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Towards net-zero compatible hydrogen from steam reformation – Techno-economic analysis of process design options

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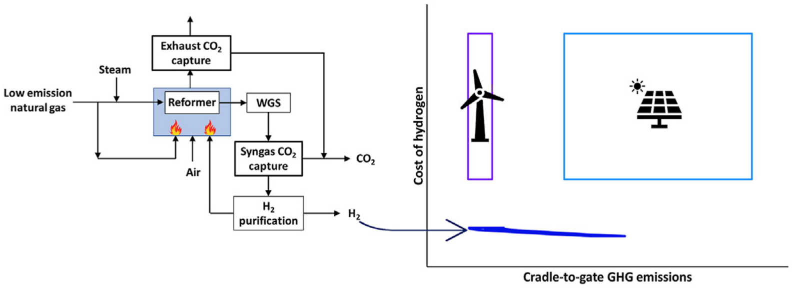
HIGHLIGHTS

- Techno-economic analysis of steam reforming natural gas with 76–99.9% CO_2 capture.
- Impact of process configuration and design choices using existing technology.
- Cradle-to-gate greenhouse gas emission intensity of 0.7–2.7 $kgCO_2e/kgH_2$.
- Levelised cost of hydrogen of US\$1.1–1.3/kgH_2.
- Emission intensity comparable to electrolytic hydrogen at significantly lower cost.

GRAPHICAL ABSTRACT



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ABSTRACT

Increased consumption of low-carbon hydrogen is prominent in the decarbonisation strategies of many jurisdictions. Yet prior studies assessing the current most prevalent production method, steam reformation of natural gas (SRNG), have not sufficiently evaluated how process design decisions affect life cycle greenhouse gas (GHG) emissions. This techno-economic case study assesses cradle-to-gate emissions of hydrogen produced from SRNG with CO_2 capture and storage (CCS) in British Columbia, Canada. Four process configurations with amine-based CCS using existing technology and novel process designs are evaluated. We find that cradle-to-gate GHG emission intensity ranges from 0.7 to 2.7 $kgCO_2e/kgH_2$ – significantly lower than previous studies of SRNG with CCS and similar to the range of published estimates for hydrogen produced from renewable-powered

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Blue hydrogen
CCS

electrolysis. The levelized cost of hydrogen (LCOH) in this study (US\$1.1–1.3/kgH_2) is significantly lower than published estimates for renewable-powered electrolysis.

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Introduction

Urgent and widespread decarbonisation of the world energy supply is required to limit global warming in accordance with the Paris Agreement [1] – a massive undertaking that will require rapid growth in low-carbon energy production [2]. Meanwhile, governments have many considerations in developing plans to mitigate global warming such as employment, public support, fiscal limitations, and energy security [3]. Decarbonisation strategies commonly include hydrogen as a low-carbon energy carrier and feedstock, and hydrogen consumption is expected to increase significantly by 2050 [2]. However, almost all existing hydrogen production is derived from fossil fuels using processes that have significant GHG emissions (900 MtCO_2 in 2020), with SRNG being the most common process [4].

Numerous studies have evaluated life cycle GHG emissions of hydrogen produced from renewable energy ("green hydrogen") [5], but green hydrogen production is currently much more expensive than conventional processes (US\$3–8/kgH_2 vs. US\$1–2/kgH_2) [4,6]. Furthermore, fossil fuels are expected to provide 66% of world energy supply in 2050 based on current policies [2]. Yet relatively few studies have thoroughly investigated life cycle GHG emissions of hydrogen produced from fossil fuels utilising CCS [7]. Policymakers and prospective hydrogen suppliers could benefit from an improved understanding of opportunities to reduce cradle-to-gate GHG emissions of hydrogen from SRNG.

SRNG produces hydrogen (H_2), carbon monoxide (CO), and CO_2 by catalytically reacting NG with steam at elevated temperature (c. 730–950 °C) and pressure (c. 1800–3000 kPa) [8,9]. NG is predominantly (c. 75–99%) methane (CH_4), but also contains heavier hydrocarbons and trace impurities [10]. SRNG typically includes (Fig. 1a): pre-treatment of the NG feedstock to remove sulphur impurities which deactivate downstream catalysts, pre-reforming to decompose hydrocarbons heavier than methane to reduce carbon formation in the primary reformer, methane reforming according to reactions (1) and (2), water gas shift (WGS) according to reaction (2), and hydrogen purification [7–9,11]. Supplementary Note 2 has a detailed process description.

$$CH_4 + H_2O \rightleftharpoons CO + 3H_2 \qquad \Delta H^0_{298} = 206\ kJ/mol \tag{1}$$

$$CO + H_2O \rightleftharpoons CO_2 + H_2 \qquad \Delta H^0_{298} = -41.1\ kJ/mol \tag{2}$$

WGS can be configured with separate high-temperature (HT) and low-temperature (LT) stages to increase hydrogen yield [8]. Active cooling has also been used in WGS reactors to achieve near isothermal conditions and increase conversion to CO_2 [12]. Pressure swing adsorption is the most common purification technique and provides near 100% pure hydrogen [8]. Since the overall reaction is endothermic and requires steam as an input, the residual gas from the purification process and additional NG is typically combusted as a heat source [7].

Conventional SRNG has direct CO_2 emissions of approximately 9 kgCO_2/kgH_2 [9,10]. However, commercially available technologies such as amine absorbents could capture CO_2 from the process [13]. Monoethanolamine (MEA), a primary amine, has very high reactivity with CO_2, while tertiary amines, such as methyldiethanolamine (MDEA), require less energy to regenerate [13,14]. MDEA alone has low reactivity with CO_2 but can be used with chemical promoters (e.g., piperazine) to achieve very low residual CO_2 concentration – e.g., 50 ppmv at NG liquefaction facilities [15]. Amines are typically employed in a regenerative cycle (Fig. 1b) with CO_2 absorbed from the gas stream into the amine solution in a contactor tower at low temperature (c. 40–70 °C) and then liberated from the amine solution in a regenerator tower at higher temperature (c. 100–130 °C) [13,14].

CO_2 capture from SRNG is often considered between WGS and hydrogen purification ("syngas capture") because this location has the highest CO_2 partial pressure and avoids issues with contaminants, dilution, and low pressure that affect CO_2 capture from the burner exhaust stream ("exhaust capture") [16]. However, syngas capture is unable to mitigate the CO_2 produced downstream in the burner (c. 42% in conventional SRNG) [7,9].

Numerous studies have assessed cradle-to-gate GHG emissions and cost of producing hydrogen from SRNG with CCS. Salkuyeh et al. showed higher cradle-to-gate GHG emissions and production cost for SRNG (3.5 kgCO_2e/kgH_2 and US\$2.16/kgH_2) compared to autothermal reforming and two chemical looping processes [17]. Their study was based on average Canadian NG supply, assumed process design parameters, and 90% CO_2 capture rate from SRNG. Timmerberg et al. assumed parametric factors from prior studies in a techno-economic comparison of SRNG with electrolysis and thermal decomposition of NG [6]. Their results showed cradle-to-gate GHG emissions are strongly dependent on NG supply chain emission intensity – a finding confirmed by subsequent studies [10,18]. Khan et al. also assumed SRNG process parameters from a prior study in their economic comparison of CCS with CO_2 utilisation [19].

Oni et al. evaluated processes for producing hydrogen from NG in Alberta, Canada based on assumed design parameters, 52–85% CO_2 capture rate from SRNG, and average NG supply chain emissions for Alberta [20]. Relatively high cradle-to-gate GHG emissions and hydrogen production cost for SRNG with CCS in their study (6.7–8.2 kgCO_2e/kgH_2 and Can\$1.69–2.36/kgH_2) can be attributed to high steam-carbon ratio compared to other studies (c. 5 vs. 2.7 typical), their assumption that the burner flue gas is compressed to 3500 kPa upstream of exhaust

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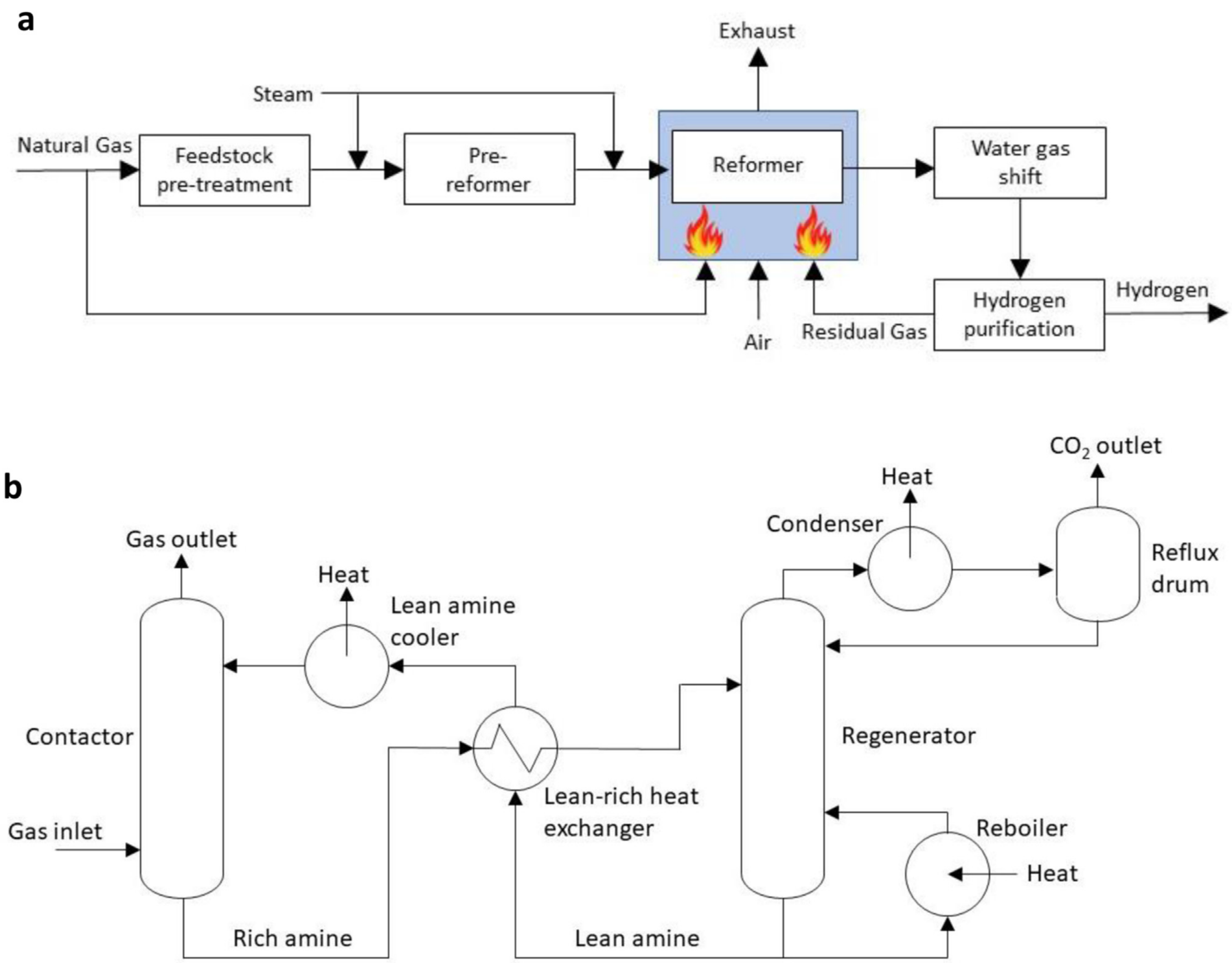

Fig. 1 – General process schematics. a, conventional SRNG process. b, regenerative amine-based CO_2 capture process. Contactor and regenerator towers contain internals (e.g., packing) to promote energy and mass transfer between countercurrent flow of liquid and vapour.

capture, and high emission intensity electricity (544 gCO_2e/kWh).

Antonini et al. assumed process parameters from conventional SRNG and average European NG supply in their LCA which considered 8 environmental impact categories [7]. Since their analysis only considered syngas capture, cradle-to-gate GHG emissions for SRNG with CCS were relatively high (4.6 $kgCO_2e/kgH_2$) while the other environmental impact categories were found to increase slightly compared to SRNG without CCS.

Prior research assessing life cycle GHG emissions from SRNG with CCS has either used simplified calculations or assumed design parameters from conventional SRNG plants (refs. [6,7,9,10,16–18,20–23]). IEAGHG considered combusting decarbonised syngas (primarily hydrogen) as the heat source; however, they only considered direct CO_2 emissions and single-stage HT WGS led to higher emissions than could have been achieved with LT WGS [9]. Pruvost et al. showed that higher steam-carbon ratio than conventional SRNG could significantly reduce direct CO_2 emissions for SRNG with syngas capture at the expense of lower process efficiency and higher cost [24]. No studies, to the knowledge of the authors, have optimised process parameters to reduce life cycle GHG emissions or assessed the effect of isothermal WGS on SRNG life cycle GHG emissions.

Prior studies (refs. [6,7,9,10,16–18,21–23]) have also usually assumed CO_2 capture rates markedly lower than is possible with promoted MDEA for syngas capture (c. 100%) [15] or MEA for exhaust capture (>99%) [25–27]. Exhaust capture rates near 100% have been demonstrated using MEA in large-scale pilot testing (126 tCO_2/d) [28]. Most studies did not consider combining syngas capture and exhaust capture (refs. [6,7,9,10,16,18,20,21]); however, this has been identified as a promising alternative to reduce the cost of achieving high CO_2 capture rate for SRNG [24].

Although cradle-to-gate GHG emissions of SRNG with CCS are strongly dependent on the NG supply chain emission intensity, a wide range of assumptions have been made in prior studies of SRNG based on national or international averages (refs. [6,7,10,17,18,22,23]) rather than production practices implemented to reduce emissions. Fugitive methane can significantly increase supply chain emissions and may be

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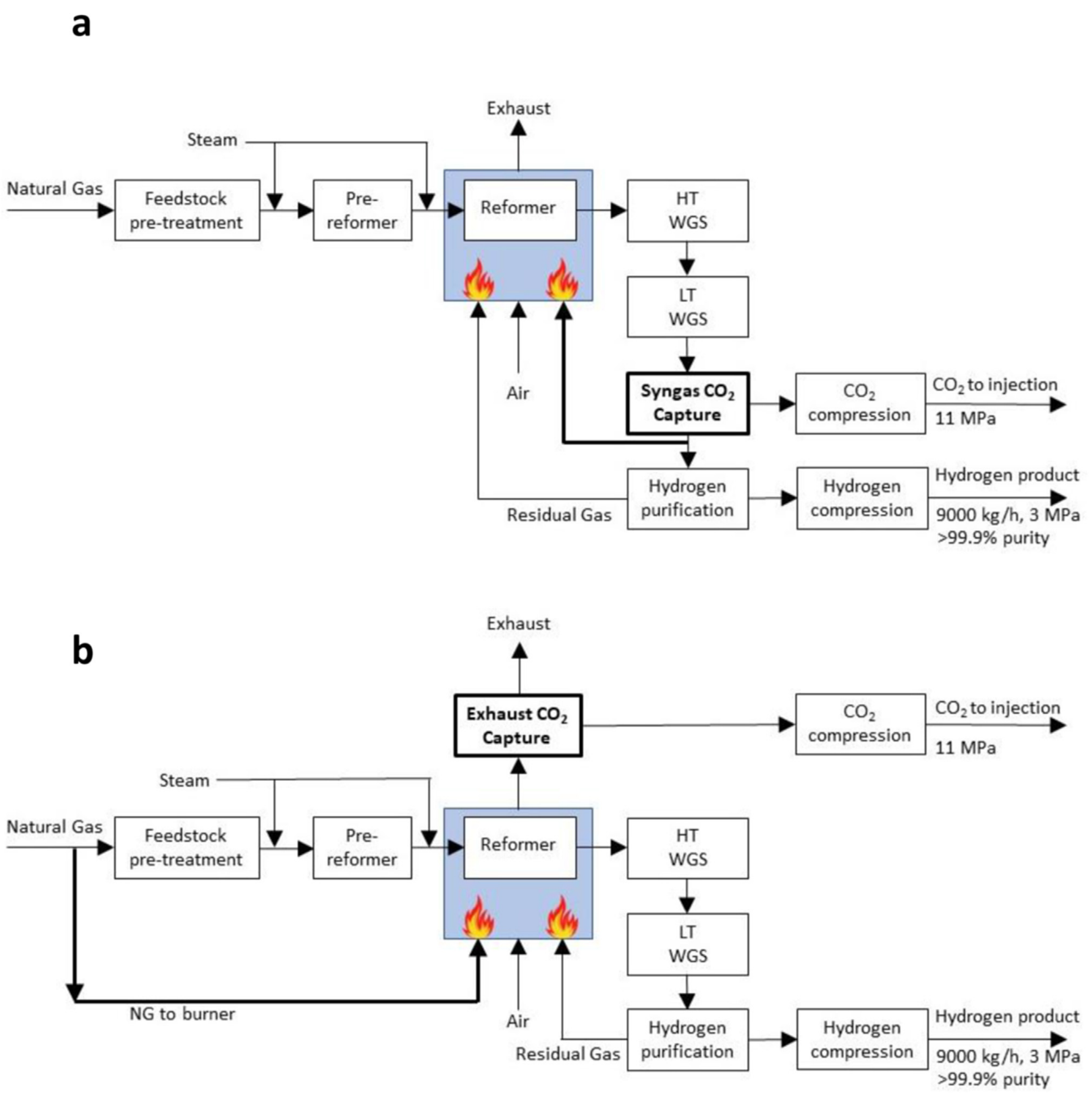

Fig. 2 – Schematics of SRNG configurations assessed in this study. a, Syngas-only CO_2 capture and syngas fueled burner. b, Exhaust-only CO_2 capture and NG fueled burner. c, Syngas-and-exhaust CO_2 capture and NG fueled burner. d, Syngas-and-exhaust CO_2 capture and syngas fueled burner.

higher than previously estimated in some jurisdictions [29]. However, leakage rates differ substantially between countries (c. 0–6%) [18], and NG-supply emission factors can vary by an order of magnitude (e.g., 3.4–32 $kgCO_2e/MJ_{LHV}$ in Ref. [10]).

British Columbia (BC) has regulations to limit methane emissions from NG production [30]. BC reported 100-year global warming potential (GWP100) emissions intensity of 5.4 gCO_2e/MJ_{LHV} for NG production and 0.61 gCO_2e/MJ_{LHV} for transmission (based on energy allocation, Supplementary Note 1) [31,32]. Similarly, Seven Generations Energy (7 GE) implemented production practices to achieve cradle-to-gate GWP100 emissions intensity of 3.1 gCO_2e/MJ_{LHV} for NG production and 1.3 gCO_2e/MJ_{LHV} for transmission from its 2016 Montney field production in Alberta, Canada [30]. Methane emission GWP100 CO_2 equivalence has been adjusted to 30 for comparability where required.

Studies assessing methane emissions from BC NG production have reported conflicting results. Tyner and Johnson [33] extrapolated measurements from 167 facilities (1.3% sample) in 2019 to estimate province-wide methane emissions 80% higher than reported. However, their data included one

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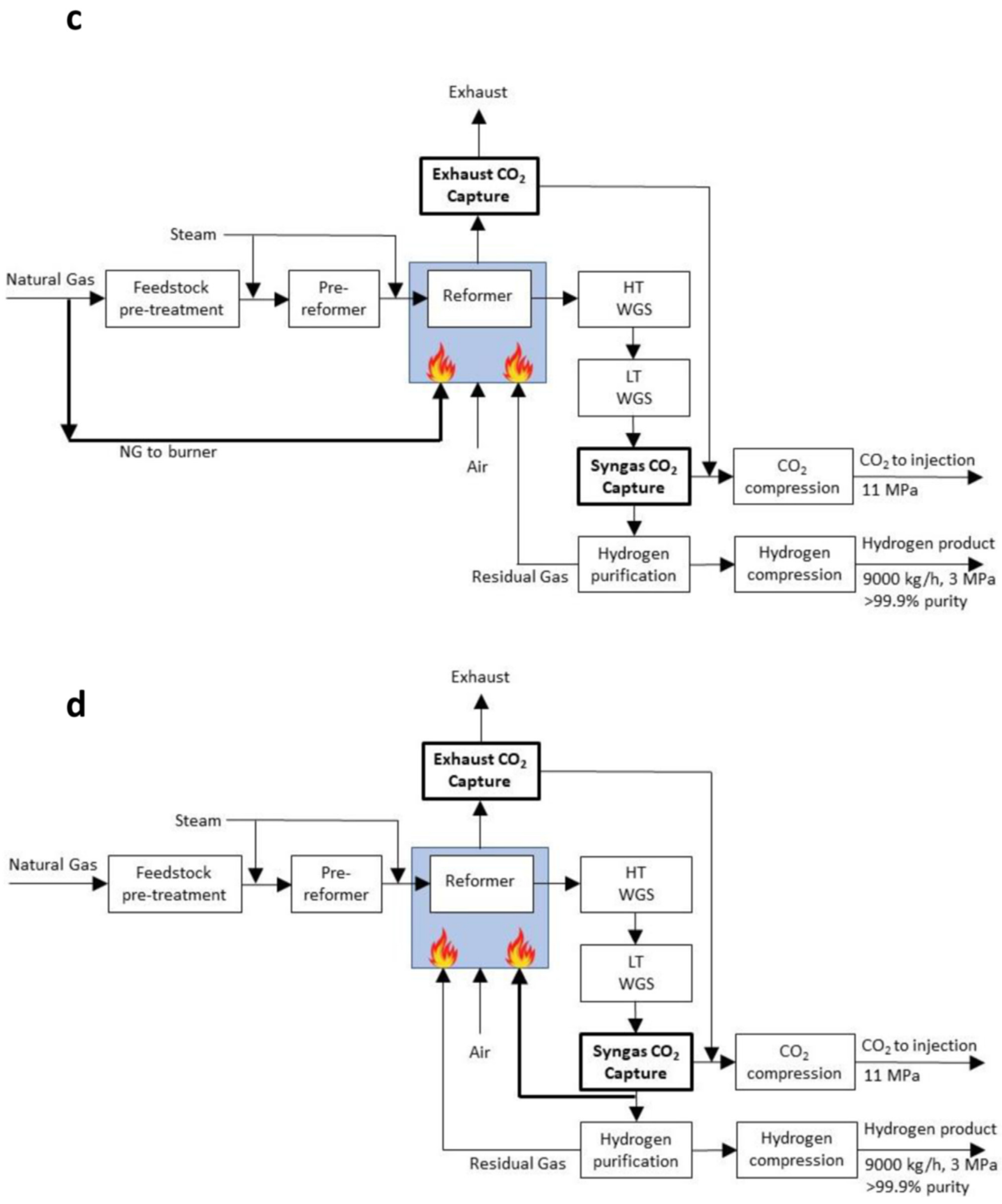

Fig. 2 — ***Continued***

"super-emitter" multi-well battery that accounted for 52% of measured emissions – 95 times higher than the average of the other 56 multi-well batteries surveyed. Methane emission studies in other jurisdictions have shown similar positively skewed distributions and cautioned against extrapolation from small samples [34,35]. MacKay et al. [36] found methane emission intensity varied substantially between different fields in western Canada (6650 sites between 2015 and 2018), with the Montney field in BC (1.0 gCO_2e/MJ_{LHV}) lower than 7 GE (1.2 gCO_2e/MJ_{LHV}) [30].

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Contribution and aims of this study

We combine detailed process modelling with an integrated life cycle assessment (LCA) and cost model to show that process design choices dramatically affect cradle-to-gate emissions intensity of hydrogen production from SRNG. We assess four combinations of CO_2 capture and burner fuel (Fig. 2).

- syngas-only capture with decarbonised syngas as fuel
- exhaust-only capture with NG as fuel
- syngas-and-exhaust capture with NG as fuel
- syngas-and-exhaust capture with decarbonised syngas as fuel

We find that existing SRNG technologies, combined with existing low-carbon electricity supply and low-emission NG production practices in western Canada (see Methods), could be configured to produce hydrogen with cradle-to-gate GHG emission intensity comparable to published estimates of wind-powered electrolysis but with significantly lower cost. Furthermore, we find that discrepancies between reported and measured methane emission rates from BC NG production affect SRNG cradle-to-gate emissions much less significantly than process design choices.

Methods

This techno-economic case study evaluated construction of a new industrial-scale SRNG plant (9000 kg/h hydrogen product). The plant size was similar to prior studies and cost data were publicly available for the technologies used [6,9,17]. BC was selected as the plant location due to established regulations to limit methane emissions from NG production [37], recent peer-reviewed studies of nearby NG production emissions [30,38], and availability of grid-supplied low-carbon electricity for industrial activity [39]. The analysis was completed by linking the output from detailed process simulations to an integrated LCA and cost model.

Process model

The SRNG mass and energy balance was modelled using UniSim process simulation software [40]. Similar software programmes were used in prior studies of SRNG which included detailed process modelling (Supplementary Note 2).

In all cases in this study, the syngas capture absorbent was MDEA promoted with piperazine, and the exhaust capture absorbent was MEA. MDEA/piperazine is commonly used for deep CO_2 removal in NG processing [15] and exhaust capture with MEA is common in prior studies and pilot testing (e.g., Ref. [41]). Many other potential absorbents and blends are possible [13] but were not included in the scope of this study.

Energy was recovered from the burner exhaust stream to provide steam for the process, pre-heat combustion air, and regenerate the amine(s). The feed rate of NG was adjusted in each case to achieve the desired rate of hydrogen product and the burner fuel flow rate was adjusted to maintain adequate energy to sustain the process. Electric drives were assumed for all compressors, pumps, and blowers to minimise cradle-to-gate emissions. Electricity consumption, water requirements, and direct CO_2 emissions were calculated by the process model. Electricity was assumed to be supplied from the BC grid.

Key process design parameters were identified and adjusted within typical ranges based on existing literature [8,13,42] to determine the impact on cradle-to-gate GHG emissions intensity and cost of producing hydrogen with each configuration.

- molar ratio of water to carbon atoms in the reformer inlet ("steam-carbon ratio"),
- primary reformer equilibrium temperature and pressure,
- molar ratio of CO_2 to amine in the absorbent solution entering the contactor tower ("lean amine loading", adjusted by regenerator reboiler temperature and pressure),
- molar ratio of CO_2 to amine in the absorbent solution leaving the contactor tower ("rich amine loading", adjusted by amine circulation rate),
- number of theoretical absorber stages (equilibrium flash calculations between liquid and vapour phases, used to model effect of absorber height), and
- amine concentration.

In addition to cradle-to-gate GHG emissions intensity and cost of producing hydrogen, performance of the configurations was compared based on process efficiency and electricity consumption per unit of hydrogen produced. Process efficiency is defined as energy content of hydrogen product divided by total NG supply on LHV basis.

Supplementary Note 2 provides a detailed description of the process design and assumptions made for other process design parameters. The process simulation framework used in this study was validated by modelling case 1B (syngas-only capture) from Ref. [9] and comparing the results with that study (see Supplementary Note 3 for details). The model results matched very closely except for mole fraction of hydrogen in the amine regenerator vapour outlet stream. While the relative difference was very high, it was not deemed significant to the overall system mass-energy balance as it represented less than 0.1% of the hydrogen passing through the absorber. Experimental data quantifying the solubility of hydrogen in MDEA solutions were not available to check the model predictions against, and further studies are recommended to quantify hydrogen absorption in MDEA, piperazine, and other prospective syngas CO_2 absorbents as data were only available for MEA [43].

LCA model

Cradle-to-gate GHG emissions for produced hydrogen (functional unit of 1 kgH_2 at 3 MPa and >99.9% purity) were calculated for the system boundary (Fig. 3) in accordance with ISO 14067 [44] using a hybrid attributional LCA model which combined process-based emission factors with environmentally extended input-output (EEIO) emission factors. LCA is the most common methodology used to evaluate potential environmental effects of hydrogen energy systems [5] and has been used previously to assess SRNG (e.g., Ref. [18]). A hybrid

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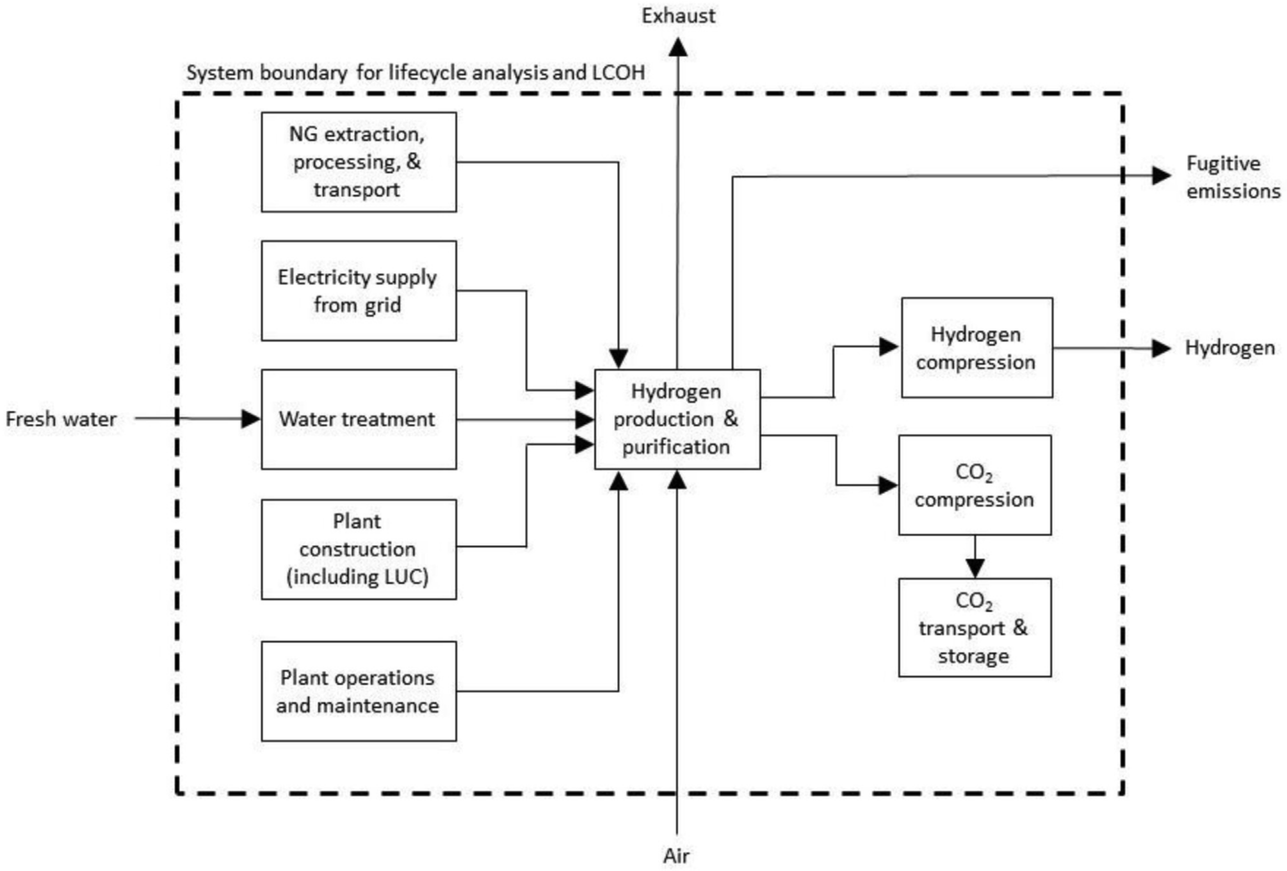

Fig. 3 – Schematic of system boundary for the LCA model in this study.

attributional approach has been used for LCA of CO_2 capture at a refinery [45] and facilitates inclusion of indirect emissions associated with expenses (e.g., labour) not captured by material estimates used in other SRNG studies (e.g., Ref. [10]). There are numerous approaches available to assess the combined climate impact of different GHGs, each with individual attributes and limitations [46]. This study used GWP100 as the baseline for comparability with prior studies; however, recent research suggests that GWP may overestimate warming caused by methane emissions if the global emission rate is constant or falling [47,48]. The impact of GWP20 (20-year time horizon) characterisation factors was also assessed.

Three different GHG emission factors for NG supply were evaluated based on published literature and government data (Supplementary Note 1): 7 GE production practices [30] (3.1 gCO_2e/MJ), average BC production [31,32] (5.6 gCO_2e/MJ), and average BC production with reported methane emissions increased by 80% (6.6 gCO_2e/MJ) based on the findings of Tyner and Johnson [33]. 7 GE production emission intensity was used as the baseline for evaluating the impact of process design parameters as representative of low-emission production practices that have been implemented at industrial scale in western Canada [30]. The SRNG plant was assumed to be co-located with upstream NG processing to eliminate emissions associated with NG transmission and utilise nearby geologic reservoirs for CO_2 sequestration [49].

Life cycle emissions associated with electricity, process water, absorption solvents, CO_2 sequestration, fugitive emissions, and land use change (LUC) were based on process data and assumed emission factors (Table 1). A 10-year average of the BC electricity grid emission factor was used to minimise the effect of annual fluctuations in hydroelectric generation. Refer to Supplementary Note 1 for further details/rationale and corresponding GWP20 emission factors. Further study is required to accurately quantify the climate effects of fugitive hydrogen [50] and nitrogen oxide pollution [51] so these were not considered in this study.

Life cycle emissions associated with SRNG plant construction, operating labour, and plant maintenance were calculated using US EEIO emission factors [57] based on the capital and operating expenses estimated from the cost model for each case. Comparable data for Canada were not available and US data were believed to be a reasonable proxy given the integrated supply chains and substantial bilateral trade shared between the countries [58]. Calculation details and emission factors are presented in Supplementary Note 1.

Cost model

Similar to Refs. [6,19], LCOH was calculated based on discounted cash flow and hydrogen production (H) using equation (3):

Table 1 – Process data and emission factors used for the LCA model in this study.

Emission source	Process data	GWP100 Emission factor
Electricity	Process model	34.2 $kgCO_2e/MWh$ [52]
Softened water	Process model	3.42E-5 $kgCO_2e/kgWater$ [53]
MEA	0.3 $kgMEA/tCO_2$ [41]	2.82 $kgCO_2e/kgMEA$ [53]
MDEA and piperazine	Process model (vapourisation) plus 7.54E-3 $kgSolvent/tCO_2$ (thermal degradation) [54]	3.08 $kgCO_2e/kgMDEA$ 9.11 $kgCO_2e/kgPiperazine$ [53,55]
CO_2 disposal	Two wells, 2370 m deep (Supplementary Note 1)	2063 $kgCO_2/m$ [53]
CO_2 pipeline	20 km pipeline (Supplementary Note 1)	1165 $kgCO_2/m$ [53]
SRNG fugitive	0.264 g/m^3 (Supplementary Note 1)	Composition from process model using IPCC AR5 GWP100 characterisation [56]
LUC	30 ha (Supplementary Note 1)	407 tCO_2e/ha [38]

$$\mathrm{LCOH} = \frac{\sum_{n=1}^{N}\left\{\frac{C_n+O_n}{(1+r)^n}\right\}}{\sum_{n=1}^{N}\left\{\frac{H_n}{(1+r)^n}\right\}} \tag{3}$$

where C is capital costs, O is operating and maintenance costs, r is the discount rate, and N is life of the plant (3 years construction plus 25 years operation). The baseline discount rate was 7% – the real discount rate used for cost-benefit analysis of Canadian regulations which reflects the weighted average cost of investment funds in Canada [59]. The discount rate in this study was the same as Khan et al. [19], lower than IEAGHG [9] (8%), but higher than Timmerberg et al. [6] (5%). Sensitivity of LCOH to discount rate was investigated (range 3–11%). The social discount rate used to assess Canadian regulations, which considers factors other than economic opportunity cost, is 3% [59]. Cost of capital during construction was included in capital cost estimates at the discount rate. Other financial assumptions are detailed in Supplementary Note 4.

Capital cost estimates for each case were adapted from IEAGHG [9] by scaling the cost estimates for SRNG subsystems by individual power factor models (Supplementary Note 4) – an approach commonly used in process engineering to develop approximate cost estimates for plants based on capacity [60]. Similar to Khan et al. [19], the estimates were updated to a base year of 2020 using the Chemical Engineering Plant Cost Index [61]. All capital and operating costs were converted to US$ using applicable purchasing power parity (PPP) exchange rates [62]. The approaches used for scaling and inflation adjustment were validated by comparing prior published estimates between cases/studies as described in Supplementary Note 4.

The IEAGHG [9] capital cost estimates are frequently cited (e.g., Refs. [6,19]) as publicly available information on recent capital costs for SRNG plants is very limited. However, there are some limitations with this data including original accuracy (+35%/-15%), base year (2014), location (Netherlands), and differences in process design (see Supplementary Note 4 for more detailed discussion). Thus, capital cost estimates in this study should be considered only for their intended purpose – directional assessment of SRNG process configurations and design parameters and high-level comparisons with other technologies.

The baseline scenario for NG cost was US\$2.04/GJ_{HHV} – the 10-year average (2011–2020) AECO-C price [63] – and impacts of 50% and 100% increases from the baseline were assessed. AECO-C is a virtual pricing point used for NG trading in western Canada. The baseline NG price used in this study was lower than Salkuyeh et al. [17] (US\$2.65/GJ_{HHV}, also based in Canada) and near the low end of the range considered in other studies based on jurisdictions with much higher NG prices [6,19]. The cost of electricity supply was based on the standard rate schedule for industrial customers of BC Hydro [64]. Other operating and maintenance expenses were estimated based on IEAGHG [9] as described in Supplementary Note 4.

Abatement cost per unit of reduced GHG emissions is a common metric for comparing policy or investment options [65]. Average abatement cost was calculated for each case relative to two baselines: SRNG without CCS and substitution of hydrogen for NG consumption. Marginal abatement cost (MAC) between SRNG process cases in this study was also calculated to assess the cost of incremental process changes to reduce GHG emissions. Similar to Timmerberg et al. [6], average and marginal abatement costs were calculated based on the change in LCOH using equation (4):

$$\mathit{Abatement\ cost} = \frac{\Delta\mathrm{LCOH}}{\Delta\mathrm{GHG}} \tag{4}$$

where ΔGHG is the change is life cycle emissions per unit of hydrogen between two cases. Abatement cost was calculated based on the change in life cycle emissions rather than amount of CO_2 captured because additional emissions are typically created in the process of capturing CO_2 [66]. In the case of NG substitution, the reference LCOH was the NG cost (US\$/GJ_{HHV}) converted to hydrogen mass equivalent (US\$/$kg_{eq}H_2$) on an energy (HHV) basis and the baseline emissions were cradle-to-gate NG production plus unabated combustion of NG to CO_2.

Impact of process design parameters

Direct emissions with syngas-only capture are strongly dependent on reformer design conditions – higher reformer equilibrium temperature, lower pressure, and higher steam-carbon ratio reduce direct emissions by increasing conversion of methane to CO_2 upstream of syngas capture (Supplementary Fig. 6). However, increased indirect emissions, primarily due to higher NG consumption, partially offset reduced direct emissions. LCOH is strongly dependent on steam-carbon ratio and moderately dependent on reformer equilibrium conditions resulting in a wide range of potential

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cradle-to-gate emissions (0.99–2.7 $kgCO_2e/kgH_2$) and associated LCOH (US\$1.06–1.45/kgH_2) (Fig. 4a).

For syngas-only capture, increasing theoretical absorption stages up to 30 reduces cradle-to-gate emissions without significantly impacting LCOH (Supplementary Fig. 8). 30 stages capture 99.8% of the CO_2 from the syngas so further increases have little effect on cradle-to-gate emissions. Amine loading and MDEA/piperazine concentration do not significantly affect cradle-to-gate emissions or LCOH within a significant range around the baseline design assumptions (Supplementary Figs. 9–12).

Exhaust rich amine loading and number of theoretical exhaust absorption stages have the most significant effect on cradle-to-gate emissions for the three configurations which include exhaust capture (Supplementary Figs. 13 and 19). While more theoretical stages and lower rich amine loading (increased MEA circulation rate) reduce direct emissions, the effects of increased equipment size and reduced process efficiency lead to a minimum achievable cradle-to-gate emissions intensity of 0.79 and 0.71 $kgCO_2e/kgH_2$ for exhaust-only capture and process-and-exhaust capture respectively.

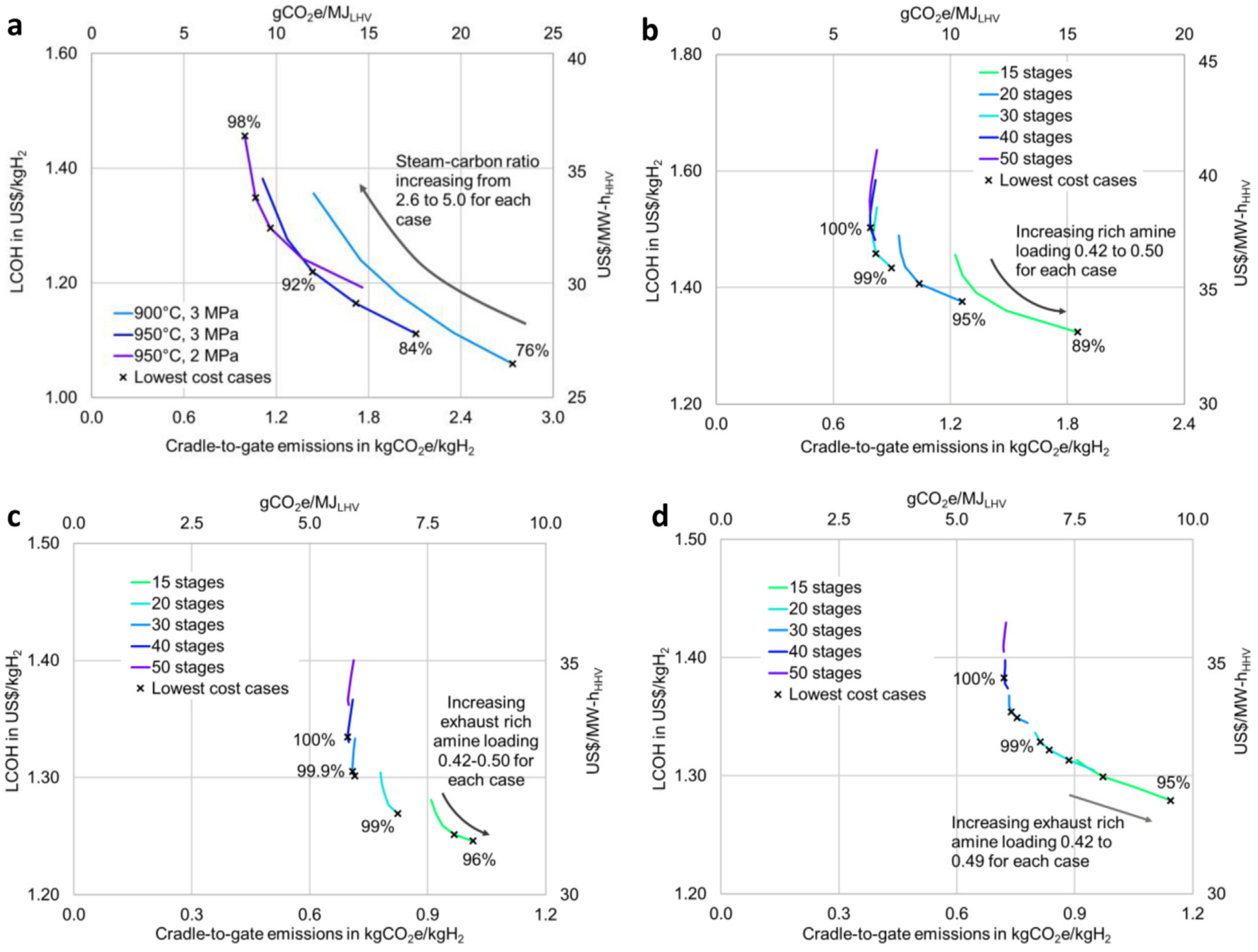

Fig. 4 – Impact of process design conditions on LCOH and cradle-to-gate GHG emissions. "Lowest cost cases" represent the progression from minimum LCOH to minimum cradle-to-gate emissions (GWP100) with the lowest MAC between each step. Overall fossil-CO_2 capture rates (%) of selected points labelled. Upstream NG supply based on 7 GE scenario. a, Impact of reformer equilibrium conditions for syngas-only capture based on 46%wt MDEA, 4%wt piperazine, 30 theoretical absorption stages, 0.06 lean loading, and 0.50 rich loading. b, Impact of rich amine loading and number of theoretical absorption stages for exhaust-only capture based on 30%wt MEA, 0.26 lean amine loading, 2.6 steam-carbon ratio, and 900 °C/3 MPa reformer equilibrium. c, Impact of exhaust rich amine loading and number of theoretical exhaust absorption stages for syngas-and-exhaust capture with NG burner. Based on 30%wt MEA, 0.21 exhaust lean amine loading, 2.6 steam-carbon ratio, and 950 °C/3 MPa reformer equilibrium. Syngas absorption with 46%wt MDEA, 4%wt piperazine, 0.06 lean amine loading, 0.50 rich amine loading, and 30 theoretical stages. d, Impact of exhaust rich amine loading and number of theoretical exhaust absorption stages for syngas-and-exhaust capture with syngas burner. Based on 30%wt MEA, 0.20 exhaust lean amine loading, 2.6 steam-carbon ratio, and 950 °C/3 MPa reformer equilibrium. Syngas absorption with 46%wt MDEA, 4%wt piperazine, 0.06 lean amine loading, 0.50 rich amine loading, and 30 theoretical stages.

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Exhaust lean amine loading between 0.24 and 0.29 (0.26 baseline) results in similar cradle-to-gate emissions for exhaust-only capture, while emissions for the process-and-exhaust capture configurations are minimised at slightly lower exhaust lean amine loading (0.21/0.20 for NG/syngas burner) due to lower CO_2 concentration in the exhaust stream (Supplementary Figs. 15 and 20). Amine intercooling (40 °C) at the midpoint of the exhaust absorption tower reduces cradle-to-gate emissions for all configurations including exhaust capture.

Cradle-to-gate emissions for configurations with exhaust capture are relatively insensitive to reformer equilibrium conditions but positively correlated with steam-carbon ratio due to higher NG consumption. Similarly, cradle-to-gate emissions and LCOH are both lower with MEA concentration of 30%wt compared to 25%wt due to reduced regeneration energy (Supplementary Fig. 14). MEA concentrations higher than 30%wt were not investigated because that was the upper limit of the recommended range for the available amine property package [40]; however, it is anticipated that combinations of higher MEA concentration and lower lean amine loading than those evaluated in this study could provide lower LCOH and cradle-to-gate emissions for configurations including exhaust capture.

The lowest LCOH for each number of theoretical exhaust absorption stages is with exhaust rich amine loading of 0.5 (Fig. 4b, c, 4d). Emissions intensity increases significantly for higher rich amine loading because 0.5 is the stoichiometric limit for the predominant reaction between CO_2 and MEA [14].

Comparing process configurations

Syngas-only capture has the lowest LCOH for emission intensity >1.3 $kgCO_2e/kgH_2$ (US$1.06–1.22/$kgH_2$ for 2.7–1.4 $kgCO_2e/kgH_2$), but below this threshold syngas-and-exhaust capture achieves lower LCOH at lower cradle-to-gate emissions (US$1.25–1.33/$kgH_2$ for 0.97–0.71 $kgCO_2e/kgH_2$) (Fig. 5a). Syngas-and-exhaust capture provides lower LCOH than exhaust-only capture as the second absorption system is justified by less absorbent regeneration energy and reduced size of the exhaust absorption system. Using syngas rather than NG as burner fuel with the syngas-and-exhaust capture configuration increases LCOH approximately US$0.05/$kgH_2$ as reduced amine regeneration energy is offset by increased energy for syngas production and overall capital is 5–8% higher. LCOH is sensitive to both NG cost and discount rate, but the relative positioning of LCOH curves for different process configurations is not materially impacted (Supplementary Note 5).

Utilising BC average NG emission intensity increases cradle-to-gate emissions by 0.40–0.51 $kgCO_2e/kgH_2$ compared to 7 GE low-emission production practices over the range of lowest cost cases for all configurations (Fig. 5b). Assuming BC average methane emissions 80% higher than reported increases cradle-to-gate emissions by an additional 0.15–0.20 $kgCO_2e/kgH_2$ (Fig. 5c). The effect of GWP20 characterisation factors was assessed in the sensitivity studies (Supplementary Note 5).

Syngas-only capture with 900 °C/3 MPa reformer equilibrium and 2.6 steam-carbon ratio (US$1.06/$kgH_2$, 2.74 $kgCO_2e/kgH_2$) has an average abatement cost of US$48/$tCO_2e$

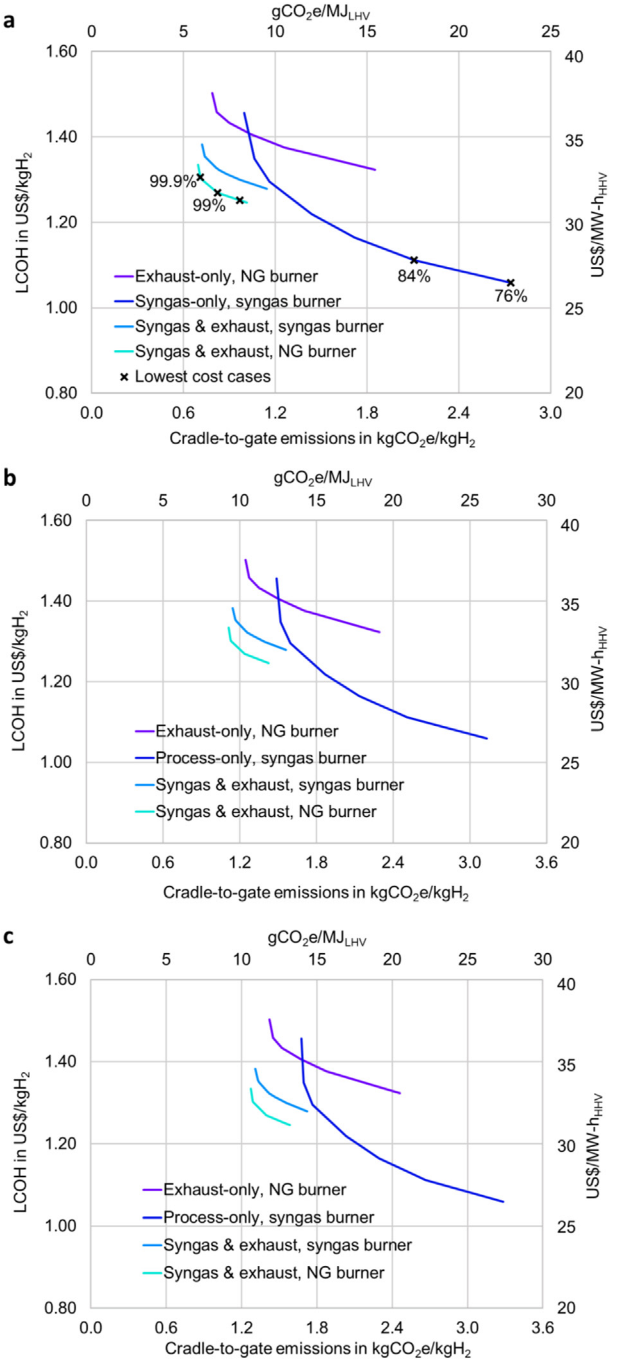

Fig. 5 – Impact of NG-supply emission intensity on the relationship between LCOH and cradle-to-gate emissions. LCOH as a function of cradle-to-gate GHG emissions (GWP100) for lowest cost cases of each process configuration with NG-supply emission factor based on (a) 7 GE NG production, (b) BC average emission intensity as reported, and (c) BC average emission intensity with methane emissions increased 80%. "Lowest cost cases" progress from minimum LCOH to minimum cradle-to-gate emissions with the lowest MAC between each step. Fossil-CO_2 capture rates (%) of selected points labelled.

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compared to SRNG without CCS (US\$0.76/kgH$_2$ and 8.95 kgCO$_2$e/kgH$_2$ in this model) and US\$169/tCO$_2$e compared to unmitigated NG combustion when substituted on an energy basis (HHV) (Fig. 6a). The options with lowest MAC to further reduce emissions are increasing reformer equilibrium temperature to 950 °C followed by switching to syngas-and-exhaust capture. Syngas-and-exhaust capture with increasing exhaust absorption stages provides MAC less than US\$125/tCO$_2$e to reduce emission intensity to 0.82 kgCO$_2$e/kgH$_2$ (99% fossil-CO$_2$ capture), but MAC increases considerably to reach 0.71 kgCO$_2$e/kgH$_2$ (US\$315/tCO$_2$e, 99.9% fossil-CO$_2$ capture). MAC to achieve >99.9% fossil-CO$_2$ capture using 30% wt MEA for exhaust capture is extremely high while the difference in cradle-to-gate emissions is <2%.

Upstream NG supply is the most significant source (c. 73%) of cradle-to-gate emissions in configurations with low direct emissions (Fig. 6b). CO$_2$ accounts for 68% of indirect emissions associated with upstream NG supply in the 7 GE production scenario, while methane accounts for 32% (74% CO$_2$ and 26% methane in the BC average NG emission intensity scenario). Other sources of indirect emissions in decreasing order of significance are plant construction, electricity supply, operating/maintenance labour, maintenance materials, and SRNG fugitive emissions. Water treatment, absorbent supply, and CO$_2$-transport fugitive emissions are insignificant sources (<0.1%).

Comparisons to prior studies

Lower cradle-to-gate GHG emission intensities are calculated in this study compared to prior studies (Fig. 7a) due to process configuration, the range of process design parameters considered, low-emission NG supply, and low-carbon grid electricity. Significantly lower cradle-to-gate emissions for

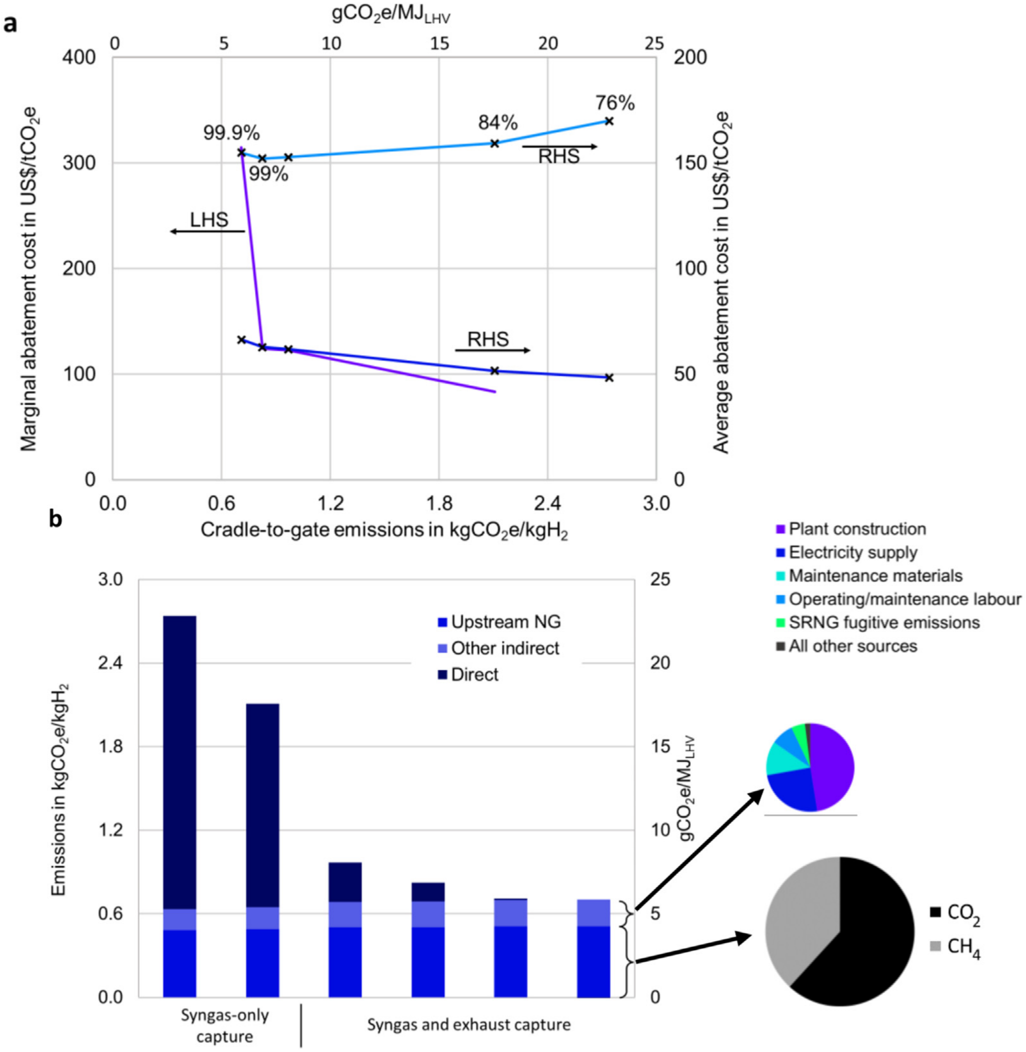

Fig. 6 – Comparing GHG abatement of process configurations. a, Average abatement cost relative to SRNG without CCS (dark blue, right axis) and substitution of hydrogen for unabated NG combustion on HHV energy basis (light blue, right axis) for the lowest cost cases in Fig. 3a as a function of cradle-to-gate emissions (GWP100). MAC (purple, left axis) is based on the change in LCOH and cradle-to-gate emissions between lowest cost cases. Fossil-CO$_2$ capture rates (%) of selected points labelled. b, Breakdown of emission sources (GWP100) for the lowest cost cases in Fig. 3a. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

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syngas-only capture are calculated in this study by incorporating piperazine-promoted MDEA and the combination of isothermal LT WGS with syngas burner fuel. Timmerberg et al. [6] and Hermesmann and Müller [10] reported cradle-to-gate emission intensity for SRNG with CCS within the range of this study at the low end of their sensitivity analyses. However, this study includes a range of CO_2 capture scenarios while those studies assumed 90% CO_2 capture.

Previous SRNG studies [6,7,10,17,18] did not consider life cycle emissions from labour, maintenance, SRNG fugitive emissions, or SRNG facility LUC (Supplementary Note 1). Although small compared to conventional SRNG emissions, these sources are significant in deep decarbonisation cases (12% combined in lowest emission case).

The span of LCOH calculated in this study is within the broad range of prior studies (Fig. 7b). LCOH estimates in NETL [23], Khan et al. [19], and IEAGHG [9] agree closely with this study. The higher range calculated by Salkuyeh et al. [17] was due to very low process efficiency and higher assumed interest rate (12%). The lower range calculated by Timmerberg et al. [6] was due to assumed capital costs, discount rate (5%), and operating and maintenance costs that were substantially lower. The assumed cost for CO_2 transport and storage in this study is similar to prior analyses [9,19,23] and accounts for US\$0.09–0.13/kgH_2. LCOH may be reduced if opportunities are available to sell the CO_2 (e.g., for enhanced oil or NG recovery).

Comparisons to green hydrogen

The ranges of cradle-to-gate emission intensities in this study are within the range of published estimates for green hydrogen (Fig. 7a). LCOH from SRNG with all NG cost scenarios in this study (Supplementary Note 5) are well below current estimates for green hydrogen [4,6]. While there is high uncertainty in future NG prices [63] and cost of green hydrogen [4], a sustained scenario of high NG prices and low-cost green hydrogen is unlikely given the proposed role for hydrogen in reducing NG consumption [70]. LCOH in this study based on historical western Canadian NG prices is in the low end of the forecast range for green hydrogen production in 2050 (US\$1.00–3.40/kgH_2) [4]. Long-range cost forecasts for green hydrogen have large uncertainty and typically do not include costs associated with smoothing out the intermittent supply of renewable power [4].

Another consideration should be scalability of hydrogen production. There are practical limitations and barriers to increasing supply of low-carbon electricity such as availability of raw materials [71] and human capital [72]. Furthermore, electrification is a key GHG mitigation strategy to address many different emission sources, and global demand for electricity is expected to increase significantly (+76% from 2020 to 2050) [2]. SRNG with CCS produces about 38–66 times more hydrogen per unit of electricity consumed than electrolysis: 690–1200 kgH_2/MWh in this study versus approximately 18 kgH_2/MWh for electrolysis [10]. Therefore, SRNG with CCS powered by low-carbon electricity could provide significantly faster scale-up of hydrogen production and facilitate more rapid systemic emissions abatement.

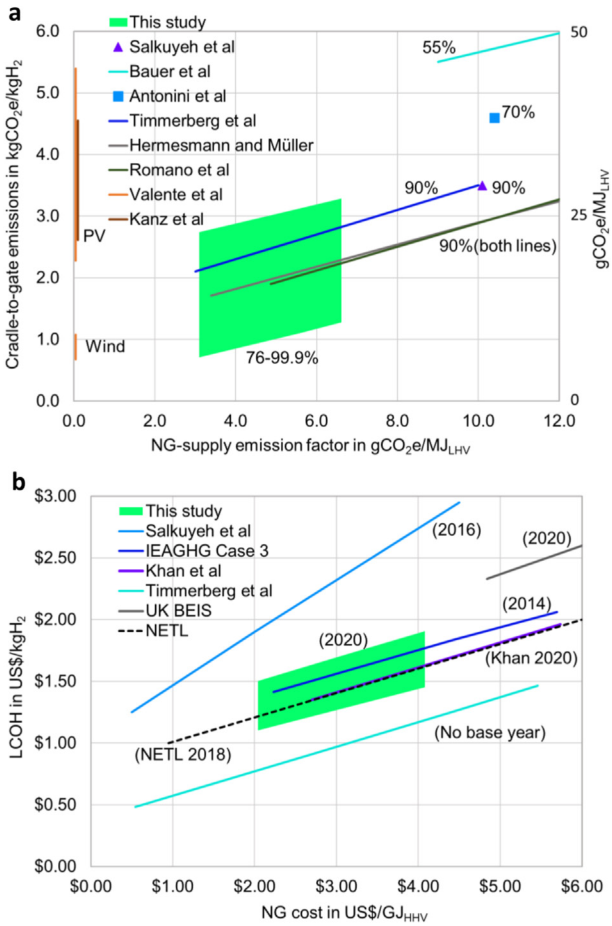

Fig. 7 – Results from this study compared with prior studies. a, Cradle-to-gate GHG emissions (GWP100) as a function of NG-supply emission intensity (GWP100) for the range of design conditions and process configurations in this study (green area) compared with previous studies of SRNG with CCS [6,7,10,17,18,67]. Overall fossil-CO_2 capture rates (%) for each study noted. Studies shown with single point indicate that sensitivity to NG-supply emission intensity was not considered. Values shown for photovoltaic and wind-powered electrolysis are interquartile ranges of the studies reviewed in Valente et al. [5] and Kanz et al. [68] b, LCOH as a function of NG cost for the range of lowest cost cases for the four process configurations in this study (green area) compared with previous values published for SRNG with CCS [6,9,17,19,23,69]. Original data converted to US\$ where necessary using purchasing power parity (PPP) exchange rates [62]. Base year of each study noted in parentheses. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Meanwhile, green hydrogen may become an attractive option for balancing electricity demand and intermittent renewable power generation [73] because electrolysers are more modular and have greater operational flexibility than SRNG [74]. Another consideration will be the availability of suitable geologic formations and infrastructure required for CCS [75].

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Low-carbon hydrogen and energy trade

NG prices vary significantly between jurisdictions due to the high cost of long-distance and *trans*-oceanic transportation. Low baseline LCOH is calculated in this study partly due to low historical NG prices in western Canada. Some jurisdictions are interested in utilising low-carbon hydrogen for energy trade [76]. However, hydrogen is more expensive to transport than NG and there are significant cost and efficiency barriers to storing hydrogen at sufficient density or producing liquid derivatives, such as ammonia, to make *trans*-oceanic shipping feasible [77,78]. This is likely to constrain broad applicability of cost projections for hydrogen, regardless of the production method, because cost of delivered hydrogen will be highly dependent on locally available resources and/or proximity to production. Regions with access to good wind/solar resources may be able to produce hydrogen more economically from renewable energy while processes such as SRNG with CCS may be favoured in regions with access to low-cost NG.

NG supply chain emissions

Prior studies correctly identified the importance of low fugitive methane emissions as a requisite for low-carbon hydrogen production from NG [18,67]. Regulations and leak detection/repair programmes have successfully reduced methane emissions in some jurisdictions [34,36]. While there is variability in estimates of methane emissions in BC, the impact on calculated SRNG cradle-to-gate emissions was attenuated because methane accounts for a relatively small portion of reported NG production emissions in BC [32]. Nonetheless, a requirement to demonstrate low fugitive methane emissions for NG supplied to produce hydrogen would assure compatibility with climate stabilisation goals.

However, in deep decarbonisation scenarios assessed in this study, SRNG cradle-to-gate emissions were dominated by CO_2 emissions from NG production. Thus, in addition to continuing efforts to minimise fugitive methane emissions, efforts should also be made to reduce upstream CO_2 emissions. 53% of cradle-to-gate emissions associated with 7 GE NG production came from stationary combustion during processing (e.g., compressor drives, dehydration, and sweetening) [38]. Further studies could assess the opportunity to mitigate upstream emissions through low-carbon electrification (particularly compressor drives) or hydrogen fuel supply. NG processing CO_2 emissions accounted for 0.25–0.28 $kgCO_2e/kgH_2$ of the cradle-to-gate emission intensity calculated in this study based on 7 GE production data.

Climate-neutral hydrogen

For all hydrogen production methods, residual emissions need to be offset with atmospheric CO_2 removal (CDR) to produce climate-neutral hydrogen. Options for CDR have different socio-economic/environmental impacts and permanence of storage [79], and there are large uncertainties in the cost and potential capacity of large-scale deployment of long permanence CDR (e.g., direct air CCS) [79–81]. The economically optimal quantity of CDR will depend on the future cost of CDR relative to abatement of direct/indirect emissions; however, SRNG designs with lower cradle-to-gate emissions could reduce the economic risk of achieving climate-neutral hydrogen production if the cost of long permanence CDR remains high.

Broader environmental impacts

Absorption solvent degradation rates/products and ambient air pollution have been identified as specific concerns related to exhaust CO_2 capture; however, long-term testing has been limited and mostly based on coal power plant flue gas [82,83]. Significant variability has been found in emission/degradation rates and degradation mechanisms are not well understood [82,84]. Water wash on the outlet from exhaust capture has been found to effectively limit atmospheric emissions of MEA and vapour phase degradation products [82,85]. Based on reported pilot data [82,83] and the results of this study, amine degradation rates and losses for SRNG with CCS are not likely to materially impact LCOH or life cycle GHG emissions. Nonetheless, further testing is required to ensure facilities will meet applicable regulations and undesired environmental impacts are minimised. Availability of life cycle data from SRNG, CCS, and specific NG production practices would facilitate LCA including a broader range of environmental impact categories.

Heterogeneous perspectives

Public support for specific approaches to mitigate global warming is likely to be heterogenous within and between jurisdictions [86]. There are vast regional differences in current energy sources [87], forecast energy demand [2], seasonal fluctuations in energy consumption [88], potential for renewable energy production [72], and available fossil fuel reserves [89]. Furthermore, jurisdictions, and their citizens, have different relationships with existing and future energy systems – e.g., importers, exporters, consumers, producers, and equipment manufacturers [76,87,90] – and a diverse range of ideological worldviews [91]. Jurisdictions are likely to pursue different energy sources depending on specific local circumstances which will affect cost, economic development, and energy security [72]. In regions historically connected with fossil fuel extraction, approaches such as SRNG with CCS could be perceived as more aligned with existing emotional attachments and place-based identities than renewable energy and, therefore, receive greater public support [92].

Conclusions

Low emission NG supply is a requisite for SRNG to be compatible with the Paris Agreement climate stabilisation objectives and cradle-to-gate emissions in this study were dominated by NG supply in deep decarbonisation scenarios. While this study found that reported discrepancies in fugitive methane emissions in BC had a smaller effect on SRNG cradle-to-gate emissions than process design choices, further study is crucial to ensure confidence in reporting and improve regulations/enforcement to reduce those emissions. Proactively addressing this issue would be in the interest of proponents seeking a long-term role for NG production as economies decarbonise. Meanwhile, this study identified a significant opportunity to reduce SRNG cradle-to-gate emissions through abatement of CO_2 emissions from upstream NG processing. This study also estimated the contribution of previously overlooked sources of indirect SRNG emissions (e.g., operating and maintenance expenses and SRNG fugitive emissions) and determined they were non-negligible in deep decarbonisation scenarios.

Cradle-to-gate emission intensity for SRNG with CCS in this study was within the range of published estimates for green hydrogen and syngas-and-exhaust capture was comparable to wind-powered electrolysis. Meanwhile, LCOH estimates in this study were significantly lower than current estimates for green hydrogen and in the low end of the range of future projections. This important finding shows that, rather than just a bridging technology, SRNG with CCS could be a long-term energy solution consistent with climate stabilisation. Having a diverse range of viable abatement options is critical to support the social, political, economic, and security needs of different jurisdictions. Furthermore, SRNG with CCS produces considerably more hydrogen per unit of electricity consumed and could facilitate more rapid expansion of low-carbon hydrogen supply than electrolysis.

LCOH was negatively correlated with cradle-to-gate emission intensity due to increased equipment expenses and NG consumption, and MAC increased considerably for near-zero direct emissions. This leads to important considerations for policymakers seeking to balance the urgent need for GHG emissions abatement with expectations of affordable energy. SRNG with CCS, like green hydrogen, will have residual cradle-to-gate emissions that will require CDR to achieve net-zero emissions for climate neutrality. Adaptive emissions targets within hydrogen strategies may help to encourage displacement of higher carbon fuels while ensuring that future emissions associated with long-lived capital assets are consistent with net-zero emission ambitions.

Data availability statement

The data that support the results of this study are provided in the main text and Supplementary Notes 1–5. Source data are provided with this paper.

Author contributions

Ryan Cownden: conceptualization, methodology, investigation, validation, formal analysis, data curation, visualization, writing – original draft, writing – review and editing. Daniel Mullen: methodology, validation, writing – review and editing. Mathieu Lucquiaud: conceptualization, methodology, writing – review and editing, supervision, resources.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ijhydene.2022.12.349.

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