

Life Cycle Assessment of Methanol from Fossil, Biomass, and Waste Sources, and Its Use as a Marine Fuel in Dual-Fuel Engines

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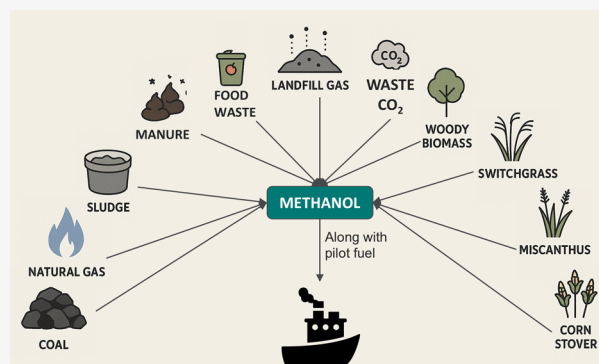
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ABSTRACT: Methanol is gaining interest in the marine sector from energy security and reducing emissions perspective. This study provides a comparative life cycle assessment of methanol as a marine fuel, across GHG and criteria air pollutant emission metrics, when it is used in a dual-fuel engine. Twelve methanol pathways from four different feedstock categories were considered, including (1) cellulosic biomass—forest residues and clean pine mix, corn stover, switchgrass, and miscanthus; (2) organic wastes—renewable natural gas from wastewater sludge, swine manure, food waste, and landfill gas; (3) fossil resources—coal and natural gas (NG); and (4) e-methanol using captured carbon dioxide. When used in a dual-fuel engine with pilot fuel, life cycle GHG emissions for woody biomass-based methanol were approximately 19 gCO₂e MJ⁻¹, while emissions from waste-based sources ranged between −154 and 31 gCO₂e MJ⁻¹. Methanol from renewable sources showed a GHG reduction potential between 58 and 226% compared to conventional NG-based methanol (122 gCO₂e MJ⁻¹), primarily due to the avoided emissions from conventional waste management. When carbon from process emissions were captured, the reduction could be up to 327%. All pathways exhibited lower NO_x and particulate matter emissions compared to the baseline marine fuel (MGO 0.1% sulfur), while woody biomass and coal pathways had higher SO_x emissions.

KEYWORDS: environmental sustainability, biomass gasification, pyrolysis, marine shipping, maritime transport



1. INTRODUCTION

The importance of the marine shipping industry is undeniable, as over 80% of global merchandise is transported via marine shipping.¹ Maritime trade is projected to grow by 2.4% annually until 2029.² However, increased shipping activity could lead to emissions rising by as much as 250% by 2050.³ On the other hand, the International Maritime Organization (IMO) envisions a carbon-neutral shipping sector by 2100 and has set an initial goal to reduce GHG emissions by 50% by 2050, compared to 2008 levels.⁴ Replacing marine gas oil (MGO, 0.1% sulfur) with alternative fuels would be necessary to comply with international shipping regulations. Alternative fuels, such as methanol, can provide a means to achieve these compliances with minimum changes to ship design, engine modifications, and fuel handling systems compared to those systems that require broader ship redesign or drivetrain overhaul. In addition to environmental benefits, there are also economic incentives to explore alternative fuels. The global bunker fuel market is currently valued at \$136 billion in 2025 and is expected to grow to \$199 billion by 2034.⁵ There are approximately 100,000 ocean-going vessels in service, which consume about 400 million tonnes of bunker fuel annually.⁶ For the sake of energy sustainability and security,

renewable fuels are becoming increasingly of interest to the shipping industry.

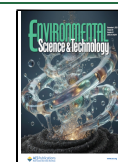
Among other alternative fuels, methanol is gaining attention in the shipping sector for several reasons. It is the simplest alcohol (CH₃-OH) with a one-carbon chain, making it easy to produce, store, and handle. Methanol remains liquid over a wide range of temperatures, from −98 to 65 °C. It has the highest hydrogen-to-carbon ratio of any liquid fuel, meaning that it releases less CO₂ per MJ in the use/combustion phase than other fuels. Additionally, methanol produces no sulfur emissions during combustion.^{7–10} The rising interest in methanol-fueled ships can be attributed to the lower costs associated with retrofitting existing vessels or constructing new ones. Unlike other fuels, methanol does not require expensive cryogenic fuel tanks or pressurization systems.¹¹ Additionally, methanol is also known to dissolve quickly and undergo rapid

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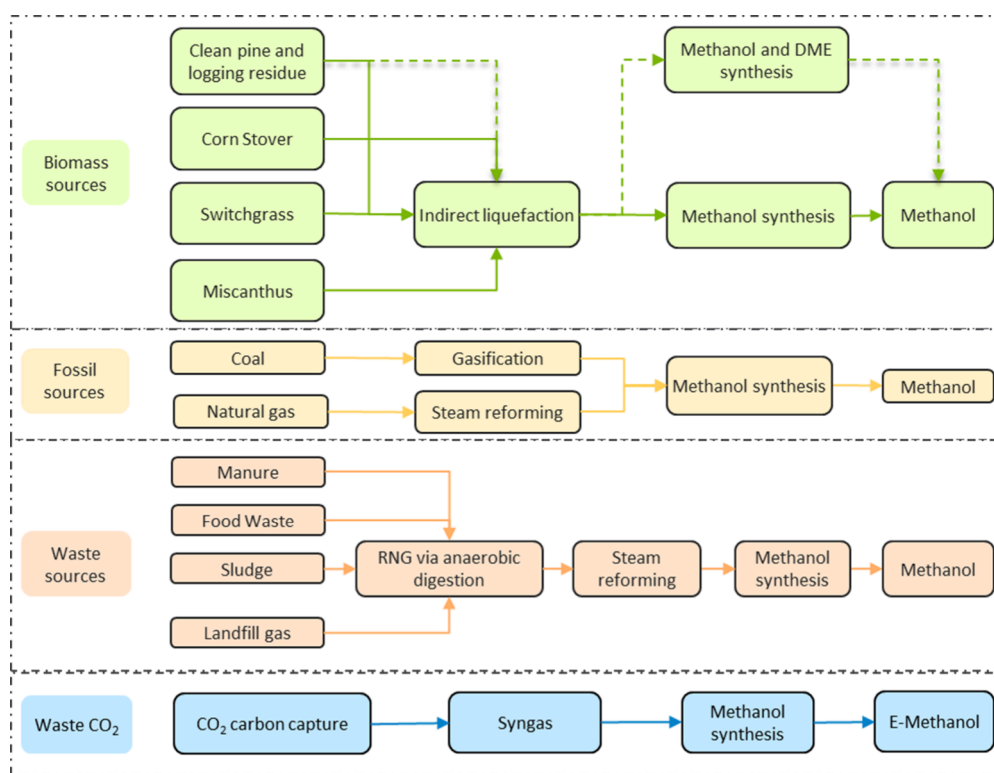


Figure 1. Pathways and system boundaries for methanol production. RNG = renewable natural gas, DME = dimethyl ether.

biodegradation in water in the event of a surface water spill, making it a safer option for the environment.¹²

Engine manufacturers such as MAN, Weichai, and Scania have demonstrated methanol's technical viability as a fuel through successful marine trials.^{13,14} The interest in methanol can be observed by the incorporation of methanol-ready and methanol-powered ships over the past few years.¹⁵ Among the ships in the order book in 2024, about 10% are either methanol-powered or methanol-ready.¹⁶ The world's first methanol-powered ferry was introduced in 2015 by Stena Line, in partnership with the Canadian chemical company Methanex, the Finnish manufacturing company Wärtsilä, and the Ports of Gothenburg in Sweden and Kiel in Germany.¹⁷ In 2016, the first methanol-powered ocean-going vessel, named Lindanger, was launched in Ulsan, South Korea.¹⁸ This cargo ship can run on marine diesel oil, fuel oil, and marine gas oil alongside methanol and has a capacity of 50,000 deadweight tonnes (DWT). Danish shipping and logistics company Maersk has ordered seven methanol-enabled container ships by 2023, six of which has 9000 TEU (20 ft equivalent unit) capacity.¹⁹ Interest in methanol increased even more after IMO provided the safety guidelines for using methanol as a maritime fuel in December 2020.²⁰ Methanol is the first alternative fuel planned to receive ISO standards for bunkering, which includes the transfer system, operational procedures, risk assessment, safety precautions, and personnel training.²¹ In an effort to create the next generation of environmentally friendly vessels and to adapt to the long-distance transportation needs, China Merchants Energy Transport Company signed a letter of intent with China Merchants Industry to develop methanol dual-fuel engine-based 9000 CEU (container equivalent unit) pure car and truck carriers (PCTC).²² The first two such vessels are expected to be delivered by 2025, with the possibility of four more vessels by 2026. In late 2022

(October), China's COSCO Shipping Holdings announced that they had ordered 12 new 24,000 TEU containerships powered by methanol dual fuel that cost \$239.85 million per ship.²³ HMM, a South Korean carrier company, has expressed interest in purchasing methanol-powered vessels as they invited tenders for nine dual-fuel ships in 2022, each with 8000 TEU.²⁴

Methanol from renewable sources has the potential to reduce up to 80% of carbon emissions by replacing MGO, the primary fuel used in the marine sector.²⁵ GHG intensity of methanol from biomass feedstocks ranges from 10 to 25 g CO₂e MJ⁻¹ depending on the feedstock;^{25–27} lower estimates were for forest residues and higher for dedicated herbaceous bioenergy crops such as poplar. Methanol derived from waste sources, such as cow manure (−55 g CO₂e MJ⁻¹)²⁵ and waste hydrogen and carbon dioxide (−66 g CO₂e MJ⁻¹),²⁶ displays negative GHG intensity. This is due to diverting waste for fuel production from conventional waste management system-generated avoided emissions credits.²⁵ However, methanol from fossil resources can exceed the GHG emissions of traditional MGO (88 g CO₂e MJ⁻¹).²⁶ The GHG intensity of coal-based methanol was 126 g CO₂e MJ⁻¹ in the USA,²⁶ while it can be as high as 300 in China.²⁵ Methanol derived from natural gas, the historically dominant way to produce methanol, was reported to have a GHG intensity ranging from 94 to 110 g CO₂e MJ⁻¹, which is similarly, if not more, GHG intensive than LSFO.^{25,26,28} This emphasizes the need for methanol production from renewable energy sources.

Modifications to ship engines and fuel handling systems will be necessary to use methanol, along with pilot fuel. Primarily, its higher octane number, lower calorific value, and higher heat of vaporization compared to conventional diesel indicate a higher heat requirement for the combustion of methanol.²⁹ To address this concern, a minimum of 6 to 17% marine gas oil or

Table 1. Conversion Methods, Co-products, Allocation Ratios, LCA Types, and Avoided Emission Scenarios for Methanol Production Pathways^a

feedstock	conversion and upgrading	coproducts	energy allocation ^b	avoided emissions
coal	gasification	sulfur	100%	N/A
natural gas	SMR to syngas and methanol synthesis	hydrogen	50%	
biomass	IDL and methanol synthesis	electricity, sulfur	100%	
	IDL and DME synthesis	DME, sulfur	11%	
RNG from wet wastes	SMR to syngas and methanol synthesis	hydrogen	50%	emissions from conventional waste management
waste CO ₂	syngas production and methanol synthesis		100%	emissions from releasing CO ₂ to atmosphere

^aNote: SMR = steam methane reforming, IDL = indirect liquefaction, DME = dimethyl ether. ^bPercentage of emissions allocated to methanol.

marine diesel oil is required as a pilot fuel, on an energy basis under various engine loads, for efficient combustion. In addition, methanol's lower energy density (approximately 20 MJ kg⁻¹ or 15.9 MJ L⁻¹) compared to marine gas oil (MGO, 42.8 MJ kg⁻¹ or 35.8 MJ L⁻¹) or low sulfur fuel oil (LSFO, 39.4 MJ kg⁻¹ or 39.1 MJ L⁻¹)²⁶ means that more fuel is required for the same trip and, therefore, a larger fuel tank is required. There could be a trade-off between fuel and cargo, i.e., the amount of cargo may need to be reduced for additional fuel. Additionally, even though the price of methanol (\$477 t⁻¹)³⁰ is lower compared to the LSFO (\$612 t⁻¹)³¹ on a mass basis, it is higher in terms of required energy due to the lower energy density of methanol.

In this study, our goal was to conduct a full life cycle analysis of methanol as a marine fuel, from multiple feedstocks and conversion methods, used in a dual-fuel engine, along with pilot fuel. There is a gap in the current literature on the use of methanol in a dual-fuel engine. This article reduces this gap by incorporating the latest engine testing data for methanol along with pilot fuel from a major marine engine manufacturer (MAN). Another novel contribution of this article is to include the impact of sequestering the process carbon emissions. Finally, we used the latest data set from GREET to conduct life cycle assessment of methanol and described the process and emission factors in great detail. We created a comparable system boundary for these pathways, created a cradle-to-grave life cycle inventory, discussed the life cycle emissions, and compared them with their fossil counterparts, as well as the IMO targets. We also analyzed the trade-off between additional fuel required and reduced cargo capacity and determined whether it is a significant reduction. This study will feed into the growing field of sustainable shipping and help improve the emission profiles of the shipping industry. Results from this study will support informed policy and investment decisions regarding methanol as a marine biofuel.

2. METHODS

2.1. Goal and Scope Definition. The purpose of this study is to conduct a full life cycle assessment of methanol as a marine fuel. We analyzed 12 methanol pathways from four different feedstock categories: fossil, biomass, organic waste, and captured CO₂ (see Figure 1). The GHG metrics included in the study are CO₂, VOC (volatile organic compound), CO, CH₄, and N₂O and are combinedly presented as CO₂e using Intergovernmental Panel on Climate Change Sixth Assessment Report (IPCC AR6) GWP (Global Warming Potential) definitions.³² Criteria air pollutants included in the study are oxides of sulfur (SO_x), oxides of nitrogen (NO_x), and particulate matter (PM10 and PM2.5). Details on coproducts, energy allocation ratios, and avoided

emissions scenarios—for RNG-based methanol and e-methanol—are listed in Table 1.

Produced methanol was considered for use in a marine dual-fuel engine. Engine testing data were acquired from the engine manufacturer, MAN Energy Solutions (www.man.eu). The functional unit of this study was emissions per combined unit energy, e.g., grams of CO₂e MJ⁻¹, at the end-use location. Combined unit energy refers to the energy derived from both methanol and pilot fuel necessary for easy ignition and efficient combustion. More information about the combination of these two fuels is discussed in Section 2.3.

2.2. Life Cycle Inventory of Methanol Production Pathways. The cradle-to-grave life cycle inventory for methanol production from biomass, coal, and natural gas was estimated using a bottom-up approach, relying on process designs.³³ To illustrate the process of converting woody biomass and bituminous coal to syngas via indirect steam gasification and steam reforming of natural gas, Figure S1, in the Supporting Information, provides a simplified block flow diagram. The key common processing areas include indirect gasification, syngas cleaning and conditioning, acid gas removal, and methanol synthesis, with detailed descriptions given in the earlier studies.^{34,35} The methanol plants process the following amounts of feedstock per day: 2000 dry metric tons (DMT) of biomass, 579 DMT of bituminous coal, and 544 t of natural gas. It is important to note that according to classical LCA methodology, the size of the plant does not influence the LCA results, as LCA is fundamentally linear and normalized to a functional unit (see Table S3).

2.2.1. Indirect Liquefaction of Biomass. Indirect liquefaction was used for biomass to methanol and biomass to DME and methanol pathways. We analyzed four biomass feedstocks—woody biomass (a mix of 50% clean pine and 50% logging residue), corn stover, switchgrass, and miscanthus. Before entering the gasifier, the biomass is sized to 2 cm (average size) and preheated using a cross-flow dryer that utilizes process waste heat. Additionally, the dryer serves as a contingency measure during wet weather, ensuring efficient feed drying. Details on the modeled feedstocks, such as carbon, hydrogen, nitrogen, sulfur, oxygen, ash content, and heating values are provided in the Supporting Information.³⁶

2.2.1.1. Gasification. The biomass gasification process involves employing preheated synthetic olivine sand in a char combustor to provide the necessary heat for the reaction.³⁴ To stabilize the flow of biomass and olivine, steam is injected into the gasifier. At a temperature of 1598 °F (870 °C), the biomass undergoes thermal deconstruction, resulting in a syngas mixture comprising CO, H₂, CO₂, CH₄, tars, and solid char containing residual carbon and coke deposited on the olivine. Cyclones are used to separate the char and olivine from the

syngas at the gasifier exit. Subsequently, the char is burned in the fluidized char combustor, while hot olivine, along with residual ash, is separated using a pair of cyclones. The hot olivine is recycled back into the gasifier to provide the necessary heat for the reaction. The ash and olivine fines are cooled and moistened to minimize dust before being disposed of as waste.

2.2.1.2. Synthesis Gas Cleanup and Syngas Compression. The syngas cleanup process in this design includes the reforming of tars, methane, and other hydrocarbons, followed by cooling, quenching, and scrubbing to prepare the syngas for downstream operations.³⁴ The reformer facilitates the water–gas shift reaction. Raw syngas is reacted with the tar reforming catalyst (Ni/Mg/K supported on alumina) in an entrained flow reactor at a temperature of (1670 °F/910 °C) and at a gas hourly space velocity of approximately 2500 h⁻¹. The catalyst is then separated from the effluent syngas in a cyclone. The spent catalyst is sent to the catalyst regenerator vessel to remove residual coke from reforming reactions through combustion. The hot catalyst is then separated from the combustion flue gas in the regenerator cyclone and returned to the tar reformer reactor to provide energy for the reforming reactions. The hot reformed syngas is further cooled through heat exchange with other process streams and scrubbed with water to remove impurities, such as particulates, ammonia, halides, and recalcitrant tars. Scrubber water is treated continuously at an on-site wastewater treatment facility. The low-quality heat in the flue gas from the catalyst regenerator is utilized for feedstock preheating. After quenching and removing condensable materials and solids, the low-pressure cooled scrubbed syngas is compressed using a three-stage centrifugal compressor with interstage cooling, increasing the pressure to approximately 430 psia (2.96 MPa).

2.2.1.3. Acid Gas Removal and Methanol Synthesis. The compressed fresh syngas undergoes acid gas removal in an amine-based acid gas enrichment unit and a Merichem LO-CAT sulfur recovery unit to eliminate CO₂ and H₂S.³⁴ The H₂S-rich acid gas stream is then directed to the Merichem LO-CAT sulfur recovery unit, where H₂S is converted to elemental sulfur and stored for disposal. The remaining CO₂ is vented to the atmosphere. After acid gas removal, the cleaned syngas is divided into two streams. A smaller portion (about 6%) of the cleaned syngas is directed to a pressure swing adsorption (PSA) system to separate hydrogen for hydrocarbon synthesis in the methanol to the high-octane gasoline area. Most of the cleaned and conditioned syngas is further compressed to 735 psia (5.07 MPa) for methanol synthesis, via commercial Lurgi's methanol technology, in a tubular, fixed-bed reactor containing a copper/zinc oxide/alumina catalyst. The methanol and unconverted syngas mixture is then cooled through heat exchange with the steam cycle and other process streams to recover methanol while allowing recycling or purging of unconverted syngas and inert gaseous species (CO₂, CH₄). To control the exothermic reaction, heat is removed from the methanol synthesis reactors by producing steam on the shell side of the tubular reactor. Temperature control and heat removal are achieved by back-pressure control at the outlet of the steam drum.

2.2.1.4. Dimethyl Ether Synthesis. Methanol can also undergo dehydration to make DME, which takes place in an adiabatic packed bed reactor with the commercially available γ -alumina (γ -Al₂O₃) catalyst at 482 °F (250 °C) and 140 psia (0.965 MPa).³⁵ The alumina-based catalyst is highly effective

and stable when it comes to catalyzing the vapor-phase dehydration of methanol to DME. This reaction is exothermic, and the heat produced by the reactor is recovered through an intercooler to generate steam. The reactor's temperature can reach a maximum of 482 °F (250 °C) due to the adiabatic temperature rise. DME is expected to exit the methanol-to-DME reactor in equilibrium with methanol at the reactor exit temperature, which results in an 88.5% conversion rate of methanol.

2.2.1.5. Methanol Conditioning. The crude methanol stream, condensed at elevated pressure and containing a substantial quantity of gas (mostly CO₂), is reduced to a lower temperature (110 °F/43 °C) and pressure (98 psia/0.68 MPa).³⁵ It is then sent to a distillation column to degas the methanol before reaching the intermediate storage phase.

2.2.2. Steam Reforming of Natural Gas or Renewable Natural Gas and Methanol Synthesis. Natural gas to methanol production includes supplying natural gas to the reformer. The syngas cleanup and compression process are similar to the description provided in Section 2.2.1.2. During the acid gas removal and methanol synthesis stage (Section 2.2.1.3), excess hydrogen is produced—about 0.99 MJ of H₂ per MJ of methanol—and can be sold as a coproduct.

Renewable natural gas (RNG) to methanol pathways were developed by replacing natural gas with RNG from waste feedstocks. Feedstock choices for the RNG were manure, sludge, food waste, and landfill gas. Water scrubbing was considered for all waste feedstock upgrading. For manure-based RNG production, animal waste feedstock was diverted from the conventional waste management system to produce RNG. This resulted in avoided emissions from business-as-usual waste management practices. For example, a conventional manure management system in open pits or lagoons releases methane, which is avoided when manure is diverted for RNG production. On the other hand, conventional manure management systems produce organic fertilizers, which need to be replaced with inorganic fertilizers. Therefore, manure-based RNG production included avoided emissions credit, while adding an emissions burden for producing inorganic fertilizers. Overall, RNG from manure has net negative GHG emissions (−130 g CO₂e MJ⁻¹).

For sludge-based RNG production, sludge is diverted from a conventional waste management system in which sludge is routed for anaerobic digestion and flaring. This process produces less GHG emissions compared to manure management and therefore generates lower credit when diverted for methanol production.

For the landfill gas (LFG) to methanol pathway, LFG was collected in a collection system that otherwise would have been flared, which would release GHGs. However, LFG upgrading for RNG production releases about 17.4 g of CO₂e MJ⁻¹ as methane. Overall, RNG production from LFG has net positive GHG emissions, approximately 20.3 g CO₂e MJ⁻¹ of RNG. Details on the RNG pathways are in R&D GREET 2024,^{26,37–39} and the supply chain emission estimates for RNG are reported in the Supporting Information (Table S2). The subsequent processes remain the same as natural gas to methanol.

2.2.3. Coal Gasification. When bituminous coal is the feedstock, the modeled coal feed is mixed with water and the resulting slurry undergoes crushing and screening.^{40,41} During the gasification stage, the coal slurry after the pretreatment step is fed to a high-temperature entrained flow slagging gasifier

under conditions similar to those of a shell gasifier. The coal gasification requires pure oxygen in which its flow rate is regulated by the desired gasifier temperature (2470 °F/1354 °C). Thus, an air separation unit (ASU) is required. The modeled ASU produces nearly pure oxygen (95 mol %). Both the air separation unit and high-temperature gasification were modeled using conditions specified in the Aspen Plus's Integrated Gasification Combined Cycle (IGCC) model.⁴⁰

During the acid gas removal and methanol synthesis stage in the coal-to-methanol pathway, additional hydrogen is required to achieve the H₂-carbon ratio of 2, according to (H₂-CO₂)/(CO+CO₂), in the methanol synthesis reactor feed since the initial hydrogen-to-carbon ratio is low (Table S1). This additional hydrogen is produced from coal gasification with carbon capture and storage (CCS). This is in contrast with natural gas or RNG to methanol pathways in which excess hydrogen is produced.

2.2.4. E-Methanol Synthesis. E-methanol was produced from the reverse water gas shift (RWGS) process with H₂ recycle. Waste CO₂ from industrial processes (such as natural gas processing, methanol, and/or ammonia production) was captured and used as feedstock. The feedstock hydrogen is produced via SMR of natural gas following the pathway presented in R&D GREET 2024.²⁶ Approximately 62% of the energy required for e-methanol production comes from electricity, while the remaining energy comes from fuel gas. US standard grid electricity emissions are considered for electricity used in the RWGS process. Full inventory is presented in the Supporting Information (Table S3).

For e-methanol produced from waste CO₂, it is assumed that the use of CO₂ avoids the release of that CO₂ into the atmosphere. Therefore, diverting CO₂ for methanol production will provide feedstock CO₂ credit. However, the e-methanol pathway does not receive the biogenic CO₂ credit during combustion to avoid double counting of any CO₂ credit. The required hydrogen to react with CO₂ and produce methanol came from natural gas. Even though blue hydrogen—produced from natural gas with CO₂ captured—and renewable electricity could have been considered, we assumed natural gas-based hydrogen and grid electricity to produce e-methanol. This assumption was made because the energy carriers for other pathways primarily came from fossil resources except for the RNG to methanol pathways.

2.3. Life Cycle Impact Assessment. We conducted the life cycle assessment using the Excel module of the 2024 R&D GREET database.²⁶ Developed and annually updated by the Argonne National Laboratory, R&D GREET is an LCA tool where emission factors for most major fuels and inputs are provided. We calculated the emissions (g CO₂e MJ⁻¹) using the following equation:

$$\text{emissions} = (\text{FP} + E + \text{M \& CI} + C + \text{PE}) \times \text{EAR} - \text{DC} + \text{MGO}_{\text{SC}} + \text{T \& D} + \text{use} \quad (1)$$

FP represents emissions related to feedstock production. Biomass feedstock production includes site preparation, farming inputs, diesel use for input application, and collection, handling, preprocessing, and storage. For woody biomass, preprocessing includes chipping, while grass or cellulosic feedstocks are baled. When NG is used as feedstocks, the production process includes NG recovery, conventional NG processing, shale gas recovery, shale gas processing, and NG transmission and distribution. Details on these processes,

including processes required for coal mining, can be found in the R&D GREET 2024 database.²⁶ Most feedstock productions had positive GHG emissions. However, RNG derived from sludge, manure, and food waste has negative GHG emissions due to the credit for emissions that are avoided from conventional waste management practices. This is discussed in the Methods section (Section 2.2.2). Emission parameters for feedstock production were from the R&D GREET 2024 database²⁶ and are presented in the Supporting Information (Table S2).

E represents the emissions related to the supply chain emissions and use of energy products, such as natural gas, coal, and electricity, during the fuel conversion process. M&CI are emissions from the use of materials and chemical inputs such as magnesium oxide, fresh olivine, cooling tower chemicals, boiler feedwater chemicals, lo-cat chemicals, dimethyl disulfide, and amine makeup; C are the emissions from the use of catalysts such as tar reformer catalyst, methanol synthesis catalyst, DME catalyst, and zinc oxide; and PE is the process emissions. Emission factors for M&CI and C are presented in the Supporting Information (Table S5). Noncombustion process emissions are presented in Table S4. EAR is the energy allocation ratio, reported in Table 1. DC is the displacement credit for sulfur and electricity (US grid average) displacement. Table S2 presents the emission parameters for the displacement credit (DC) for electricity and sulfur.

MGO_{SC} represents the upstream or supply chain emissions for marine gas oil, which is used as pilot fuel along with methanol in a dual-fuel engine. T&D represents the methanol transportation and distribution-related emissions. For the coal-to-methanol pathway, T&D also includes coal transportation. For biomass and RNG to methanol pathways, transportation of feedstock is included in the feedstock supply chain. MGO_{SC} and T&D-related emissions are presented in Table S6. Use is the combined emissions from using methanol and pilot fuel in a dual-fuel engine. Combustion emissions at every energy load with a 5% interval are presented in the Supporting Information (Table S7). The tested engine was a two-stroke, slow-speed engine with a specified maximum continuous rating of 46,300 kW at 75.5 rpm (RPM). This RPM information was used to compare the IMO NO_x emission limit,⁴² 14.4 g NO_x kWh⁻¹, or 4 g NO_x MJ⁻¹. Pure MGO combustion estimates were also provided by MAN (Table S8). The IMO emission profile of the engine load and percentages of a trip spent with the respective engine load was used to calculate the emission estimates (Table S9).

Capture and storage of CO₂ from methanol production processes is considered for all of the pathways. CO₂ in methanol production is assumed to be pure, and 99% of all emissions during the conversion stage, including combustion of energy carriers and noncombustion emissions, were captured from the exhaust system without any further input using the Shell Cansolv Post-Combustion CO₂ capture system.⁴³ Fossil, biogenic, and noncombustion emissions during methanol production are presented in the Supporting Information (Table S4). The electricity requirement for CO₂ compression was 351 MJ t⁻¹, similar to the CO₂ compression condition for the ammonia or natural gas processing industry.

2.4. Scaling-up Consideration. Availability of different feedstocks in the US was considered for scaling up of methanol production. We used the US DOE Billion Ton study estimates for most feedstock availability,⁴⁴ while captured CO₂

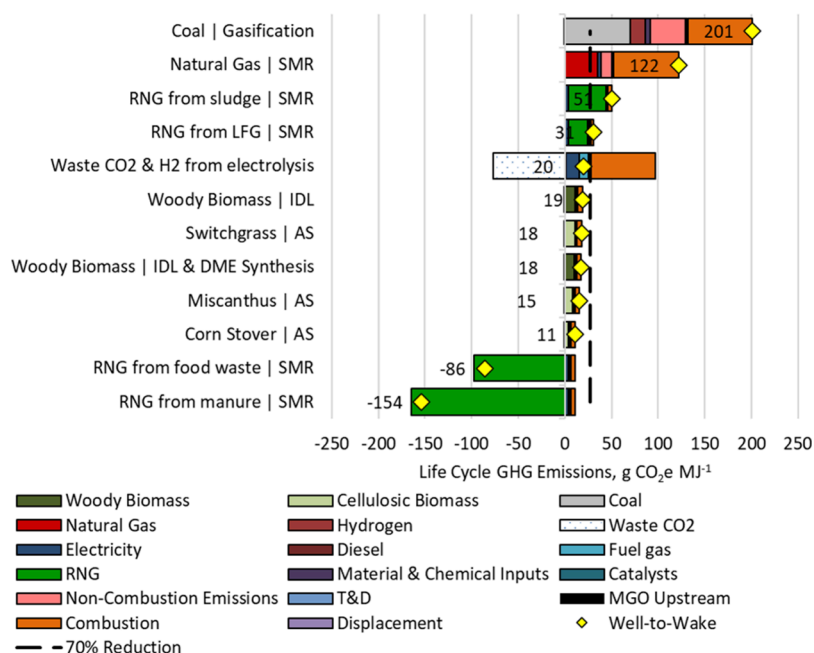


Figure 2. Life cycle GHG emissions of methanol as a marine fuel used in a dual-fuel engine with marine gas oil (MGO) as the pilot fuel. RNG = renewable natural gas; SMR = steam methane reforming; IDL = indirect liquefaction; AS = alcohol synthesis; LFG = landfill gas. 70% reduction line refers to reduction compared to MGO.

availability data was acquired from the US Congressional Budget Office.⁴⁵

Although an important consideration with methanol is its energy density, which is approximately half that of marine diesel oil, we did not perform any impact study on cargo carrying capacity using methanol. Even though some tank volume adjustments and associated cost are necessary, other ship operations, such as cargo carrying capacity, are not expected to experience any impact. For example, a cargo ship with 1109 m³ of fuel storage volume (1099 t) used for MGO would require 2718 m³ of methanol (2160 t). If the tank size is fixed, then the vessel range would be reduced by approximately 43%. However, increasing the tank size would be a better option from a logistics perspective since the increase in fuel storage volume for methanol to the total ship storage volume will be insignificant (0.5% for methanol and 0.2% for MGO).

3. RESULTS AND DISCUSSION

3.1. GHG Emissions. The life cycle GHG results of methanol, along with pilot fuel for dual-fuel combustion, show 270% lower to 122% higher emissions compared to conventional MGO (Figure 2), highlighting the critical importance of understanding the feedstocks and processes used to produce methanol when considering its suitability for contributing to GHG goals. The life cycle GHG emissions from methanol without the consideration of pilot fuel are provided in the Supporting Information (Figure S2).

Methanol from cellulosic feedstocks such as switchgrass, miscanthus, and corn stover provided 80 to 88% GHG reduction as well. It is important to note the higher potential of cellulosic feedstocks, as quantified by the 2023 Billion Ton Report, and therefore their potential to produce impactful fuel volumes. Additionally, forest biomass pathways provided approximately 78% GHG reduction compared with marine gas oil. According to the Methanol Institute, the GHG emission of methanol from forest residues was 10 g CO₂e

MJ⁻¹. Our estimate was 90% higher. Part of the increased emission was due to the supply chain-related emissions of the pilot fuel, and the remaining emissions were due to the mix of clean pine (50% of the total feedstock mix) along with logging residues.

Producing methanol from the RNG produced from organic waste such as manure and food waste offers the potential for large life cycle GHG reductions on an energy basis, in some cases offering net negative opportunities. For example, manure- and food waste-based methanol achieve 270% and 195% GHG reduction, respectively. These results should be interpreted in the context of the lower resource potential of organic waste feedstocks, the significant competition for negative CI fuels, and the large existing market for natural gas, which could readily accept all the available RNG. It is important to consider that using RNG for methanol production represents an expansion of the natural gas market amid an energy transition, where reducing the use of conventional natural gas is already a major challenge.

It is important to note that the net negative results for wet waste feedstock pathways presented here are based on emissions avoided relative to the current waste management practices, while the negative emissions from the pathways based on cellulosic biomass with capture and storage reflect a physical flow of CO₂ from the atmosphere into storage (which is discussed in Section 3.1.1). Presumably, the standard management practices for organic wastes will be improved as the economy moves toward net zero emissions, and as this happens, the avoided emissions benefit will decrease and eventually go away entirely.

Landfill gas to renewable natural gas as an intermediate fuel has positive GHG emissions, primarily due to methane emissions during RNG production. Methanol from other waste feedstocks—manure, food waste, and sludge—receives the avoided emissions credit for producing RNG with diverted waste feedstock from conventional waste management. Our

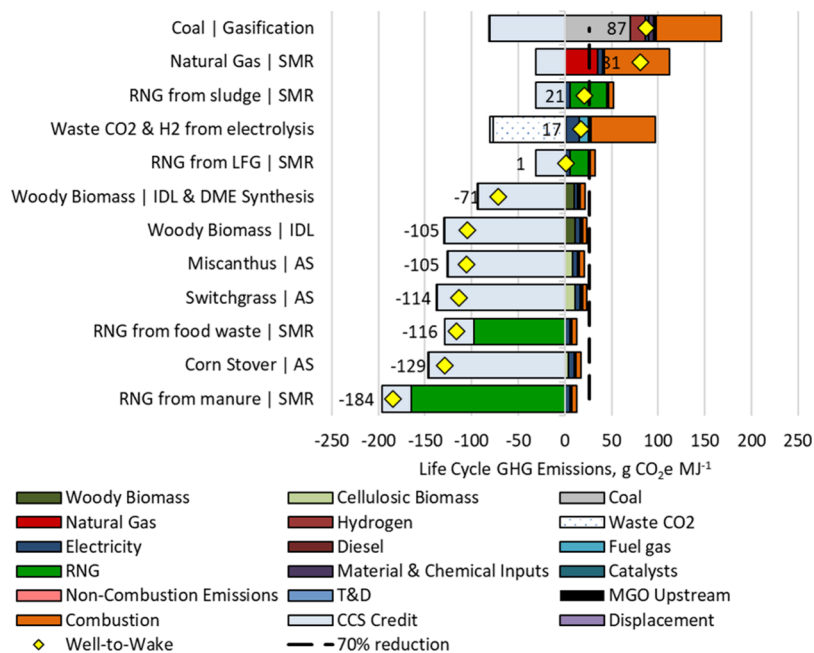


Figure 3. Life cycle GHG emissions of methanol as a marine fuel used in a dual-fuel engine with marine gas oil (MGO 0.1% sulfur) as pilot fuel with carbon capture. RNG = renewable natural gas; SMR = steam methane reforming; IDL = indirect liquefaction; AS = alcohol synthesis; LFG = landfill gas.

estimate was about 208% lower for methanol from swine manure compared to the estimate reported by the Methanol Institute, primarily due to the differences in the avoided emissions credit received by avoiding business-as-usual waste management. It is important to note that in this study, RNG production from waste feedstocks uses RNG as a process fuel. Using fossil resources as process fuel, such as natural gas, will increase the GHG estimates reported in this study.

These waste pathways and the biomass pathways also receive the biogenic CO₂ credit added to the combustion estimate, which e-methanol from the waste CO₂ and electrolysis pathways does not. Despite that, e-methanol provided 76% GHG reduction compared to the baseline. If renewable electricity (not accounting for embodied emissions) is used for hydrogen production or methanol synthesis, GHG emission from e-methanol goes from 17 to 3 g of CO₂e MJ⁻¹.

Fossil-based methanol, from coal or natural gas, shows higher GHG emissions compared to MGO. Coal used for gasification and as an energy carrier was the most GHG-intensive stage (36% of the total) in the coal-to-methanol pathway, followed by the combined combustion of methanol and MGO (34%). Methanol Institute's GHG emissions estimate for methanol from coal was 50% higher compared to this study.²⁵ Their coal production takes place in China, where coal mining is more GHG-intensive than in the USA, which is used for this study. Most of China's methanol production is currently derived from coal, and its methanol production is projected to increase.^{46,47} Even though US coal mining is less GHG intensive compared to China, there was a 128% increase compared to the life cycle GHG emissions of MGO. The other fossil fuel feedstock considered in this study was natural gas, which shows approximately 39% higher GHG emissions compared to MGO. Our estimate was about 12% higher than the estimates reported by Methanol Institute for the NG-to-methanol pathway,²⁵ primarily due to the differences in life cycle inventory. For example, in this study,

methanol is produced with hydrogen in an approximately 50:50 ratio. As hydrogen is gaining attention and is considered a high-value product, this pathway may become lucrative to fuel producers and other industry stakeholders. Nevertheless, fossil-based methanol without any carbon capture will not be an effective option for the international shipping sector's GHG emissions reduction objective. When pilot fuel is not considered, GHG from these fossil sources were even higher (Figure S2) since their GHG emissions are higher compared to the pilot fuel (MGO 0.1% sulfur).

3.1.1. GHG Emissions with Carbon Capture. Capturing the emissions from the fuel production stage offers a significant opportunity for reducing GHG emissions and creating net negative or biomass with carbon removal and storage (BiCRS) opportunities. In biological processes, these emissions can come from fermentation gas, which typically has a high CO₂ concentration and is therefore easier to capture and purify for transport and storage. Thermochemical processes require significant amounts of heat, which can be produced by combusting biomass.

Methanol from manure-based RNG remained the least GHG-intensive option when carbon capture was implemented in the conversion stage (Figure 3). However, cellulosic biomass options showed the highest emission reduction due to carbon capture. Life cycle GHG emissions were reduced by approximately 140, 121, and 132 gCO₂e MJ⁻¹ for cellulosic biomass options—corn stover, miscanthus, and switchgrass, respectively. This is primarily due to the reduction of biogenic CO₂ emissions—ranging from 126 to 146 g of CO₂ MJ⁻¹—during the conversion stage of cellulosic biomass to methanol that was considered neutral due to their biogenic nature without the carbon capture consideration. With carbon capture turned on, these emissions were captured and thus provided large CCS credit. The woody biomass to methanol pathway provided about 124 g of CO₂e MJ⁻¹ reduction due to a large CCS credit. Even though the coal-to-methanol pathway

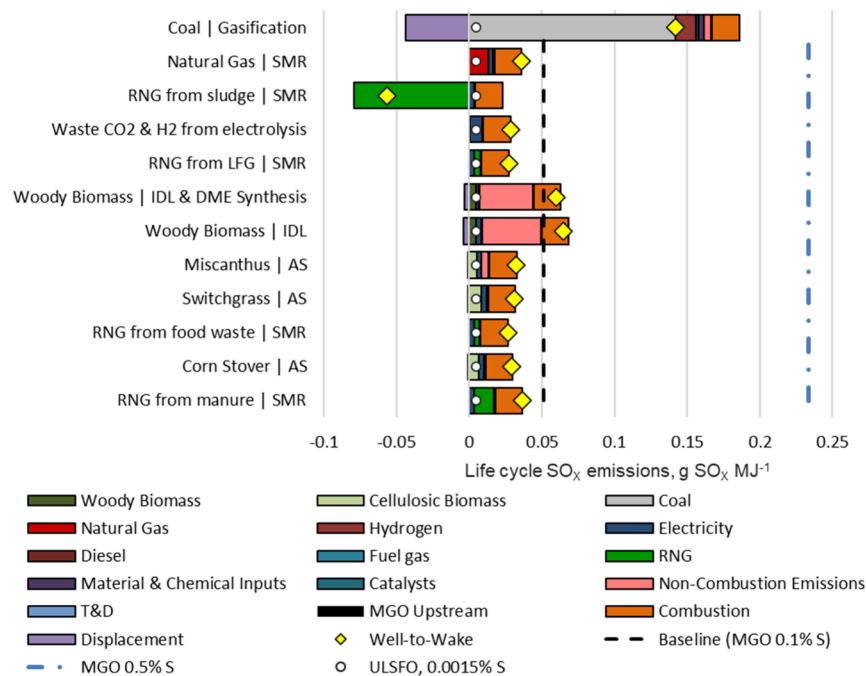


Figure 4. Life cycle SO_x emissions of methanol as a marine fuel used in a dual-fuel engine with marine gas oil (MGO 0.1% sulfur) as pilot fuel. RNG = renewable natural gas; SMR = steam methane reforming; IDL = indirect liquefaction; AS = alcohol synthesis; LFG = landfill gas.

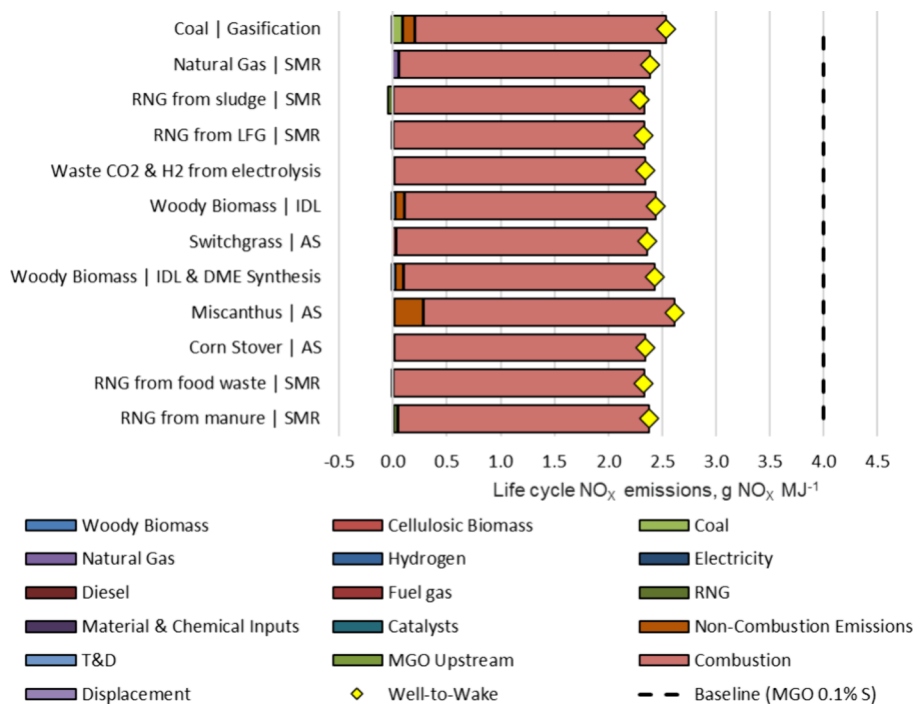


Figure 5. Life cycle NO_x emissions of methanol as a marine fuel used in a dual-fuel engine with marine gas oil (MGO 0.1% sulfur) as pilot fuel; the IMO Tier II limit is for engines running under 130 rpm. RNG = renewable natural gas; SMR = steam methane reforming; IDL = indirect liquefaction; AS = alcohol synthesis; LFG = landfill gas.

generated about 81 g of CO₂ MJ⁻¹ of CCS credit—due to capturing both the combustion of coal and the noncombustion process emissions—which is lower compared to MGO, it remained the most GHG-intensive option among other methanol pathways.

3.2. SO_x Emissions. Even though methanol does not have sulfur molecules, pilot fuel (MGO, 0.1% S) does. The production process also generates sulfur over the life cycle.

Coal gasification to methanol, the most SO_x-intensive pathway, emitted over 200% more SO_x over its life cycle compared to MGO (Figure 4). Coal gasification is a SO_x-intensive process, when coal is used as an energy carrier. Approximately 0.2 g SO_x MJ⁻¹ is emitted into the atmosphere when coal is used in an industrial boiler.²⁶ Even though the coal-to-methanol production process generates displaced sulfur and electricity credits, life cycle emissions still exceeded other

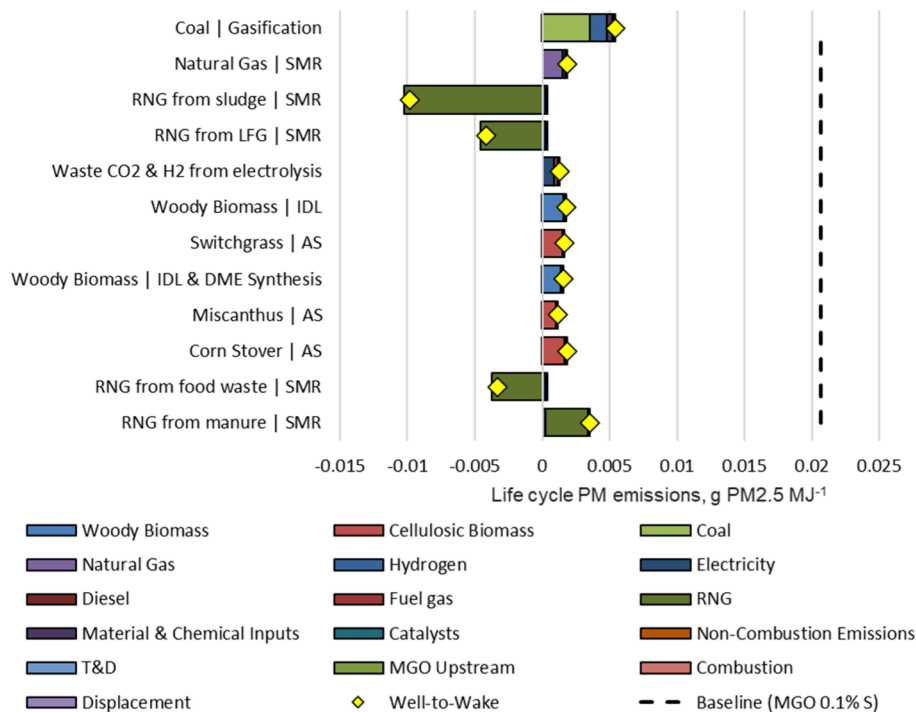


Figure 6. Life cycle particulate matter (PM_{2.5}) emissions of methanol as a marine fuel used in a dual-fuel engine with marine gas oil (MGO 0.1% sulfur) as pilot fuel. RNG = renewable natural gas; SMR = steam methane reforming; IDL = indirect liquefaction; AS = alcohol synthesis; LFG = landfill gas.

pathways. Woody biomass liquefaction is associated with some noncombustion SO_x emissions; however, life cycle emissions were lower compared to the baseline. All other pathways, except for sludge to methanol, had similar SO_x emissions as MGO 0.1% S. RNG production from sludge includes credit for fertilizer displacement and avoidance of emissions by diverting waste from BAU practices. About 92% of the SO_x credits come from the fertilizer displacement stage.

We also compared our life cycle SO_x emission results with two common marine fuels—MGO 0.5% sulfur and ultralow sulfur diesel with 0.0015% sulfur (Figure 4). It should be noted here that sulfur in the lubricants used in the engine during combustion produces SO_x. This is not explored in this study because of the lack of comparative data. Also, because both combustion scenarios—pure MGO combustion and dual-fuel combustion with methanol and MGO—will require lubricants that contain sulfur. Since the sulfur content of methanol is insignificant, the main source of engine-out SO_x emissions is from the pilot fuel and the lubricating oil. Thus, the use of methanol in a dual-fuel engine can potentially reduce (or eliminate) scrubber costs by significantly decreasing engine-out SO_x emissions.

3.3. NO_x Emissions. Combustion NO_x emissions from methanol dual-fuel operation were under the current IMO NO_x emissions limit for engines with less than 130 rpm engine speed and provided approximately a 40% life cycle NO_x emission reduction compared to MGO (Figure 5).⁴² Almost all of the NO_x emissions occurred during the fuel combustion stage. Even though methanol does not have nitrogen in the fuel, combustion along with atmospheric nitrogen can produce NO_x.

3.4. Particulate Matter Emissions. All pathways emitted less PM into the atmosphere compared to MGO life cycle PM emissions (Figure 6). Combustion accounted for 90% or more

of the PM emissions among all categories. RNG from sludge provided the highest PM reduction (68%) compared to that of the baseline fuel. While RNG production from sludge reduces PM emissions due to fertilizer displacement and avoided energy and emissions, RNG from LFG reduces PM emissions only due to avoided energy and emissions. On the other hand, manure to RNG production emits PM in the anaerobic digestion phase, resulting in the most PM-intensive among all renewable pathways. However, coal-methanol had the highest PM emission among all pathways. Total PM₁₀ emissions are provided in the Supporting Information (Figure S3).

As a fuel, methanol has advantages compared to conventional MGO in terms of its use-phase emissions of a criteria pollutants, specifically SO_x, NO_x, and PM. This shows that methanol can reduce life cycle GHG emissions while simultaneously reducing local criteria pollutant emissions.

3.5. Scaling-up Consideration. Although biomass-to-methanol pathways provided significant GHG emission reduction, biomass availability could be an issue in scaling up the production of biomass-based methanol. According to the Billion Ton 2023 report, 131 million tonnes of forest and woody biomass were used for energy and biobased chemical production in the US.⁴⁴ If the entire amount is used for methanol production, approximately 58 million tonnes or 73 billion liters of methanol can be produced. However, this biomass feedstock will be used for various other purposes. On the other hand, total distillate fuel and residual fuel sales in 2019 were approximately 250 billion liters. Since methanol's energy density is approximately half of distillate and residual fuel, approximately 500 billion liters of methanol will be necessary to replace the fuel.

The geographical distribution of biomass and waste feedstock is also an issue from a logistics perspective. The supply centers of feedstock and demand centers of marine fuel

may require more optimization to reduce transportation distance and, therefore, costs and emissions.

According to the Congressional Budget Office, the total carbon capture capacity in the USA was approximately 22 million metric tons of CO₂ per year, which was 0.4% of the nation's annual CO₂ emissions.⁴⁵ If all captured CO₂ was used only for methanol production, 16.7 billion liters of e-methanol could be produced. Low-carbon electricity requirement is another important consideration for e-methanol as its electricity requirement for water electrolysis will be much higher compared to other pathways. This study does not utilize projected lower grid emissions in the future. This study did not consider low-carbon electricity sources such as nuclear- or hydropower. As increased renewable and nuclear electricity generation goals are realized,⁴⁸ e-methanol production is expected to increase considerably, and associated GHG emissions will decrease.

As discussed in Section 2, methanol's energy density was not considered as a concern for other ship operations, such as cargo carrying capacity since the ratio of tank size to total cargo carrying capacity is insignificant.

A limitation of the study is the omission of energy requirements of transporting and injecting CO₂ in the carbon capture and storage scenario. Another limitation was the assumption of uniform transportation and distribution-related emissions across all feedstocks, except coal. Realistically, these feedstocks would have different feedstock collection radii and, therefore, spatially varied transportation distances.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.est.5c08873>.

Feedstock specifications for woody biomass and bituminous coal; emission factors of feedstock supply chain and displacement credit; detailed life cycle inventory for all pathways presented in the study; fossil, biogenic, and noncombustion emissions during methanol production; emissions factors for all inputs and transportation processes; combustion characteristics of marine gas oil and methanol with pilot fuel in dual-fuel engine; time share and engine load for a standard trip; simplified process flow diagram; life cycle GHG emissions results of methanol without pilot fuel; and life cycle PM10 emissions results of methanol as a marine fuel with MGO as pilot fuel (XLSX)

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Notes

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■ ABBREVIATIONS

USD United States dollar
CO₂e carbon dioxide equivalent
g grams
GHG greenhouse gases
LCA life cycle assessment
LFG landfill gas
LSFO low sulfur fuel oil
NO_x oxides of nitrogen
PM particulate matter
SO_x oxides of sulfur
t tonne or metric ton
TEU twenty-foot equivalent unit

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