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# Comparative life cycle assessment of sustainable energy carriers including production, storage, overseas transport and utilization

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ABSTRACT

Countries are under increasing pressure to reduce greenhouse gas emissions as an act upon the Paris Agreement. The essential emission reductions can be achieved by environmentally friendly solutions, in particular, the introduction of low carbon or carbon-free fuels. This study presents a comparative life cycle assessment of various energy carriers namely; liquefied natural gas, methanol, dimethyl ether, liquid hydrogen and liquid ammonia that are produced from natural gas or renewables to investigate greenhouse gas emissions generated from the complete life cycle of energy carriers accounting for the leaks as well as boil-off gas occurring during storage and transportation. The entire fuel life cycle is considered consisting of production, storage, transportation via an ocean tanker to different distances, and finally utilization in an internal combustion engine of a road vehicle. The results show that using natural gas as a feedstock, total greenhouse gas emissions during production, ocean transportation (over 20,000 nmi) by a heavy fuel oil-fueled ocean tanker, and utilization in an internal combustion engine are 73.96, 95.73, 93.76, 50.83, and 100.54 g $CO_2$ eq. $MJ^{-1}$ for liquified natural gas, methanol, dimethyl ether, liquid hydrogen, and liquid ammonia, respectively. Liquid hydrogen produced from solar electrolysis is the cleanest energy carrier (42.50 g $CO_2$ eq. $MJ^{-1}$ fuel). Moreover, when liquid ammonia is produced via photovoltaic-based electrolysis (60.76 g $CO_2$ eq. $MJ^{-1}$ fuel), it becomes cleaner than liquified natural gas. Although producing methanol and dimethyl ether from biomass results in a large reduction in total greenhouse gas emissions compared to conventional methanol and dimethyl ether production, with a value of 73.96 g $CO_2$ eq. per MJ, liquified natural gas still represents a cleaner option than methanol and dimethyl ether considering the full life cycle.
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## 1. Introduction

International initiatives are driving the research towards reducing greenhouse gas (GHG) emissions by alternatives to fossil fuels. A variety of alternative fuels that have low carbon or carbon-free are being discussed to improve the environmental footprint. In particular, there is an ambitious goal, which is set by the International Maritime Organization (IMO) to reduce GHG emissions by 50% until 2050 and to limit the sulfur content of used fuels by 0.5% starting from January 2020 (IMO, 2019). This means that low GHG emission fuels will take over the role of today's environmentally harmful fuels.

The increased demand for natural gas and the use of natural gas as fuel gained a growing interest in the past decade (Corbin et al., 2020). Previous studies have emphasized that the use of liquefied natural gas (LNG) reduces peak temperatures required in an engine chamber during combustion, which results in a reduction of $NO_x$ emissions (Mokhatab et al., 2013; Woodyard, 2009). However, LNG may be a counterproductive fuel and can cause climate change by increasing GHG emissions, particularly $CH_4$ and $CO_2$. This is due to its high global warming potential and the possible losses in the supply chain, specifically when considering the full life cycle (IPCC, 2007).

From this perspective, many existing studies in the recent literature have examined GHG emissions of alternative fuels to meet the IMO target. Bicer and Dincer presented a comparative life cycle assessment of a complete transport life cycle of ammonia and hydrogen as alternative fuels to replace heavy fuel oils (HFO) in ocean vehicles (Bicer and Dincer, 2018). El-Houjeiri et al. compared

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generated GHG emissions of the fuel supply chain produced from Saudi crude oil (heavy fuel oil and marine gas oil) to LNG in various regions around the world (El-Houjeiri et al., 2019). Peng et al. estimated energy consumption and GHG emissions for more than 20 fuel pathways in China, including coal and gas-based vehicle fuel, diesel and gasoline fuel, and electric vehicles (Peng et al., 2017). Several studies are reported in the literature to address GHG emissions concerning losses in the entire life cycle. Bengtsson et al. presented comparative analysis in GHG emissions including upstream emissions, utilization emissions, and unburned methane (methane slip) between LNG, Marine Gas Oil (MGO), and HFO in maritime transportations (Bengtsson et al., 2013). As it has been previously reported in the literature, DME can be used as clean ignition fuel with reduced emissions such as nitrogen dioxide ($NO_x$), sulfur $SO_{x,}$ and particular matter (PM) (Semelsberger et al., 2006). Whereas Tomatis et al. evaluated the environmental performance of DME production through $CO_2$ enhanced gasification process by using SimaPro software and found out that the LCA results revealed a 20% reduction in emissions on climate change and eco-toxicity compared to the conventional approach. This decrement was due to low feedstock consumption, utilization of $CO_2$, and high energy recovery (Tomatis et al., 2019). A series of recent studies have indicated that the usage of ammonia fuel is favored compared to fossil fuels. These studies concluded that ammonia fuel can achieve higher sustainability and low carbon emissions because of its chemical structure of not containing any carbon atoms and its ability to store hydrogen as the cost of hydrogen storage is commonly higher than the cost of ammonia storage (Yapicioglu and Dincer, 2019; Zamfirescu and Dincer, 2008). Furthermore, a prior study established an LCA for the coal-derived methanol transportation pathway and found out that the total $CO_2$ emissions of the whole life cycle is 892.27 g $MJ^{-1}$. Production of methanol accounts for 63.58% whereas the utilization process accounts for 34.04% of total emitted $CO_2$ (Wang et al., 2014). Reno et al. examined GHG emissions through LCA for methanol production from sugarcane bagasse, the results proved that sugarcane bagasse methanol was a feasible alternative for substitution and less emitter of $CO_2$ compared to methanol produced from natural gas (Zhen and Wang, 2015). There have been numerous studies to investigate GHG emissions emitted in the LNG complete life cycle. A study implemented LCA from power generation systems to understand these systems from an environmental perspective. Energy production process by coal, oil, LNG, nuclear, hydropower geothermal, wind, and solar photovoltaic (PV) releases GHG emissions of about 975.2, 742.1, 607.6, 24.2, 11.3, 15.0, 29.5, and 53.4 g $CO_2$ $kWh^{-1}$, respectively (ACIL Allen Consulting, 2018).

Storing energy using chemical energy carriers in liquid form has the capability of reducing the storage volume drastically compared to gaseous forms. Hence, transporting energy in liquified forms results in massive volume shrinkage, which enables more feasible delivery of energy to demanded regions (Han and Lim, 2012). However, liquid energy carriers have an unavoidable problem specifically when they are stored in cryogenic temperatures, a temperature lower than the boiling temperature of a liquid carrier. This problem is called boil-off gas (BOG), and it occurs due to temperature differences between liquid temperature and environment temperature. BOG generation occurs when energy is stored in tanks or injected into or out of tanks and it appears in different phases of a liquid energy carrier supply chain, including storage, loading, unloading, and transportation (Dobrota et al., 2013). BOG can be handled by the reliquefaction facility or flared to the environment (Kwak et al., 2018). The handling pathway of BOG is decided based on the economics of a project since the BOG reliquefaction facility is an energy-intensive process (Liu et al., 2010). When BOG is flared to the environment, these emissions negatively affect the environment and increase GHG emissions. Therefore, some studies in the literature examined the environmental burdens of fuels considering BOG generation. For instance, Arteconi et al. investigated GHG emissions generated from utilizing LNG as a fuel for heavy vehicles in Europe, and they reported that identifying GHG emissions from fuel consumption for driving motors and turbines, combustion of waste gases in flares, and gas losses from venting and losses from pipes and equipment results in more realistic estimation of GHG emissions (Arteconi et al., 2010). Tamura et al. estimated the GHG emissions from exported LNG to Japan. The estimated emissions include the amount of LNG loaded, the amount of BOG released, fuel-consuming, and cargo handling (Tamura et al., 2001). The previous studies reveal that most studies related to BOG generation emissions were focusing on only LNG. However, studies analyzing BOG generation emissions from various liquefied energy carriers, namely; liquid ammonia, liquid hydrogen, methanol, and DME are rarely available in the literature. Most early studies as well as current work focus on technical or economical aspects of BOG generation on liquefied energy carriers. For instance, McKinlay et al. compared liquid ammonia and liquid hydrogen as future long-distance shipping fuels and stated that BOG losses are simply fuel lost, which results in decreased economic cost (McKinlay et al., 2020). Therefore, the novelty of this work is assessing GHG emissions of a complete life cycle of variety of sustainable energy carriers by accounting the emissions associated with BOG generation in all phases.

In this context, although many studies are focusing on a complete life cycle of various energy carriers, the research, which accounts for emissions produced due to leaks and BOG remains limited. To fill this literature gap, more specific research questions will be introduced and addressed in the estimation of the total produced GHG emissions during a complete life cycle, including production, transportation, utilization, and emissions from leaks and BOG. The main goal of this work is to perform a life cycle assessment to quantify GHG emissions of the full supply chain of various energy carriers produced in Qatar. This study represents a comparative analysis of GHG emissions of energy carriers produced from natural gas and renewables, namely: LNG, methanol, dimethyl ether (DME), liquid hydrogen, and liquid ammonia. The specific objectives of this study are listed as follows:

- Comparative assessment of total GHG emissions from production, long-distance overseas transportation, and utilization in the internal combustion engine of a road vehicle for LNG, DME, methanol, liquid ammonia, and liquid hydrogen.
- Estimation of GHG emissions per unit mass (kg $CO_2$ eq. $kg^{-1}$ of energy carrier) and per unit energy (kg $CO_2$ eq. $MJ^{-1}$ of energy carrier) from production, transportation, and utilization of energy carriers for short distances (5000 nmi) and long-distances (20,000 nmi).
- Comparing total produced GHG emissions from various liquid hydrogen production methods such as water electrolysis via PV, water electrolysis via wind power, steam methane reforming with CCS, steam methane reforming without CCS, and from transportation (20,000 nmi) by HFO-fueled ocean tanker, and from utilization in an internal combustion engine.
- Assessing NOx and SOx emissions (in mg of emission/MJ of energy carrier) during the production, transportation, and utilization in an internal combustion engine of various energy carriers produced from natural gas.
- Performing sensitivity analysis to estimate GHG emissions of a full life cycle covering production, transportation, and utilization of energy carriers derived from either renewable (solar PV, wind and biomass) or natural gas over distances ranging from 0 to 20,000 nmi.

- Assessment of different production methods of energy carriers such as solar PV, wind, and biomass to investigate the reduction potential in GHG emissions during the complete life cycle by accounting generated BOG.

## 2. Methodology

LCA is a methodology used to assess environmental burdens of products, processes, or services from a cradle-to-grave perspective (International Organization for Standardization, 2006). LCA study contains four stages, namely: goal and scope definition, life cycle inventory (LCI), life cycle impact assessment (LCIA), and interpretation. The methodology flowchart followed in this study is presented in Fig. 1 (Curran, 2008).

The goal and scope stage explains a product system in terms of a functional unit and system boundaries. The functional unit is the equivalent basis, which enables different products to be examined and compared. In this study, the functional unit is 1 kg and 1 MJ of energy carrier produced, transported to various distances, and utilized in an internal combustion engine. The scope of the study is to compare the life cycle environmental performance of five different energy carriers produced from natural gas or renewables in different phases of their supply chains. The system boundary covers raw materials extraction, feedstock transportation, liquefied energy carrier production, storage, transportation, and utilization. Since Qatar's main source of energy is natural gas, Qatar is taken as the geographical region of the study. Therefore, natural gas is used as feedstock to produce fuels in most of the cases. Natural gas is recovered from the North Field in Qatar and transported 80 km to Ras Laffan industrial city (RLIC) by pipeline (Korre et al., 2012). The study assumed that the five fuels are produced in RLIC and transported to different regions globally. A fuel production plant consists of machinery to convert feedstock, equipment to move the product, storage tanks, and loading facility to load the product into transported tanks (Dobrota et al., 2013). Hence, this study covers the production of fuels from natural gas, storing and loading them into transported tanks, transporting them to different distances ranging from 0 to 20,000 nautical mile (nmi), and finally utilizing these fuels in an internal combustion engine. A sensitivity analysis is also carried out for examining emissions released from transporting fuels by an ocean tanker that is fueled by either heavy fuel oil (HFO) or LNG. Here, LNG is also considered as alternative bunker fuel for propulsion.

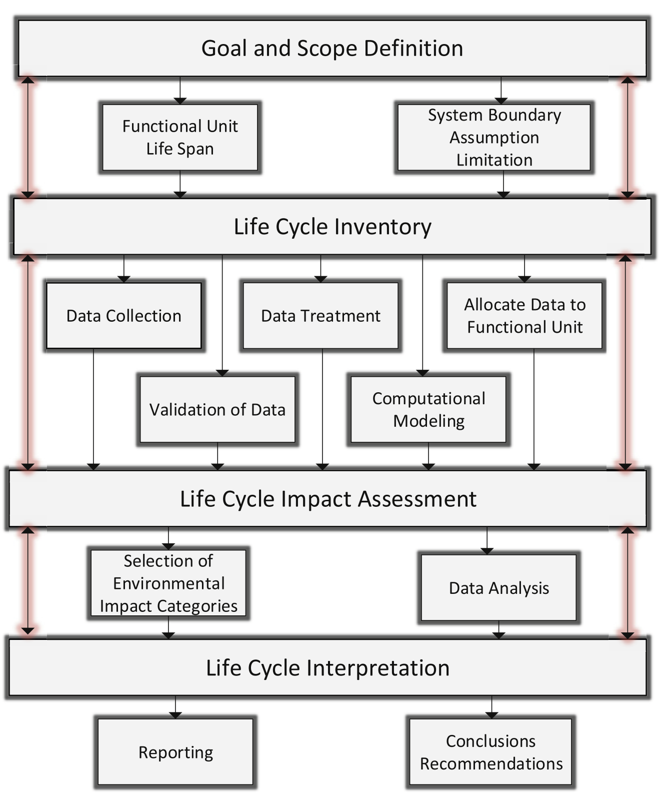



**Fig. 1.** Life cycle assessment framework and methodology flowchart.

The second stage of LCA is LCI, which is a description of energy and material flows within the production system and its interaction with the environment. The inputs and outputs of a process are identified and quantified in this stage (e.g., energy, water, materials, and environmental releases). The Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET) model are used to implement this study (Argonne National Laboratory, 2012). The mathematical model used in GREET calculates emissions such as $CO_2$, $CH_4$, and $N_2O$, and other pollutants emitted from transportation life cycles. GREET inventory data contain an extensive set of predefined process parameters specific to North America. For example, a defined pathway which shows $CO_2$ produced from the production of LNG from natural gas. Hence, GREET inventory data is used to implement this study. However, since the inventory data is specific to North America (U.S.A), process parameters and associated emissions have been updated to reflect the same processes used in Qatar. The inventory data used in this study including energy and materials flows are given in supplementary file. The updated processes are mentioned as follows:

- Only natural gas is used as feedstock for electricity production.
- Pipeline distance from the natural gas field to RLIC is adjusted to 80 km.

LCIA covers impact assessment and consists of several steps such as weighting, classification, and normalization. Determination of pollution types and size is assessed in this step. LCIA covers environmental indicators that address generated emissions released to air, water, or soil. For instance, global warming potential (GWP), acidification potential (AP), and ozone depletion potential (ODP) address effect on air or climate whereas, eutrophication potential (EP) and marine aquatic ecotoxicity potential (MAEP) address effect on water (Herva et al., 2011). The impact indicator investigated in this study is global warming potential (GWP) to give a comparative evaluation among the energy carriers. GREET software is used to simulate and characterize the generated emissions from the energy carriers in their supply chain. It calculates emissions in terms of the GWP indicator only. Therefore, GWP is implemented to classify the impacts associated with emissions, which are equivalent to $CO_2$. 100-year time frame GWP is adapted from the Fifth Assessment Report (AR5) issued by the Intergovernmental Panel on Climate Change (IPCC) to calculate ($CO_2$ equivalent) (IPCC, 2013). The last step is interpretation, which provides an evaluation of results from LCI and LCIA to identify major contributions to total pollution.

The proposed supply chain for the energy carriers consists of common processes such as production of feedstock, production of fuels, storage, transportation, and utilization as illustrated in Fig. 2. Generated emissions from (i) combustion of process fuels that provides energy for the processes, (ii) leaks and BOG, which are associated with production, loading, storage, transportation, and utilization of energy carriers are accounted for in the study. The well-to-use emissions for each considered method in this study are given in detail in supplementary file.

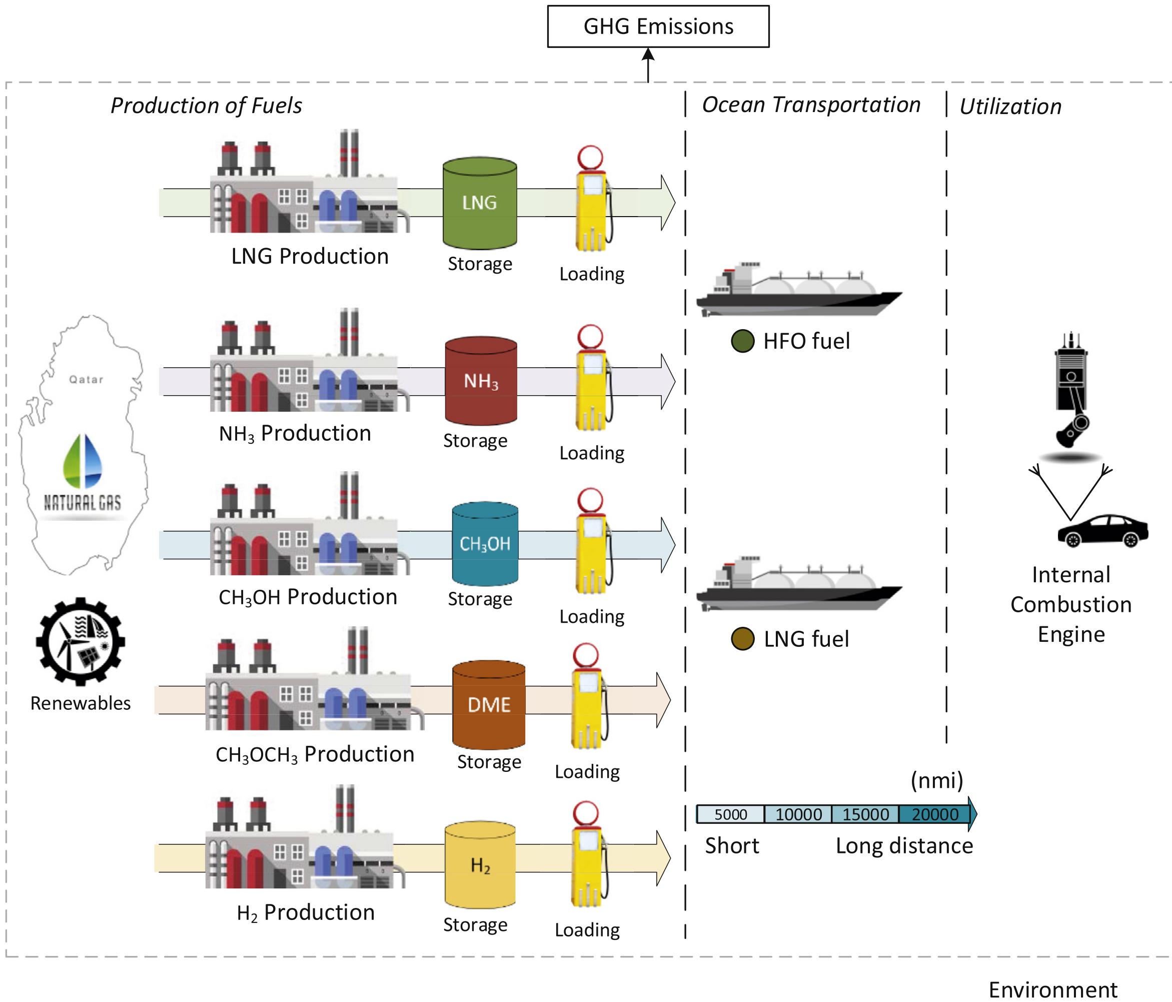



**Fig. 2.** Process flow chart showing various stages within the system boundary.

To improve applicability and validity of the study, empirical data are used. Regarding the limitation of the proposed approach whether information given by LCA provides enough guidance for decision makers, there is a list of factors that can be considered in this matter such as infrastructure requirements, maintenance, fuel availability, and fuel cost. These factors are not included in LCA. However, there can be additional tools that can cover more aspects is a multi-criteria decision such as life cycle costing and sustainability assessment (Belton and Stewart, 2002). However, LCA is commonly the initial step for that type of analysis. Therefore, the LCA approach can be complemented with other tools for comprehensive assessments. LCA results might not have a direct effect on the final decision of energy carrier to choose but it can be used as arguments for the decision.

## 3. Analysis

All data for energy requirements and leakage rates in processes are reported from GREET inventory data whereas BOG generation rates are taken from our detailed previous publications (Al-Breiki and Bicer, 2020a, 2020b). The following subsections represent supply chains for the fuels, mentioning inputs and outputs resources, electricity requirements, leakage rates, and BOG rates in each process.

### 3.1. Production of energy carriers

Since natural gas is used as the main feedstock, emissions from recovering and processing of natural gas are accounted for in the total emissions of production of fuels. Fig. 3 presents inputs and outputs in each subprocess from extraction to pipeline transportation. The inputs and outputs for natural gas recovery and processing are natural gas flaring, electricity, and other resources. The resources are water, residual oil for boilers, diesel for engines and turbines, and gasoline blend-stock. The electricity required for producing 1 GJ of natural gas during natural gas recovery and processing is 270 kJ and 862 kJ, respectively (Seddon, 2006). Due to the internal processes, some unrecovered leaks occur. The emissions created in these processes from used resources, electricity, and leaks are accounted for. Once natural gas is produced, it is transported 80 km by pipeline to different industrial processing plants. Emissions generated from electricity requirement to pump natural gas for this distance are also accounted for. These released emissions are added into total generated emissions from the production of LNG, liquid ammonia, methanol, DME, and liquid hydrogen.

#### 3.1.1. LNG

The supply chain of LNG starts from liquefying natural gas feedstock. The main cause of emissions in the LNG production

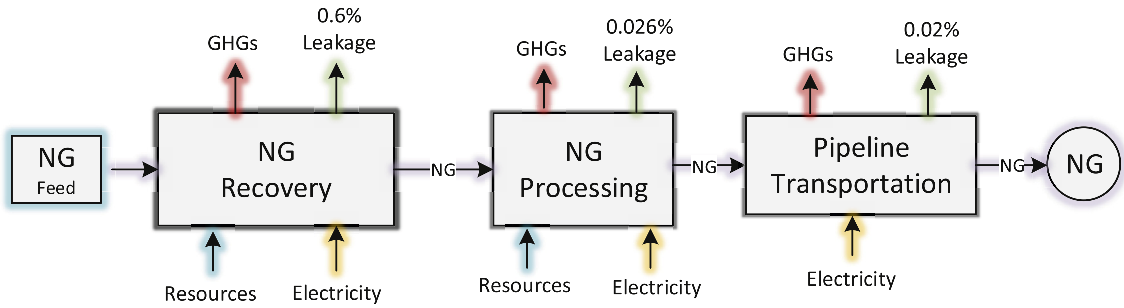



**Fig. 3.** Natural gas production process main inputs and outputs.

phase is fuel consumption for operating equipment, which is presented as electricity input in Fig. 4 and gas losses from venting, which is associated with losses from pipes and equipment (Arteconi et al., 2010). Once LNG is produced, it is sent to storage tanks. These storage tanks are located on-site and they are used to store LNG for large quantities before loading into ocean tankers. The generation of BOG in storage and loading phases is considered to occur at constant rates. The inventory data of GHG emissions of these processes are built-in GREET. The summation of emissions released from fuel requirements, leaks, and BOG determines total emissions of the LNG supply chain from production to loading phase.

### 3.1.2. Liquid ammonia (L-$NH_3$)

The liquid ammonia production pathway from reforming of natural gas or renewables is presented in Fig. 5. In this study, GHG emissions are investigated from different L-$NH_3$ production methods. The first method is the Haber-Bosch process, which produces ammonia by combining nitrogen from the air and hydrogen derived from either natural gas or water electrolysis. The Haber-Bosch method is the most common method for ammonia production and it consumes about 3–5% of all-natural gas produced (Ashida et al., 2019). Moreover, implementing carbon capture and storage (CCS) facility in ammonia production plants has been introduced extensively in the literature and in industrial scale (Gonzalez-Diaz et al., 2020). Therefore, this study examines GHG from L-$NH_3$ with and without CCS facility. Implementing CCS facilities into conventional ammonia production from natural gas results in about a 25% reduction in GHG emissions (Mehmeti et al., 2018). This percentage reduction is used in L-$NH_3$ with CCS to estimate total GHG emission in this method. The inventory data to estimate GHG emissions for ammonia production by reforming of natural gas can be found in GREET database. However,production of ammonia by water electrolysis via PV, water electrolysis via wind energy, and concentrated light photoelectrochemical (PEC)-based electrochemical method are taken from previous publications as summarized in Table 1. After the production of ammonia from these different methods, it is sent to storage tanks then loaded into a ship for transportation. GHG emissions of L-$NH_3$ and L-$H_2$ from renewables are not available in GREET inventory data so previous publications are used to obtain these data.

These emissions are then built in the software to estimate total GHG emissions from the whole supply chain. The high-temperature difference between storage temperatures and average environment temperature (25 °C) causes an increment in heat transfer into the tanks, which results in higher losses. Thus, the leakage rate and BOG rate in the L-$NH_3$ storage phase are 2.5 and 5 times lower compared to LNG storage due to the lower storage temperature of LNG. The storage temperature of L-$NH_3$ is −34 °C whereas LNG storage temperature is −160 °C. The emissions caused by these losses and required fuel to produce and load ammonia into storage tanks are accounted for in the estimations of total GHG emissions.

### 3.1.3. Methanol ($CH_3OH$) and DME ($CH_3OCH_3$)

GREET software is used to quantify and identify emissions in different phases of methanol and DME production as illustrated in Fig. 6. In this study, GHG emissions of methanol and DME production pathways from natural gas by steam reforming process and biomass by gasification process are estimated. GHG emissions of these processes are available in GREET inventory data. The formation of methanol starts with the production of syngas via a steam reformer. The syngas is then compressed with required compression ranges from 30 bar to 100 bar. The compressed gas then passes through a converter. The conversion temperature is around 300 C. The pass conversion of product gas moves to a condenser. The condenser typically raises steam whereas the condensed product is removed in a knockout pot, which releases methanol (Seddon, 2006). DME can be produced by either a single-stage route or a two-stage route. The single-stage route is to produce DME directly from synthesis gas via methanol dehydration whereas the two-stage process is to produce methanol first then DME (Solomon, 2017). The two-stage process is used in this study to estimate

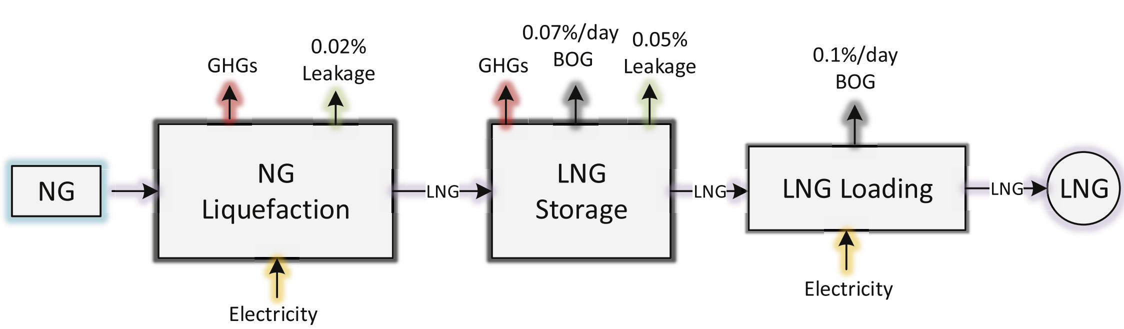



**Fig. 4.** Sources of emissions, leaks, and BOGs during LNG production.

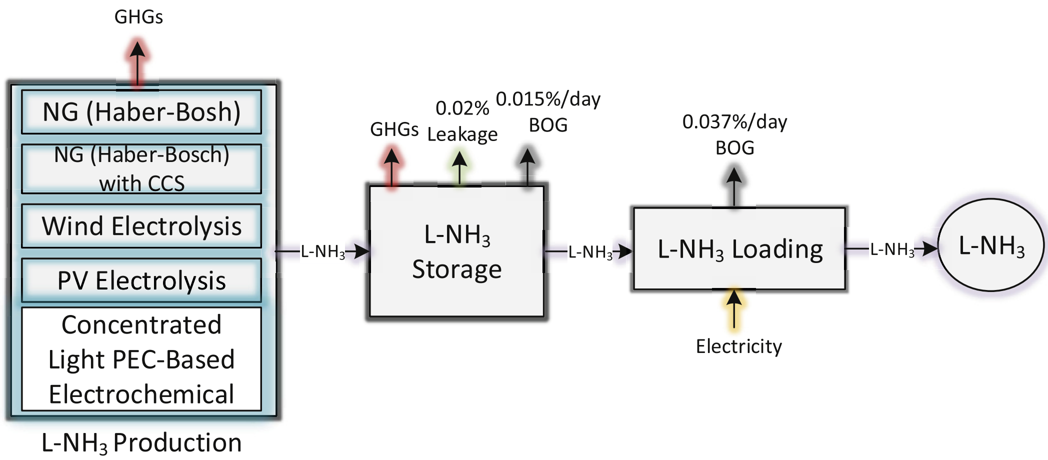



**Fig. 5.** Ammonia production methods considered in this study.

**Table 1**
Summary of GHG emissions of renewable-based ammonia production methods.

| Method | GHG Emissions (kg of $CO_2$ eq/kg of $NH_3$) | Reference |
|---|---|---|
| Water electrolysis via wind energy and Haber-Bosch method | 0.4 | (Amponsah et al., 2014; Bicer et al., 2016) |
| Water electrolysis via PV energy and Haber-Bosch method | 0.9 | (Bicer et al., 2016; Ozawa et al., 2019) |
| Concentrated light PEC-based electrochemical | 1.06 | Bicer and Dincer (2017) |

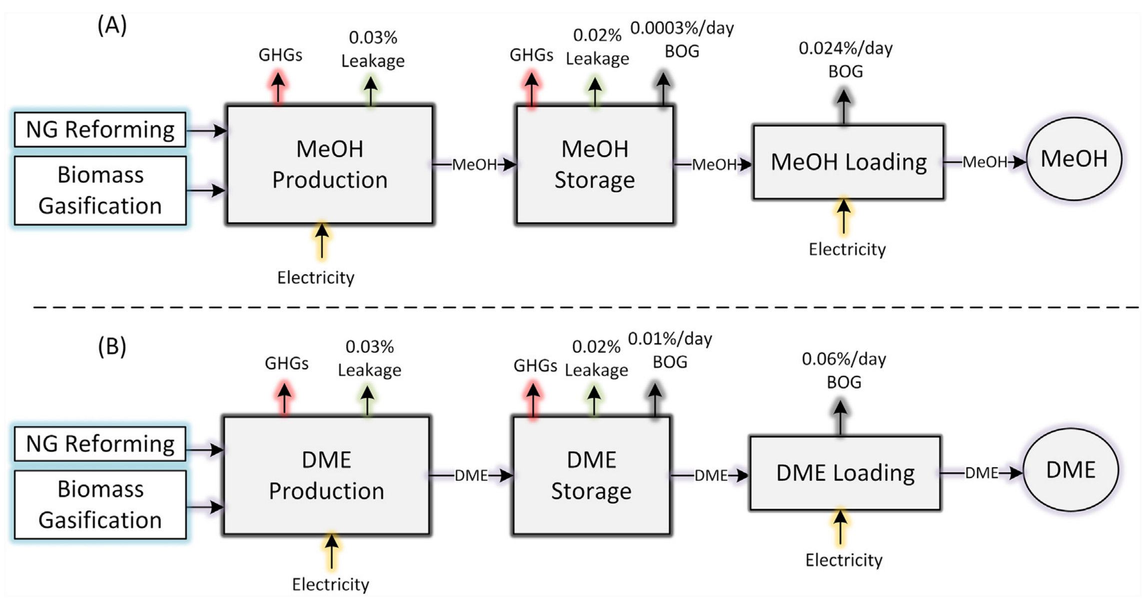



**Fig. 6.** Sources of emissions in (A) methanol and (B) DME production pathways.

GHG emissions produced in the DME life cycle.

### 3.1.4. *Liquid hydrogen (L-$H_2$)*

A similar approach to L-$NH_3$ is used for L-$H_2$. Hydrogen is produced either from reforming of natural gas or renewable sources (wind and solar energy) as presented in Fig. 7. Regarding liquid hydrogen production process from natural gas, this process is a built-in GREET model and the process does not contain $CO_2$ sequestration. In this study, GHG emissions from the hydrogen production process by reforming of natural gas is taken directly from GREET inventory data. Accounted emissions from this process are coming from fuel used to run turbines and motors and fuel leakages. However, generated GHG emissions from the production phase of L-$H_2$ from renewables, water electrolysis via wind and PV energy is retrieved from previous studies. GHG emissions of L-$H_2$ by water electrolysis via wind energy is 0.97 kg of $CO_2$ eq/kg of $H_2$ and by water electrolysis via PV energy is 2.4 kg of $CO_2$ eq/kg of $H_2$ (Bhandari et al., 2012). The relationship between hydrogen and GHG emissions is that increment of emitted gaseous hydrogen to the atmosphere can consider an indirect greenhouse gas because its emissions influence the tropospheric free radical species ($CH_4$, OH, and $H_2$). An increase in these emissions leads to an increase in global radiative forcing which harms the environment. Therefore, using hydrogen as a fuel to replace fossil fuel without considering

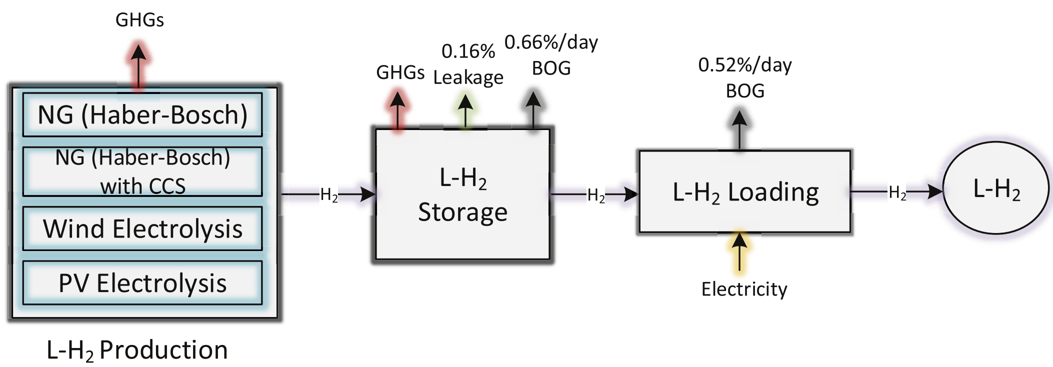



**Fig. 7.** Sources of emissions in L-$H_2$ production.

an efficient system to capture $H_2$ leaks can also affect the environment negatively (Derwent et al., 2006). Once liquid hydrogen is produced, it is sent into a storage tank and then loaded into a shipping tanker. In storage and loading processes, some portion of liquid hydrogen is lost as BOG and leaks, which are higher than other energy carriers due to very low storage temperature.

### 3.2. Transportation and utilization of energy carriers

GHG emissions are estimated from raw material extraction to loading into ship tankers as previously shown. The phase after production and on-site storage is the ocean transportation phase. Large quantities of fuels are transported by ocean tankers. For example, Qatar has managed to transport 77.9 million tonnes of natural gas in LNG form by ocean tankers in 2019 (John, 2020). Hence, this study considers transporting the produced fuels via ocean tankers. Table 2 presents ocean tanker specifications, which transports fuels to different distances. The payload is the load for each resource transported by ocean tanker. The average speed represents average speed from the originating location to the destination location. 'Load factor to' is the percentage of power used overpower installed from originating location to the destination location whereas 'load factor from' represents the percentage of power installed from the destination location to originating location. Brake specific fuel consumption and typical horsepower parameters are used to estimate the energy intensity of the ocean tanker. The formula is built-in GREET software which calculates energy intensity required by ocean ship fuel. Generated emissions from used fuel to power ocean tanker is taken into account in this phase.

Moreover, BOG generation rates for different fuels in ship tanks are reported from our previous publication (Al-Breiki and Bicer, 2020a). Table 3 shows BOG rates in the ocean transportation phase. These BOG rates are time-dependent loss rates which means the time necessary for travel is dependent on the distance and the average speed of the ocean tanker. The emissions generated due to BOG losses are accounted for in the total GHG emissions for each supply chain.

**Table 2**
Ocean tanker specific parameters.

| Ocean Tanker Specific Parameters | Value |
|---|---|
| Payload (kg) | 60,000,000 |
| Average Speed (m/s) | 8.94 |
| Load Factor to | 0.83 |
| Load Factor From | 0.70 |
| Brake Specific Fuel Consumption (g/kWh) | 195 |
| Typical Horsepower (hp) | 9070 |

**Table 3**
Daily BOG rates in ocean tankers during ocean transportation for various energy carriers.

| Energy Carrier | BOG/day (%) |
|---|---|
| LNG | 0.12 |
| L-$NH_3$ | 0.025 |
| MeOH | 0.005 |
| DME | 0.017 |
| L-$H_2$ | 1.06 |

Source: (Al-Breiki and Bicer, 2020a)

Summation of fuel requirements by each ocean tanker and mass losses as BOG and leak gives the total GHG emissions in the transportation phase. Moreover, a sensitivity analysis is implemented to estimate total GHG emissions from the production of fuels and transportation to various distances by different fueled ocean tankers. Diesel and heavy fuel oil are usually used as fuel for shipping tankers (Oeder et al., 2015). To reduce emissions, LNG is now increasingly used as a bunker fuel for ships (Spoof-Tuomi and Niemi, 2020). A comparative analysis is also presented here between heavy fuel oil (HFO) and LNG used as fuel for ocean tankers.

Once the liquefied energy carriers are transported to the demanded region, they are considered to be utilized in internal combustion engine vehicle (ICEV) for propulsion. The combustion and emission characteristics of LNG, DME, methanol, L-$H_2$ are taken from GREET software in which there are fueled internal combustion engines for each type except for L-$NH_3$. L-$NH_3$ combustion is calculated based on its chemical reaction and L-$H_2$ data. The generated emissions from the utilization phase are then added into emissions from production and transportation to estimate total GHG emissions of the full supply chain of these fuels.

## 4. Results and discussion

The inventory data in GREET contains GHG emissions emitted from various fuel pathways. After modifying these pathways to meet the study parameters; specifying feedstock sources, electricity source, travel distance, and entering BOG rates, GHG emissions were estimated. Table 4 presents a validation study of generated GHG emissions during the production phase by comparing it with previous studies.

A comparison of GHG emissions generated from LNG, DME, and methanol in production and transportation phases is presented in Fig. 8. Among the three fuels, methanol has the lowest GHG emissions in terms of kg of $CO_2$ $kg^{-1}$ of fuel at the production phase. When 1 kg of methanol is produced from natural gas, a combination of steam reforming and partial oxidation is applied, and it emits around 0.4 kg of $CO_2$ eq. This quantity of emissions is 25% and

**Table 4**
Literature comparison of GHG emissions of energy carriers produced from natural gas in their production and utilization phases.

| Production Phase | GHG Emissions (This Study) | Reference 1 | Reference 2 | Reference 3 |
|---|---|---|---|---|
| LNG (g/MJ) | 10.29 | 9 (Kandijk, 2015) | 10.70 (DNV GL - Maritime, 2019) | 13.3 (Edwards et al., 2007) |
| Methanol (g/MJ) | 20.10 | 21.97 (Aronoff et al., 2014) | 24.0 (Semelsberger et al., 2006) | 20 (Brynolf et al., 2014) |
| DME (g/MJ) | 21.11 | 12.2–36.7 (Higo and Dowaki, 2010) | 16.3–47.2 (Higo and Dowaki, 2010) | |
| Liquid Hydrogen (g/MJ) | 119.92 | 120.4 (Baptista et al., 2011) | | |
| Liquid Ammonia (g/MJ) | 41.66 | 29.7 (Kongshaug and Jenssen, 2003) | 48 (Spoof-Tuomi and Niemi, 2020) | 23.25–33.48 (Wood and Cowie, 2004) |
| **Utilization Phase** | | | | |
| LNG (g/MJ) | 57.5 | 56 (Peng et al., 2017) | 60 (Pehnt, 2000) | |
| Methanol (g/MJ) | 68.64 | 69 (Pehnt, 2000) | 68 (Peng et al., 2017) | 68 (Lee et al., 2016) |
| DME (g/MJ) | 67.81 | 66 (Peng et al., 2017) | 67 (Lee et al., 2016) | 67 (Wang, 1999) |
| Liquid Hydrogen (g/MJ) | 0.54 | 0 (Pehnt, 2000) | 0 (Lewis, 2018) | 0 (Wang, 1999) |
| Liquid Ammonia (g/MJ) | 0.54 | 0 (Brohi, 2014) | 0 (Lewis, 2018) | |

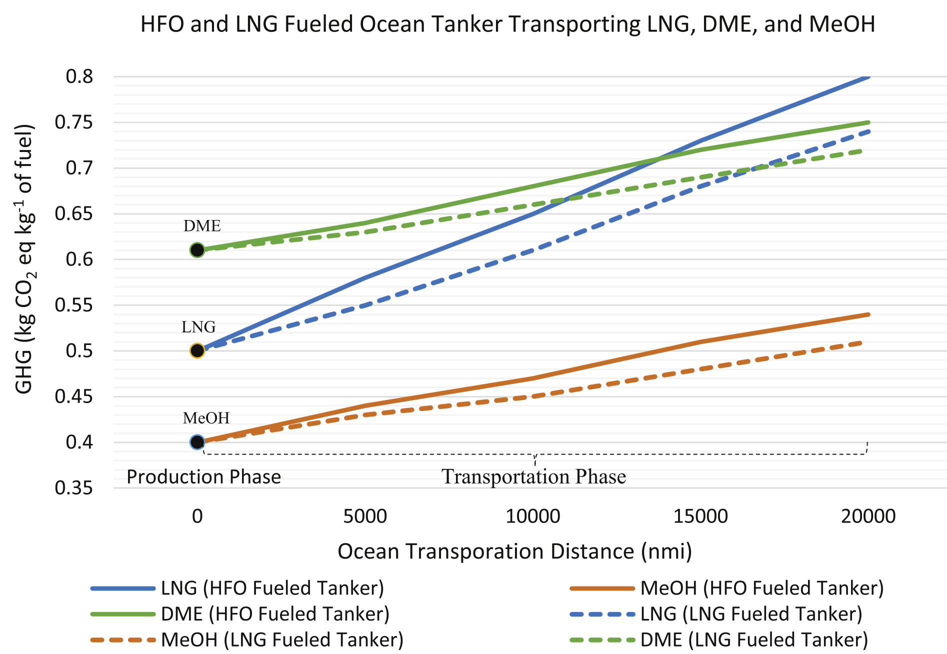



**Fig. 8.** GHG emissions generated from production and transportation of LNG, DME, and MeOH by two ocean tankers: (–) HFO fueled tanker and (–) LNG fueled tanker.

50% less than LNG and DME, respectively. After the production of these fuels, they are usually transported overseas by an ocean tanker. Emissions in the transportation phase are produced from required fuel for the ocean tanker and generation of BOG. When the used fuel for the ocean tanker is HFO, generated emissions are greater than the LNG fueled ocean tanker. Total well-to-propeller global warming potential of HFO is 23% higher than LNG when used as fuel for a ship for the same distance (Bengtsson et al., 2013). Furthermore, the daily BOG generation of LNG is 0.12%, therefore LNG emits more emissions as ocean transportation distance increases compared to methanol and DME. Even though DME starts with greater emissions compared to LNG, as ocean distance length passes 14,000 nmi, emissions from LNG becomes greater. In summary, methanol and DME are less harmful to the environment compared to LNG considering production and transportation phases only. However, there will be additional emissions in the utilization phase as they are also discussed in the forthcoming sections.

To comply with environmental regulations, addressing the complete life cycle of fuels is required. Therefore, Fig. 9A and Fig. 9B represent GHG emissions of compeleted life cylce of fuels per kg and MJ, including production, transportation, and utilization phases of LNG, DME, and methanol. The utilization phase of these fuels is accounted for most of the GHG emissions. Taking LNG supply chain as an example, emissions from the utilization phase of 1 kg of LNG for a distance of 20,000 nmi accounts for more than 77% of the total emissions. Whereas emissions from the production phase account for 15% and the rest from the transportation phase. Moreover, considering complete life cycle, GHG emissions per unit mass (kg of $CO_2$ $kg^{-1}$ of fuel) starting from producing methanol and DME from natural gas, transporting them over 5000 nmi distance by HFO fueled ocean tanker, and utilization in internal combustion engine results in GHG reduction of 45% and 20%, respectively compared with LNG. On the other hand, LNG contains less carbon per unit energy which means that when burning LNG, it emits fewer emissions. Hence, LNG emits 70 g of $CO_2$ eq $MJ^{-1}$ during combustion. In contrast, methanol and DME emit 90 g of $CO_2$ eq $MJ^{-1}$ and 89 g of $CO_2$ eq $MJ^{-1}$ respectively. Methanol emits more GHG emissions per unit energy compared with LNG and DME due to lower heating value and greater density. To sum up, considering total GHG emissions from a complete life cycle of LNG, DME, and methanol, LNG emits the lowest of emissions per unit energy nevertheless methanol emits the lowest per unit mass.

Among the proposed energy carriers, L-$H_2$ and L-$NH_3$ are two of the promising carbon-free alternatives. Since Qatar has abundant

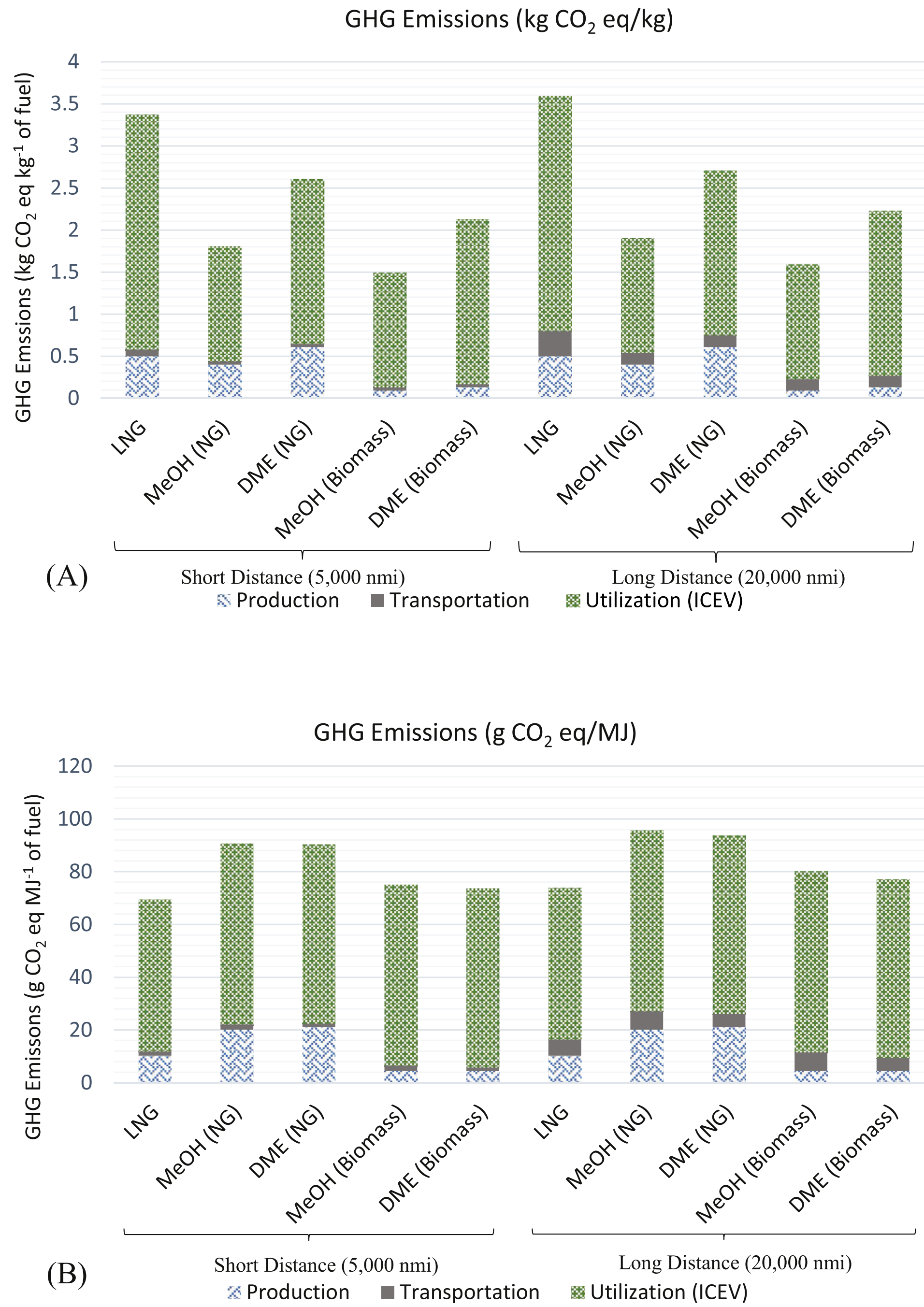



**Fig. 9.** Total GHG emissions from production, transportation, and utilization of LNG, DME, and methanol for short distances and long-distances, (A) per unit mass (kg $CO_2$ eq kg$^{-1}$of fuel) and (B) per unit energy (g $CO_2$ eq MJ$^{-1}$ of fuel).

sources of natural gas, L-$H_2$ and L-$NH_3$ can be produced by reforming natural gas (including CCS), by cracking natural gas with the help of concentrated solar energy or by water/seawater electrolysis using renewable matter. In this study, water electrolysis by PV and by wind power produces gaseous hydrogen under the renewable scenario. For the liquefaction of hydrogen process, the source of electricity is natural gas. Thus, Fig. 10 represents GHG emissions from various L-$H_2$ production methods that use natural gas as a source of electricity in the liquefaction process. Steam reforming with and without CCS uses electricity, which is produced from natural gas in production and liquefaction of liquid hydrogen. Steam reforming without CCS emits around 14.39 kg of $CO_2$ eq kg$^{-1}$ of L-$H_2$. When CCS facilities are implemented into steam reforming process, 59% of the total GHG emissions are reduced. Production of hydrogen from wind energy emits the lowest GHG emissions of about 3.6 kg of $CO_2$ eq kg$^{-1}$ of L-$H_2$. Emissions difference between production of hydrogen from renewables (wind energy) and production from natural gas reforming with CCS is about 40%, whereas the difference is 75% without CCS. Thus, implementing CCS facility is essential in plants where natural gas is used as feedstock for hydrogen production to reduce GHG emissions.

Observing the L-$H_2$ supply chain, the majority of emissions are produced during the production phase. Reforming and liquefaction processes are the main emissions producers in the production

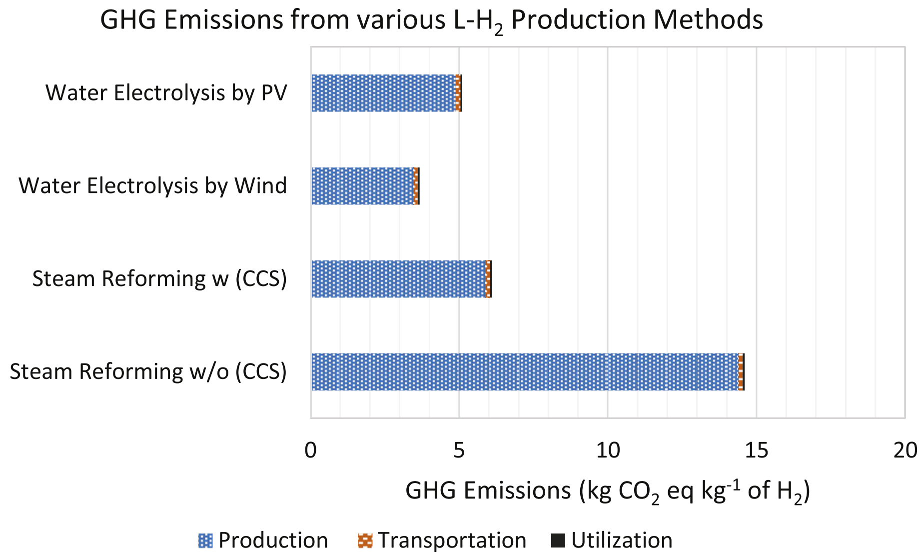



**Fig. 10.** Total GHG emissions from different L-$H_2$ production methods covering emissions in production, 20,000 nmi transportation by HFO fueled ocean tanker, and utilization in internal combustion engine processes.

phase of L-$H_2$. Required electricity to produce 1 kg of gaseous hydrogen is 50 kWh and an additional 10 kWh of electricity for the liquefaction process (Elgowainy et al., 2012). Therefore, a clean electricity generation source plays a significant role in the reduction of emissions. In a country with an abundant source of natural gas, electricity generation source is usually natural gas. Hence, Fig. 11 illustrates GHG emissions of L-$H_2$ various production methods that use produced electricity by natural gas to function reforming and liquefaction processes. It also shows GHG emission of these production methods which use produced electricity from renewables (solar energy) in the liquefaction process only. When solar PV is used to generate electricity, GHG emissions reduce by 2.3 kg of $CO_2$ eq $kg^{-1}$ of L-$H_2$ in hydrogen production methods compared with electricity produced from natural gas. In Qatar, production of L-$H_2$ by steam reforming with CCS (using natural gas electricity), and transportation of the L-$H_2$ by HFO fueled tanker for a distance of 20,000 nmi generates 5.89 of $CO_2$ eq $kg^{-1}$ of L-$H_2$. In contrast, using electricity, which is produced by solar PV for the same supply chain results in the reduction of GHG emissions of about 39%. Therefore, the desire to minimize GHG emissions in hydrogen production methods is likely achievable using renewably generated electricity.

L-$NH_3$ can be produced either by reforming of natural gas or renewables. Fig. 12 represents GHG emissions of L-$NH_3$ from different production pathways and from ocean transportation, which ranges from 0 nmi to 20,000 nmi. As the distance increases by 5000 nmi, the total GHG emissions increase by 1% only. These emissions are mainly coming from HFO that is used to fuel the tanker. Moreover, producing L-$NH_3$ in Qatar by reforming of natural gas without CCS emits around 2.24 $CO_2$ eq $kg^{-1}$ of L-$NH_3$, however implementing CCS results in GHG reduction by 24%. In comparison, GHG emissions via electrolysis of water using wind and solar PV are

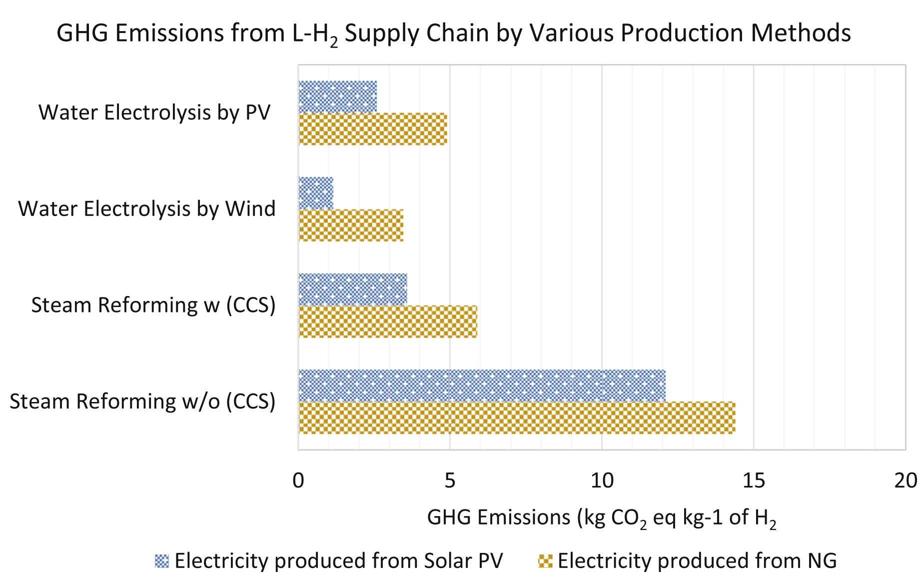



**Fig. 11.** Comparison of produced GHG emissions from the L-$H_2$ supply chain (production, storage, and 20,000 nmi ocean transportation) that uses generated electricity either by natural gas or by solar PV.

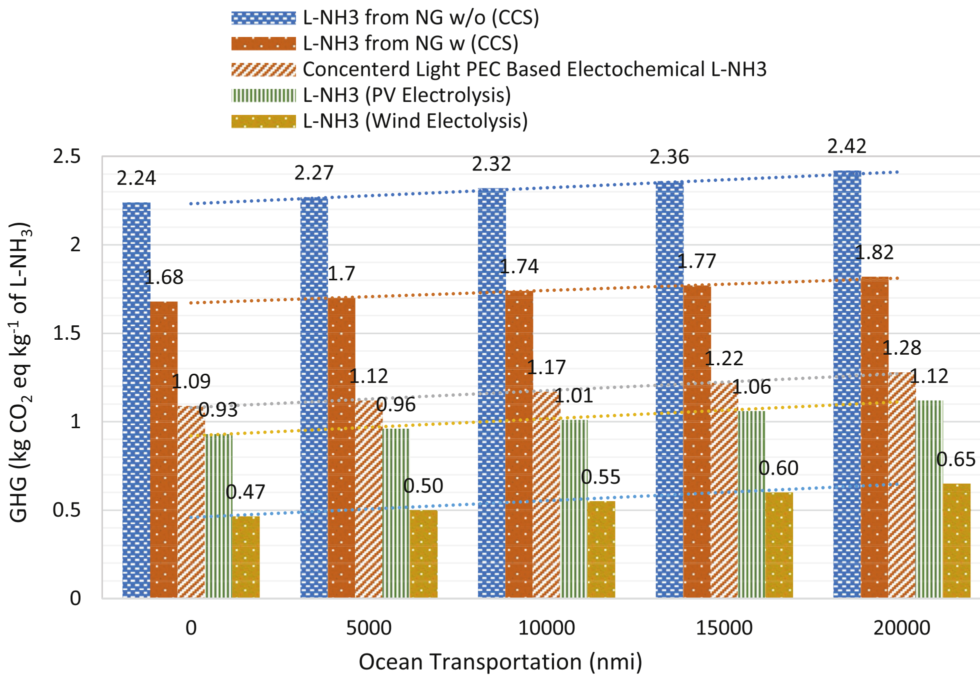



**Fig. 12.** GHG emissions generated from the production of L-$NH_3$ by different production methods and transportation over various distances by HFO fueled ocean tanker.

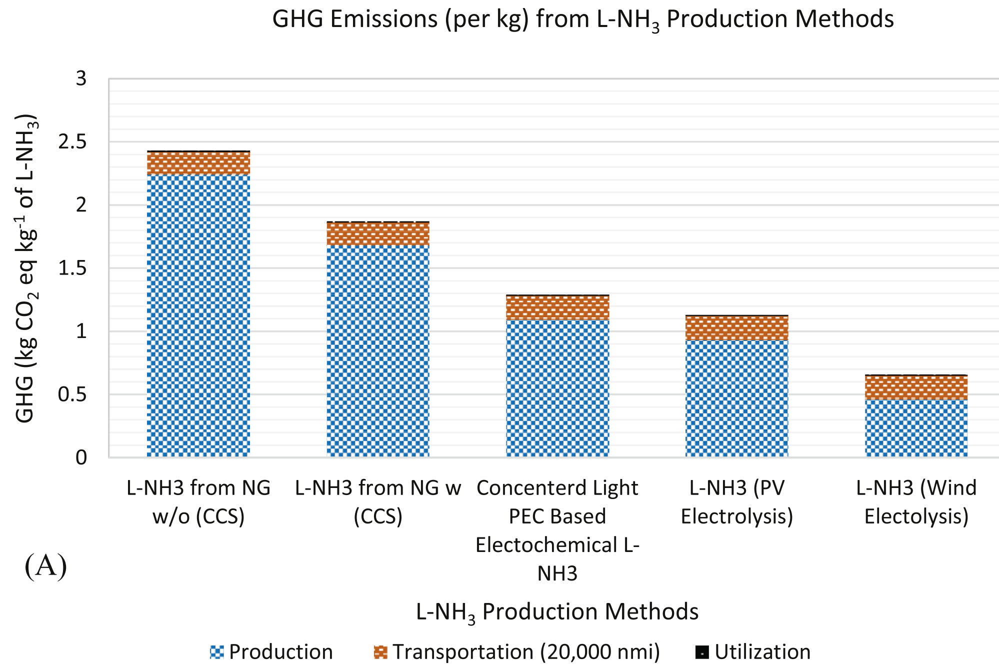
0.47 and 0.93 $CO_2$ eq $kg^{-1}$ of L-$NH_3$ accordingly. Concentrated light PEC based electrochemical method is a modern process to produce ammonia. GHG emissions of producing 1 kg of L-$NH_3$ was calculated as 1.09 kg of $CO_2$ eq. (Bicer et al., 2020). GHG emissions of this method are 35% less compared with natural gas reforming with CCS, and 15% higher than electrolysis of water using solar PV.

Accounting for the full life cycle is essential to achieve decarbonization as stated by IMO (IMO, 2009). Therefore, GHG emissions produced from L-$NH_3$ must be estimated based on a complete life cycle. Fig. 13A and Fig. 13B represent GHG emissions in different processes of L-$NH_3$ supply chain per unit mass (kg) and per unit energy (MJ), respectively. Emissions are mainly generated during the production phase even if different production methods are employed. GHG emissions generated in the production phase accounts for 90%, whereas transportation of L-$NH_3$ by HFO ocean tanker for long distances of about 20,000 nmi and utilization in internal combustion engine accounts for 8% and 2% accordingly of total emissions of L-$NH_3$. Embedding CCS facility into conventional ammonia production method (natural gas) emits about 100 g of $CO_2$ eq $MJ^{-1}$ of L-$NH_3$, which is 23% lower than a plant without CCS facility. On the other hand, the supply chain that uses electrolyzers for L-$NH_3$ production results in a large reduction of GHG emissions. Wind electrolysis-based ammonia production yields the lowest emissions of about 35.48 g of $CO_2$ eq $MJ^{-1}$ of L-$NH_3$.

Furthermore, other emissions are released when producing, transporting, and utilizing these fuels. $NO_x$ and $SO_x$ are hazardous emissions and authorities are paying attention to the consequences of these emissions. These emissions are obtained from GREET inventory data as they can be tracked in the provided supplementary file. Thus, Table 5 specifically shows $NO_x$ and $SO_x$ generated in the supply chain phases of various fuels produced from natural gas. The full supply chain of methanol has the lowest $NO_x$ emissions of about 76.35 mg $MJ^{-1}$. This makes methanol a promising alternative for complying with environmental regulations that aim to reduce hazardous $NO_x$ emissions.

Fig. 14 presents a comparative analysis in GHG emissions of methanol, DME, L-$H_2$, and L-$NH_3$ produced from renewables or natural gas over a variety of distances. The straight lines represent fuels produced from renewable energy sources. The results indicate a reduction in GHG emissions compared to fuel derived from natural gas. L-$H_2$ full life cycle emitted about 41.29 g of $CO_2$ $MJ^{-1}$ of L-$H_2$, and it is the cleanest fuel. When producing methanol, DME, L-$NH_3$ from natural gas, total GHG emissions of the full life cycle are 88.7, 88.9, 90.9 g of $CO_2$ $MJ^{-1}$ of fuel correspondingly. These emissions are nearly close to each other and ammonia has a slightly higher value. In contrast, the production of these fuels renewably shows that total GHG emissions L-$NH_3$ is about 50.5 g of $CO_2$ $MJ^{-1}$ of L-$NH_3$. This value is lower compared to methanol and DME.

To find out the lowest GHG emitting energy carrier in Qatar, the comparison is performed as shown in Fig. 15A and Fig. 15B in g of $CO_2$ eq $kg^{-1}$ and g of $CO_2$ eq $MJ^{-1}$, respectively. When comparing these fuels, generated GHG emissions from the utilization phase account for the most of emissions in LNG, DME, and MeOH supply chains, whereas utilization emissions of L-$H_2$ and L-$NH_3$ supply chain represent the least emissions. Even though LNG loses more of its energy due to high BOG rates, GHG emissions of the full supply chain of LNG emits fewer emissions compared to MeOH and DME produced from the reforming of natural gas, which have lower BOG rates. This is related to lower emissions at the production phase of LNG. However, the production of DME and methanol from biomass gasification have lower GHG emissions in the production phase compared to the LNG production phase which makes renewably produced DME and methanol more environmentally friendly. Moreover, implementing the CCS facility in the L-$H_2$ and L-$NH_3$ production phase reduces GHG emissions by 35% and 25% respectively which can make GHG emissions of full supply chain lower. Production of L-$NH_3$ from reforming of natural gas emits the most of GHG emissions however when producing $NH_3$ by renewables (PV), total GHG emissions of the full supply chain become lower compared to LNG, MeOH, and DME. Besides, the supply chain including production, 20,000 nmi transportation by HFO fueled ocean tanker and utilization in an internal combustion engine of L-$H_2$ produced from solar PV has the lowest GHG among these fuels of about 42.5 g of $CO_2$ eq $MJ^{-1}$ of L-$H_2$.



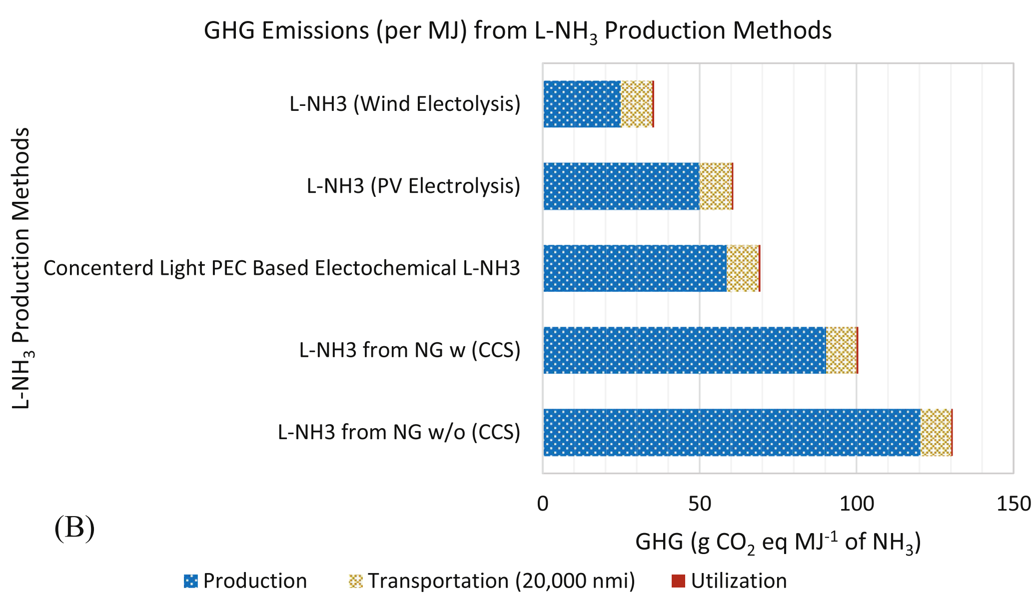




**Fig. 13.** Estimation of GHG emissions from different L-$NH_3$ production methods covering emissions in production, 20,000 nmi transportation by HFO fueled ocean tanker and utilization in an internal combustion engine, (A) per unit mass (kg), (B) per unit energy (MJ).

**Table 5**
$NO_x$ and $SO_x$ emissions in the production, transportation (by HFO tanker for 20,000 nmi distance), and utilization (in internal combustion engine) phases of various fuels produced from natural gas.

| Fuel Type | L-$NH_3$ (NG) | | LNG | | MeOH (NG) | | DME (NG) | | L-$H_2$ (NG) | |
|---|---|---|---|---|---|---|---|---|---|---|
| Unit | mg/MJ | | mg/MJ | | mg/MJ | | mg/MJ | | mg/MJ | |
| Emission Types | $NO_x$ | $SO_x$ | $NO_x$ | $SO_x$ | $NO_x$ | $SO_x$ | $NO_x$ | $SO_x$ | $NO_x$ | $SO_x$ |
| Production | 90.03 | 28.5 | 22.17 | 12.04 | 30.65 | 22.19 | 31.04 | 23.66 | 56.06 | 28.5 |
| Transportation | 34.9 | 15.5 | 35.1 | 21.04 | 37.1 | 20.24 | 41.24 | 25.99 | 34.9 | 15.5 |
| Utilization | 31.83 | 0.0 | 25.13 | 0.0 | 8.6 | 0.0 | 34.75 | 0.0 | 31.83 | 0.0 |

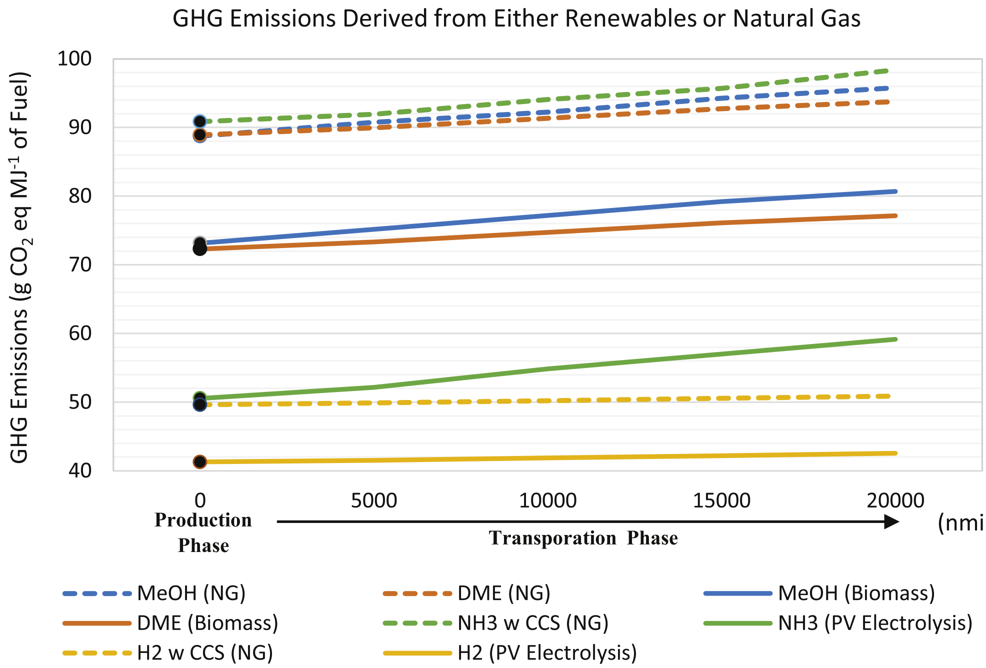



**Fig. 14.** Sensitivity analysis on various distances of a full life cycle of energy carriers derived from either renewables (solar PV and biomass) or natural gas covering production, transportation, and utilization phases.

## 5. Conclusions and recommendations

Investigating GHG emissions from the full life cycle of energy carriers covering production, transportation, and utilization phases by accounting for leaks and BOG generations can provide a full environmental assessment. Therefore, this study shows a comparison of GHG emissions from the full life cycles of energy carriers namely; LNG, DME, methanol, L-$H_2$, and L-$NH_3$ produced either from natural gas or renewables in Qatar. The main findings of this study are listed as follows:

- Methanol and DME are less harmful to the environment compared to LNG considering production and transportation phases only. However, the utilization phase has an important contribution to overall emissions. Considering the full life cycle, methanol and DME from the reforming of natural gas emits about 95.8 g of $CO_2$ eq $MJ^{-1}$ of methanol and 93.7 g of $CO_2$ eq $MJ^{-1}$ of DME. These values are 30% and 27% higher compared to the full life cycle of LNG. However, the production of methanol and DME from renewables (biomass) results in a reduction of GHG emissions by 75% and 80% respectively in their production phase, which makes DME and methanol alternative fuels besides LNG.
- Regarding generated emissions during the transportation phase, even though DME starts with greater GHG emissions at the production phase compared to LNG, as ocean distance length exceeds 14,000 nmi, emissions from LNG becomes greater.
- The desire to minimize GHG emissions in L-$H_2$ production methods is likely achievable using renewably generated electricity. When used electricity is produced from natural gas, the production of L-$H_2$ by steam reforming with CCS emits about 5.89 kg of $CO_2$ eq $kg^{-1}$ of L-$H_2$. Using renewables (solar PV) to produce electricity instead of natural gas results in a reduction of emissions by 40% compared to that method.
- Producing L-$NH_3$ by reforming of natural gas without CCS emits around 2.24 kg $CO_2$ eq $kg^{-1}$ of L-$NH_3$, however implementing CCS results in GHG reduction of about 24%. For the complete life cycle of L-$NH_3$, total GHG emissions of L-$NH_3$ by reforming of natural gas is 1.87 kg of $CO_2$ eq $kg^{-1}$ of L-$NH_3$, which is lower compared to LNG, DME, and methanol full life cycles.

A list of recommendations for decision-makers are presented here to comply with environmental regulations and meet UNEP standards as follows:

- Since Qatar has an abundant source of natural gas, converting natural gas energy into different fuels, instead of extensively focusing on LNG, will reduce international pressures toward environmental attitudes and result in making the country environmentally oriented.
- Converting natural gas into L-$NH_3$, methanol, and DME for long storage can potentially reduce GHG emissions instead of storing natural gas as LNG, which emits more GHG emissions during the storage phase.
- Increased utilization of L-$NH_3$ and L-$H_2$ in combustion processes for power generation and transportation can have direct environmental benefits.
- Producing and exporting natural gas as methanol and DME results in a reduction in GHG emissions compared to LNG production and transportation pathway, however the pattern is inverted when utilization in an internal combustion engine is added.

Future studies can include considering other environmental impact categories, such as ozone layer depletion, acidification, and marine sediment ecotoxicity. Since energy carriers differ in their chemical characteristics, released emissions as BOG are different in type and quantity. Hence, these impact categories can address the effects of a variety of emissions, which can provide a full environmental picture of an energy carrier life cycle. Moreover, assessing GHG emissions when the BOG is reused in a combustion process in an ocean tanker during the transportation phase can enhance the environmental performance of these energy carriers.

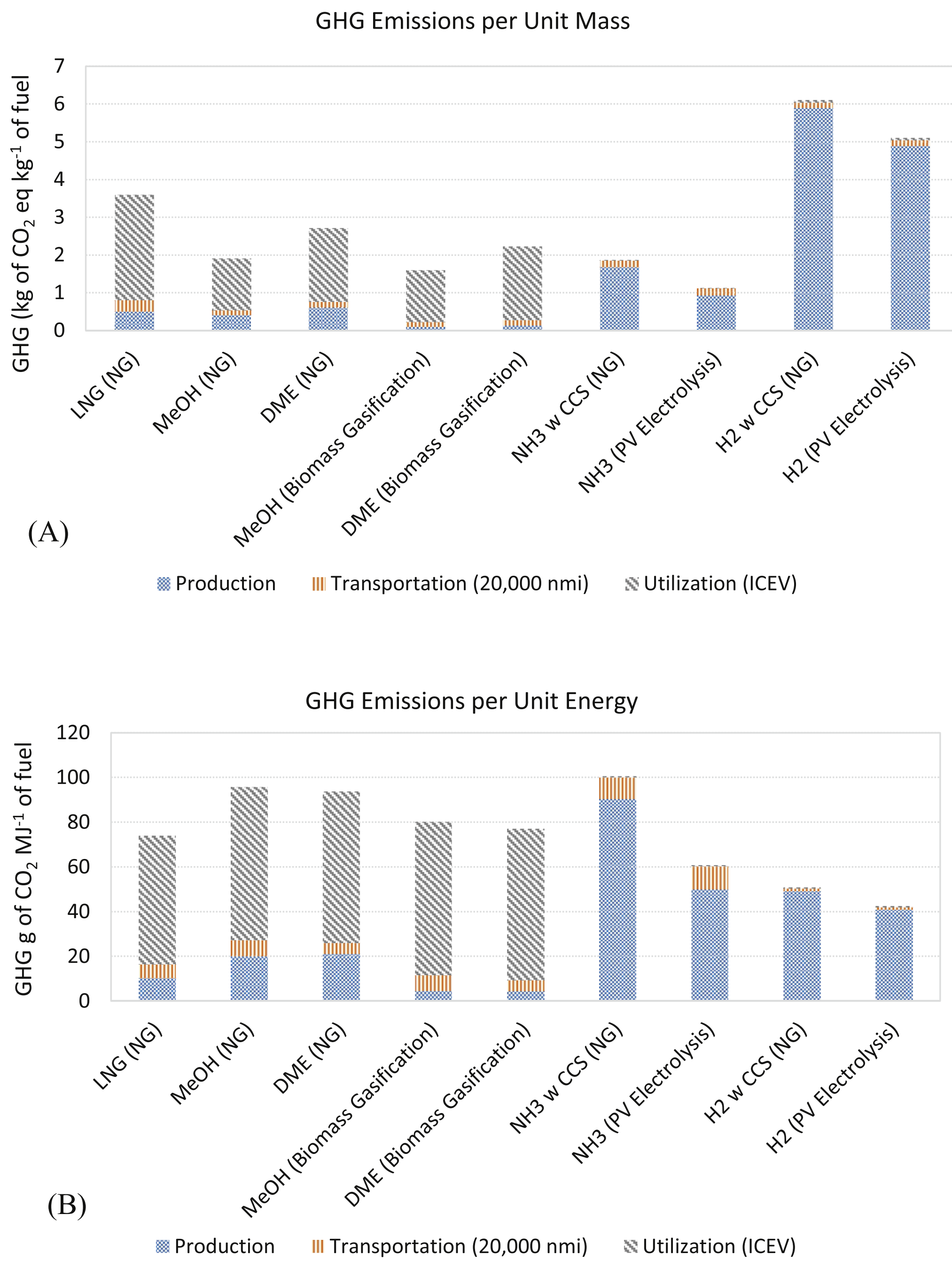



**Fig. 15.** B: Comparison of GHG emissions of different fuel supply chains, (A) per unit mass (kg), (B) per unit energy (MJ).

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## Declaration of competing interest

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

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2020.123481.

## Abbreviations

| | |
|---|---|
| BOG | Boil off-gas |
| DME | Dimethyl ether |
| eq | Equivalent |
| GREET | The Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation |
| GWP | Global Warming Potential |
| IMO | International Maritime Organization |
| L-$H_2$ | Liquid hydrogen |
| L-$NH_3$ | Liquid ammonia |
| nmi | Numerical mile |
| PV | Photovoltaic |

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