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Assessing Uncertainties of Well-To-Tank Greenhouse Gas Emissions from Hydrogen Supply Chains

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Abstract: Hydrogen is a promising energy carrier in the clean energy systems currently being developed. However, its effectiveness in mitigating greenhouse gas (GHG) emissions requires conducting a lifecycle analysis of the process by which hydrogen is produced and supplied. This study focuses on the hydrogen for the transport sector, in particular renewable hydrogen that is produced from wind- or solar PV-powered electrolysis. A life cycle inventory analysis is conducted to evaluate the Well-to-Tank (WtT) GHG emissions from various renewable hydrogen supply chains. The stages of the supply chains include hydrogen being produced overseas, converted into a transportable hydrogen carrier (liquid hydrogen or methylcyclohexane), imported to Japan by sea, distributed to hydrogen filling stations, restored from the hydrogen carrier to hydrogen and filled into fuel cell vehicles. For comparison, an analysis is also carried out with hydrogen produced by steam reforming of natural gas. Foreground data related to the hydrogen supply chains are collected by literature surveys and the Japanese life cycle inventory database is used as the background data. The analysis results indicate that some of renewable hydrogen supply chains using liquid hydrogen exhibited significantly lower WtT GHG emissions than those of a supply chain of hydrogen produced by reforming of natural gas. A significant piece of the work is to consider the impacts of variations in the energy and material inputs by performing a probabilistic uncertainty analysis. This suggests that the production of renewable hydrogen, its liquefaction, the dehydrogenation of methylcyclohexane and the compression of hydrogen at the filling station are the GHG-intensive stages in the target supply chains.

Keywords: hydrogen; supply chain; hydrogen carrier; life cycle inventory analysis; GHG emissions; probabilistic uncertainty analysis

1. Introduction

Hydrogen has been attracting attention as a clean energy source in part because of its flexibility as an energy carrier; it can be produced from a variety of industrial processes and consumed by a variety of end-users. For example, hydrogen can be produced from hydrocarbon feedstocks via chemical processes (e.g., steam reforming of natural gas (NG) and coal gasification), and by using electricity to power the electroylysis of water [1,2]. "Renewable hydrogen" is produced using renewable energy or electricity sources and can usually be expected to decrease the greenhouse gas (GHG) emissions associated with its use [3,4]. At the other end of the supply chain, hydrogen can be consumed by diverse end-use applications as it can be transformed into kinetic, electric and thermal energy [3,4].

In the transportation sector, for example, hydrogen fuel cell vehicles (FCVs) have received attention globally as one of the next-generation clean vehicles that major motor companies are developing and releasing in the passenger car market [5–7]. Other types of FCVs, such as forklifts [8–10] and buses [11–13], have also been commercialized. Regarding residential energy applications, micro fuel cell combined heat and power (FC-CHP) systems could decrease household energy use owing to their high gross energy efficiencies (the sum of power generation efficiency and heat production efficiency) [14]. Megawatt-class hydrogen energy systems have also been installed around the world in data centers, hotels and many other commercial facilities to combine distributed renewable energy resources—such as solar photovoltaics (PV) and wind turbines—and measures to ensure a stable energy supply—such as battery storage, and water electrolyzers, storage tanks and fuel cells for hydrogen production, storage and use, respectively [15–18]. In terms of power generation, a commercially operated 12 MW hydrogen turbine began operating in Italy in 2009 [19,20].

Given this flexibility, hydrogen is regarded as useful for increasing energy diversity and mitigating GHG emissions in Japan. To support future increases in its supply and demand, various technologies related to the production, transport and storage of hydrogen are being developed in Japan [21]. Liquid hydrogen (LH_2) and methylcyclohexane (MCH) are promising hydrogen carriers that are suitable for long-term storage and long-distance transport owing to their relative ease of handling [22]. LH_2 and MCH have therefore been considered for a private-sector-led national demonstration project focusing on an international hydrogen supply chain that transports hydrogen produced from renewable or excess energy to Japan [23–25].

In parallel with research and development (R&D) surrounding hydrogen technologies, designing the future hydrogen economy requires understanding the potential socio-economic and environmental properties of the hydrogen supply chain. A common approach to help decision makers with the design and development of products and processes is to evaluate their environmental influences throughout their life cycle [26]. Many studies have been published that report life cycle approaches to the hydrogen production process. For example, life cycle inventory (LCI) analyses have been published that calculate the life cycle GHG emissions for hydrogen produced from steam reforming of NG [27–32], coal gasification [29,31,32], and renewably powered electrolysis of water [30–33]. Specifically, Kato [34] took an economic approach and estimated the final cost in Japan of hydrogen produced in South Australia, Norway and the Middle East. Meanwhile, other studies have conducted life cycle sustainability assessments to simultaneously evaluate the economic, social and environmental impacts [35–37].

A number of LCI studies have also been published that compare the environmental benefits of using hydrogen instead of conventional energy sources. Examples of such comparative studies in the automotive sector using Well-to-Wheel (WtW) and Well-to-Tank (WtT) frameworks to estimate GHG emissions have been carried out for Japan, the US and Europe [38–42]. Regarding the use of hydrogen in local energy systems, Usui and Hondo [43] compared the life cycle CO_2 emissions from electricity storage systems for distributed wind power, including hydrogen storage using LH_2 and MCH that was then used in FC-CHP. Mori et al. [44] compared the life cycle environmental emissions from a renewable hydrogen-powered uninterruptible power system (UPS) and a UPS powered by an internal combustion engine. However, it should be noted that most of these LCI studies were conducted based on specific assumptions and the mere use or the combinations of the results in previous studies does not make sense due to the differences of methodologies, system boundaries and databases used in each analysis. These Japanese studies were also conducted under the energy supply and demand structure during the first decade of the 21st century. After the Great East Japan earthquake occurred in 2011, our energy supply structure has drastically changed so that the LCI results should be different if conducted under the current energy supply. As far as the authors recognize, the WtW study conducted by Mizuho Information & Research Institute [45] was the only study that assessed the life cycle GHG profiles of various hydrogen supply chain under the current energy supply but most of the data used for the assessment are not provided in their report. In the same manner as the EU's CertifHy project [46], LCI

analysis should be conducted to understand the environmental profile of the potential international hydrogen supply chain and extract the technical issues related to hydrogen carrier to make the supply chain low-carbon.

Using LCI analysis as a decision-making tool requires considering the range of possible consequences of a given technological pathway [47]. Once the pathway is identified, remaining uncertainty in the LCI is usually attributed to the use of inaccurate or unrepresentative data [48,49]. One method of mitigating these uncertainties is to conduct sensitivity analyses. The ISO 14044 guidelines suggest that sensitivity analyses should include a wide range of factors to determine the influence of variation in assumptions, methods and data [50]. An alternative option, uncertainty analysis, employs probabilistic simulations based on the Monte Carlo method to evaluate the combined influence of multiple uncertain factors on the results. Here, probability distributions are assumed for the system's input parameters. Repeated calculations with different input values then yield a probability frequency distribution of total GHG emissions from the whole system [49]. The application of uncertainty analysis using Monte Carlo simulation and LCI has been demonstrated for WtW emissions [41].

As awareness of unintended and unwanted side-effects has increased, it has become common practice during the early stages of new technology development to carry out ex ante assessments of potential consequences that the widespread implementation of the new technologies may create [51,52]. Most components in the hydrogen supply chain are immature and still developing, which creates uncertainties regarding their real-world performance and could affect the supply chain's total GHG emissions. These uncertainties have not be fully considered in previous Japanese WtW studies [38–40] and addressing this knowledge gap is a key focus of this study.

Foreground inventory data related to the hydrogen supply chains are collected by literature surveys, and the Japanese life cycle inventory database is used as the background data of the analysis. A probabilistic uncertainty analysis of the LCI results is then conducted and the results used to highlight GHG hotspots in the supply chain to promote a discussion of the technical opportunities available for reducing GHG emissions across the different supply chains.

2. Methods and Assumptions

2.1. Overview

Figure 1 shows the processes that are within the study's system boundary. The renewable hydrogen supply chain comprises domestic and overseas stages. The overseas stages are: the generation of renewable power; the production of renewable hydrogen by water electrolysis; the production and storage of the hydrogen carrier (LH_2 or MCH); and the ocean transport of the hydrogen carrier to Japan. The domestic stages comprise: the storage of the hydrogen carrier and its distribution to hydrogen filling stations by tank truck; the release of hydrogen from the energy carrier (restoration) followed by its compression; and finally the filling of FCVs with hydrogen. A reference supply chain of hydrogen produced domestically from the steam reforming of natural gas (NG) was also analyzed. This supply chain comprises: the overseas extraction and liquefaction of NG; the ocean transport of liquefied natural gas (LNG) to Japan; the production of hydrogen by steam reforming of NG which is obtained by LNG regasification; the compression and distribution of hydrogen as compressed gaseous hydrogen (CGH_2) to hydrogen filling stations by tank truck; and finally the compression and filling of FCVs stages.

According to Kato [34], Australia and Norway both have the potential to supply large amounts of low-cost hydrogen using wind power. In Australia, solar PV is also a potential power source for hydrogen production and the feasibilities of exporting solar PV-generated hydrogen from Australia to Japan have been investigated [53,54]. For these reasons, Australia and Norway were chosen as the renewable hydrogen producing countries in this study. The one-way transport distance to Japan was set as 10,000 km from Australia and 20,000 km from Norway.

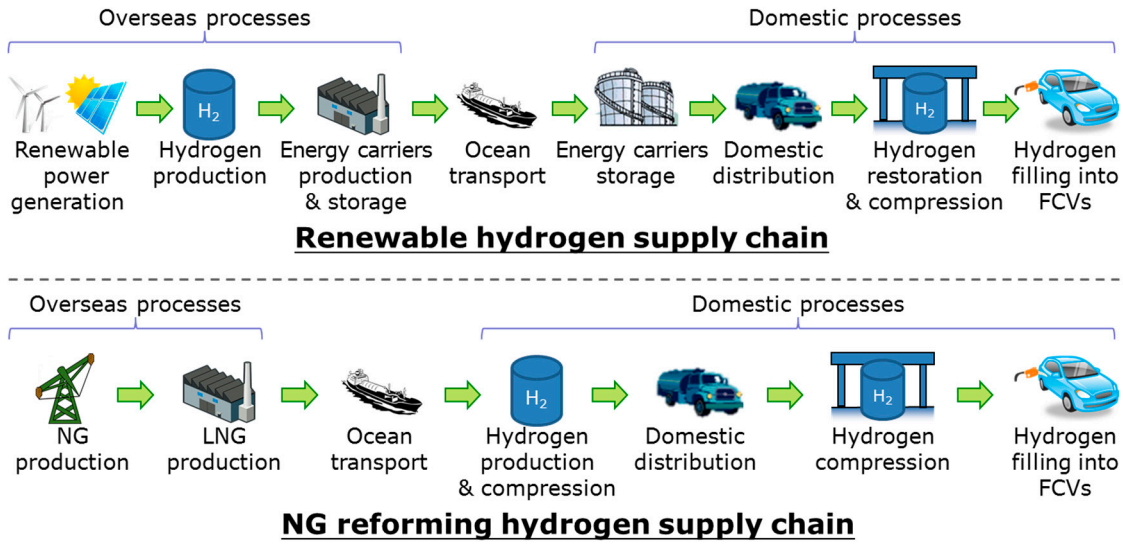

Figure 1. Process flow charts showing stages within the life cycle inventory (LCI) system boundary.

Table 1 shows the case settings configured for hydrogen from Australia. It was typically assumed that the electricity used in overseas processes other than hydrogen production was supplied using the country's standard electricity grid (referred to as "base case" hereafter). Electricity from the Australian grid has a high GHG emissions intensity owing to the grid's strong dependency on coal-fired power generation. Thus, the use of low-carbon electricity, as investigated in the "low-carbon case" where all electricity inputs were from the same low-carbon source, was investigated for its potential to significantly decrease GHG emissions from Australian renewable hydrogen supply chain.

Table 1. The base and low-carbon cases.

Case	Description
Base Case	Electricity for hydrogen production via water electrolysis was supplied with renewable electricity, while the electricity used in the other Australian processes was supplied by Australian grid electricity.
Low-Carbon Case	Electricity used in all the Australian processes was supplied with the same renewable electricity as water electrolysis.

Inventory data were collected from previous WtW and LCI studies on hydrogen supply chains. Where more than two process options were found (owing to differences in equipment specifications), the mean and standard deviation were also calculated. The Japanese Inventory Database for Environmental Analysis (IDEA) ver. 2.0 [55] was used as the background data.

2.2. GHG Emissions Calculation Using IDEA

GHG emissions were defined as the sum of the 100-year CO_2-equivalent global warming potentials of emitted CO_2, CH_4, N_2O, HFCs, PHCs and SF_6 [56].

The IDEA inventory database was developed by National Institute of Advanced Industrial Science and Technology [55]. Figure 2 illustrates IDEA's structure, which uses a unit process to relate input flows (raw materials, energies, and resources) to output flows (products, wastes, emissions to air, water and soil) [57].

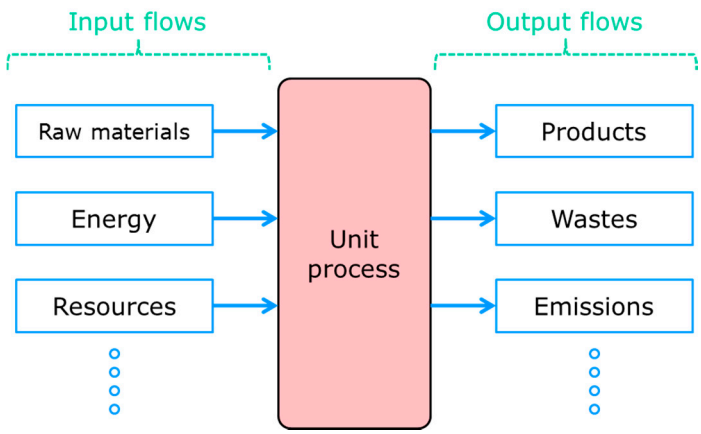

Figure 2. Data structure of Inventory Database for Environmental Analysis (IDEA) [57].

The GHG emissions of a product were calculated using IDEA as follows. First, a matrix (the IDEA input coefficient table) was configured by tabulating the input flows. Each element of the table (a_{ij}) represented the amount of product i required to produce one unit of product j. Letting x_i and f_i be the amount of production and final demand of product i, respectively, the supply–demand balance for product i could be expressed using Equation (1):

$$\boldsymbol{A}\boldsymbol{x} + \boldsymbol{f} = \boldsymbol{x} \tag{1}$$

where $\boldsymbol{A} = \begin{pmatrix} a_{11} & \cdots & a_{1n} \\ \vdots & \ddots & \vdots \\ a_{n1} & \cdots & a_{nn} \end{pmatrix}$; $\boldsymbol{x} = \begin{pmatrix} x_1 \\ \vdots \\ x_n \end{pmatrix}$; and $\boldsymbol{f} = \begin{pmatrix} f_1 \\ \vdots \\ f_n \end{pmatrix}$.

Then, the amount of i produced (x_i) was obtained using a matrix operation. Here, the inverse matrix was approximated by the sum of the power series, as shown in Equation (2):

$$\boldsymbol{x} = (\boldsymbol{I} - \boldsymbol{A})^{-1}\boldsymbol{f} = \left(\boldsymbol{I} + \boldsymbol{A} + \boldsymbol{A}^2 + \boldsymbol{A}^3 + \cdots\right)\boldsymbol{f} \tag{2}$$

where $\boldsymbol{I}$ denotes the $n \times n$ identity matrix.

By defining e_i as the GHG emissions from the unit process for product i, the embodied GHG emissions were then obtained from Equation (3):

$$GHG = \boldsymbol{E}\boldsymbol{x} \tag{3}$$

where $\boldsymbol{E} = (e_1 \ \cdots \ e_n)$.

IDEA ver. 2.0, released in May 2016, includes inventory data of more than 3800 unit processes. Because it covers all of the items in the Japan Standard Commodity Classification, the GHG emissions of any product produced in Japan in the year 2014 are included in IDEA. However, IDEA's embodied GHG emissions data are limited to Japanese economic activities and thus require assumptions to approximate the emissions attributed to Australian or Norwegian economic activity. For estimating overseas activity using IDEA, a new matrix $\boldsymbol{A}'$ was created and used in Equation (2) instead of $\boldsymbol{A}$ $\boldsymbol{A}'$ was configured from the IDEA input coefficient table $\boldsymbol{A}$ by employing the following steps:

- All of the elements of $\boldsymbol{A}'$ were set equal to those in $\boldsymbol{A}$. This assumed that the economic activity in Australia and Norway was the same as that in Japan. (For example, if the same product is produced in Japan and Australia, it requires the same inputs in both countries).
- The $\boldsymbol{A}'$ elements for oversea transport of resources were set to zero to ensure that the approximated emissions of a product did not include those generated by overseas transport of the constituent inputs.

- The A' elements referring to grid electricity in Japan were changed to reflect grid electricity in Australia or Norway.

2.3. Renewable Power Generation in Hydrogen-Producing Country

Renewable power plants (wind and solar PV in Australia, and wind in Norway) were assumed to provide dedicated power to parts of the hydrogen supply chains. This meant that the power plants' life cycle GHG emissions (for example, those associated with materials production, construction, transport and operation) were then used whenever renewable electricity was consumed.

The input data used by Imamura et al. [58,59] were used to calculate life cycle GHG emissions using IDEA. The assumptions made by Imamura et al. [58,59] were to evaluate the Japanese renewable power plants. In order to reflect the wind conditions and insolation in hydrogen producing countries, the Australian and Norwegian load factors for wind and solar PV [34,60] were used in our calculation. Table 2 shows the specifications of wind and solar PV power plants assumed in this study while Table 3 shows the calculated life cycle GHG emissions of wind and solar PV power generation. The reason for the large difference in Australian and Norwegian wind power emissions could be attributed to the GHG emissions intensity divergence in grid electricity of the two countries.

Table 2. Specifications of wind and solar PV power plants used in the model.

Parameter	Wind	Solar PV
Capacity (MW)	40	10
Load factor (%)	35	18
Auxiliary power ratio (%)	10	3
Amount of electricity generated (MWh/year)	122,640	14,532
Life time (year)	30	30

Table 3. Calculated life cycle GHG emissions for wind and solar PV power generation (g-CO_2eq./kWh).

Hydrogen-Producing Country	Wind	Solar PV
Australia	15.3	66.4
Norway	10.8	n/a

2.4. Renewable Hydrogen Production by Water Electrolysis

Water electrolysis is a popular method of producing hydrogen which has been known for around 200 years [61]. Hydrogen was assumed to be produced using renewable power via electrolysis using either polymer electrolyte membranes or alkaline water. The mean electricity input was calculated as 5.19 kWh/Nm^3-H_2 [62–74]. The pure water requirement was set to the theoretical value of 0.804 kg/Nm^3-H_2.

2.5. Hydrogen Energy Carriers

2.5.1. Liquid Hydrogen (LH_2)

Hydrogen liquefaction has long been the preferred method of increasing hydrogen density for transport as the liquid has approximately one eight-hundredth of the volume of gaseous hydrogen [22]. This preference is despite hydrogen having the second lowest boiling point (−253 °C) after helium and the large amount of electricity that the process requires. Figure 3 shows the renewable hydrogen supply chain using LH_2 that was modeled in this study. Assumptions for each of the supply chain components are described below.

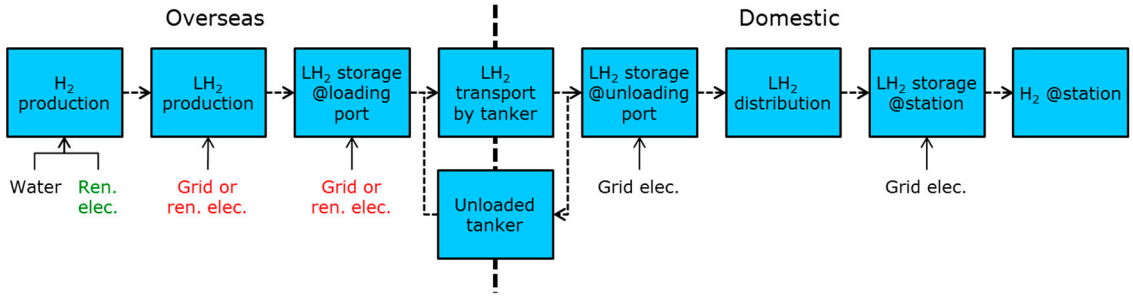

Figure 3. Renewable hydrogen supply chain using LH_2. Only renewable electricity was used for hydrogen production (expressed in green font). For electricity inputs for production and storage of liquid hydrogen (red font), the base cases assumed the input was equal to grid electricity. However, for the Australian low-carbon cases, the same renewable electricity input was assumed as that used for hydrogen production.

- LH_2 production

LH_2 was produced by cooling gaseous hydrogen. The mean electricity input for LH_2 production was calculated as 0.906 kWh/Nm^3-H_2 [65,66,75–78].

- LH_2 storage at loading port

LH_2 was stored in stationary insulation tanks at a loading port. It was assumed that the boil-off of hydrogen gas during this stage was subsequently re-liquefied. The electricity input for LH_2 storage at the loading port was set to 0.055 kWh/Nm^3-H_2 [39].

- LH_2 ocean transport by tanker

A LH_2 tanker (160,000 m^3 tank capacity, 16 knots sailing speed) [66] was assumed for transport of LH_2 to Japan. However, because LH_2 tankers are still being developed, exact data were not available. Thus, the emissions data were estimated using data for LNG tankers in IDEA under the assumption that the GHG emissions of a LH_2 tanker per transport volume of LH_2 expressed in ton-kilometer unit were equal to those for a LNG tanker per ton-kilometer of LNG. The emissions from laden and ballast voyages were both included in the estimation. The mean boil-off rate of LH_2 during the voyage was calculated as 0.3%/day [65,66].

- LH_2 storage at unloading port

LH_2 was assumed to be transferred from the tanker to stationary tanks at an unloading port. It was assumed that the gas resulting from hydrogen boil-off was subsequently re-liquefied. The electricity input for LH_2 storage at the unloading port was set to 0.055 kWh/Nm^3-H_2 [39].

- Domestic distribution of LH_2 by tank truck

LH_2 tank trucks (23 kL tank capacity, 3.5 km/L-diesel oil fuel economy) [45,79] were assumed to be responsible for the domestic distribution from the LH_2 storage terminal to the hydrogen filling stations. The one-way distance was set to 50 km [66], and both the emissions by the laden and empty journeys were included to the inventory.

- LH_2 storage at hydrogen filling stations

LH_2 was assumed to be transferred from tank trucks to stationary tanks at hydrogen filling stations. The electricity input required for LH_2 storage at a filling station was set to 0.055 kWh/Nm^3-H_2 [39].

2.5.2. Methylcyclohexane (MCH)

MCH ($CH_3C_6H_{11}$) is another promising hydrogen carrier. It is produced by the hydrogenation of toluene (TOL; $CH_3C_6H_5$) and releases hydrogen via catalytic dehydrogenation:

$$CH_3C_6H_5 + 3H_2 \rightleftharpoons CH_3C_6H_{11} \quad (4)$$

MCH was first proposed as a hydrogen storage system for use as a vehicle fuel in 1980 [80] under what is now known as the methylcyclohexane-toluene-hydrogen (MTH) system [81,82]. After hydrogenation of TOL at the hydrogen supply site, MCH is transported by tanker and dehydrogenated at the demand site to yield H_2 and the original TOL, which is then returned to the supply site and reused [83]. The dehydrogenation of MCH to TOL is important to the process owing to its large endothermic heat of reaction (205 kJ/mol-MCH = 68.3 kJ/mol-H_2) [84]. A number of catalysts, including those that are platinum [85–88], palladium [88–90] and nickel [91,92] based, have been investigated for their ability to facilitate an efficient dehydrogenation of MCH. Figure 4 shows the renewable hydrogen supply chain using MCH that was assumed in this study.

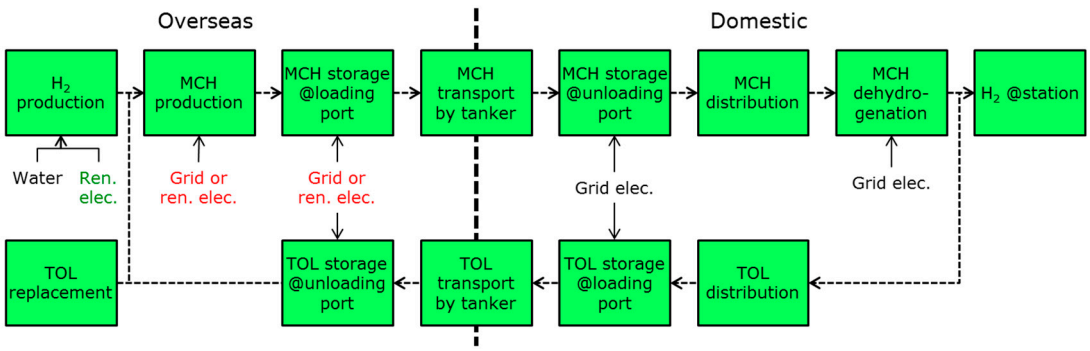

Figure 4. Renewable hydrogen supply chain using MCH. Only renewable electricity was used for hydrogen production (expressed in green font). For electricity inputs for production and storage of MCH and TOL (red font), the base cases assumed the input was equal to grid electricity. However, for the Australian low-carbon cases, the same renewable electricity input was assumed as that used for hydrogen production.

- MCH production

MCH was produced by the chemical reaction between TOL and hydrogen. The reaction yield of hydrogen addition to TOL and the hydrogen consumption rate were set to 99.8% and 97.9%, respectively [65]. The mean electricity input for MCH production was calculated as 40.68 kWh/t-MCH [65,66,93].

- MCH storage at loading port

MCH was assumed to be stored in cone roof tanks at an overseas loading port. The mean electricity input for MCH storage at the loading port was calculated as 0.915 kWh/t-MCH [65,66].

- MCH ocean transport by tanker

A chemical tanker was assumed to be used for the ocean transport of MCH to Japan. The associated GHG emissions were calculated using IDEA's emissions data for a chemical tanker.

- MCH storage at unloading port

MCH was assumed to be transferred from the chemical tanker to cone roof tanks at a domestic unloading port. The electricity input for MCH storage at the unloading port was assumed to be the same as that required for storage at the loading port.

- Domestic distribution of MCH/TOL by tank trucks

Tank trucks (20 kL tank capacity, 2.34 km/L-diesel oil fuel economy) [45,79] were assumed to be used for the domestic distribution of MCH to and TOL from the hydrogen filling stations. The one-way distance was set as 50 km [66], and both the emissions associated with the MCH- and TOL-laden trucks were included in the inventory.

- Dehydrogenation of MCH at hydrogen filling stations

At the hydrogen filling stations, MCH was assumed to be converted to hydrogen and TOL by the dehydrogenization reaction. The conversion rate, selectivity and hydrogen yield of the reaction were set to 95.0%, 99.9% and 90.0%, respectively [65]. It was assumed that heat required by the dehydrogenization was supplied by combustion of city gas. The mean electricity input for MCH dehydrogenization was calculated as 0.310 kWh/Nm^3-H_2 [65,66,93].

- TOL storage at loading port

TOL was assumed to be stored in cone roof tanks at a domestic loading port. The mean electricity input for TOL storage at the loading port was calculated as 0.915 kWh/t-TOL [65,66].

- TOL ocean transport by tanker

A chemical tanker was assumed for transport of TOL from Japan. The associated GHG emissions were calculated using IDEA's emissions data for a chemical tanker.

- TOL storage at unloading port

TOL was assumed to be transferred from the chemical tanker to cone roof tanks at a domestic unloading port. The electricity input for TOL storage at the unloading port was assumed to be the same as the input at the loading port.

- TOL replacement

Owing to unwanted side chemical reactions (including demethylation, isomerization, cycloreversion and dimerization) that also occur during the hydrogenation and dehydrogenation stages, a portion of the TOL must be replaced with virgin TOL to maintain the MTH cycle efficiency [94]. It was assumed that 3% of the initial TOL-loading was replaced every year.

2.6. Supply Chain for Hydrogen Produced by Natural Gas (NG) Reforming

Figure 5 shows the supply chain for hydrogen produced by NG reforming assumed in this study. IDEA's GHG emissions associated with the combustion of LNG were used to calculate the emissions associated with the production of NG, its compression to produce LNG and LNG transport by tanker to Japan. Other stages in the supply chain are described below.

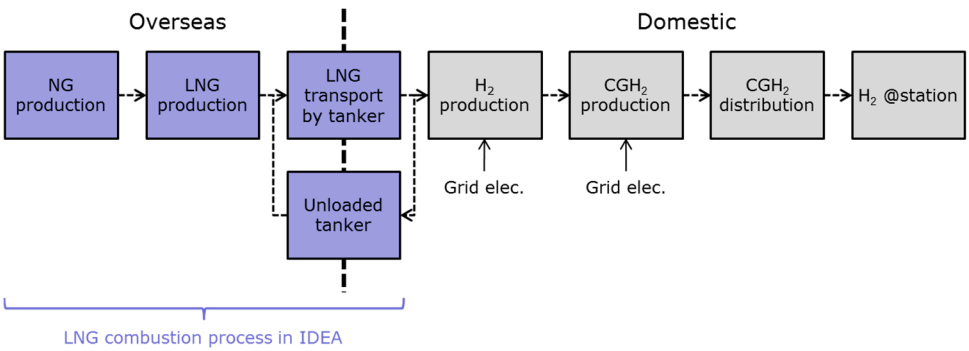

Figure 5. Supply chain for hydrogen production by NG reforming.

- Hydrogen production via steam reforming of NG

Gaseous hydrogen was assumed to be produced in Japan by reforming NG derived from imported LNG. The mean NG and electricity inputs were calculated as 15.04 MJ(LHV)/Nm^3-H_2 and 0.211 kWh/Nm^3-H_2, respectively [38,39,95–97]. The process water requirement was set to 1.964×10^{-3} m^3/Nm^3-H_2 [68,97].

- Hydrogen compression at production plant

The produced hydrogen was assumed to be compressed to 20 MPa and loaded into gas tank trucks for distribution. The electricity input for the compression stage at the production plant was set to 0.272 kWh/Nm^3-H_2 [39].

- Domestic distribution of CGH_2 by tank trucks

Gas tank trucks were assumed to carry out the domestic distribution of CGH_2. The mean tank capacity and fuel economy of the trucks were calculated as 2330 Nm^3-H_2 and 2.75 km/L-diesel oil, respectively [40,98]. The one-way distance was set to 50 km [66], and both the emissions associated with laden and empty trucks were included to the inventory.

2.7. Hydrogen Compression and Filling of FCVs at Hydrogen Filling Stations

At the hydrogen filling stations, hydrogen was assumed to be compressed to a pressure of 70 MPa before the FCVs were filled. The electricity consumption for the compression and filling activities were set to 0.282 kWh/Nm^3-H_2 and 0.093 kWh/Nm^3-H_2, respectively [39].

2.8. Uncertainty Analysis

A range of parameters (listed in Table A1) were considered as part of the uncertainty analysis carried out on the WtT GHG emissions resulting from the various hydrogen supply chains. To determine the probability distribution for each parameter, the WtW analysis methodology adopted by General Motors et al. [95] was applied. Most parameters were assumed to follow a normal distribution curve according to the mean and standard deviation values presented in Table A1. Based on the literature values for inventory data quality, the standard deviation of GHG emissions attributed to renewable power generation was assumed to be 10% of the mean value [99,100].

As for MCH production, hydrogen production by NG steam reforming and hydrogen compression at the NG steam reforming plant, electricity input might show negative if a normal distribution was assumed for the parameters. Hence, in these processes triangle distribution functions using the mean, minimum and maximum data in the inventory were assumed instead of a normal distribution.

Monte Carlo simulations were performed using Oracle Crystal Ball [101]. A total of 10,000 iterations was used for each simulation to ensure the analysis was reproducible [102].

3. Results and Discussion

3.1. WtT GHG Emissions for Hydrogen Carriers

3.1.1. Liquid Hydrogen (LH_2)

Figure 6 shows the mean WtT GHG emissions from the renewable hydrogen supply chains using LH_2. The emissions from the NG reforming hydrogen supply chain are also shown for reference. Except for the Australian solar PV base case, all supply chains that combined LH_2 with renewable power-produced hydrogen showed lower GHG emissions than those from reforming of NG.

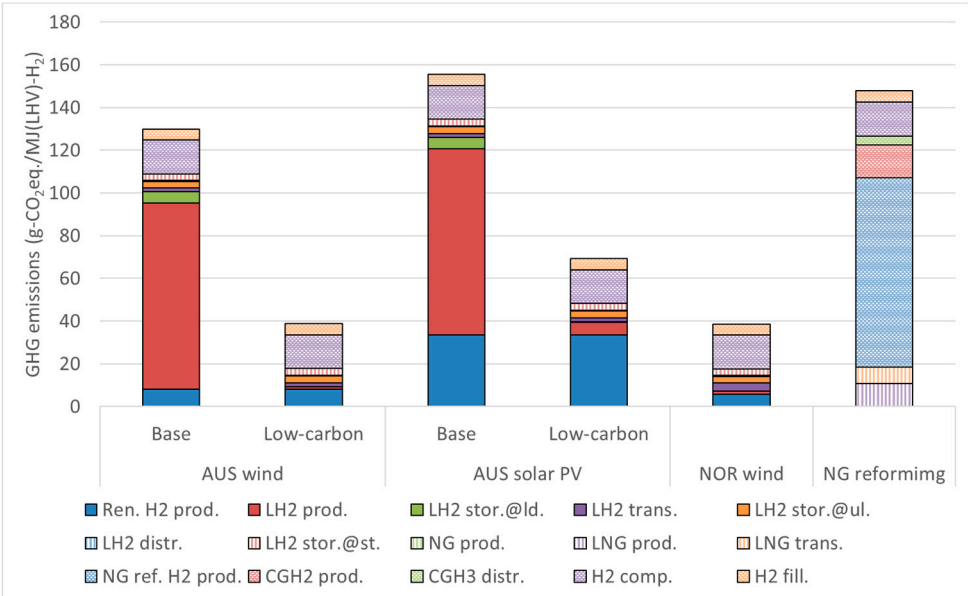

Figure 6. Mean WtT GHG emissions for renewable hydrogen supply chains using LH_2 as the hydrogen carrier.

In the Australian base cases, cooling hydrogen to produce LH_2 was responsible for the largest portion of GHG emissions, accounting for 67% and 56% of the total for the wind and solar PV supply chains, respectively. This is explained by considering the large electricity consumption of the hydrogen liquefaction process owing to hydrogen's very low boiling point (−253 °C). Possible options for decreasing electricity consumption include improving process efficiency by implementing new liquefiers [103,104] or combining the liquefaction with a LNG re-gasification process [105]. Alongside decreasing the electricity requirement, the associated GHG emissions could be decreased by employing a low-carbon power source: using wind and solar PV power in Australia rather than grid electricity decreased the total GHG emissions by 70% and 55%, respectively.

Other GHG hotspots of the LH_2 supply chain were the original hydrogen production step and hydrogen compression at domestic filling stations. Continued R&D for technologies related to the components in water electrolysis—such as electrodes, electrolytes and membranes—is important in further decreasing the carbon-intensity of renewable hydrogen production [106,107]. Meanwhile, employing domestic renewable energy sources to power the hydrogen compression process at the filling stations could decrease GHG emissions from this stage.

Regarding the NG reforming hydrogen supply chain, hydrogen production via steam reforming of NG was the most GHG-intensive process which account for 60% of the total GHG emissions.

3.1.2. Methylcyclohexane (MCH)

Figure 7 shows the mean WtT GHG emissions from the renewable hydrogen supply chains that used MCH as the hydrogen carrier. The associated emissions for hydrogen produced by NG reforming are again also shown for reference. All of the WtT GHG emissions presented lie in the range 130–170 g-CO_2eq./MJ(LHV)-H_2 which indicates that the GHG emissions were relatively insensitive to both where the hydrogen was produced and the renewable power technology employed. Unlike the large decreases observed for hydrogen supplied using LH_2, the WtT GHG emissions for the low-carbon cases for Australian wind and solar PV power were only 5% and 4%, respectively, lower than the emissions in the base case.

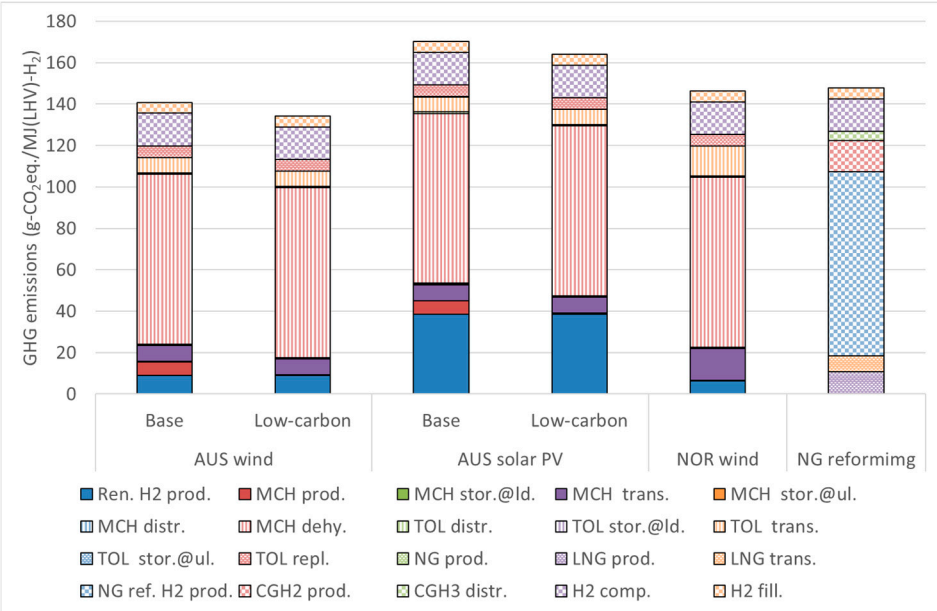

Figure 7. Mean WtT GHG emissions from the renewable hydrogen supply chains that use MCH as the hydrogen carrier.

For all cases, the dehydrogenation of MCH at the hydrogen filling stations was responsible for the largest portion—approximately half—of the total GHG emissions. Because the dehydrogenation of MCH is considerably endothermic—and therefore energy-intensive—the energy source and process efficiency for this step have a large influence on the supply chain's overall life cycle emissions. Given that it was assumed that the heat required for the dehydrogenation was obtained by the combustion of city gas, this stage's GHG emissions could be decreased by using waste heat from nearby plants. For example, if all of this heat could be supplied from waste sources, the total emissions for the Australian base cases for wind and solar PV power could be 58 and 48%, respectively, lower than when burning city gas. Notably, Cresswell and Metcalfe demonstrated a MCH dehydrogenation system in which the reaction heat was supplied by waste heat from a solid oxide fuel cell [108]. R&D that focuses on improving the dehydrogenation catalysts is also important for advancing the process efficiency [109,110]. In this manner, catalysts that facilitate the dehydration using low-quality waste heat have been reported by Chaouki and Klvana [111] and Hodoshima et al. [112].

As with the LH_2 process, the GHG emissions attributed to the initial production of renewable hydrogen and the compression of hydrogen at filling stations were also found to have a moderate impact on the total. Finally, the two-way long-distance ocean transport (of MTH and TOL) was particularly notable in the Norwegian case where the combined emissions attributed to ocean transport of MTH and TOL accounted for 20% of the total WtT GHG emissions.

3.2. WtT GHG Emissions Based on Uncertainty Analysis

Figure 8 shows the uncertainty analysis results for the WtT GHG emissions from renewable and NG reforming hydrogen supply chains. From the comparison of all results and their variability analyses, it is clear that the GHG emissions from renewable hydrogen using LH_2 as the hydrogen carrier are significantly lower than those from hydrogen produced by NG reforming for the Australia low-carbon cases and the Norway case. Conversely, no significant decrease was observed for the MCH cases. Nonetheless, with future technical developments, such as those described in the previous section, the GHG emissions from the MCH process are expected to decrease.

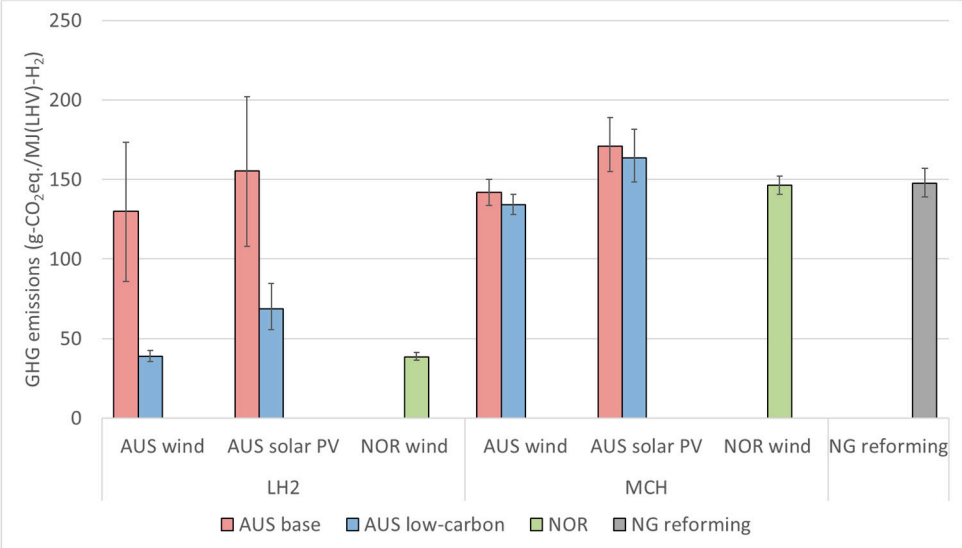

Figure 8. Uncertainty analysis results for WtT GHG emissions from renewable and NG reforming hydrogen supply chains. Bar graphs show the mean values; error bars represent the 95% confidence interval. Probability histograms of the WtT GHG emissions obtained by Monte Carlo simulation are shown in Appendix B.

4. Conclusions

To better understand the potential role of hydrogen energy in decreasing GHG emissions in Japan, a WtT LCI analysis was carried out for renewable hydrogen supply chains originating in Australia and Norway. Wind- or solar PV-powered electrolysis generated hydrogen that was transported to Japan using a hydrogen carrier (LH_2 or MCH), before being distributed to domestic hydrogen filling stations where the hydrogen was restored and pressurized for FCV applications. Data were drawn from literature surveys and IDEA, the Japanese life cycle inventory database. A Monte Carlo-based uncertainty analysis was performed to investigate the impact of variations in the supply chain's energy and material inputs. The LCI analyses showed that the initial hydrogen production, its liquefaction to produce LH_2, the dehydrogenation of MCH and the compression of hydrogen at the filling stations were particularly GHG-intensive activities in the respective supply chains. A number of technological options to decrease GHG emissions in these areas were discussed, as summarized in Figure 9. With a 95% confidence interval, renewable hydrogen produced in Australia (low-carbon cases) or Norway and transported to Japan as LH_2 exhibited significantly lower WtT GHG emissions than those calculated for hydrogen produced by NG reforming. Although the technology options modeled herein suggested the MCH pathway exhibited similar GHG emissions to those from the NG reforming production supply chain, the uncertainty analysis results suggested that feasible technical developments could result in significantly lower emissions from the MCH pathway.

This study mainly focused on the GHG emissions of hydrogen produced from renewable energy resources. However, other potentially low-carbon supply chains exist, such as hydrogen produced from gasification of lignite combined with carbon capture and storage (CCS), which shall be evaluated in future work.

Similarly, we note that extending the LCI analysis to include economic, social and other environmental impacts as part of multiple criteria analyses will also be increasingly important for the deliberation of hydrogen's potential role as an energy medium.

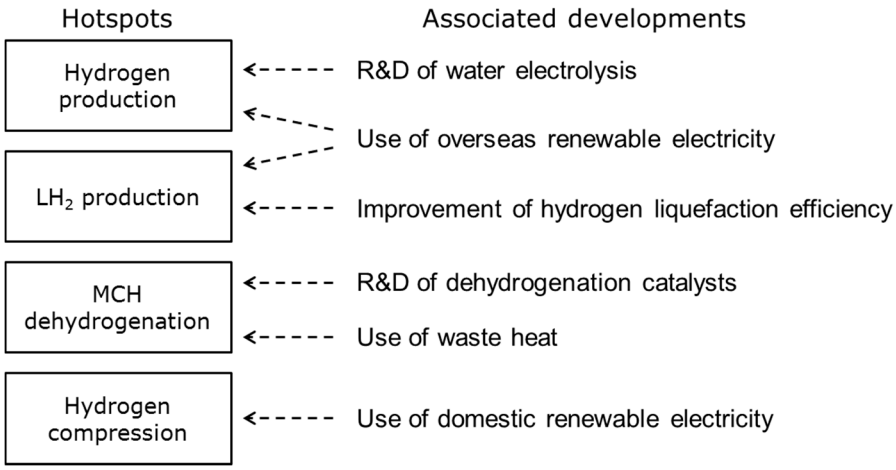

Figure 9. GHG hotspots in the hydrogen supply chains, and associated developments that could decrease GHG emissions.

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Author Contributions: Akito Ozawa performed the LCI and uncertainty analyses and, together with Mai Inoue, interpreted the results and wrote the manuscript under the supervision of Yuki Kudoh and Yutaka Genchi. Naomi Kitagawa, Ryoji Muramatsu and Yurie Anzai helped collect inventory data. All authors approved the final version of the manuscript.

Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

AUS	Australia
CGH_2	compressed gaseous hydrogen
FC-CHP	fuel cell combined heat and power unit
FCVs	fuel cell vehicles
GHG	greenhouse gas
IDEA	Inventory Database for Environmental Analysis
ISO	International Organization for Standardization
LCI	life cycle inventory
LHV	lower heating value
LH_2	liquid hydrogen
LNG	liquefied natural gas
MCH	methylcyclohexane
MTH	methylcyclohexane-toluene-hydrogen
NG	natural gas
NOR	Norway
PV	photovoltaics
R&D	research and development
TOL	toluene
UPS	uninterruptible power system
WtT	Well-to-Tank
WtW	Well-to-Wheel

Appendix A

Table A1. The probability distribution function, mean, standard deviation (SD), minimum (min.) and maximum (max.) values for various uncertainty parameters.

Parameter (Unit)	Function	Mean	SD	Min.	Max.	Reference
GHG emissions for wind power generation in Australia (g-CO_2eq./kWh)	Normal	15.3	1.53			[34,58–60,99,100]
GHG emissions for solar PV power generation in Australia (g-CO_2eq./kWh)	Normal	66.4	6.64			[34,58–60,99,100]
GHG emissions for wind power generation in Norway (g-CO_2eq./kWh)	Normal	10.8	1.08			[34,58–60,99,100]
Electricity input for hydrogen production via water electrolysis (kWh/Nm^3-H_2)	Normal	5.19	0.94			[62–74]
Electricity input for liquefaction of gaseous hydrogen to produce LH_2 (kWh/Nm^3-H_2)	Normal	0.906	0.233			[65,66,75–78]
LH_2 Boil-off rate during ocean transport (%/day)	Normal	0.3	0.1			[65,66]
Electricity input for MCH production (kWh/t-MCH)	Triangular	40.68	37.62	7.54	93.30	[65,66,93]
Electricity input for MCH storage at loading/unloading port (kWh/t-MCH)	Normal	0.915	0.085			[65,66]
Electricity input for MCH dehydrogenization (kWh/Nm^3-H_2)	Normal	0.310	0.047			[65,66,93]
Electricity input for TOL storage at loading/unloading port (kWh/t-TOL)	Normal	0.915	0.085			[65,66]
NG input for hydrogen production via NG steam reforming (MJ(LHV)/Nm^3-H_2)	Normal	15.04	0.24			[95,97]
Electricity input for hydrogen production via NG steam reforming (kWh/Nm^3-H_2)	Triangular	0.211	0.110	0.100	0.323	[95,97]
Electricity input for hydrogen compression at NG steam reforming hydrogen production plant (kWh/Nm^3-H_2)	Triangular	0.272	0.135	0.119	0.440	[39]
Tank capacity of CGH_2 tank trucks (Nm^3-H_2)	Normal	2,330	130			[40,98]
Fuel economy of CGH_2 tank trucks (km/L-diesel oil)	Normal	2.75	0.25			[40,98]

Table A2. Electricity input for hydrogen production via water electrolysis; individual values of inventory data from each literature are shown side by side.

Reference	Electricity (kWh/Nm^3-H_2)						
Asaoka et al. [62]	4.5	5.1	4.85				
Hydrogenics [63,64]	5.4	4.9	6.7				
IAE (2010) [65]	6.5	4.3	5.3				
IAE (2016) [66]	4.8						
Japan Petroleum Energy Center [67]	4.8	5.3	6.5	4.5			
Kawasaki Heavy Industries [68]	4.8						
Kobelco Eco-Solutions [69]	6.5						
Mitsusima and Matsuzawa [70]	4.3	4.6	3.9	4.2	4.6	4.8	4.3
	4.3	4.5	4.7	4.4	5.8	6.0	4.5
	4.5	3.9	3.7	5.6	5.2		
NEDO [71]	4.3	5.3	6.5				
NREL [72]	6.3	6.1					
Proton On-Site [73,74]	7.3	7.0	6.8	6.2			
Mean	5.19						
Standard deviation	0.94						

Table A3. Electricity input for liquefaction of gaseous hydrogen to produce LH_2; individual values of inventory data from each literature are shown side by side.

Reference	Electricity (kWh/Nm^3-H_2)			
IAE (2010) [65]	1.059	0.958	0.900	
IAE (2016) [66]	0.550	0.690		
IAE et al. [75]	0.970			
JRC [76]	0.900			
JHFC [77]	1.259			
Kawasaki Heavy Industries [78]	1.079	1.259	0.692	0.555
Mean	0.906			
Standard deviation	0.233			

Table A4. LH_2 boil off rate of liquid hydrogen while ocean transport.

Reference	Boil off Rate (%/day)
IAE (2010) [65]	0.4
IAE (2016) [66]	0.2
Mean	0.3
Standard deviation	0.1

Table A5. Electricity input for MCH production.

Reference	Electricity (kWh/t-MCH)
IAE (2010) [65]	7.54
IAE (2016) [66]	93.3
Hondo et al. [93]	21.2
Mean	40.68
Standard deviation	37.62

Table A6. Electricity input for MCH storage at loading/unloading port.

Reference	Electricity (kWh/t-MCH)
IAE (2010) [65]	0.83
IAE (2016) [66]	1.00
Mean	0.915
Standard deviation	0.085

Table A7. Electricity input for MCH dehydrogenization.

Reference	Electricity (kWh/Nm^3-H_2)
IAE (2010) [65]	0.335
IAE (2016) [66]	0.352
Hondo et al. [93]	0.244
Mean	0.310
Standard deviation	0.047

Table A8. Electricity input for TOL storage at loading/unloading port.

Reference	Electricity (kWh/t-MCH)
IAE (2010) [65]	0.83
IAE (2016) [66]	1.00
Mean	0.915
Standard deviation	0.085

Table A9. NG input for hydrogen production via NG steam reforming.

Reference	Natural Gas (MJ(LHV)/Nm^3-H_2)
General Moters et al. [95]	15.27
Ramsden et al. [97]	14.80
Mean	15.04
Standard deviation	0.24

Table A10. Electricity input for hydrogen production via NG steam reforming.

Reference	Electricity (kWh/Nm^3-H_2)
General Moters et al. [95]	0.323
Ramsden et al. [97]	0.100
Mean	0.211
Standard deviation	0.110

Table A11. Electricity input for hydrogen compression at NG steam reforming hydrogen production plant; individual values of inventory data from each literature are shown side by side.

Reference	Electricity (kWh/Nm^3-H_2)	
JHFC (2011) [39]	0.119	0.440
Mean	0.272	
Standard deviation	0.135	

Table A12. Capacity and fuel economy of CGH_2 tank trucks.

Reference	Capacity (Nm^3-H_2)	Fuel Efficiency (km/L-diesel oil)
Toyota & Mizuho [40]	2460	3.00
JPEC [98]	2200	2.50
Mean	2330	2.75
Standard deviation	130	0.25

Appendix B

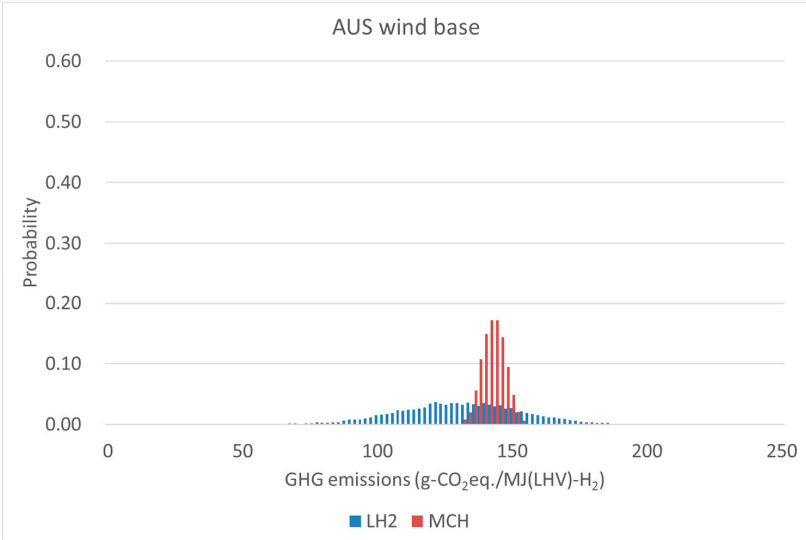

Figure A1. Probability histograms of WtT GHG emissions from hydrogen supply chains for hydrogen produced using wind power in Australia (base case).

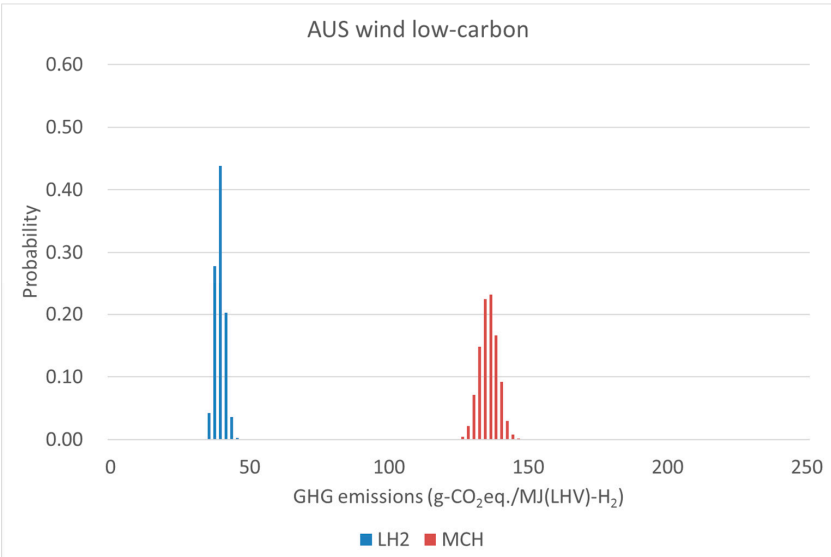

Figure A2. Probability histograms of WtT GHG emissions from hydrogen supply chains for hydrogen produced using wind power in Australia (low-carbon case).

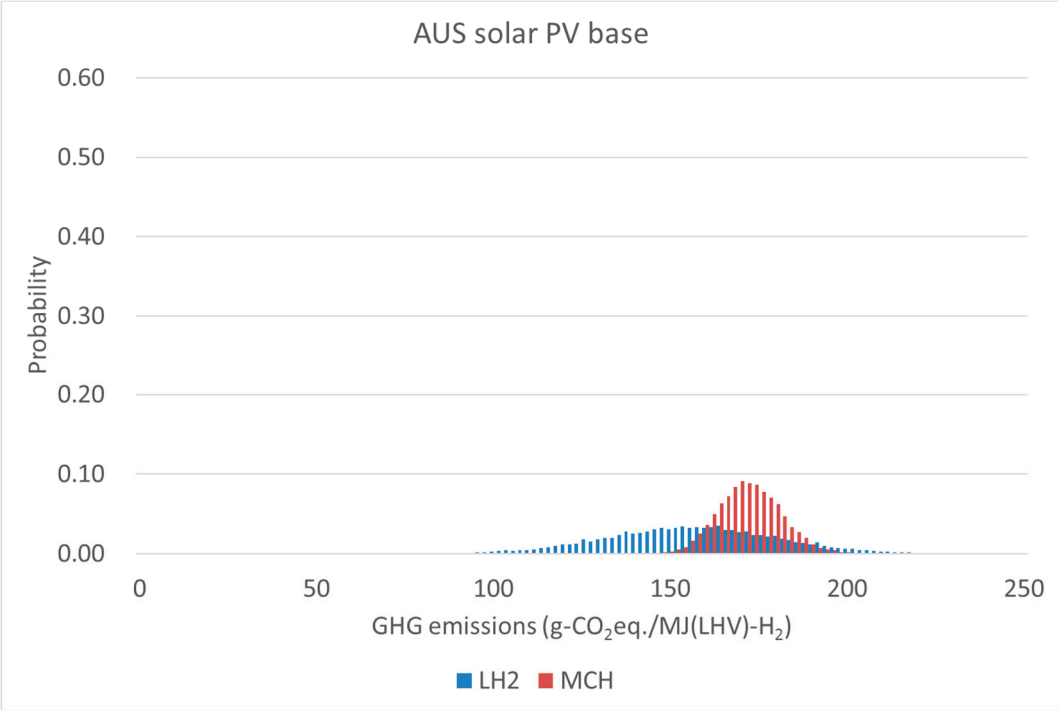

Figure A3. Probability histograms of WtT GHG emissions from hydrogen supply chains for hydrogen produced using solar PV power in Australia (base case).

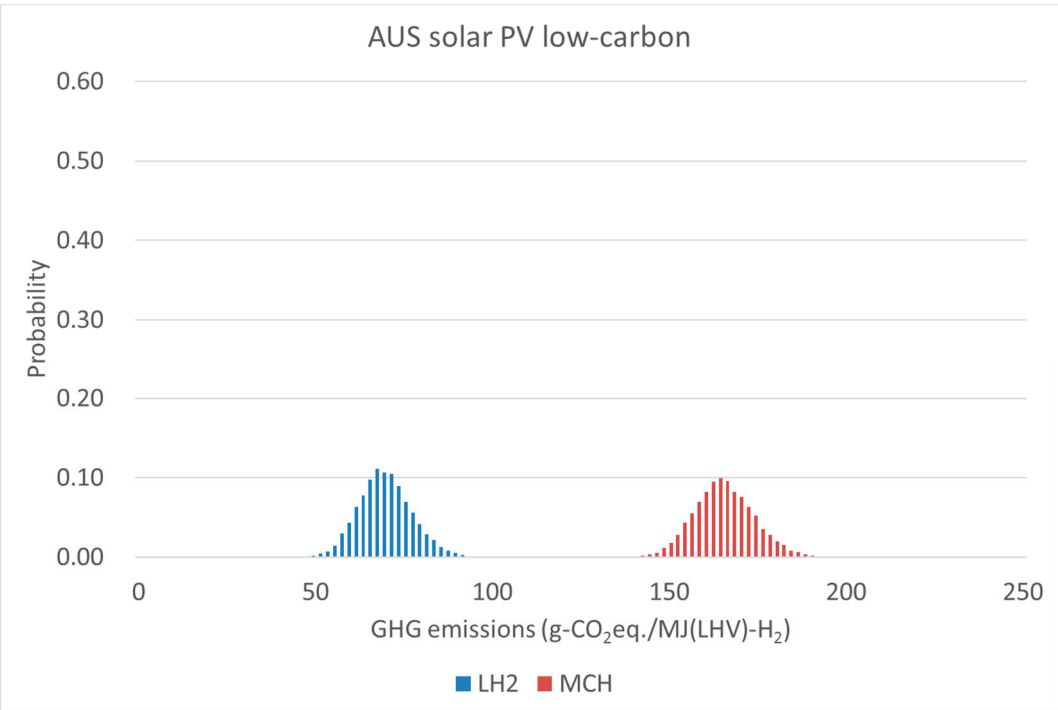

Figure A4. Probability histograms of WtT GHG emissions from hydrogen supply chains for hydrogen produced using solar PV power in Australia (low-carbon case).

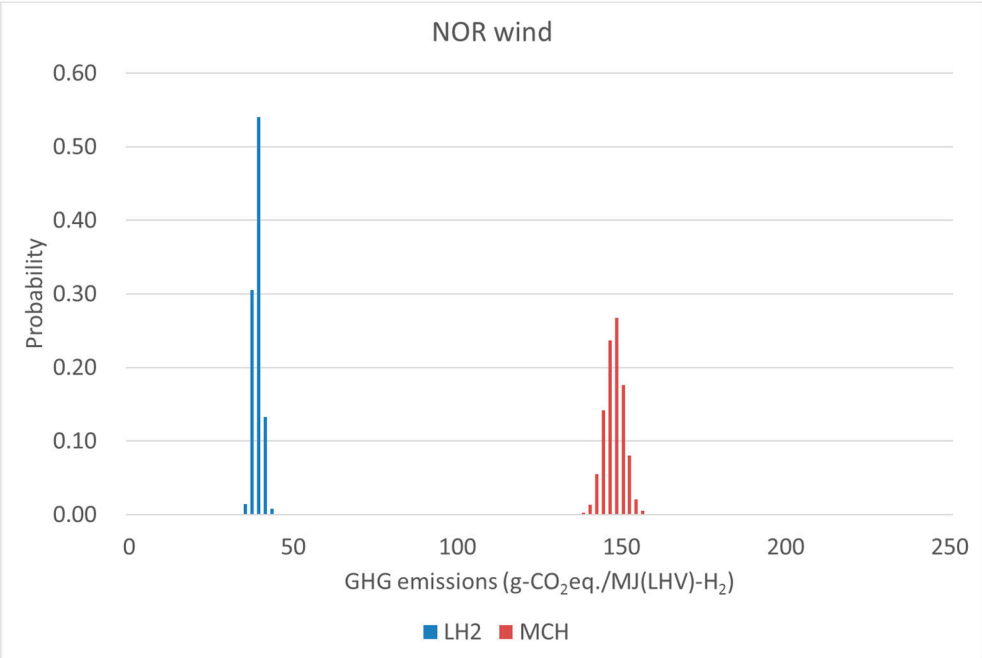

Figure A5. Probability histograms of WtT GHG emissions from hydrogen supply chains for hydrogen produced using wind power in Norway.

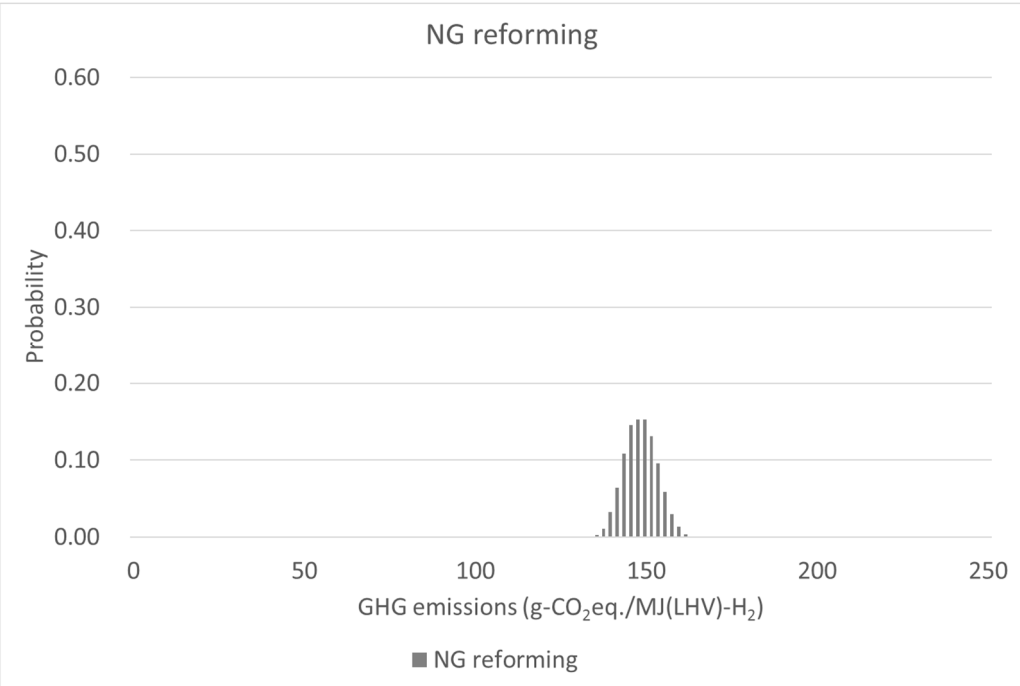

Figure A6. Probability histogram of WtT GHG emissions for supply chain for hydrogen produced using NG reforming.

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