2.

2.7. Model description
A MS Excel/VBA tool was developed to conduct Monte Carlo simulations with 10 000 runs. This tool is accessible in a GitHub repository and can be easily adjusted for use in a range of different scenarios and climate goals.

The upper and lower bounds presented throughout this study correspond to the 10th and 90th percentiles, respectively. A summary of the model input parameters used in this study is provided in table 2.

3. Results
3.1. Energy consumption of natural gas pyrolysis and electrolysis
Based on the discussion of process efficiencies in section 2.6 and the resulting CDF’s shown in the appendix, the consumption of natural gas and electricity of the pyrolysis process and the electricity consumption of the electrolysis process can be obtained.
From the specific emissions profiles, shown exemplarily in figure 4, the deployment year of natural gas pyrolysis and water electrolysis is decided (cf figure 2). From the so-obtained time-resolved specific emission profiles of all three pathways, the resulting specific temperature changes in 2050 are calculated according to equation (5). In the following subsections, the temperature changes are presented and discussed.

3.3. Contributions of CH4 and CO2 to warming in 2050

The specific temperature changes in 2050 resulting from the three different pathways for US average conditions, and under all three CH4 mitigation scenarios, can be broken down into contributions from CO2 and CH4, as summarized in table 4. As expected, for the direct use of natural gas, the warming effect is dominated by combustion CO2 emissions; however, there are also some substantial CH4 contributions if CH4 emissions are not abated rapidly. Similarly, warming is dominated by the contribution of CO2 from the power generation in the water electrolysis pathway. In contrast, in the natural gas pyrolysis pathway, warming contributions are predominantly from CH4 emissions, if they are not rapidly abated. The characterization of natural gas pyrolysis as a high-CH4/low-CO2 technology is thus justified. It is noteworthy that in the more ambitious CH4 mitigation scenarios, the global warming impact of natural gas pyrolysis decreases substantially, whereas the global warming impact of electrolysis does not decrease that much due to the relatively smaller contribution of CH4 compared to CO2. A more detailed breakdown of the contributions of CO2 and CH4 from each stage is provided in table S2.

3.4. State-specific impact on warming in 2050

In the preceding subsection, the effect on global warming was shown for US average conditions. However, global warming effects are starkly different when compared regionally because of the large differences in consumption-based, state-specific CH4 emissions along the natural gas supply chain, as well as in the grid-regional timeline of the phase-out of coal and natural gas use in power generation. In figures 5–7, the density plots of state-specific estimates for the 11 states with the largest consumption of natural gas in the US are shown for all three technological pathways and CH4 mitigation scenarios.

The density plots show a large degree of uncertainty in the warming impact of natural gas pyrolysis, particularly in states where CH4 emissions are high (e.g., California, Indiana and Oklahoma) and when these emissions are not well abated (figure 5). The uncertainty of the global warming effect of the water electrolysis pathway is mainly a result of uncertainty of future electrolyzer efficiency, as well as how rapid

3.2. Projected specific emissions over time

The calculated CO2-equivalent emission profiles towards a 2050 temperature endpoint obtained from equation (4), are shown for US average conditions under all three scenarios in figure 4.

Initially, in 2025, emissions from H2 produced through water electrolysis are the highest but then reduce drastically due to the projected progression of the decarbonization of power generation. The emissions from H2 produced through natural gas pyrolysis decrease significantly only in the ‘rapid action’ scenario. The ‘EPA rule’ scenario assumes a substantial decrease of CH4 emissions from 2025 to 2026 (in accordance with the EPA regulatory impact analysis), however CO2-equivalent emissions remain almost constant afterwards due to a balance between stagnating CH4 emissions with an increasing GTP2050(t) (cf figure 2), and decreasing emissions from power consumption. In the ‘rapid action’ scenario, a deeper emission reduction for pyrolysis is achieved until 2030, but also in this scenario emissions similarly level out from 2030 onwards.

The years when the specific emissions of water electrolysis become lower than those of natural gas pyrolysis are summarized in table 3 for all studied locations and under the three CH4 mitigation scenarios. As can be clearly observed, there is a large spread with electrolysis being favorable as early as 2026 in Oklahoma in the ‘current regulations’ scenario, and as late as 2047 in New York in the ‘rapid action’ scenario.

Figure 3. Consumption of natural gas and electricity for H2 production from natural gas pyrolysis and electrolysis. Note: only consumption during operation considered, embodied energy of plant construction etc not included.
Figure 4. Specific emissions for US average conditions and under all three CH\textsubscript{4} mitigation scenarios for the three technologies investigated. Shaded areas indicate the 10\%-90\% confidence intervals. Superimposed is the probability distribution of the year by which specific emissions of H\textsubscript{2} produced from electrolysis become lower than H\textsubscript{2} produced from natural gas pyrolysis. State-specific results are provided in tables S3 to S4 in the supporting information.

Table 3. Projected year by which specific emissions of H\textsubscript{2} produced from electrolysis become lower than H\textsubscript{2} produced from natural gas pyrolysis.

| State | Current regulations | EPA rule | Rapid action |
|-------|-------------------|----------|-------------|
| US avg. | 2032 (2030–2035) | 2033 (2031–2036) | 2036 (2034–2040) |
| TX | 2029 (2028–2032) | 2030 (2029–2034) | 2033 (2031–2036) |
| CA | 2034 (2031–2037) | 2034 (2031–2037) | 2037 (2034–2042) |
| LA | 2029 (2028–2029) | 2029 (2029–2030) | 2030 (2029–2030) |
| PA | 2035 (2033–2039) | 2037 (2034–2040) | 2041 (2037–2045) |
| FL | 2040 (2037–2043) | 2041 (2039–2044) | 2043 (2041–2046) |
| NV | 2041 (2036–2046) | 2044 (2038–2048) | 2047 (2043–2050) |
| OH | 2035 (2033–2040) | 2037 (2034–2041) | 2041 (2038–2045) |
| IL | 2036 (2033–2040) | 2037 (2034–2041) | 2041 (2037–2045) |
| MI | 2036 (2033–2040) | 2037 (2034–2041) | 2041 (2038–2045) |
| IN | 2034 (2031–2037) | 2035 (2032–2038) | 2038 (2035–2042) |
| OK | 2026 (2026–2028) | 2028 (2027–2029) | 2029 (2027–2030) |

Table 4. Specific temperature increases in 2050 resulting from emissions between 2025 and 2050 for US average conditions, broken down into contributions from CO\textsubscript{2} and CH\textsubscript{4} under the three investigated CH\textsubscript{4} mitigation scenarios.

| | Direct natural gas use | Natural gas pyrolysis | Water electrolysis |
|---|----------------------|-----------------------|-------------------|
| CO\textsubscript{2} | 0.77 | 0.31 (0.28–0.36) | 0.42 (0.36–0.51) |
| CH\textsubscript{4}—current regulations | 0.36 (0.30–0.43) | 0.57 (0.46–0.70) | 0.17 (0.13–0.23) |
| CH\textsubscript{4}—EPA rule | 0.28 (0.24–0.33) | 0.45 (0.36–0.54) | 0.13 (0.10–0.18) |
| CH\textsubscript{4}—rapid action | 0.14 (0.11–0.16) | 0.21 (0.17–0.26) | 0.07 (0.06–0.09) |

the power grid decarbonization progresses. In many locations, across all three CH\textsubscript{4} mitigation scenarios, there is a significant overlap between the pyrolysis and electrolysis temperature change pdfs, which illustrates the difficulty of predicting the technological pathway with the lowest climate impact. Only in a few states (Louisiana, Oklahoma, Texas), electrolysis is clearly predicted to have the lowest climate impact.

From figure 6, it can be observed that the impact of a federal EPA rule on the warming effect of natural gas pyrolysis is large in many states (e.g. Oklahoma), but almost marginal in other states (e.g. California, due to ambitious regulation already in place in-state and in Canada, where much of the consumed natural gas is sourced).

To assess the merit of H\textsubscript{2} from natural gas pyrolysis as a potential technology for mid-century global warming mitigation, it is useful to consider the extent to which warming can be reduced compared to the business-as-usual, direct utilization of natural gas and to the use of H\textsubscript{2} from water electrolysis (figure 8). In states where natural gas and coal-fired power generation are projected to be rapidly phased out (e.g. Oklahoma, Louisiana, and Texas), water electrolysis is preferable.

However, in states with a longer projected dependence on natural gas for power generation and lower CH\textsubscript{4} supply chain emissions, such as New York, pyrolysis appears to be an attractive option if CH\textsubscript{4} emissions are abated rapidly. Even in New York, in the ‘current regulations’ scenario, the mid-century warming effect will be reduced by only −19% (−28%, −9%) compared to the continuing direct use of natural gas. However, if CH\textsubscript{4} emissions are rapidly reduced, a more substantial reduction of −38% (−47%, −28%) compared to direct use is achievable.
3.5. Effect of CO₂ equivalency metric
Throughout this study, the GTP in 2050, GTP_{2050}(t), has been used to obtain the CO₂ equivalency of CH₄ emissions (see section 2.2). Sensitivity tests were conducted to determine the deviation when GWP_{100} or GWP_{20} are used instead, and the results are presented in tables S6 to S11, and summarized in table 5. The use of the 100 year global warming potential GWP_{100} substantially underestimates the effect of methane emissions from natural gas pyrolysis, while the use of GWP_{20} yields similar results compared to the use of GTP_{2050}(t).

3.6. What if decarbonization of the power grid progresses more slowly?
Throughout this study, it is assumed that net zero emissions from power generation are achieved by 2050, however, it is uncertain whether this will be achieved. Thus, a sensitivity test was conducted, where the decarbonization progresses more slowly towards net zero in 2060 instead of 2050. Results from this sensitivity test are provided in tables S12 to S14.

As expected, such a slower grid decarbonization makes H₂ from electrolysis less attractive; the specific effect on global temperature in 2050 in the ‘water electrolysis’ pathway increases from ∆T_{2050} = 0.59 * 10^{-15} K (MJ_{HHV} y^{-1})^{-1} to ∆T_{2050} = 0.79 * 10^{-15} K (MJ_{HHV} y^{-1})^{-1} (U.S. average, central estimate).

4. Discussion
In this analysis, it is shown that the merit of natural gas pyrolysis for mid-century global warming mitigation depends considerably on the location of the plant and future progress in natural gas supply chain CH₄.
mitigation. In addition, even in a well-specified CH₄ mitigation scenario, there is considerable uncertainty regarding its climate impact, both due to uncertainty in present-day CH₄ emissions and achievable process efficiency. It is noted that in this analysis, uncertainty related to the climate sensitivity and the CO₂ equivalency of CH₄ were not included, which, if included, would further add to the uncertainty in the projected climate impact.

In the following, I discuss some of the underlying assumptions of this analysis grouped by whether they would positively or negatively affect the climate benefit of natural gas pyrolysis if violated.

In favor of natural gas pyrolysis:

- The analysis presented here assumed that the decarbonization of power generation progresses rapidly toward net-zero emissions by 2050, which is ambitious and requires immediate action. Should this decarbonization progress become significantly slower, natural gas pyrolysis would become considerably more attractive than water electrolysis with grid electricity to limit global warming.

- In this analysis, it was assumed that electrolyzers will be available without restrictions from 2025. However, it is possible that the demand for H₂ could exceed the installed capacity of electrolyzer manufacturing, either because of a shortage of production capacity or a shortage of critical metals, such as iridium (Minke et al 2021). In that case, an all-of-the-above strategy could require the deployment of natural gas pyrolysis, even if its climate benefit would be smaller than that of water electrolysis.

In opposition to natural gas pyrolysis:

- For large-scale applications of natural gas pyrolysis, it is likely necessary to landfill the produced solid...
carbon, as global markets for carbon would not be able to take up the large volume of carbon produced (Parkinson et al 2017). The landfilling/disposal of very large quantities of solid carbon can incur further direct or indirect greenhouse gas emissions; for example, for transport to the final disposal site. The technical and commercial viability of large-scale pyrolysis for hydrogen production has not yet been demonstrated.

- In this analysis, it was assumed that water electrolysis is operated continuously from grid electricity, and with average specific emissions associated with its power consumption. However, water electrolysis can also be operated in part-load mode, thereby providing an additional benefit to the energy system in the form of grid balancing (Williams et al 2021). In such a case, lower-than-average specific emissions should be ascribed to the power consumption of water electrolysis.

- The present-day estimates of the CH$_4$ emission factors of the natural gas supply chain used in this analysis may need to be corrected upward in the future. Recent satellite data show that CH$_4$ emissions may be higher than previously known (Zhang et al 2020, Lauvaux et al 2022). This could substantially increase the impact of natural gas pyrolysis on global warming.

This analysis did not provide estimates of the projected costs of water electrolysis and natural gas pyrolysis. It is very challenging to predict future electricity prices, natural gas prices, disposal costs for solid carbon, and carbon taxes (on CO$_2$ and CH$_4$ emissions). However, this work may provide a basis for further techno-economic assessment of high-CH$_4$/low-CO$_2$
Figure 8. State-specific relative change in effect on 2050 temperature for natural gas pyrolysis pathway compared to both direct natural gas use and water electrolysis: (a) ‘current regulations,’ (b) ‘EPA rule,’ and (c) ‘rapid action’ scenarios. Error bars indicate the 10%–90% confidence intervals.

Table 5. Specific temperature increases in 2050 resulting from emissions between 2025 and 2050 for US average conditions under the ‘current regulations’ CH₄ mitigation scenario, with three different CO₂ equivalency metrics used.

| Process                  | ΔT₂₀₅₀ [K (MJ₁Hᵥ⁻¹ · 10¹⁵)] |
|--------------------------|-------------------------------|
| Direct natural gas use   |                               |
| GTP₂₀₅₀(t)               | 1.13 (1.06–1.20)             |
| GWP₁₀₀                   | 0.89 (0.86–0.91)             |
| GWP₂₀                   | 1.13 (1.06–1.19)             |
| Natural gas pyrolysis    |                               |
| GTP₂₀₅₀(t)               | 0.89 (0.77–1.03)             |
| GWP₁₀₀                   | 0.50 (0.45–0.56)             |
| GWP₂₀                   | 0.87 (0.75–1.00)             |
| Water electrolysis       |                               |
| GTP₂₀₅₀(t)               | 0.59 (0.49–0.74)             |
| GWP₁₀₀                   | 0.49 (0.41–0.59)             |
| GWP₂₀                   | 0.62 (0.51–0.76)             |

processes. It also provides an indication of how large the uncertainty of potential climate benefits of such processes can be.

Besides natural gas pyrolysis, blue hydrogen produced from steam methane reforming (SMR) of natural gas with CO₂ capture and geological storage is
frequently discussed as a bridge technology. However, the natural gas consumption, the feasibility of auto-thermal reforming vs. SMR, and especially the realistically achievable carbon capture rate from a blue H\textsubscript{2} plant are unclear and estimates vary widely (Howarth and Jacobson 2021, Bauer et al. 2022). Nevertheless, preliminary estimates with the model developed herein (assuming SMR + carbon capture and storage (CCS), 1.37 MJ\textsubscript{NG} MJ\textsubscript{H2}\textsuperscript{-1}, ∼73% CO\textsubscript{2} capture rate based on an ammonia plant configuration) indicate that the warming effect of blue hydrogen could be quite similar to that of natural gas pyrolysis.

5. Conclusion

In this paper, the climate benefit of H\textsubscript{2} production from natural gas pyrolysis in comparison to water electrolysis and unabated direct natural gas utilization toward a mid-century temperature target and corresponding net-zero-by-2050 decarbonization scenarios were studied.

The time-resolved emission profiles indicate that the year in which emissions from electrolysis are lower than those from natural gas pyrolysis depends highly on the state where the plant is located and on the timeline of CH\textsubscript{4} emission mitigation efforts. States that (a) consume natural gas from basins that already have a comparatively low CH\textsubscript{4} emission factor and (b) are projected to depend for a longer time on natural gas use for power generation, such as New York, Pennsylvania, and Ohio, appear to be the most attractive locations for natural gas pyrolysis plants. However, it was found that the uncertainty of current CH\textsubscript{4} emission factors as well as the magnitude and speed of future CH\textsubscript{4} emission abatements make it challenging to judge the climate benefits of natural gas pyrolysis.

This study was carried out with the aim to inform the debate surrounding high-CH\textsubscript{4}/low-CO\textsubscript{2} technologies such as natural gas pyrolysis. I thus provide a simple, easy-to-use/modify Excel VBA tool to assess the climate impact of these technologies; the tool can be easily updated whenever better information on CH\textsubscript{4} emissions, CH\textsubscript{4} mitigation, and power generation decarbonization pathways becomes available. Policymakers, businesspeople, and scholars can use it freely to estimate the climate impact within their own scenarios and locations, which may aid them in their decision-making.

Data availability statement

The MS Excel sheet for the Monte Carlo simulations can be obtained from the following GitHub repository: https://github.com/Keller-Martin/Pyrolysis-Electrolysis-Emissions.
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