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MODELLING AND ANALYSIS

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How green is blue hydrogen?

Robert W. Howarth[1] | **Mark Z. Jacobson**[2]

[1]Department of Ecology & Evolutionary Biology, Cornell University, Ithaca, New York, USA

[2]Department of Civil & Environmental Engineering, Stanford University, Stanford, California, USA

Correspondence
Robert W. Howarth, Department of Ecology & Evolutionary Biology, Cornell University, Ithaca, NY 14853, USA.
Email: howarth@cornell.edu

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Abstract

Hydrogen is often viewed as an important energy carrier in a future decarbonized world. Currently, most hydrogen is produced by steam reforming of methane in natural gas ("gray hydrogen"), with high carbon dioxide emissions. Increasingly, many propose using carbon capture and storage to reduce these emissions, producing so-called "blue hydrogen," frequently promoted as low emissions. We undertake the first effort in a peer-reviewed paper to examine the lifecycle greenhouse gas emissions of blue hydrogen accounting for emissions of both carbon dioxide and unburned fugitive methane. Far from being low carbon, greenhouse gas emissions from the production of blue hydrogen are quite high, particularly due to the release of fugitive methane. For our default assumptions (3.5% emission rate of methane from natural gas and a 20-year global warming potential), total carbon dioxide equivalent emissions for blue hydrogen are only 9%-12% less than for gray hydrogen. While carbon dioxide emissions are lower, fugitive methane emissions for blue hydrogen are higher than for gray hydrogen because of an increased use of natural gas to power the carbon capture. Perhaps surprisingly, the greenhouse gas footprint of blue hydrogen is more than 20% greater than burning natural gas or coal for heat and some 60% greater than burning diesel oil for heat, again with our default assumptions. In a sensitivity analysis in which the methane emission rate from natural gas is reduced to a low value of 1.54%, greenhouse gas emissions from blue hydrogen are still greater than from simply burning natural gas, and are only 18%-25% less than for gray hydrogen. Our analysis assumes that captured carbon dioxide can be stored indefinitely, an optimistic and unproven assumption. Even if true though, the use of blue hydrogen appears difficult to justify on climate grounds.

KEYWORDS

blue hydrogen, decarbonization, greenhouse gas footprint, hydrogen, methane, methane emissions

1 | INTRODUCTION

Hydrogen is widely viewed as an important fuel for a future energy transition. Currently, hydrogen is used mostly by industry during oil-refining and synthetic nitrogen fertilizer production, and little is used for energy because it is expensive relative to fossil fuels.[1] However, hydrogen is increasingly being promoted as a way to address climate change, as

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indicated by a recent article in the New York Times.[2] In this view, hydrogen is to be used not only for hard to decarbonize sectors of the economy such as long-distance transportation by trucks and airplanes but also for heating and cooking, with hydrogen blended with natural gas and distributed to homes and business through existing pipeline systems.[2] Utilities are also exploring the use of hydrogen, again blended with natural gas, to power existing electric generating facilities.[3] In Europe, a recent report from Gas for Climate, an association of natural gas pipeline companies, envisions large scale use of hydrogen in the future for heating and electricity generation.[4] The Hydrogen Council, a group established in 2017 by British Petroleum, Shell, and other oil and gas majors, has called for heating all homes with hydrogen in the future.[5]

The vast majority of hydrogen (96%) is generated from fossil fuels, particularly from steam methane reforming (SMR) of natural gas but also from coal gasification.[6] In SMR, which is responsible for approximately three quarters of all hydrogen production globally,[7] heat and pressure are used to convert the methane in natural gas to hydrogen and carbon dioxide. The hydrogen so produced is often referred to as "gray hydrogen," to contrast it with the "brown hydrogen" made from coal gasification.[8] Production of gray hydrogen is responsible for 6% of all natural gas consumption globally.[7] Hydrogen can also be generated by electrolysis of water. When such electricity is produced by a clean, renewable source, such as hydro, wind, or solar, the hydrogen is termed "green hydrogen." In 2019, green hydrogen was not cost competitive with gray hydrogen,[9] but that is changing as the cost of renewables is decreasing rapidly and electrolyzers are becoming more efficient. Still, the supply of green hydrogen in the future seems limited for at least the next several decades.[2,5]

Greenhouse gas emissions from gray hydrogen are high,[10,11] and so increasingly the natural gas industry and others are promoting "blue hydrogen".[5,8,9] Blue hydrogen is a relatively new concept and can refer to hydrogen made either through SMR of natural gas or coal gasification, but with carbon dioxide capture and storage. As of 2021, there were only two blue-hydrogen facilities globally that used natural gas to produce hydrogen at commercial scale, as far as we can ascertain, one operated by Shell in Alberta, Canada, and the other operated by Air Products in Texas, USA.[12] Often, blue hydrogen is described as having zero or low greenhouse gas emissions.[8,9] However, this is not true: not all of carbon dioxide emissions can be captured, and some carbon dioxide is emitted during the production of blue hydrogen.[1] Further, to date no peer-reviewed analysis has considered methane emissions associated with producing the natural gas needed to generate blue hydrogen.[1] Methane is a powerful greenhouse gas. Compared mass-to-mass, it is more than 100-times more powerful as a warming agent than carbon dioxide for the time both gases are in the atmosphere and causes 86-times the warming as carbon dioxide over an integrated 20-year time frame after a pulsed emission of the two gases. Approximately 25% of the net global warming that has occurred in recent decades is estimated to be due to methane.[13] In a recent report, the United Nations Environment Programme concluded that methane emissions globally from all sources need to be reduced by 40%-45% by 2030 in order to achieve the least cost pathway for limiting the increase in the Earth's temperature to 1.5°C, the target set by COP 21 in Paris in December 2015.[14]

Here, we explore the full greenhouse gas footprint of both gray and blue hydrogen, accounting for emissions of both methane and carbon dioxide. For blue hydrogen, we focus on that made from natural gas rather than coal, that is gray hydrogen combined with carbon capture and storage. In China, brown hydrogen from coal now dominates over gray hydrogen from natural gas, due to the relative prices of natural gas and coal, but globally and particular in Europe and North America, gray hydrogen dominates.[1]

2 | ESTIMATING EMISSIONS FROM PRODUCING GRAY HYDROGEN

Greenhouse gas emissions from the production of gray hydrogen can be separated into two parts: (a) the SMR process in which methane is converted to carbon dioxide and hydrogen; and (b) the energy used to generate the heat and high pressure needed for the SMR process. For blue hydrogen, which we discuss later in this paper, emissions from the generation of electricity needed to run the carbon dioxide capture equipment must also be included. In this analysis, we consider emissions of only carbon dioxide and methane, and not of other greenhouse gases such as nitrous oxide that are likely to be much smaller. For methane, we consider the major components of its lifecycle emissions associated with the mining, transport, storage, and use of the natural gas needed to produce the hydrogen and power carbon capture. Emissions are expressed per unit energy produced when combusting the hydrogen, to aid in comparing the greenhouse gas footprint with other fuels.[15,16] In this paper, we use gross calorific values.

We start by estimating how much methane is consumed and how much carbon dioxide is produced in the two aspects of production of gray hydrogen. From this information, we can subsequently below estimate emissions of unburned methane.

2.1 | Consumption of methane and production of carbon dioxide in SMR process

In the SMR process, 1 mole of carbon dioxide and 4 moles of hydrogen gas (H_2) are produced per mole of methane consumed, according to this overall reaction:

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$$CH_4 + 2H_2O \rightarrow CO_2 + 4H_2 \quad (1)$$

The gross caloric calorific heat content of hydrogen is 0.286 MJ per mole,[17] or inverting this value, 3.5 moles H_2 per MJ. The carbon dioxide produced during the SMR process is given by:

$$(3.5 \text{ moles } H_2/\text{MJ}) * (1 \text{ mole } CO_2/4 \text{ moles } H_2) = 0.875 \text{ moles } CO_2 \text{ per MJ} \quad (2)$$

With a molecular weight of 44.01 g per mole, the amount of carbon dioxide produced during the SMR process is 38.51 g CO_2 per MJ (Table 1). The amount of methane consumed is given by:

$$(3.5 \text{ moles } H_2/\text{MJ}) * (1 \text{ mole } CH_4/4 \text{ moles } H_2) = 0.875 \text{ moles } CH_4 \text{ per MJ} \quad (3)$$

With a molecular weight of 16.04 g per mole, 14.04 g CH_4 per MJ is consumed during the SMR process (Table 1). There is essentially no uncertainty in these estimates of how much methane is consumed, and how much carbon dioxide is produced during the SMR process: the relationship is set by the chemical stoichiometry shown in Equation (1).

2.2 | Consumption of methane and production of carbon dioxide from energy needed to drive SMR process

The production of hydrogen from methane is an endothermic reaction and requires significant input of energy, between 2.0 and 2.5 kWh per m^3 of hydrogen, to provide the necessary heat and pressure.[18] This energy comes almost entirely from natural gas when producing gray hydrogen, and therefore, also presumably when producing blue hydrogen proposed for Europe or North America.[1] Using a mean value of 2.25 kWh per m^3 of hydrogen, we estimate the energy in natural gas (methane) required to produce a mole of hydrogen as follows:

$$(2.25 \text{ kWh}/\text{m}^3 \text{ of } H_2) * (3.6 \text{ MJ/kWh}) * (1 \text{ m}^3/1000 \text{ L}) * (22.4 \text{ L/mol}) = 0.1814 \text{ MJ per mole } H_2 \quad (4)$$

That is, 0.1814 MJ of energy from burning methane is required per mole of hydrogen produced. When burning natural gas for heat, 50 g CO_2 per MJ in emissions are produced, using gross calorific values.[19] Note that higher carbon dioxide emission values are reported when using net calorific values.

Therefore,

$$(0.1814 \text{ MJ/mole } H_2) * (50 \text{ g } CO_2/\text{MJ}) = 9.07 \text{ g } CO_2 \text{ per mole } H_2. \quad (5)$$

As noted above, the gross calorific heat content of hydrogen is equivalent to 3.5 moles H_2 per MJ. Therefore,

$$(9.07 \text{ g } CO_2/\text{mole } H_2) * (3.5 \text{ moles } H_2/\text{MJ}) = 31.8 \text{ g } CO_2/\text{MJ} \quad (6)$$

So 31.8 g of carbon dioxide are produced to generate the heat and pressure to drive the SMR process per MJ of hydrogen produced (Table 1). Since one mole of methane in natural gas is burned to produce one mole of carbon dioxide emissions, we can estimate the methane consumed as follows:

$$(31.8 \text{ g } CO_2/\text{MJ}) * (1 \text{ mole } CO_2/44.01 \text{ g } CO_2) * (16.04 \text{ g } CH_4/\text{mole } CH_4) * (1 \text{ mole } CH_4/\text{mole } CO_2) = 11.6 \text{ g } CH_4/\text{MJ} \quad (7)$$

See Table 1.

2.3 | Total carbon dioxide and methane emissions for gray hydrogen

The sum of the carbon dioxide from the SMR process (38.5 g CO_2 per MJ) and from the energy used to generate the heat and electricity for the SMR (31.8 g CO_2 per MJ) is 70.3 g CO_2 per MJ. Additionally, it takes energy to produce, process, and transport the natural gas used to generate the hydrogen. Using the analysis of Santoro et al.[20] as reported in Howarth et al,[21] these indirect upstream emissions are approximately 7.5% of the direct carbon dioxide emissions for natural gas, or an additional 5.3 g CO_2 per MJ (7.5% of 70.3 g CO_2 per MJ). Therefore, the total quantity of carbon dioxide produced is 75.6 g CO_2 per MJ (Table 1).

The total quantity of methane in natural gas consumed to generate gray hydrogen is the sum of that used in the SMR process (14.04 g CH_4 per MJ) and the amount burned to generate the heat and high pressure needed for the process (11.6 g CH_4 per MJ) or 25.6 g CH_4 per MJ. It is not possible to produce and use natural gas without having some methane emitted unburned to the atmosphere, due both to leaks and to purposeful emissions including venting.[21,22] Below, we briefly discuss the recent literature that characterizes methane emissions from natural gas operations, and use a range of values in a sensitivity analysis. Here, for our default estimation of the greenhouse gas footprint of gray hydrogen, we rely on a recent synthesis on "top–down" emission studies.[16] Top–down estimates use information such as from satellites or airplane flyovers that characterize an integrated flux. The mean value of estimates from 20 different studies in 10 major natural gas fields in the United States, normalized to gas production in those fields, indicates that 2.6% of gas production is emitted to the atmosphere.[16] This is a good estimate for the upstream emissions that occur in the gas fields. Methane is also emitted from storage and transport to consumers, and the data in the top–down study of Plant et al[23] suggests this is an additional

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TABLE 1 Comparison of methane that is consumed, of carbon dioxide that is produced, and of emissions of both methane and carbon dioxide for each step in the processing of methane to hydrogen for gray hydrogen, blue hydrogen with carbon dioxide capture from the SMR process but not from the exhaust flue gases created from burning natural gas to run the SMR equipment, and blue hydrogen with carbon dioxide capture from both the SMR process and from the exhaust flue gases

	Gray H_2	Blue H_2 (w/o flue-gas capture)	Blue H_2 (w/flue-gas capture)
SMR process			
CH_4 consumed (g CH_4/MJ)	14.0	14.0	14.0
CO_2 produced (g CO_2/MJ)	38.5	38.5	38.5
Fugitive CH_4 emissions (g CH_4/MJ)	0.49	0.49	0.49
Fugitive CH_4 emissions (g CO_2eq/MJ)	42.1	42.1	42.1
Direct CO_2 emissions (g CO_2/MJ)	38.5	5.8	5.8
CO_2 capture rate	0%	85%	85%
Energy to drive SMR			
CH_4 consumed (g CH_4/MJ)	11.6	11.6	11.6
CO_2 produced (g CO_2/MJ)	31.8	31.8	31.8
Fugitive CH_4 emissions (g CH_4/MJ)	0.41	0.41	0.41
Fugitive CH_4 emissions (g CO_2eq/MJ)	35.3	35.3	35.3
Direct CO_2 emissions (g CO_2/MJ)	31.8	31.8	11.1
CO_2 capture rate	0%	0%	65%
Energy to power carbon capture			
CH_4 consumed (g CH_4/MJ)	0	3.0	6.0
CO_2 produced (g CO_2/MJ)	0	8.2	16.3
Fugitive CH_4 emissions (g CH_4/MJ)	0	0.11	0.21
Fugitive CH_4 emissions (g CO_2eq/MJ)	0	9.5	1
Direct CO_2 emissions (g CO_2/MJ)	0	8.2	16.0
Indirect upstream CO_2 emissions (g CO_2/MJ)	5.3	5.9	6.5
Total CH4 consumed (g CH_4/MJ)	25.6	28.6	31.6
Total CO_2 emitted (g CO_2/MJ)	75.6	51.7	39.7
Total fugitive CH_4 emissions (g CO_2eq/MJ)	77.4	86.9	95.4
Total emissions (g CO_2eq/MJ)	153	139	135

Note: The methane leakage rate is 3.5%.

0.8%.[16,24] Combined with the 2.6% for field-level emissions, we estimate a total of 3.4% of production is emitted to the atmosphere overall. Note that in addition to some methane being lost between production and consumption due to leaks, methane is also burned by the natural gas industry to power natural gas processing and transport. This is important to consider, since we want to evaluate how much methane is emitted for the methane in natural gas that is consumed in producing hydrogen. In 2015, natural gas production in the United States was 817 billion m^3, while consumption was 771 billion m^3,[25,26] (converting cubic feet to cubic meters). Using this information, we can estimate the methane emission as a percentage of gas consumption as follows:

$$(3.4\%\ \text{of production}) * (817 \times 109\ \text{m}^3 / 771 \times 109\ \text{m}^3) = 3.5\%\ \text{of consumption} \quad (8)$$

With this value and the quantity of methane consumed to produce gray hydrogen, we can estimate the upstream emissions of methane:

$$(3.5\%\ \text{of consumption}) * (\text{consumption of } 25.6\ \text{g CH}_4\ \text{per MJ}) = 0.90\ \text{g CH}_4\ \text{per MJ} \quad (9)$$

To compare methane emissions with carbon dioxide emissions requires a specified time frame, since the half-life of methane in the atmosphere is only 12 years or so, far less than that of carbon dioxide.[13] Greenhouse gas inventories often compare methane with carbon dioxide for an integrated period of 100 years following pulsed emissions of both gases. However, this underestimates the role of methane in global warming over shorter time periods. An increasing number of scientists have called for using a 20-year integrated time period instead of or in addition to the 100-year period.[15,21,24,27,28]

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The 20-year time frame is now mandated by law in the State of New York, as part of the Climate Leadership and Community Protection Act of 2019.[24] And a 20-year period is more appropriate than a 100-year time frame given the urgency of reducing methane emissions globally over the coming decade.[14] Here, we use the 20-year time frame using the Global Warming Potential (GWP) for 20 years of 86.[13] We also consider other GWP values in a sensitivity analysis presented below. Using the 86 value, we estimate upstream methane emissions associated with the production of gray hydrogen in units of carbon dioxide equivalents (CO_2eq) thus:

$$(0.90 \text{ g } CH_4 \text{ per MJ}) * (86 \text{ g } CO_2\text{eq/g } CH_4) = 77.4 \text{ g}CO_2\text{eq per MJ} \quad (10)$$

The sum of emissions of carbon dioxide (75.6.0 g CO_2 per MJ) and unburned methane (77.4 g CO_2eq per MJ) for the production of gray hydrogen is 153 g CO_2eq per MJ (Table 1).

There are remarkably few published peer-reviewed papers with which to compare our estimate. Many non peer-reviewed reports give estimates for carbon dioxide emission from gray hydrogen that are in the range of 10 tons carbon dioxide per ton of hydrogen,[1,7] although data in support of these values are generally absent, perhaps because they are based on confidential information.[11] Since the gross calorific heat energy content of hydrogen is 0.286 MJ per mole,[17] 10 tons of carbon dioxide per ton of hydrogen corresponds to 70 g CO_2 per MJ. This is similar to but somewhat lower than our value of 75.6 g CO_2 per MJ. Most of these non peer-reviewed reports do not include methane in their estimates,[1] or if they do, they provide no detail as to how they do so. The most thorough peer-reviewed analysis of carbon dioxide emissions for gray hydrogen is that of Sun et al[11] who obtained data on both rates of hydrogen production and emissions of carbon dioxide from many individual facilities across the United States. They concluded that on average, carbon dioxide emissions for gray hydrogen are 77.8 g CO_2 per MJ, remarkably close to our value of 75.6 g CO_2 per MJ. They did not estimate methane emissions.

3 | ESTIMATING EMISSIONS FOR BLUE HYDROGEN

Blue hydrogen differs from gray hydrogen in that, with blue hydrogen, some of the carbon dioxide released by the SMR process is captured. In another version of the blue-hydrogen process, additional carbon dioxide is removed from the flue gases created from burning natural gas to provide the heat and high pressure needed to drive the SMR process. A third set of emissions, not usually captured, is the carbon dioxide and methane from the energy used to produce the electricity for the carbon-capture equipment.

3.1 | How much carbon dioxide is emitted after carbon capture?

As noted above, only two facilities that produce blue hydrogen from natural gas are in commercial operation in 2021. Thus, only limited data are available on the percentage of carbon dioxide that can be captured. For the carbon dioxide generated during SMR, the reported capture efficiencies range from 53% to 90%.[29] Actual data from one of the two commercially operating facilities, the Shell plant in Alberta, show a capture a mean capture efficiency of 78.8%, with daily rates varying from 53% to 90% except for one outlier of 15%.[30] For our baseline analysis, we use a capture rate of 85%, roughly half way between the 78.8% for the Shell plan and the best-case of 90%. Applying 100% minus the capture efficiency to the carbon dioxide produced in SMR:

$$(15\%) * (38.5 \text{ g } CO_2 \text{ per MJ}) = 5.8 \text{ g } CO_2 \text{ per MJ} \quad (11)$$

That is, 5.8 g CO_2 per MJ are emitted from the SMR process after emissions are treated for carbon capture (Table 1).

For the blue-hydrogen facilities so far in commercial operation, carbon capture has focused only on the SMR process, and no attempt has been made to capture the carbon dioxide generated from the combustion of natural gas used to provide the heat and high pressure. If these combustion emissions are captured, the carbon dioxide capture efficiency may be lower than that from the SMR process because the carbon dioxide is more dilute in the former case. We are aware of no data on carbon-capture efficiency from any plant, including any electric power plant, that combusts natural gas, but capture efficiencies of carbon dioxide from the exhaust stream of two coal-burning power plants are reported in the range of 55%-72%.[31-33] Note that efficiencies of up to 90% have been observed in one of the plants when running at full load. However, this does not reflect long-term performance, which is evaluated at average load. Load is less than full load either when the carbon-capture equipment is down for repair or when the demand for carbon dioxide is lower than it is at full load. In this analysis, we use a value of 65% capture efficiency from flue gases for our baseline analysis. Applying 100% minus this factor for emissions from the natural gas burned to produce the heat and pressure:

$$(35\%) * (31.8 \text{ g } CO_2 \text{ per MJ}) = 11.1 \text{ g } CO_2 \text{ per MJ} \quad (12)$$

Therefore, total carbon dioxide emissions from the SMR process, including the energy used to drive the process, are in the range of 16.9 g CO_2 per MJ if the combustion flue is captured (5.8 g CO_2 per MJ plus 11.1 g CO_2 per MJ) to 37.6 g CO_2 per MJ (5.8 g CO_2 per MJ plus 31.8 g CO_2 per MJ) if the flue gases are not treated (Table 1).

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3.2 | Consumption of methane and production of carbon dioxide from electricity used to capture carbon dioxide

Energy is required to capture the carbon dioxide, and often this is provided by electricity generated from burning additional natural gas.[7] The existing blue-hydrogen facilities make no effort to capture the carbon dioxide from the fuel burned to generate this electricity, nor has there been any effort to do so in the case of carbon capture from coal-burning power plants.[31] Often, an energy penalty of 25% is assumed for this additional electricity.[34-36] However, this estimate is based on very little publicly available, verifiable information and may be optimistically low. A recent analysis of carbon capture from the flue gases of a coal-burning power plant, where the electricity for carbon capture came from a dedicated natural gas plant, found that the carbon dioxide emissions from the natural gas were 39% of the carbon dioxide captured from the coal-flue gases.[31] Carbon dioxide is more concentrated in the gases produced through SMR than in the flue exhaust from combustion, suggesting that it can be captured more easily.

For this analysis, we assume that the energy used in the carbon-capture results in carbon dioxide emissions equal to 25% of the carbon dioxide captured from the stream reforming process, based on IPCC,[34] Jacobson,[35] and Sgouridi et al.[36] Therefore,

$$(25\%) * [(38.5 \text{ g } CO_2 \text{ per MJ}) - (5.8 \text{ g } CO_2 \text{ per MJ})] = 8.2 \text{ g } CO_2 \text{ per MJ} \quad (13)$$

That is, emissions from the energy used to drive the carbon captured from the SMR process are themselves an additional 8.2 g CO_2 per MJ (Table 1).

If carbon dioxide is also captured from the flue gases used to generate heat and pressure, we assume the emissions from the energy cost is equal to 39% of the emissions captured, based on Jacobson.[31] That is,

$$(39\%) * [(31.8 \text{ g } CO_2 \text{ per MJ}) - (11.1 \text{ g } CO_2 \text{ per MJ})] = 8.1 \text{ g } CO_2 \text{ per MJ} \quad (14)$$

Therefore, the carbon dioxide emissions from the energy used to drive the carbon capture is between 8.2 g CO_2 per MJ if only emissions from the SMR process are captured or an additional 8.1 g CO_2 per MJ for a total of 16.3 g CO_2 per MJ if emissions from the energy source used for heat and pressure are also captured (Table 1).

As above for Equation 7, one mole of methane is burned for every mole of carbon dioxide emitted from the burning. Therefore, we can estimate the methane burned to produce the electricity required for the carbon dioxide capture as follows, for the case where only the SMR carbon is captured:

$$(8.2 \text{ g } CO_2/\text{MJ}) * (1 \text{ mole } CO_2/44.01 \text{ g } CO_2) * (16.04 \text{ g } CH_4/\text{mole } CH_4) * (1 \text{ mole } CH_4/1 \text{ mole } CO_2) = 3.0 \text{ g } CH_4/\text{MJ} \quad (15)$$

That is, 3.0 g CH_4 per MJ are consumed to generate the electricity used for carbon capture if only the reforming process emissions are captured (Table 1). Similarly, if the emissions from the energy used for the heat and pressure are also captured,

$$(8.1 \text{ g } CO_2/\text{MJ}) * (1 \text{ mole } CO_2/44.01 \text{ g } CO_2) * (16.04 \text{ g } CH_4/\text{mole } CH_4) * (1 \text{ mole } CH_4/1 \text{ mole } CO_2) = 3.0 \text{ g } CH_4/\text{MJ} \quad (16)$$

Therefore, the quantify of methane used to drive carbon capture when the flue gases from the combustion of the gas used to generate heat and pressure for the SMR process are 3.0 g CH_4 per MJ plus 3.0 g CH_4 per MJ, for a total of 6.0 g CH_4 per MJ when carbon capture is applied both to SMR and exhaust flue gases (Table 1).

If we again assume that 3.5% of the natural gas that is consumed is emitted unburned to the atmosphere (as in Equation 9), then for the case where only carbon dioxide emissions from SMR are captured, upstream methane emissions are:

$$(3.5\%) * (3.0 \text{ g } CH_4/\text{MJ}) = 0.11 \text{g } CH_4/\text{MJ} \quad (17)$$

For the case where flue gases are also treated for carbon capture, the upstream methane emissions are:

$$(3.5\%) * (6.0 \text{ g } CH_4/\text{MJ}) = 0.21 \text{ g } CH_4/\text{MJ} \quad (18)$$

Converting these methane emissions to carbon dioxide equivalents:

$$(0.11 \text{ g CH4 per MJ}) * (86 \text{ g } CO_2\text{eq/g } CH_4) = 9.5 \text{ g } CO_2\text{eq per MJ} \quad (19)$$

And

$$(0.21 \text{ g } CH_4\text{per MJ}) * (86 \text{ g } CO_2\text{eq/g } CH_4) = 18 \text{ g } CO_2\text{eq per MJ} \quad (20)$$

Therefore, upstream emissions of unburned methane from the energy used to drive carbon capture are between 9.5 g CO_2eq per MJ if only the SMR carbon is captured and 18 g CO_2eq per MJ if the flue-gas emissions are also captured (Table 1).

3.3 | Total carbon dioxide and methane emissions for blue hydrogen

The total emission of carbon dioxide for the production of blue hydrogen is the sum of the emissions from the SMR process after carbon capture, emissions from the energy used for heat and pressure to drive SMR, emissions from the energy used to power the carbon capture, and the indirect upstream emissions associated with producing and transporting natural gas. The indirect upstream carbon dioxide emissions result from the activity needed to provide the natural gas, and so should be applied as a percentage to the carbon dioxide

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produced from using natural gas, and not simply the carbon dioxide emitted after carbon capture. Using the approach of Howarth et al,[21] this is 7.5% of the carbon dioxide produced in the SMR process plus energy needed to fuel that process as for gray hydrogen (70.3 g CO_2 per MJ) plus the emissions from the energy needed to drive the carbon capture (8.2-16.3 g CO_2 per MJ depending on whether or not the flue gases from the SMR-energy source is captured). Therefore, these indirect upstream carbon dioxide emissions are between 5.9 g CO_2 per MJ and 6.5 g CO_2 per MJ depending on whether or not the flue-gas emissions are captured (Table 1). For the case where only the emissions from the SMR processes are treated for carbon capture, total emissions of carbon dioxide are:

$$(5.8\text{ g CO2 per MJ}) + (31.8\text{ g CO2 per MJ}) + (8.2\text{ g } CO_2\text{ per MJ}) + (5.90\text{ g } CO_2\text{ per MJ}) = 51.7\text{ g } CO_2\text{ per MJ} \quad (21)$$

When the emissions from exhaust flue gases are also treated for carbon capture:

$$(5.8\text{ g } CO_2\text{ per MJ}) + (11.1\text{ g } CO_2\text{ per MJ}) + (16.3\text{ g } CO_2\text{ per MJ}) + (6.5\text{ g } CO_2\text{ per MJ}) = 39.7\text{ g } CO_2\text{ per MJ} \quad (22)$$

To summarize, when only the carbon from the SMR process itself is captured, total emissions of carbon dioxide are 51.7 g CO_2 per MJ. When efforts are also taken to capture the carbon dioxide from the flue exhaust from the energy driving the reforming process, total carbon dioxide emissions are 39.7 g CO_2 per MJ (Table 1). Treating the exhaust flue gases for carbon capture reduces total lifecycle emissions of carbon dioxide by 23%, less than might have been expected. This is due both to a relatively low efficiency for the carbon capture of flue gases[31] and to the increased combustion of natural gas needed to provide the electricity for the carbon capture.

The methane emissions from blue hydrogen are the same as for gray hydrogen, except for those associated with the increased use of energy from natural gas to drive the carbon-capture process. The emissions for gray hydrogen are 77.4 g CO_2eq per MJ. The additional methane emissions from the gas used to drive carbon capture are given in Equations 19 and 20: 9.5 g CO_2eq per MJ when only SMR is treated for carbon capture and 18 g CO_2eq per MJ when the exhaust flue gases are also captured. Therefore, the total upstream methane emissions for the production of blue hydrogen are:

$$(77.4\text{ g } CO_2\text{eq per MJ}) + (9.5\text{ g } CO_2\text{eq per MJ}) = 86.9\text{ g } CO_2\text{eq per MJ} \quad (23)$$

when only emissions from the SMR process are captured (Table 1). When flue gases are also treated, total upstream methane emissions are:

$$(77.4\text{ g } CO_2\text{eq per MJ}) + (18\text{ g } CO_2\text{eq per MJ}) = 95.4\text{ g } CO_2\text{eq per MJ} \quad (24)$$

Total emissions for blue hydrogen when only the SMR process is treated are the sum of the carbon dioxide emissions and the upstream methane emissions:

$$(51.7\text{ g } CO_2\text{per MJ}) + (86.9\text{ g } CO_2\text{eq per J}) = 139\text{ g } CO_2\text{eq per MJ} \quad (25)$$

See Table 1. When the exhaust flue gases are also treated for carbon dioxide capture, total emissions for producing blue hydrogen are:

$$(39.7\text{ g } CO_2\text{per MJ}) + (95.4\text{ g } CO_2\text{eq per MJ}) = 135\text{ g } CO_2\text{eq per MJ} \quad (26)$$

We are aware of no previously published, peer-reviewed analyses on either total carbon dioxide or methane emissions associated with producing blue hydrogen. Several non peer-reviewed reports suggest that it may be possible to reduce carbon dioxide emissions for blue hydrogen by 56% (when only the SMR process is treated) to 90% (when exhaust flue gases are also treated) relative to gray hydrogen.[1,7] However, no data have been presented to support these estimates, and they apparently do not include emissions associated with the energy needed to drive carbon capture. Our results using a full lifecycle assessment show the 56% to 90% assumptions are too optimistic.

In Figure 1, we compare the greenhouse gas footprint of gray hydrogen with blue hydrogen where only the SMR process is captured and with blue hydrogen where carbon capture is also used for the exhaust flue gases. Because of the increased methane emissions from increased use of natural gas when flue gases are treated for carbon capture, total greenhouse gas emissions are only very slightly less than when just the carbon dioxide from the stream reforming process is treated, 135 vs 139 g CO_2eq per MJ. In both cases, total emissions from producing blue hydrogen are only 9% to 12% less than for gray hydrogen, 135 or 139 g CO_2eq per MJ compared with 153 g CO_2eq per MJ. Blue hydrogen is hardly "low emissions." The lower, but nonzero, carbon dioxide emissions from blue hydrogen compared with gray hydrogen are partially offset by the higher methane emissions. We further note that blue hydrogen as a strategy only works to the extent it is possible to store carbon dioxide long term indefinitely into the future without leakage back to the atmosphere.

4 | COMPARISON OF EMISSIONS WITH OTHER FUELS AND SENSITIVITY ANALYSES

4.1 | Emissions for fossil fuels

In Figure 1, we also compare greenhouse gas emissions from gray and blue hydrogen with those for other fuels per unit of energy produced when burned. The carbon dioxide emissions

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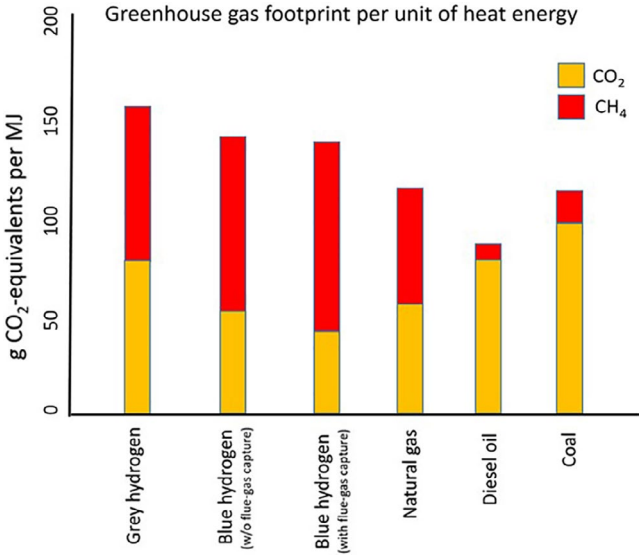

FIGURE 1 Comparison of carbon dioxide equivalent emissions from gray hydrogen, blue hydrogen with carbon dioxide capture from the SMR process but not from the exhaust flue gases created from burning natural gas to run the SMR equipment, blue hydrogen with carbon dioxide capture from both the SMR process and from the exhaust flue gases, natural gas burned for heat generation, diesel oil burned for heat, and coal burned for heat. Carbon dioxide emissions, including emissions from developing, processing, and transporting the fuels, are shown in orange. Carbon dioxide equivalent emissions of fugitive, unburned methane are shown in red. The methane leakage rate is 3.5%. See text for detailed assumptions

shown for coal, diesel oil, and natural gas include both direct and indirect emissions. The direct emissions are based on gross calorific values from EIA.[19] Indirect emissions are those required to develop and process the fuels and are based on Howarth et al.[21] These indirect carbon dioxide emissions are 4 g CO_2 per MJ for coal, 8 g CO_2 per MJ, and 3.8 g CO_2 per MJ for natural gas. Upstream fugitive emissions of unburned methane are assumed to be 3.5% for natural gas, as we have assumed for the hydrogen estimates. Methane emissions for coal and diesel oil are as presented in Howarth[24]: 0.185 g CH_4 per MJ for coal and 0.093 g CH_4 per MJ for diesel oil, corresponding to 8.0 and 15.9 4 g CO_2eq per MJ respectively based on a 20-year GWP of 86.

Combined emissions of carbon dioxide and methane are greater for gray hydrogen and for blue hydrogen (whether or not exhaust flue gases are treated for carbon capture) than for any of the fossil fuels (Figure 1). Methane emissions are a major contributor to this, and methane emissions from both gray and blue hydrogen are larger than for any of the fossil fuels. This reflects the large quantities of natural gas consumed in the production of hydrogen. Carbon dioxide emissions are less from either gray or blue hydrogen than from coal or diesel oil. Carbon dioxide emissions from blue hydrogen are also less than from using natural gas directly as a fuel, but not substantially so. Carbon dioxide emissions from gray hydrogen are somewhat larger than from natural gas (Figure 1).

4.2 | Sensitivity analyses for methane emissions

Given the importance of methane emissions to the greenhouse gas footprints of gray and blue hydrogen, we here present sensitivity analyses on our estimates. We separately consider different rates of fugitive methane emissions and different assigned GWP values.

Our default value for methane emissions used above for gray hydrogen, blue hydrogen, and natural gas is 3.5% of consumption. As noted above, this is based on top–down estimates for emissions from 20 different studies in 10 different gas fields plus a top–down estimate for emissions from gas transport and storage.[16] This is very close to an independent estimate of emissions from shale gas production and consumption estimated from global trends in the ^{13}C stable isotopic composition of methane in the atmosphere since 2005.[37] For the sensitivity analysis, we also evaluate one higher rate and two lower rates of methane emission. The higher rate is from the high-end sensitivity analysis for shale gas emissions based on the global ^{13}C data, or 4.3% of consumption.[37] The lower rates we analyze are 2.54% and 1.45% of consumption. The 2.54% value is based on Alvarez et al[22] who used "bottom–up" approaches to estimate the upstream and midstream methane emissions for natural gas in the United States as 12.7 Tg per year in 2015. This is 2.54% of consumption, based on annual gas consumption for 2015 of 771 billion m^3 of natural gas in the United States,[26] assuming methane comprises 93% of the volume of gas.[38] The bottom–up approach presented by Alvarez et al[22] likely underestimates methane emissions.[24,39,40] We also consider an even lower estimate based on Maasakkers et al.[41] Using an inverse model in combination with satellite data and the US EPA methane emissions inventory, they concluded that methane emissions from natural gas operations in the United States were 8.5 T per year in 2012. This is 1.45% of gas consumption, based on again assuming methane is 93% of gas and a national US consumption of gas of 723 billion m^3 in 2012.[26]

Our baseline analysis is based on a 20-year GWP value of 86.[13] There is uncertainty in this estimate, so here we also explore the higher 20-year GWP value of 105 presented in Shindell et al.[42] Most traditional greenhouse gas inventories use a 100-year GWP, so we explore that as well, using the latest value from the IPCC[13] synthesis report of 34. However, the IPCC[13] noted that the use of a 100-year time period is arbitrary. We prefer the use of 20-year GWP, since it better captures the role of methane as a driver of climate change over the time period of the next several decades, and the 100-year

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time frame discounts the importance of methane over these shorter time frames.[15,24]

In our sensitivity analyses, we substitute emission rates of 4.3%, 2.54%, and 1.54% for our baseline value of 3.5% in Equations 9, 17, and 18 for gray and blue hydrogen and in our estimate for natural gas presented in Figure 1. We also substitute a 20-year GWP value of 105 and a 100-year GWP value of 34 for the 20-year GWP of 86 used in Equations 10, 19, and 20. The sensitivity estimates are shown in Table 2. Across the full set of assumptions, both gray hydrogen and blue hydrogen without flue-gas capture (where only the carbon dioxide from SMR is captured) always have greater emissions than natural gas. The differences between the greenhouse gas footprint of blue hydrogen with or without the capture of carbon dioxide from the exhaust flue gases are generally small across all assumptions concerning fugitive methane emissions, with the total greenhouse gas emissions without the flue-gas treatment usually higher. The emissions from blue hydrogen with full carbon capture including the exhaust flue gases are higher than for natural gas across all set of assumptions except for the analysis with the 100-year GWP of 34 and low methane emissions, 2.54% or less (Table 2).

We also evaluate the sensitivity of our conclusions to the percentage of carbon dioxide that is captured from SMR and from the flue exhaust from the natural gas burned to power the SMR process. Our default values presented above are for 85% capture from the SMR process and 65% capture from the flue gases, if an effort were made to capture those. Our sensitivity analysis includes a low estimate for SMR capture of 78.8% based on actual data from one commercial blue-hydrogen plant[30] and a high estimate of 90%, the highest yet reported.[31] For capture of the flue gases, we explore carbon dioxide capture efficiencies of 55% at the low end and 90% at the high-end based on actual facility performance for flue gases from coal-burning electric plants.[31-33] Note that the 90% rate is the best ever observed and does not reflect likely actual performance under long-term commercial operations. We present the results of this sensitivity analysis in Table 3. Perhaps surprisingly, our conclusions are very insensitive to assumptions about carbon dioxide capture rates. This is because capture is very energy intensive: to capture more carbon dioxide takes more energy, and if this energy comes from natural gas, the emissions of both carbon dioxide and fugitive methane emissions from this increase in such proportion as to offset a significant amount of the reduction in carbon dioxide emission due to the carbon capture.

These sensitivity analyses show that our overall conclusion is robust: the greenhouse gas footprint of blue hydrogen, even with capture of carbon dioxide from exhaust flue gases, is as large as or larger than that of natural gas.

5 | IS THERE A PATH FOR TRULY "GREEN" BLUE HYDROGEN?

Some of the CO_2eq emissions from blue hydrogen are inherent in the extraction, processing, and use of natural gas as the feedstock source of methane for the SMR process: fugitive methane emissions and upstream emissions of carbon dioxide from the energy needed to produce, process, and transport the natural gas that is reformed into hydrogen are inescapable. On the other hand, the emissions of methane and

TABLE 2 Sensitivity analysis for total emissions of carbon dioxide and methane (g CO_2-equivalents per MJ of heat generated in combustion) for different upstream fugitive methane leakage rates and for either 20-year or 100-year global warming potentials (GWP20, GWP100)

	Gray H_2	Blue H_2 (w/o flue-gas capture)	Blue H_2 (w/flue-gas capture)	Natural gas
Fugitive CH_4 = 3.5%				
GWP20 = 8	153	139	135	111
GWP20 = 105	170	158	155	123
GWP100 = 34	106	86	77	76
Fugitive CH_4 = 4.3%				
GWP20 = 86	171	159	156	124
GWP20 = 105	192	182	181	139
GWP100 = 34	113	94	86	81
Fugitive CH_4 = 2.54%				
GWP20 = 86	133	115	109	95
GWP20 = 105	144	129	124	104
GWP100 = 34	98	76	67	70
Fugitive CH_4 = 1.54%				
GWP20 = 86	110	90	82	79
GWP20 = 105	117	98	91	84
GWP100 = 34	89	67	57	64

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TABLE 3 Sensitivity analysis for combined emissions of carbon dioxide and methane (g CO_2-equivalents per MJ of heat generated in combustion) while producing blue hydrogen as a function of the percent carbon dioxide captured from the SMR process and from flue gases for the energy that drives the SMR process

	Total CO_2	Total fugitive CH_4	Total emissions
Blue H_2 w/o flue-gas capture			
85% SMR capture	51.7	86.9	139
90% SMR capture	50.2	86.9	137
78.8% SMR capture	53.5	85.7	139
Blue H_2 w/flue-gas capture			
85% SMR & 65% flue-gas capture	39.7	95.4	135
90% SMR & 90% flue-gas capture	33.3	98.9	132
78.8% SMR & 55% flue-gas capture	43.4	93.2	137

Note: The methane leakage rate is 3.5%. The first row in each case is from the baseline case in Table 1.

carbon dioxide from using natural gas to produce the heat and high pressure needed for SMR and to capture carbon dioxide could be reduced if these processes were instead driven by renewable electricity from wind, solar, or hydro. If we assume essentially zero emissions from the renewable electricity, then carbon dioxide emissions from blue hydrogen could be reduced to the 5.8 g CO_2 per MJ that is not captured from the SMR process (Equation 11) plus the indirect emissions from extracting and processing the natural gas used as feedstock for the SMR process, estimated as 2.9 g CO_2 per M (7.5% of 38.5 g CO_2 per MJ; see section on "total carbon dioxide and methane emissions for gray hydrogen"), for a total of 8.7 g CO_2 per MJ. This is a substantial reduction compared with using natural gas to power the production of blue hydrogen. However, the fugitive methane emissions associated with the natural gas that is reformed to hydrogen would remain if the process is powered by 100% renewable energy. These emissions are substantial: 3.5% of 14 g CH_4 per MJ (Equation 3). Using the 20-year GWP value of 86, these methane emissions equal 43 g CO_2eq per MJ of hydrogen produced. The total greenhouse gas emissions, then, for this scenario of blue hydrogen produced with renewable electricity are 52 g (8.7 g plus 43 g) CO_2eq per MJ. This is not a low-emissions strategy, and emissions would still be 47% of the 111 g CO_2eq per MJ for burning natural gas as a fuel, using the same methane emission estimates and GWP value (Table 1). Seemingly, the renewable electricity would be better used to produce green hydrogen through electrolysis.

This best-case scenario for producing blue hydrogen, using renewable electricity instead of natural gas to power the processes, suggests to us that there really is no role for blue hydrogen in a carbon-free future. Greenhouse gas emissions remain high, and there would also be a substantial consumption of renewable electricity, which represents an opportunity cost. We believe the renewable electricity could be better used by society in other ways, replacing the use of fossil fuels.

Similarly, we see no advantage in using blue hydrogen powered by natural gas compared with simply using the natural gas directly for heat. As we have demonstrated, far from being low emissions, blue hydrogen has emissions as large as or larger than those of natural gas used for heat (Figure 1; Table 1; Table 2). The small reduction in carbon dioxide emissions for blue hydrogen compared with natural gas are more than made up for by the larger emissions of fugitive methane. Society needs to move away from all fossil fuels as quickly as possible, and the truly green hydrogen produced by electrolysis driven by renewable electricity can play a role. Blue hydrogen, though, provides no benefit. We suggest that blue hydrogen is best viewed as a distraction, something than may delay needed action to truly decarbonize the global energy economy, in the same way that has been described for shale gas as a bridge fuel and for carbon capture and storage in general.[43] We further note that much of the push for using hydrogen for energy since 2017 has come from the Hydrogen Council, a group established by the oil and gas industry specifically to promote hydrogen, with a major emphasis on blue hydrogen.[5] From the industry perspective, switching from natural gas to blue hydrogen may be viewed as economically beneficial since even more natural gas is needed to generate the same amount of heat.

We emphasize that our analysis in this paper is a best-case scenario for blue hydrogen. It assumes that the carbon dioxide that is captured can indeed be stored indefinitely for decades and centuries into the future. In fact, there is no experience at commercial scale with storing carbon dioxide from carbon capture, and most carbon dioxide that is currently captured is used for enhanced oil recovery and is released back to the atmosphere.[44] Further, our analysis does not consider the energy cost and associated greenhouse gas emissions from transporting and storing the captured carbon dioxide. Even without these considerations, though, blue hydrogen has large climatic consequences. We see no way that blue hydrogen can be considered "green."

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ORCID

Robert W. Howarth https://orcid.org/0000-0001-9531-4288

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