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Hydrogen emissions from the hydrogen value chain-emissions profile and impact to global warming

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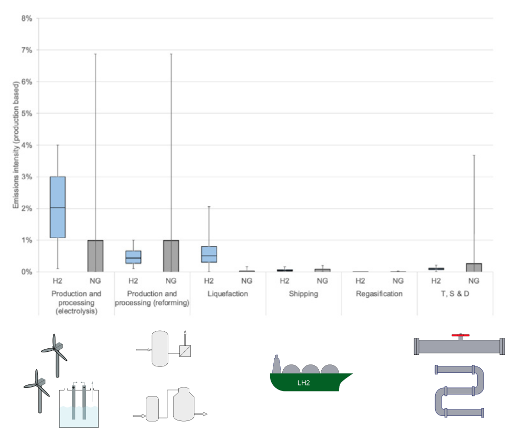
HIGHLIGHTS

- The first estimation of H_2 emissions from H_2 supply chains
- Up to 138 $MtCO_{2eq.}$ could be caused by H_2 emissions alone.
- Green H_2 appears to have higher H_2 emissions than blue H_2.
- Impacts to global warming could be more important over short time-horizons.
- Large data gap in H_2 emissions data- no measurement studies conducted

GRAPHICAL ABSTRACT



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ABSTRACT

Future energy systems could rely on hydrogen (H_2) to achieve decarbonisation and net-zero goals. In a similar energy landscape to natural gas, H_2 emissions occur along the supply chain. It has been studied how current gas infrastructure can support H_2, but there is little known about how H_2 emissions affect global warming as an indirect greenhouse gas. In this work, we have estimated for the first time the potential emission profiles (g CO_{2eq}/MJ $H_{2,HHV}$) of H_2 supply chains, and found that the emission rates of H_2 from H_2 supply chains and methane from natural gas supply are comparable, but the impact on global warming is much lower based on current estimates. This study also demonstrates the critical importance of establishing mobile H_2 emission monitoring and reducing the uncertainty of short-lived H_2 climate forcing so as to clearly address H_2 emissions for net-zero strategies.

Abbreviations: ATR, Autothermal reforming; AU, Australia; AUBH, Australia blue H_2; AUGH, Australia green H_2; CCS, Carbon capture and storage; GCC, Gulf cooperation council; GHG, Greenhouse gas; GTP, Global temerpature potential; GWP, Global warming potential; LH2, Liquid H_2; LOHC, Liquid organic H_2 carrier; NS, North Sea; PSA, Pressure swing adsorption; QA, Qatar; SA, Saudi Arabia; SMR, Steam methane reforming; T,S&D, Transmission, storage and distribution; WTG, Well to gate.

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1. Introduction

Hydrogen (H_2) could be one of the key pillars of decarbonising global energy systems because of its significant benefits in terms of reducing greenhouse gas (GHG) emissions. It shares many characteristics with natural gas but contains no carbon, allowing it to complement electrification and other approaches to decarbonisation. For this reason, it is seen as a key energy vector in meeting Paris Agreement goals (Rogelj et al., 2018). Global consumption of H_2-based fuels in 2050 is expected to be around 33 exajoule (10^{18} J, EJ) and 18 EJ in the IEA's net zero emission (IEA, 2021) and IPCC SR1.5 scenarios (IEA, 2021, Rogelj et al., 2018), respectively. These would limit temperature rises to well below 2°C compared to pre-industrial levels. According to the IEA Net-Zero by 2050 report, H_2 and H_2-based fuels can achieve 4 Gt CO_2 emissions reductions between 2046 and 2050 (IEA, 2021). As a result of this potential, several nations have recently announced ambitious H_2 strategies, attempting to define its role in their future energy systems. This has been accompanied by numerous targets, projects and policy incentives in place to directly promote it for a decarbonised energy system (BEIS, 2021, COAG Energy Council, 2019, Hydrogen Europe, 2020, Qamar Energy, 2020).

Despite the enthusiasm in its development, the effects of large-scale transitions are not fully understood (de Coninck et al., 2018) as it is an indirect GHG. As a homonuclear diatomic molecule with no dipole moment, it cannot not absorb infrared radiation (BEIS, 2019; Derwent, 2018), but can deplete stratospheric ozone by moistening the stratosphere, thus promoting the growth of methane and tropospheric ozone. Therefore, as an indirect GHG, the impacts of emissions to the atmosphere need to be understood for decarbonisation strategies to be fully effective.

There are many projects ongoing or announced which will significantly increase global H_2 production capacity. One of the world's largest green H_2 production plants (1 GW H_2 plant powered by 2 GW electricity from wind farms) is to be built on the North Sea Port to provide sustainable H_2 to the Netherlands and Belgium (TNO, 2021). In North America, the H2@Scale program aims to improve H_2 production, for domestic use, from natural gas via steam methane reforming (SMR) and biomass gasification with carbon capture and storage (CCS) (US DOE, 2020). The export of H_2 is also being explored in numerous projects; Australia and Gulf Cooperation Council (GCC) countries, such as Qatar and Saudi Arabia, have emerged as major players in a potential global hydrogen market. Australia has launched the world's first H_2 export supply chain project, with the goal of producing liquid H_2 (LH2) from coal gasification with CCS in the Latrobe Valley and exporting this to Kobe, Japan (COAG Energy Council, 2019). The GCC countries have expressed an interest in green H_2 (Qamar Energy, 2020) and in 2020 Saudi Arabia announced it was to build the world's largest green H_2 plant (4 GW renewable electricity) (Parnell, 2020).

From the number of large-scale projects, large volumes of H_2 will be produced in the near to medium term future. Consequentially there is the potential for large quantities of H_2 to be emitted into the atmosphere, which could place additional GHG burdens on the planet. GHG emissions from H_2 production and use have been extensively studied through life cycle assessment and other tools (Cetinkaya et al., 2012; Mehmeti et al., 2018; Miller et al., 2017; Ramsden et al., 2013; Reiter and Lindorfer, 2015), but these studies all omit H_2 emissions and their impacts on global warming. Studies which have estimated H_2 emissions have found that leakage rates range from 0.3 to 20% depending on the infrastructure (Colella et al., 2005; Schultz et al., 2003; van Ruijven et al., 2011; Wuebbles et al., 2009) but these did not estimate the impacts to global warming. Field and Derwent (2021) estimated the global warming impacts of replacing natural gas with H_2 but only considered leaks in downstream distribution. Derwent et al. (2020) also estimated the impacts of H_2 leaks in a system where H_2 replaces fossil fuels and found that H_2 is a good alternative fuel, provided H_2 leaks are curtailed.

If H_2 is used in ways akin to natural gas, then there is potential for emissions along the whole supply chain. Previous studies have demonstrated that H_2 production, transportation and storage result in losses to the atmosphere (BEIS, 2019; Crowther et al., 2015; Feck, 2009; Schultz et al., 2003). At the time of writing, only Derwent et al. (2006), Derwent et al. (2018) and Derwent et al. (2020) have investigated the global warming potential (GWP) of H_2 (1.9 to 9.8) and Paulot et al. (2021) have estimated the radiative forcing (0.13 mW m^{-2} $ppbv^{-1}$). Therefore, H_2 emissions from H_2 supply chains should be thoroughly addressed as it could contribute towards global warming (Rogelj et al., 2018). Investigating H_2 emissions from supply chains is crucial in light of likely future energy transitions, which will necessitate the replacement of some fossil fuels with H_2. This is the main novelty of this work; we assess, for the first time, the global warming impacts of H_2 emissions for a variety of supply chains and compare the different supply chains.

The aims of this study are to assess H_2 emissions from various supply chains (blue, biomass and green H_2 spanning domestic use and international export) and estimate the impact of these emissions on global warming. As far as the authors are aware, this is the first study to do this. This study's findings will be of interest to plant operators, supply chain investors, and policymakers. The rest of the paper is structed as follows: the methodology is presented, followed by our emissions results and discussion and limitations of the work. The paper ends with the conclusions drawn and areas for future work.

2. Methodology

In this work, H_2 emissions are estimated for blue (fossil fuel with CCS), biomass and green (renewable energy powered electrolysis) H_2 supply chains. A feedstock procurement to gas distribution, or well to gate (WTG), system is considered (Fig. 1 and Fig. 2), to be analogous to emission studies of natural gas supply chains. The supply chain stages are disaggregated as much as possible so that emission hot spots could be identified. In total we consider seven supply chains, which encompass a wide variety of synthesis and trade routes (Fig. 1 and Fig. 2) to cover likely future supply chains. These were selected based on pilot projects and trials (completed, ongoing and announced) and projected H_2 potentials detailed in national H_2 strategies. For three of the supply chains (blue and biomass H_2), methane emissions are also considered and compared with H_2 emissions to compare the impacts to global warming, hence why pre-production stages are included in the system boundary.

2.1. Supply chains considered

2.1.1. USA biomass gasification for local use

A gasifier converts biomass (woody biomass such as poplar) into biogas which is then converted into H_2 in a SMR (with water gas shift (WGS) and pressure swing adsorption (PSA)). A localised supply chain is considered where H_2 is produced for local use, hence transmission and storage are not needed. A distribution pipeline network of total length 75,000 km is considered. Please note this pipeline length is not for one distribution network, but multiple networks. The feedstock and technology are based on Ramsden et al. (2013) and a generic USA geography is considered.

2.1.2. Australian blue H_2 from coal for export to Japan

This supply chain is based on the Latrobe Valley pilot project (HESC, 2021a). Brown coal is mined in the Latrobe Valley and used in a gasifier (with CCS) to produce H_2 which is then transported via truck as compressed H_2 to a liquefaction facility in the Port of Hastings where it is liquefied to produce liquid H_2 (LH2). The LH2 is shipped to Kobe (5000 nautical miles) where it enters a H_2 gas grid.

2.1.3. Qatar blue H_2 from natural gas for export to Japan

As Qatar is a major natural gas producing country (BP, 2020), it has great blue hydrogen potential, which is why it has been considered in this work. H_2 is produced through autothermal reforming (ATR) (with CCS) which is then liquefied and shipped to Tokyo (6500 nautical miles).

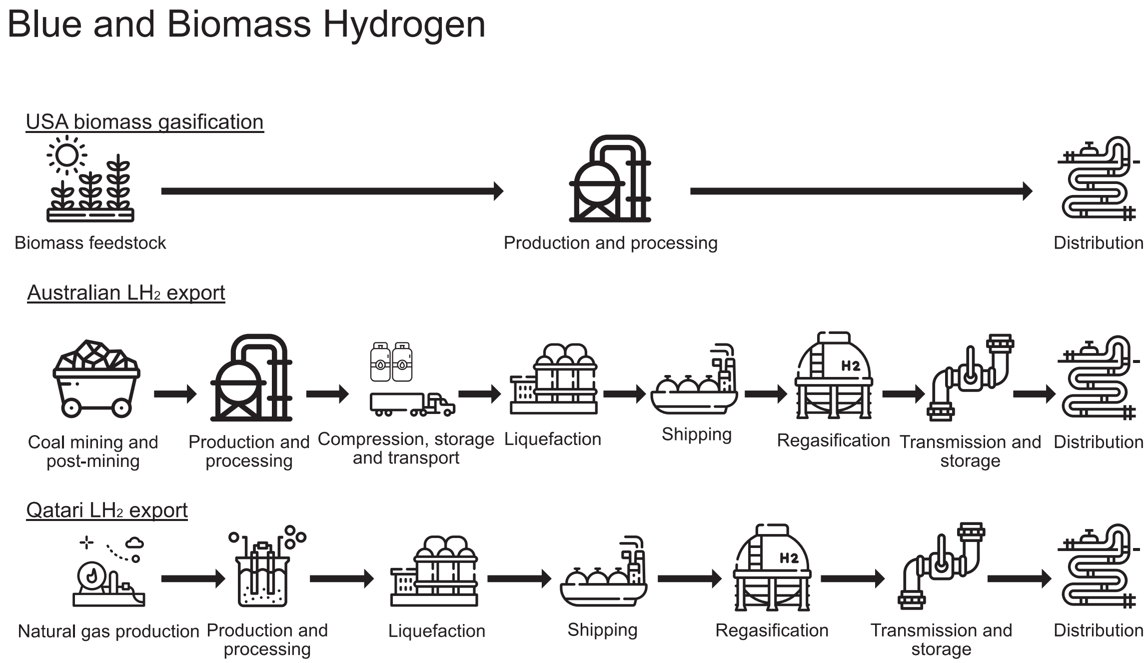

Fig. 1. Blue and biomass H_2 supply chain routes.

2.1.4. North Sea green H_2 for local use

The North Sea has great offshore wind potential and projects such as NortH2 (Laat, 2020) have been proposed to utilise this to produce H_2. Offshore wind turbines generate electricity which is used to produce H_2 in onshore electrolysers. We consider H_2 production in the Netherlands and the H_2 is fed into the Dutch transmission system (1000 km) where it is then sent for local use (10,000 km distribution pipelines).

2.1.5. Australian green H_2 for export to Japan

In 2019, Australia exported green H_2 to Japan in a research trial (Maisch, 2019). The H_2 was produced in Queensland and exported as a liquid organic H_2 carrier (LOHC). We consider H_2 produced using electricity from onshore wind turbines but consider export to Japan as LH2 rather than as LOHC. The H_2 is produced and liquefied in Gladstone where it is then shipped to Tokyo (4000 nautical miles).

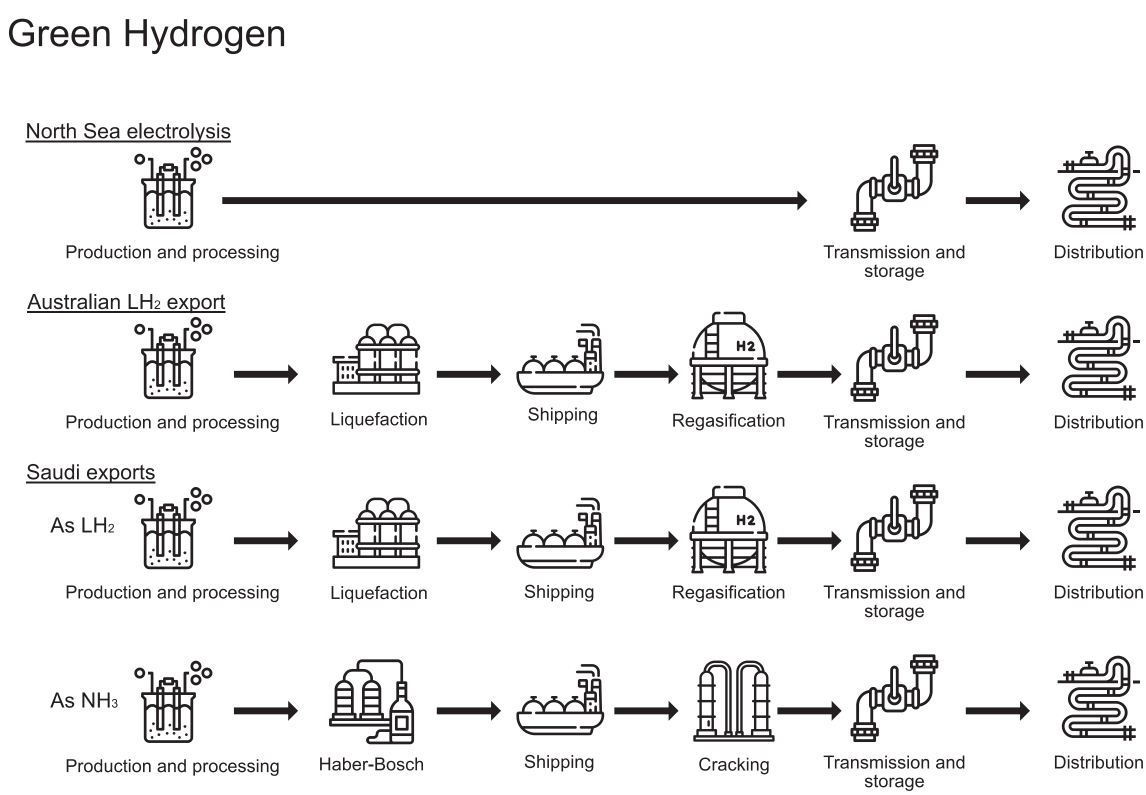

Fig. 2. Green H_2 supply chain routes.

2.1.6. Saudi Arabian green H_2 for export to Japan- as LH2

In 2020 Saudi Arabia unveiled the world's largest green H_2 project, in which solar panels and wind turbines would be used to produce H_2 (Parnell, 2020). Based on this we consider green H_2 produced and exported as LH_2 to Tokyo (7000 nautical miles).

2.1.7. Saudi Arabian green H_2 for export to Japan- as ammonia

In addition to exporting H_2 as LH2, we also consider export as ammonia (NH_3). The H_2 is converted into NH_3 through the Haber-Bosch process and then shipped to Tokyo where it is cracked to convert it back into H_2, which is then fed into the transmission system for localised distribution and use (1000 to 10,000 km).

2.2. Calculating H_2 emissions

The H_2 emissions calculated in this work are based on a combination of literature data and modelling (Section 2.3). Throughout this paper we define H_2 losses and emissions as:

- H_2 losses are H_2 consumed within a process, such as for heat and power generation or diffusion across the electrolyser membrane (Rozendal et al., 2006a, 2006b); and
- H_2 emissions are H_2 directly lost to the atmosphere (fugitives, venting, incomplete combustion).

To calculate emissions, a method akin to that used to estimate methane emissions in natural gas supply chains has been used (IPCC, 2003; IPCC, 2006):

$$E_i = A_i \times EF_i$$

Where E_i is the emissions (kg H_2, m^3 H_2 etc.) per supply chain stage i, A_i is the H_2 throughput in stage i and EF_i is the emission rate of stage i. From this, the total amount of H_2 emitted (E) across a supply chain with N stages can be calculated:

$$E = \sum_{i=1}^{N} E_i$$

From this the emissions intensity can be calculated by dividing the emissions (E or E_i) by the H_2 output of the supply chain.

2.2.1. H_2 emissions data, EF_i

In this work, EF_i is the percentage of H_2 lost to the atmosphere through venting, fugitives and incomplete combustion (Table 2). There is extremely limited data available on H_2 emissions; data were only available for the upstream stages (production only). The literature data are largely modelling studies and there have been no studies conducted to measure H_2 emissions in supply chains. For all other stages, emission rates were estimated through modelling; refer to Section 2.3.

2.2.2. H_2 throughput and losses

To compare the different supply chain routes, we consider the H_2 production volume, which ranged from 3 t to 0.8 Mt H_2 per year (see Section 4 in the Supporting Information (SI)) to calculate emissions. As H_2 is consumed by processes within the different supply chain stages, the throughput (A_i) is different in each stage. A_i is calculated by factoring in losses (Table 1). As with emissions data, this was extremely scarce, and no loss data for stages downstream of production was located. The loss rates of the stages downstream of production were estimated through modelling; refer to Section 2.3.

2.3. Modelling H_2 emissions and losses

To model emissions and losses several avenues were explored. Where H_2 specific data was available, these were used but these data were sparse and we primarily relied on using natural gas systems as a proxy with differences in physical properties between the gasses determining relative loss and leak rates. As there are similarities between the supply chains, this is viewed as a suitable placeholder until further research is carried out to determine more precise values. See Section 1 in the SI for details on the modelling.

2.3.1. Natural gas to H_2 conversion

To convert emission estimates in natural gas studies the properties of each gas were compared, and these used to calculate equivalent leakage rates. For further details on modelling and conversions, see Section 1 and 2 in the SI.

2.3.2. Incomplete combustion of H_2 and slip

The energy requirements during compression and liquefication were met using H_2 to be in line with natural gas systems. Engine slip of H_2 is reported to range from 0 to 12% (Fayaz et al., 2012) with the highest losses occurring at low engine loads. An average value of 0.5% was taken, as it is unlikely suboptimal conditions would be implemented given the high penalty for doing so. However, it is clear large uncertainties are present in engine slip. Flaring is another source of uncombusted H_2 but we do not consider any flaring in our supply chains.

2.3.3. Fugitive emissions

These are the unintentional emissions such as leaks in equipment and faulty equipment emissions. These were modelled for compressors and pipelines. Literature values of natural gas leakage rates for reciprocating compressors (Subramanian et al., 2015) and natural gas pipelines were used. USA and European data were used to model pipeline emissions (Lamb et al., 2015; MARCOGAZ, 2018; United States Environmental Protection Agency et al., 1996; Weller et al., 2020). Plastic (polyvinyl chloride, PVC, and polyethylene, PE) was chosen as the pipeline material to minimise leaks and embrittlement. The values were converted to H_2 using knowledge on the type of leak, the flow and physical properties of the gasses (Section 1 in the SI). Due to the high level of detail involved in the USA studies, these were used for pipelines in the USA, Japan, Australia, Qatar and Saudi Arabia. For Europe a literature value was taken from MARCOGAZ (2018). Emissions from embrittlement and diffusions through pipelines are assumed to be negligible (see Section 2 in the SI for more information).

2.3.4. Liquefaction

H_2 liquefaction is an extremely energy intensive process, using 15 to 41% (Yin and Ju, 2020) of the H_2 to fuel the process and therefore, and the primary source of emissions is slip. We do not consider outside energy sources to run the liquefication process, rather using H_2 to keep the process in line with LNG (Pospíšil et al., 2019). Should future processes use low carbon electricity for this process the losses and associated emissions would be reduced by approximately one-third. We used a range of possible liquefaction routes from Yin and Ju (2020) to create our uncertainty bounds. Many liquefaction processes are not yet at scale, so may encounter problems when scaling up. Thus, we chose the precooled dual pressure Linde Hampson method, which uses 12.14 kWh/kg H_2 as our average route (Yin and Ju, 2020). This is in line with the current liquefaction plants operating in the USA which range from 12.5 to 15 kWh/kg H_2 (Drnevich, 2003).

2.3.5. Loading, unloading and storage

Once the H_2 has been liquefied it has to be stored, loaded and unloaded. We assumed the boil-off from storage would be re-supplied back to the liquefaction stage to reduce losses. When loading and unloading, LNG was used as a comparative. During loading, 4% of the tanker's capacity will remain to keep the tanker temperature low to reduce boil-off (Rogers, 2018). When unloading any boil-off will be routed to a compressor to be transported via pipeline. As with LNG, when the level of boil-off exceeds the pressure capacity, the H_2 is vented. When H_2 is stored or transported as compressed H_2, it is assumed to be stored and transported in the same tube trailer. This is to minimise the losses from changing trailers. The

Table 1
H_2 loss rates. The loss rates of biomass gasification (gasification with SMR) were assumed for coal gasification. No single electrolyser technology is assumed, and the values presented are for a range of electrolysis (alkaline water to biocatalysed) technologies. Further information on how loss rate and emission rates were calculated can be found in Section 2.3.1 to 2.3.7.

	USA biomass gasification for local use	Australian blue H_2 from coal for export to Japan	Qatar blue H_2 from natural gas for export to Japan	North Sea green H_2 for local use	Australian green H_2 for export to Japan	Saudi Arabian green H_2 for export to Japan- as LH2	Saudi Arabian green H_2 for export to Japan- as NH_3
Production and processing	1.50% (1.00–2.00%)	1.50% (1.00–2.00%)	3.00% (0.10–37.00%)[c]	5.00% (0.00–43.00%)[a]	5.00% (0.00–43.00%)[a]	5.00% (0.00–43.00%)[a]	5.00% (0.00–43.00%)[a]
Compression		5.75% (0.00–11.50%)[a]				5.75% (0.00–11.50%)[a]	
Storage and transport		0.50%				0.50%	
Liquefaction[b]		31.67% (14.17–34.95%)	38.95% (17.43–42.99%)		38.95% (17.43–42.99%)	31.67% (14.17–34.95%)	
Haber-Bosch							15.00% (0.00–30.00%)[a]
Shipping		3.57%	6.04%		3.57%	6.68%	0%[a]
Regasification		2.01% (0.01–2.52%)	2.01% (0.01–2.52%)		2.01% (0.01–2.52%)	2.01% (0.01–2.52%)	
NH_3 cracking							15.00% (0.00–32.40%)[a]
Transmission and storage		1.22% (0.84–1.58%)	1.22% (0.84–1.58%)	2.42% (1.67–3.12%)	1.22% (0.84–1.58%)	1.22% (0.84–1.58%)	1.22% (0.84–1.58%)
Distribution	0.67% (0.46–0.92%)	0.26% (0.17–0.44%)	0.26% (0.17–0.44%)	0.02% (0.02–0.14%)	0.26% (0.17–0.44%)	0.26% (0.17–0.44%)	0.26% (0.17–0.44%)
Sources	De Souza-Santos (2004) and our calculation-see Section 2.3	De Souza-Santos (2004) and our calculation- see Section 2.3	Ishimoto et al. (2020), Bakkaloglu (2021) and our calculation- see Section 2.3	Stolzenburg (2007), Rozendal et al. (2006a, 2006b) and our calculation- see Section 2.3	Stolzenburg (2007), Rozendal et al. (2006a, 2006b) and our calculation- see Section 2.3	Stolzenburg (2007), Rozendal et al. (2006a, 2006b) and our calculation- see Section 2.3	Stolzenburg (2007), Rozendal et al. (2006a, 2006b) Ishimoto et al. (2020) and our calculation- see Section 2.3

[a] 0% losses- alternative fuel used to power the process.
[b] loss rates differ for liquefaction depending on whether compression occurs in the supply chain. If compression occurs, losses are lower than when liquefying without prior compression.
[c] values is for production of LH2 via ATR (ATR plant and liquefaction). A disaggregated value was not available. However, we assumed this value for ATR to explore potential high loss rates, as well as to explore loss rates in reforming which are similar to electrolysis.

trailers will be transported once each month as per the Latrobe Valley project (HESC, 2021b).

2.3.6. Shipping

The H_2 tanker ship from Yoshino et al. (2012) was used to model LH2 transport. This ship has a 160,000m^3 capacity and utilises 0.2% boil-off per day to fuel the ship. We assume engine slip is the main source of emissions and fugitives are negligible (Abrahams et al., 2015). The travel distances for each supply chain were taken from Sea Distances (2021) using speeds based on current LNG tankers- an average of 18 knot (33 km/h) (Msakni and Haouari, 2018; Raab et al., 2021). Uncertainty bounds of 10% were assumed for total journey time.

2.3.7. Regasification

The quantity of LNG used during regasification was used as a reference: 2% (Pospíšil et al., 2019). This was converted into a quantity of H_2 fuel loss using the energy density differences between H_2 and natural gas (Section 2.3.1). H_2 slip was assumed to be the main source of emissions and fugitives and venting were not included.

2.4. Methane emissions and comparison to H_2

In the blue and biomass H_2 supply chains, methane is emitted during the pre-production and production stages e.g. natural gas production and transport and agricultural activities and feedstock storage. Methane emissions were calculated for these stages and compared to H_2 emissions to gauge the magnitude of H_2's global warming impact. Emission factors from the literature (see Section 6 in the SI) were used to estimate emissions and the methane throughput of these stages estimated assuming 3.2 kg per kg H_2, including conversion and fuel usage (Budsberg et al., 2015; Kabir and Kumar, 2011).

2.5. The impact of H_2 emissions to global warming

H_2 is an indirect GHG as it interferes with the sink of other non-CO_2 GHG, most importantly methane. However there have been few studies conducted to estimate its global warming impacts. The global warming potential (GWP), in particular the GWP_{100}, is the most commonly used metric to compare GHGs and quantify their impacts to global warming. However, H_2 has a short atmospheric lifespan (two to seven years (Derwent, 2018, Paulot et al., 2021)). Therefore, it is uncertain which metric is the most appropriate to use to quantify its climate change impacts e.g., continuous emissions over emissions pulse, GWP_{20} over GWP_{100}, GTP over GWP. Derwent et al. (2001) were the first to attempt to estimate a GWP but since then there have been only four further peer reviewed studies carried out (Table S3 in the SI). There is limited literature available which quantifies (in a metric) the global warming impact of H_2 and thus we use the GWP_{100} to estimate the CO_{2eq} of H_2 emissions as only this data is available in the (open source) literature at the time of writing.

2.6. Sensitivity and uncertainty analysis

To examine the effect of uncertainties within each stage of the supply chain on the overall emission estimates we ran a sensitivity analysis. We simulated 20,000 runs of our model using the uncertainty bounds for each of the gas losses and emission rates at each stage of the supply chain. From this we could determine how influential each uncertainty was on the overall result by examining the R squared values. The uncertainty bounds were from literature where possible and where this was not

Table 2

H_2 emission rates used to calculate emissions (emission intensity of H_2 and g CO_{2eq}). The same emission intensities are assumed in T&S and distribution for H_2 exports to Japan. The emission rates of biomass gasification (gasification with SMR) were assumed for coal gasification. No single electrolyser technology is assumed, and the values presented are for a range of electrolysis (alkaline water to biocatalysed) technologies. Further information on how loss rate and emission rates were calculated can be found in Section 2.3.1 to 2.3.7.

	USA biomass gasification for local use	Australian blue H_2 from coal for export to Japan	Qatar blue H_2 from natural gas for export to Japan	North Sea green H_2 for local use	Australian green H_2 for export to Japan	Saudi Arabian green H_2 for export to Japan- as LH2	Saudi Arabian green H_2 for export to Japan- as NH_3[a]
Production and processing	0.55% (0.10–1.00%)	0.55% (0.10–1.00%)	0.55% (0.10–1.00%)	2.05% (0.10–4.00%)	2.05% (0.10–4.00%)	2.05% (0.10–4.00%)	2.05% (0.10–4.00%)
Compression		0.18% (0.15–0.27%)				0.17% (0.14–0.26%)	
Storage and transport		0.31% (0.06–0.53%)				0.31% (0.05–0.54%)	
liquefaction		0.34% (0.15–2.21%)	0.33% (0.14–0.98%)		0.32% (0.14–0.95%)	0.33% (0.01–2.04%)	
Haber-Bosch							0%
Shipping		0.03% (0.00–0.10%)	0.06% (0.01–0.17%)		0.03% (0.003–0.10%)	0.06% (0.01–0.17%)	0%
Regasification		0.002%	0.002%		0.002%	0.002%	
NH_3 cracking							0%
Transmission and storage		0.03% (0.02–0.05%)	0.03% (0.02–0.05%)	0.05% (0.04–0.06%)	0.03% (0.02–0.05%)	0.03% (0.02–0.05%)	0.03% (0.02–0.05%)
Distribution	0.08% (0.05–0.12%)	0.08% (0.05–0.16%)	0.08% (0.05–0.16%)	0.02% (0.0003–0.03%)	0.08% (0.05–0.16%)	0.08% (0.05–0.16%)	0.08% (0.05–0.16%)
Sources	De Souza-Santos (2004) and our calculation- see Section 2.3	De Souza-Santos (2004) and our calculation- see Section 2.3	Ishimoto et al. (2020), Bakkaloglu (2021) and our calculation- see Section 2.3	Stolzenburg (2007), Rozendal et al. (2006a, 2006b) and our calculation- see Section 2.3	Stolzenburg (2007), Rozendal et al. (2006a, 2006b) and our calculation- see Section 2.3	Stolzenburg (2007), Rozendal et al. (2006a, 2006b) and our calculation- see Section 2.3	Stolzenburg (2007), Rozendal et al. (2006a, 2006b), Ishimoto et al. (2020) and our calculation- see Section 2.3

[a] no H_2 emissions are considered during NH_3 conversion, shipping and cracking. This is because we assumed any NH_3 emitted to the atmosphere does not decompose to produce H_2. The same gas losses and emission factors are assumed for Japan export supply chains for T&S and distribution.

possible IPCC uncertainty bounds were used. A gaussian distribution of uncertainty was assumed for the analysis.

2.6.1. Literature quality assessment

In addition to a sensitivity analysis, a quality assessment of the literature data was conducted. This allowed for uncertainty that was not captured in the bounds of the work to be assessed. We conducted a quality assessment on each piece of data we collected based on six criteria:

- Is it a direct measurement study?
- Have the results been replicated?
- Was H_2 studied?
- Was it a large-scale study?
- Were the results supported by theory/modelling?
- Was the paper written within last five years?

Each criterion is graded on a 0 to 1 scale using 0.1 increments, with 0 indicating low and 1 as high. The scores in each criterion are then aggregated to derive an overall quality assessment score: low (0 to 2), medium (2.1 to 4), high (4.1 to 6).

3. Results and discussion

The estimates of H_2 emissions from the supply chains considered are presented first, along with the impacts of these emissions to global warming and the impacts relative to methane for the blue and biomass supply chains. The results of the sensitivity and literature data quality assessment are then presented.

3.1. H_2 emissions from supply chains and their impacts towards global warming

3.1.1. H_2 emissions- emissions intensity

The supply chain route has a significant impact on emissions. The green H_2 routes have higher emissions (2.6%, 0.1 to 6.9%) than the blue and biomass (0.1% to 4.3%, average 1.4%) (Fig. 3). This is because the data we collected (Section 2) inferred a higher emission rate for electrolysis in comparisons to gasification, SMR and ATR. The trade method also has an impact, as shipping H_2 as NH_3 instead of as LH2 has lower emissions (2.1% versus 2.9%). The LH2 supply chains, particularly the ones which transport green H_2, were found to be the highest emitting. Consequentially, pipeline transport appears to be a better method of transporting H_2. Whilst we did not consider pipeline traded H_2, it is likely that even over cross-continent pipelines, emissions and losses would be lower than LH2 because of the high losses incurred during liquefaction, as well as the losses and emissions from shipping and regasification. In comparison to past studies, our results are in line with what has been previously reported: 0.3 to 20% (Colella et al., 2005; Schultz et al., 2003; van Ruijven et al., 2011; Wuebbles et al., 2009; Bond et al., 2011). When our results are extrapolated for 80 EJ (Hydrogen Council, 2017), 2.6 to 23.8 Mt H_2 could be emitted in 2050. van Ruijven et al. (2011) estimated H_2 emissions in 2050 to range between 30 and 70 Mt H_2 (depending on the emission factor assumed), but they included end use in their emissions modelling, while this study excluded end use.

3.1.2. Impacts to global warming and comparison to natural gas supply chains

When emissions are converted into CO_{2eq} based on GWP_{100}, they range between 4×10^{-4} to 1 g CO_{2eq}/MJ $H_{2,HHV}$ with the GWP impacting the results. Previous studies which have calculated the GHG emissions from H_2 production have found emissions to vary from -103 to 178 g CO_{2eq}/MJ $H_{2,\ HHV}$ (Speirs et al., 2017). In comparison, H_2 emissions are minimal. When compared to natural gas, the CO_{2eq} of H_2 is much lower across comparable supply chain stages (Fig. 4). However, when the two are compared on emissions intensity (based on volume of H_2/natural gas produced) H_2 is similar if not higher (Fig. 5). The reason for the discrepancy is that while the GWP of H_2 is ten-times lower than methane, its density is also lower (eight-fold). Therefore, when converting from emissions intensity into grams of CO_{2eq}/MJ $H_{2,HHV}$ is lower despite having similar or higher emissions intensity.

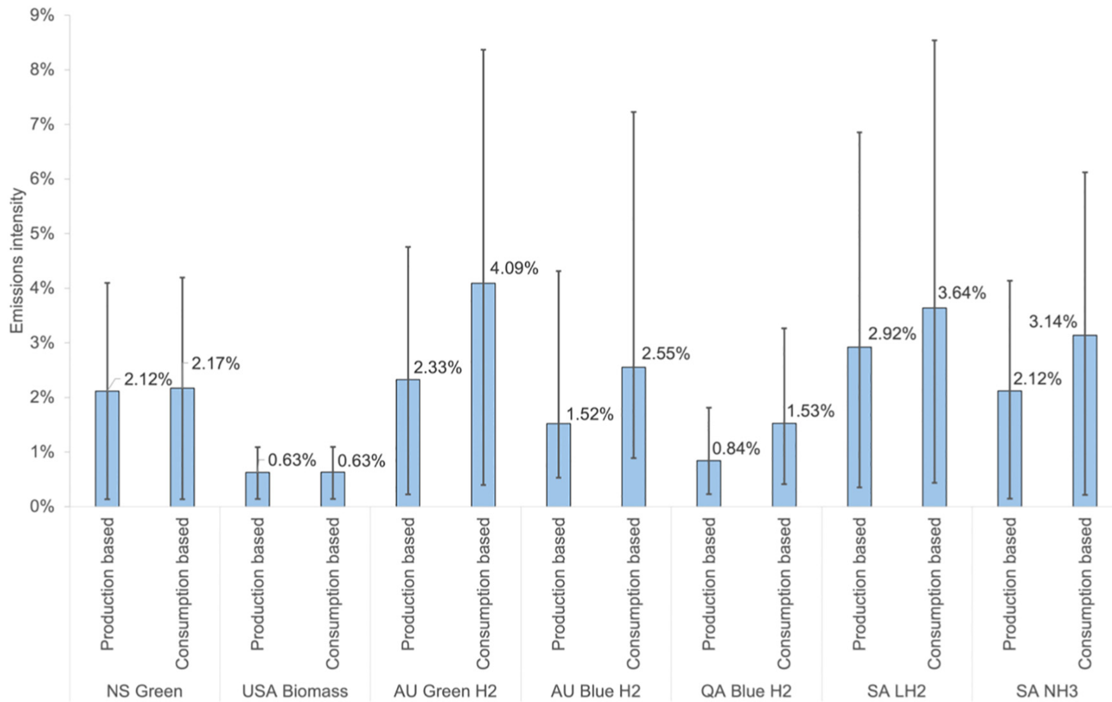

Fig. 3. Emissions intensity of H_2 supply chains. Bar represents average intensity and error bars show the minimum and maximum intensities calculated. AU: Australia; NS: North Sea; QA: Qatar; SA: Saudi Arabia.

To give an indication of the CO_{2eq} that could be emitted into the atmosphere in a world where H_2 plays a more prominent role in the global energy mix, we extrapolated our results assuming 80 EJ of H_2 in 2050 (Hydrogen Council, 2017). From this, roughly 9 to 138 Mt CO_{2eq} would be emitted (Fig. S2 in the SI) with over 10 Mt CO_{2eq} emitted from electrolysis alone (Fig. S1 in the SI). While this is small in comparison to total GHG emissions predicted for 2050 (IEA, 2021, Rogelj et al., 2018), as H_2 is not included in GHG budgets and climate modelling, there is at least 9 Mt CO_{2eq} not being accounted for.

However, as H_2 is a short-lived climate pollutant it has been argued that alternative metrics to GWP_{100} should be used to quantify its global warming impacts (Kurmayer, 2021). Specifically, metrics which better account for the relative potency of GHGs on a mass-to-mass basis, which are not time associated, such as radiative forcing. However, if comparing on a CO_{2eq} basis is important, comparing on multiple time-horizons e.g., GWP_{20}, GWP_{50}, could be beneficial to relying solely on the GWP_{100}, given the short lifespan of H_2. However, at the time of writing, there are no GWP estimates for H_2 outside of GWP_{100}, but if and when GWP over

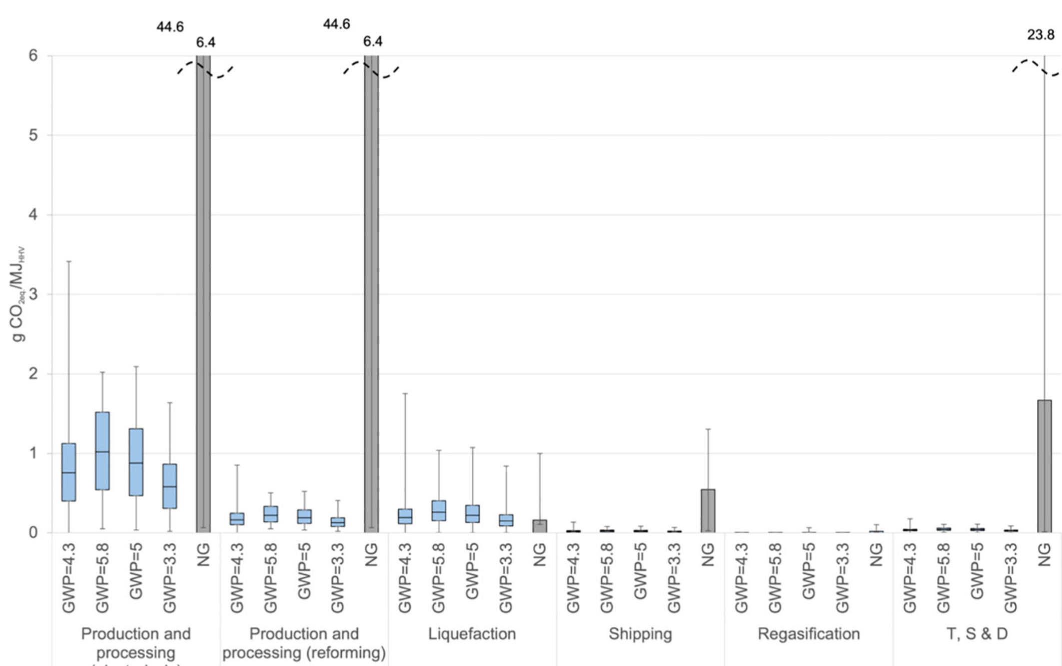

Fig. 4. Impacts to global warming (based on GWP_{100}) of H_2 emissions in comparisons to methane (from natural gas) across comparable stages. T, S & D: transmission, storage and distribution. Methane emissions data are from Alvarez et al. (2018), Balcombe et al. (2015) and Cooper et al. (2021).

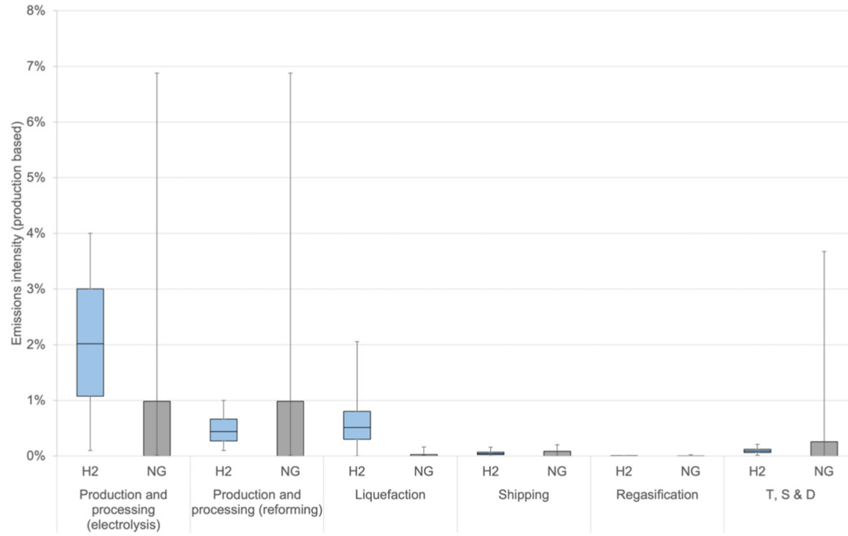

Fig. 5. Emissions intensity, volume of H_2 or methane emitted per volume of H_2 or natural gas produced, of H_2 in comparison to methane emissions from natural gas across comparable stages. T, S & D: transmission, storage and distribution. Methane emissions are from Alvarez et al. (2018), Balcombe et al. (2015) and Cooper et al. (2021).

different time-horizons are established, our results can be updated to reflect these. As H_2 contributes towards global warming by increasing the amount of time methane is present in the atmosphere, it could be as important as methane in a world where significant quantities of H_2 is blue or biomass.

The quantification of the global warming impacts of H_2 is an area that needs further research as only a handful of studies have studied it. Consequentially there is a high degree of uncertainty in the accuracy of the GWP (in comparison to established GHGs such as methane and hydrofluorocarbons). Despite this, in the long term other GHGs emitted along the H_2 supply chain will likely be more important, primarily CO_2 (from any direct or indirect energy use) and methane (for blue and biomass supply chains) because of either their long atmospheric lifetime or GHG potency. Consequentially, given current knowledge, H_2 emissions are highly unlikely to have significant impacts on reaching climate goals, provided emissions are curtailed and the sector regulated in a way akin to natural gas.

3.1.3. Impact relative to methane

For the blue and biomass supply chains, the global warming impact of methane emissions is much larger (Fig. 6). This is because both the GWP and quantity of methane emitted is higher. Therefore, it is important that methane emissions be minimised to reduce the total GHG emissions from these supply chains. If methane emission rates are high, the GHG emissions of blue H_2 could be high and less favourable for decarbonisation (Howarth and Jacobson, 2021). Given that H_2 enhances the global warming impacts of methane, cuts to H_2 emissions in blue and biomass H_2 supply chain could be as important as cuts to methane, if large cuts in GHGs (in CO_{2eq}) are needed imminently to ensure we do not overshoot 2°C or 1.5°C temperature goals.

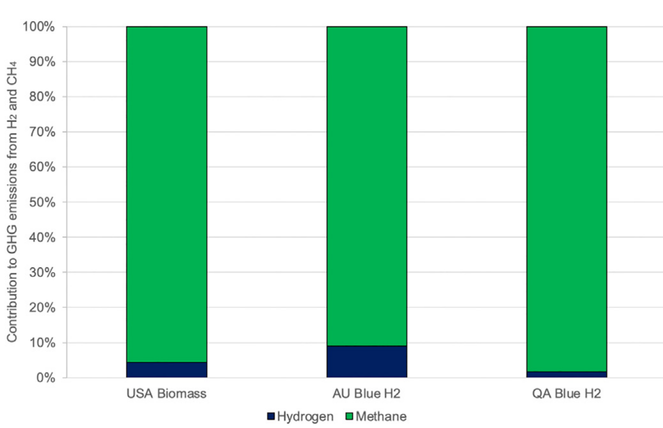

Fig. 6. Comparison of CO_{2eq} impacts of H_2 to methane in the blue and biomass supply chains. AU: Australia; QA: Qatar.

3.2. Sensitivity and uncertainty analysis

There is a significant lack of H_2 specific loss and emissions data and thus we conducted a sensitivity analysis to gauge the impacts of uncertainties in our data and modelling. We found that the production and liquefaction stages are the most important (Fig. 7) as they account for most of the uncertainties in the emission estimates and are therefore the main drivers of emissions. This indicates that these stages require the most urgent collection of data. Conversely, the downstream stages have far smaller emissions with much less impact on total supply chain emissions. The loss rates were also examined in a similar way. These showed the same areas were key to the overall loss rate and require further analysis.

3.3. Quality assessment results

Our literature quality assessment found that the majority of the data collected is low quality (Table 3). This was expected given that no large scale H_2 measurement studies have been conducted as far as the authors are aware. Overall, the production and processing stages have high levels of uncertainty, combined with high estimated emission intensities making them crucial areas for further study. Many other areas have low levels in quality, meaning significant sections of the supply chain require urgent research to determine the emission routes. The only stages with high quality data are transport (high pressure transmission) and distribution as emissions from natural gas pipelines are better understood and more studied than natural gas liquefaction, storage, regasification and loading/unloading. However, this is not to say focus should be shifted away from these stages in H_2 emission measuring and monitoring campaigns. The use of natural gas as a proxy for H_2 is not a substitute for actual H_2 emission measurements.

3.4. Limitations

There is a high level of uncertainty and reliability in our results (emissions intensities and CO_{2eq} estimates) largely due to the significant lack in data available on H_2 emission rates and losses. We have relied heavily on using natural gas supply chains emissions as a proxy and while there will be similarities between the two, they are not substitutes for one another.

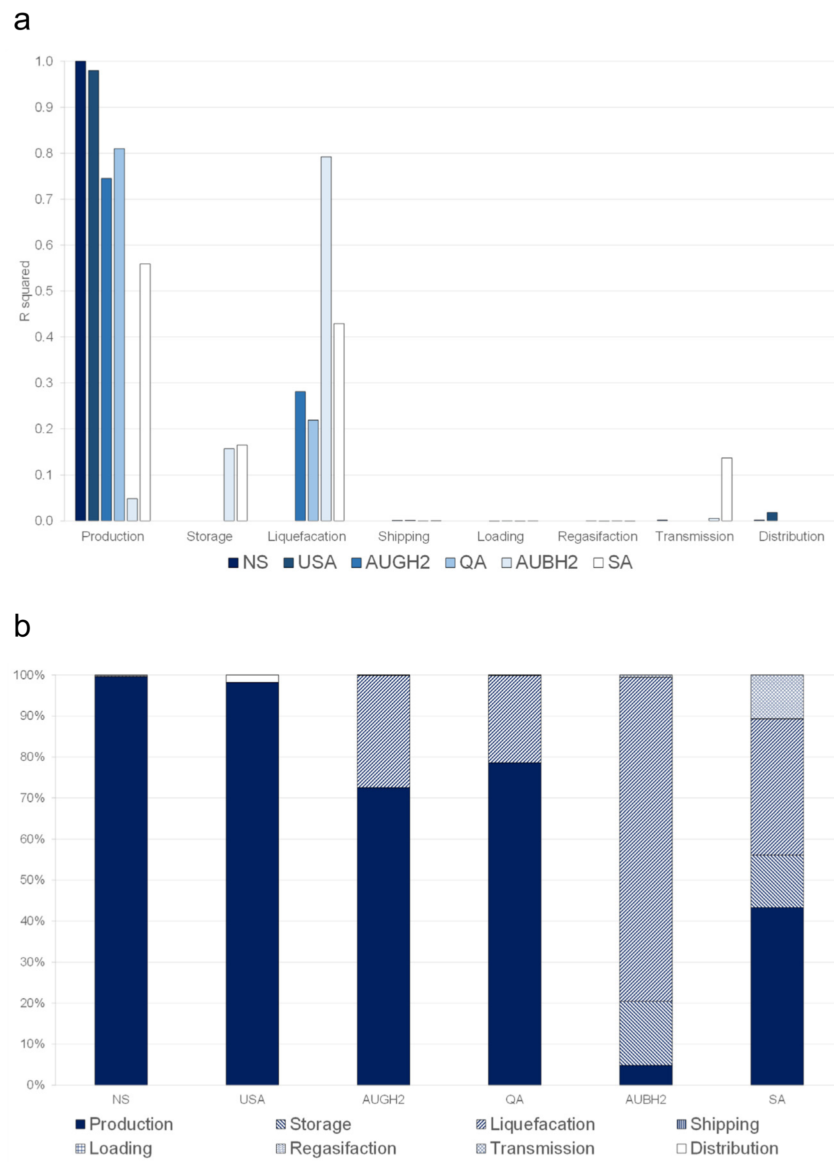

Fig. 7. Sensitivity analysis to determine which supply chain stage is the most influential on the emission estimates. a) top- R squared values per supply chain stage. b) bottom- contribution of each supply chain stage to emissions uncertainty. AUBH: Australia blue H_2; AUGH: Australia green H_2; NS: North Sea electrolysis; QA: Qatar blue H_2; SA: Saudi Arabia; USA: USA biomass.

While emissions in natural gas supply chains are better understood, there is a high degree of uncertainty in the emissions estimated due to factors such as super-emitters, discrepancies in top-down and bottom-up estimates etc. (Alvarez et al., 2018). Also, there is uncertainty around whether H_2 will leak at the same or similar rate as methane/natural gas- H_2 is a much smaller molecule but H_2 has been found to leak at the same rate as natural gas (Hormaza Mejia et al., 2020). Subsequently this is a major weakness of this work. While we have used the best available (open source) data and assumptions in our emission rates and losses modelling, we were unable to account for all emission sources, such as flaring or emissions from pipeline maintenance, in our estimates. As a result, the emissions estimated could be an underestimate as not all emission sources are accounted for but given the lack of H_2 emissions measurement studies, we are unable to verify whether our estimates are an over or underestimate.

Table 3
Literature quality assessment.

Stage	Quality score
Electrolysis	Low
Gasification	Low
SMR/ATR	Low
Processing	Low
Liquefaction	Medium
Storage	Low
Regasification	Low
Loading/unloading	Low
Transport	High
Distribution	High

There is also uncertainty around the GWP of H_2 as there are no (peer-reviewed publicly available) estimates of GWP over different time horizons. As H_2 has a short lifetime, it has been argued that other metrics should be used to estimate the global warming impacts as most of the impacts to global warming induced by H_2 are caused upon release into the atmosphere. In addition to the climate metric, another area of uncertainty in estimating global warming impacts is the predicted quantity of H_2 produced and consumed in future energy systems. We have made our estimation assuming 80 EJ in 2050 but different studies report different values.

Subsequently, our estimate of 9 to 138 Mt CO_{2eq} emitted in 2050 in highly uncertain and should be interpreted with caution.

3.5. Future work

Should H_2 become widely used, further understanding of its emissions are of paramount importance. Our sensitivity analysis and literature quality assessment have highlighted clear areas where there are large data gaps:

1. Production and processing- experiments to calculate the H_2 emissions from large scale electrolysers.
2. Uncombusted H_2 (H_2 slip)- can vary widely depending on engine loads.
3. H_2 storage- potential to be very high emitting, due gas being stored for long periods of time.
4. Loading and unloading- experiments to calculate H_2 emissions from boil-off.
5. Transmission and distribution- experiments which measure leaks in H_2 pipelines (high, medium and low pressure).
6. Climate metric of H_2- estimating the GWP over multiple time horizons calculating other climate metrics, such as GTP and radiative forcing.

Overall, more data on emissions and losses is desperately needed for all stages of the H_2 supply chain, specifically direct measurement data. These are crucial to better characterise and understand the emissions profiles, as well as to verify the results of this study and establish baseline emissions data. As technology advances it is likely that losses and emissions will reduce. Therefore, baseline data is necessary to measure whether emission abatement measures are successful. Also, further work is need in understanding/quantifying the climate change impacts of H_2, such as calculating the GWP over different timeframes, calculating the GWP under scenarios of reduced methane in the atmosphere (likely to occur given recent pledges to cut emissions (Vaughan, 2021)) and calculating other climate metrics.

4. Conclusion

In this paper we have estimated the potential H_2 emissions for a variety of H_2 supply chains. This enabled the CO_{2eq} of these emissions to be calculated and gives an understanding of their global warming impacts. The H_2 emissions were also compared to methane emissions from current gas supply chains to compare leakage rates and impacts to global warming. A literature quality assessment and sensitivity analysis were carried out to determine where future work should be focussed improve future estimates of H_2 emissions from H_2 supply chains.

In summary, we find three main conclusions:

1. If H_2 is used and traded the way natural gas is then emissions (in $CO_{2eq.}$) are considerably smaller (29 (0.6 to 66) times lower) when comparing the two. This is because H_2 has a significantly smaller GWP, and a higher mass energy density meaning a smaller mass needs to be transferred for the same end use and any emissions that do occur have a lesser effect.
2. Up to 138 Mt CO_{2eq} could be emitted in low carbon energy systems in 2050 from H_2 emissions alone. This quantity is small in comparison to estimated total GHG emission in 2050. Therefore, it is likely that CO_2 and non-CO_2 GHG emissions in some, if not all, H_2 supply chains are more important. However, if H_2 emissions are large, then the impacts to climate change become important.
3. There is a significant lack of H_2 emissions and losses literature, as well as its impacts to global warming. It is imperative that future work is carried out to measure losses and emissions from every stage of the supply chain to better understand its emission profile and impacts on global warming, as well to establish accurate and appropriate metrics to measure and compare H_2 to other GHGs.

CRediT authorship contribution statement

Jasmin Cooper: Conceptualization, Methodology, Formal analysis, Investigation, Writing- Original Draft, Writing- Review & Editing, Visualization, Project administration. **Luke Dubey:** Conceptualization, Methodology, Formal analysis, Investigation, Writing- Original Draft, Writing- Review & Editing. **Semra Bakkaloglu:** Conceptualization, Investigation, Writing- Original Draft, Writing- Review & Editing. **Adam Hawkes:** Conceptualization, Writing- Review & Editing, Project administration.

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. Supporting Information (SI)

Supporting information to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.154624.

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