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Biofuel Options for Marine Applications: Technoeconomic and Life-Cycle Analyses

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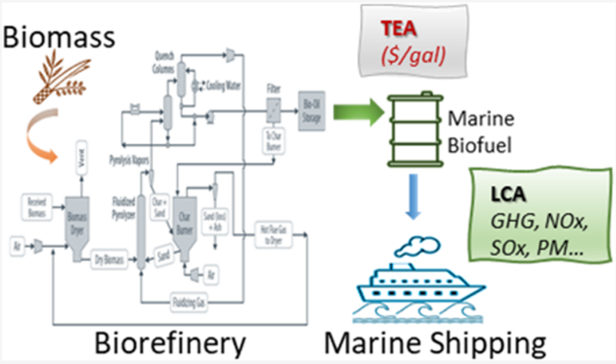

ABSTRACT: This study performed technoeconomic and life-cycle analyses to assess the economic feasibility and emission benefits and tradeoffs of various biofuel production pathways as an alternative to conventional marine fuels. We analyzed production pathways for (1) Fischer−Tropsch diesel from biomass and cofeeding biomass with natural gas or coal, (2) renewable diesel via hydroprocessed esters and fatty acids from yellow grease and cofeeding yellow grease with heavy oil, and (3) bio-oil via fast pyrolysis of low-ash woody feedstock. We also developed a new version of the Greenhouse gases, Regulated Emissions, and Energy use in Transportation (GREET) marine fuel module for the estimation of life-cycle greenhouse gas (GHG) and criteria air pollutant (CAP) emissions of conventional and biobased marine fuels. The alternative fuels considered have a minimum fuel selling price between 2.36 and 4.58 \$/heavy fuel oil gallon equivalent (HFOGE), and all exhibit improved life-cycle GHG emissions compared to heavy fuel oil (HFO), with reductions ranging from 40 to 93%. The alternative fuels also exhibit reductions in sulfur oxides and particulate matter emissions. Additionally, when compared with marine gas oil and liquified natural gas, they perform favorably across most emission categories except for cases where carbon and sulfur emissions are increased by the cofed fossil feedstocks. The pyrolysis bio-oil offers the most promising marginal CO_2 abatement cost at less than \$100/tonne CO_2e for HFO prices >\$1.09/HFOGE followed by Fischer−Tropsch diesel from biomass and natural gas pathways, which fall below \$100/tonne CO_2e for HFO prices >\$2.25/HFOGE. Pathways that cofeed fossil feedstocks with biomass do not perform as well for marginal CO_2 abatement cost, particularly at low HFO prices. This study indicates that biofuels could be a cost-effective means of reducing GHG, sulfur oxide, and particulate matter emissions from the maritime shipping industry and that cofeeding biomass with natural gas could be a practical approach to smooth a transition to biofuels by reducing alternative fuel costs while still lowering GHG emissions, although marginal CO_2 abatement costs are less favorable for the fossil cofeed pathways.

KEYWORDS: *heavy fuel oil, low-sulfur marine fuels, decarbonization, technoeconomic analysis, life-cycle assessment, sustainability, biofuels*

1. INTRODUCTION

The marine shipping sector heavily depends on fossil fuel and is one of the largest petroleum fuel consumers. The annual global marine fuel consumption was estimated to be around 400 million metric tons in 2019 (2.5 billion barrels).[1] Moreover, ocean shipping is one of the most significant contributors to emissions of sulfur oxides (SO_x), nitrogen oxides (NO_x), and particulate matter (PM). Global shipping contributes 13% of human-caused emissions of SO_x[2] and 2.6% of anthropogenic carbon dioxide (CO_2) emissions.[3] As a major source of pollutant emissions, the marine industry faces several challenges related to emission regulations. The International Maritime Organization (IMO) has set emission targets, known as IMO 2020, to reduce global marine fuel sulfur content from the current 3.5% m/m (mass by mass) to 0.5% m/m, starting on January 1, 2020.[4] Besides, the California Air Resources Board and other U.S. state agencies have established regulations limiting the sulfur content of fuel used in coastal regions to 0.1% m/m.[5,6] These regulations will require shipowners to find alternative fuel pathways. The options include low-sulfur heavy fuel oil (HFO), low-sulfur marine gas oil (MGO), installing sulfur scrubbers, or other alternative fuels/powertrains.[4] In addition to sulfur emission reduction targets, IMO has also established a framework explicitly for reducing the carbon intensity of shipping: 40% reduction relative to 2008 levels by 2030 and 70% reduction by 2050.[7,8]

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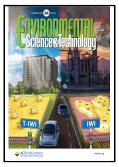

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The expected higher costs for low-sulfur marine fuels, other forthcoming emission regulations, and the additional processing associated with HFO could provide a new market opportunity for biofuels, which have inherently low-sulfur content and the potential to reduce PM emissions. Also, biofuels can reduce net CO_2 emissions due to the uptake of carbon from the atmosphere during biomass growth and play an essential role as a future marine fuel that is more renewable and lower in sulfur and other emissions.

To support this effort, we evaluate the economic potential of producing renewable fuels for marine propulsion. The selected fuel production pathways for the technoeconomic analysis (TEA) are (1) gasification and Fischer−Tropsch (FT) for a range of feedstocks including biomass, natural gas, biomass and coal, and biomass and natural gas (pathway 1); (2) conversion of extracted oils (yellow grease [YG] for this study) to marine fuels via hydrotreating with and without cofeeding of fossil feedstock (pathway 2); and (3) fast pyrolysis (FP) of low-ash woody feedstock to produce pyrolysis oil or bio-oil (pathway 3). The biofuels considered in this study are considered to be potential drop-in fuels compatible for use in marine engines; however, further work is needed to confirm compatibility and to address any potential issues that could be caused by differences in their properties.

In conjunction with the marine biofuels' TEA study, the second objective of this paper is to estimate the life-cycle greenhouse gas (GHG) and criteria air pollutant (CAP) emissions of the biofuels. As a comparison, this study also quantifies results for HFO, MGO, marine distillate oil, and their low-sulfur versions, as well as liquefied natural gas (LNG), soybean biodiesel, and straight vegetable oil (SVO). Except conventional HFO, MGO, and marine distillate oil, this study is the first time these pathways have been implemented as marine fuels in Greenhouse gases, Regulated Emissions, and Energy use in Transportation (GREET). While the models presented here leverage existing GREET supply chains, significant additions have been made to address the desulfurization of conventional fuels, detail the use of LNG as a marine fuel, and consider the effects of coprocessing biomass and fossil resources to produce marine fuels.

2. METHODS

2.1. Process Models for the Marine Biofuel Pathways. Process models for the marine biofuel pathways are developed for quantifying the product yields, raw materials, and energy consumption. Detailed process descriptions are provided in the Supporting Information. Pathway 1 is gasification and Fisher−Tropsch synthesis of gasified feedstocks. This pathway includes feedstock gasification; gas cleanup via reforming of tars and other hydrocarbons; syngas conditioning, including compression and acid gas removal; and Fisher−Tropsch synthesis, hydrotreating, and product separation (see Figure S1, Supporting Information). The pathway was modeled for biomass-to-liquid (BTL), natural-gas-to-liquid (GTL), and coal-to-liquid (CTL) options, as well as cofeeding biomass and natural gas (GBTL) and biomass and coal (CBTL). We consider four feedstock scenarios: (1) BTL with 100% biomass, (2) GTL with 100% natural gas, (3) CBTL, and (4) GBTL. For CBTL and GBTL, the fossil-to-biomass cofeeding feedstock is 51/49 by weight. The biomass pathways assume high-ash woody biomass.

Pathway 2 is a conversion of yellow grease to hydrocarbon fuels via hydroprocessed esters and fatty acids. Figure S2 in the Supporting Information provides the schematic process flow diagram. Two feedstock scenarios are considered: (1) yellow grease (100%) and (2) yellow grease and heavy fuel oil cofeed (50/50 by weight). Pathway 3 is fast pyrolysis of low-ash woody feedstock designed as a standalone process with a capacity of 2205 U.S. ton (2000 metric tons) dry biomass per day.[9] The simplified process flow diagram is provided in Figure S3 in the Supporting Information. We model low-ash woody biomass for this study. In contrast with gasification, fast pyrolysis technology is sensitive to feedstock composition, and feedstock ash and carbon content significantly impact product yield and composition. Therefore, the low-ash feedstock is typically used.[10] All feedstock costs and specifications are summarized in Tables S1 and S2 in the Supporting Information.

2.2. Technoeconomic Analysis. The technoeconomic analysis is performed based on *n*th-plant economic assumptions. The important aspect of *n*th-plant economics is that a successful industry has been established with many operating plants using similar process technologies. The TEA model encompasses a process model and an economic model. For a given set of conversion parameters, the process model solves mass and energy balances for each unit operation. These data are used to size and cost-process equipment and compute raw materials and other operating costs. Once the capital and operating costs are determined, we performed a discounted cash flow rate of return calculation to determine the minimum fuel selling price (MFSP) that meets the economic parameter using the general methodology[11,12] and the financial parameters summarized in Table S1 in the Supporting Information. The MFSP value represents the minimum selling price of fuels assuming a 30 year plant life and 40% equity financing with a 10% internal rate of return and the remaining 60% debt-financed at an 8% interest. These financial parameter assumptions were based on the U.S. Department of Energy's Bioenergy Technologies Office guidelines.[13] Sensitivity analyses on these parameters were performed. The unit for the MFSP is dollars per HFO gallon equivalent (HFOGE). HFOGE is determined using eq 1, using the lower heating value (LHV) of HFO (140 352 Btu/gal) obtained from the Greenhouse gases, Regulated Emissions, and Energy use in Transportation (GREET) model[14]

$$\text{HFOGE} = \frac{\text{LHV of fuel}}{\text{LHV of heavy fuel oil}} \tag{1}$$

2.3. Life-Cycle Analysis (LCA). Life-cycle analysis (LCA) is undertaken to screen marine fuel alternatives to gauge their potential emission benefits and identify potential tradeoffs among fuel options. The LCA results are also used together with the TEA results to estimate the marginal abatement cost (MAC) of CO_2 associated with the alternative fuel pathways. To provide context for the pathways that are the subject of the TEA, a total of nine fuel options are considered in the LCA (see Tables S3 and S4 in the Supporting Information). Typical HFO is represented by having a sulfur content of 2.7% by weight,[15−17] which reflects the industry average before the IMO limits that became effective in January 2020. We also consider HFO with sulfur contents of 0.5 and 0.1%. In addition, we consider MGO, a distillate fuel, with sulfur contents of 1.0, 0.5, and 0.1% by weight and marine diesel oil (MDO), which is a mixture of HFO and MGO. For this study, we approximate MDO as a 50/50 blend of HFO and MGO, for which we model three sulfur levels: 1.92, 0.5, and 0.1%

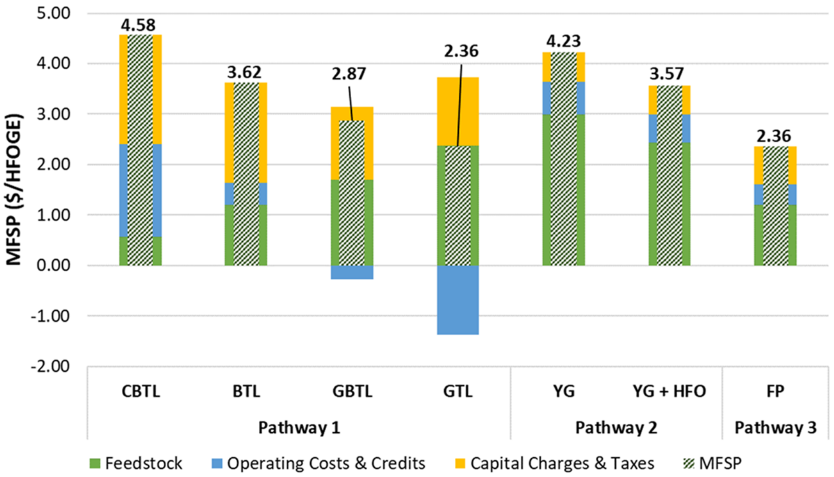

Figure 1. Comparative TEA result summary. One metric ton is approximately equal to 285.05 HFOGE, which can be used for the MFSP unit conversion from $/HFOGE to $/metric ton.

(based on blends of HFO 2.7%S (by weight) with MGO 1.0% S, HFO 0.5%S and MGO 0.5%S, and HFO 0.1%S and MGO 0.1%S, respectively). Beyond these petroleum-based marine fuels, we consider LNG, and soybean oil-derived biodiesel and straight vegetable oil (SVO). These were selected because LNG use as a marine fuel has recently expanded due to its competitive price and low-sulfur content, biodiesel produced from soybean oil is a commonly available biofuel in the United States, and SVO is included as a bounding case for a minimally processed biofuel option.

The scope includes the full life cycle of each fuel option. Supply-chain emissions are included for petroleum extraction and refining; biofuel feedstock growth, harvesting, and conversion; natural gas extraction and liquefaction; and fuel distribution operations. Fuel combustion emissions are estimated based on the best available data for the various fuels. Infrastructure associated with fuel production and distribution is outside the scope of this LCA. This study's scope also omits some fuel-specific onboard handling considerations, such as the effect of fuel storage space on ship cargo capacity and the effect of biofuel blends on HFO preheating requirements, which are the subject of ongoing research.

The GREET model[14] is used to evaluate the life-cycle GHG and CAP emissions for the fuel pathways based on a new version of the marine fuel module developed for this study and is publicly available. The module includes several new marine biofuel pathways and updated pathways for conventional marine fuels and natural gas as a marine fuel, together with updated combustion emission factors. The GREET model provides background data sets with a consistent and comprehensive system boundary, including all significant processes associated with raw material extraction, feedstock production, waste recovery logistics, fuel production, transportation along supply chains, and fuel combustion.[18] Data sets for the new marine fuel pathways presented here are summarized in Tables S2–S6 and Figure S4 in the Supporting Information, with some contribution to the publicly available in GREET 2019.[14] Detailed description of the GREET modeling of the evaluated marine fuels (production, consumption, and emissions) is provided in the Supporting Information.

Global warming potential for GHG emissions is calculated using the characterized factor for a 100 year time horizon from the Intergovernmental Panel on Climate Change Fifth Assessment Report from the life-cycle inventory values.[19]

Marginal abatement costs for GHGs and SO_x are calculated as the difference between the GHG and SO_x emissions for HFO and biofuel alternatives divided by the difference between the costs of HFO and biofuel alternatives. MACs for GHGs are presented relative to HFO 0.5%S, assuming it is the business-as-usual fuel following the IMO 2020 sulfur limits, while MACs for SO_x are calculated relative to HFO 2.7%S, as biofuels offer an alternative to HFO 0.5%S to meet the IMO 2020 sulfur limits. The values used to calculate MAC are found in Table S9 in the Supporting Information and Figure 1 (MFSPs).

3. RESULTS AND DISCUSSION

3.1. Process Performance. Product yields for the three pathways are summarized in Table S7 in the Supporting Information. For Pathway 1, the products include naphtha-, jet-, and marine-/diesel-range hydrocarbons. The product distribution is similar for all scenarios, roughly 20% for marine/diesel, 37% for jet, and 43% for naphtha. The FT process is based on the Anderson–Schulz–Flory (ASF) value of 0.84 with a H_2/CO ratio of 2.1 in syngas, which provides the maximum HFOGE yield.[12] Varying the ASF value affects the product distribution and therefore the MFSP. As the marine fuels are typically distillate and residual bunker fuel, the FT crude hydrocarbon separation process can be easily tailored to meet the desired properties for blending. Marine biofuels do not need intensive refining, and the cost of separation is relatively lower.

For pathway 2, the liquid fuel products also include naphtha-, jet-, and marine-/diesel-range hydrocarbons (Table S7, Supporting Information). The YG-only scenario's product distribution is roughly 11% for marine/diesel, 59% for jet, and 22% for naphtha. The product distribution for the case with HFO cofeed (YG + HFO) is roughly 18% for marine/diesel, 48% for jet, and 26% for naphtha. It is noteworthy that the hydrocarbon product distribution, including increasing the marine/diesel slate, can be manipulated at the product separation and fractionation step. The current process is modeled to produce jet fuel that meets the jet fuel specifications (e.g., high flash point and good cold flow properties), and these are accomplished by the hydrocracking and isomerization steps. The current processes can be further optimized for marine fuel-range hydrocarbon production. Additionally, cofeeding yellow grease with heavy oil provides another process parameter for achieving the targeted product distribution, which can be tailored by varying the two cofeeding feedstocks' ratios.

For pathway 3, the condensed bio-oil is the only hydrocarbon product (see Table S7, Supporting Information). As a potential drop-in fuel compatible for marine engines, bio-oil is not further upgraded via hydrotreating to transportation fuels (diesel- and naphtha-range blendstocks). Further work is needed to confirm compatibility and to address any potential issues that differences in their properties could cause. Substantial capital and operating expenditures are avoided, as hydrotreating is a multistep process involving a stabilizer, a low-temperature hydrotreater, a high-temperature hydrotreater, and even a hydrocracker and requires a substantial amount of hydrogen.[9]

3.2. Capital Investment. Capital investment for the three pathways is presented in Table S8 in the Supporting Information. For pathway 1, the total capital investment for the four feedstock scenarios increases in the order: GTL ($422 million) < GBTL ($455 million) < BTL ($633 million) < CBTL ($717 million). CBTL exhibits the highest total capital investment, mainly attributed to the air separation unit requirement and the more expensive high-temperature slagging gasifier. BTL and GBTL do not need an air separation unit to produce pure oxygen for the less expensive low-temperature entrained flow gasifier.

For pathway 2, the fuel upgrading area, which includes hydrogenation, propane cleave, and decarboxylation, and hydrocracking and hydroisomerization, exhibits the highest capital expenditure for both scenarios. The total capital investment for the case with YG only and the case with YG and HFO cofeeds is similar, at $257 and $263 million, respectively.

The total capital investment for pathway 3 is the lowest among all of the evaluated pathways, at $249 million. The fast pyrolysis reactor system represents approximately 90% of the total installed equipment cost ($138 million). The remaining 10% is attributed to the bio-oil filtration, char combustor, and utility system. The assumptions for capital cost estimates are provided in the Supporting Information.

3.3. Operating Costs and Coproduct Credits. Operating costs and coproduct credits for the three pathways are presented in Table S8 in the Supporting Information. Variable operating costs, including the annualized costs for catalysts, olivine, and waste disposal, are determined based on raw materials, waste-handling charges, and byproduct credits incurred only during the process operation. Fixed operating costs are generally incurred in full whether the plant is producing at full capacity. These costs include labor and various overhead items following previously published assumptions.[9,20,21] General overhead equals 90% of total salaries, maintenance equals 3% of fixed capital investment, and insurance and taxes equal 0.7% of fixed capital investment. For pathway 1, the fixed operating costs range from $0.50/HFOGE (GTL) to $0.75/HFOGE (CBTL). Similarly, the total operating costs vary from $1.02/HFOGE (GTL) to $2.40/HFOGE (CBTL). For GTL, the coproduct credits are largely attributed to excess hydrogen (owing to the inherently high hydrogen-to-carbon ratio of natural gas). Conversely, hydrogen import is required for the CBTL case.

The total operating costs for the case with YG only and the YG and HFO cofeed cases are $3.63/HFOGE and $3.00/HFOGE, respectively. The feedstock costs represent the most significant contribution (approximately 82%) to the operating costs. The lower operating cost for the HFO cofeed case is largely attributed to the lower HFO cost ($0.56/kg vs $0.61/kg for the YG) and the lower hydrogen cost.

The total operating cost for pathway 3 is $1.61/HFOGE. Variable costs (feedstock, chemical, utilities, and waste disposal) are about 80% of the total operating cost, while fixed costs (labors) are about 20% of the total operating cost. Woody biomass feedstock is the most expensive variable cost at $1.21/HFOGE or 75% of the total operating cost.

3.4. Minimum Fuel Selling Price. Minimum fuel selling price (obtained by performing the discounted cash flow analysis via iterating the selling cost of fuel until the net present value of the project is zero) across the pathways ranges between $2.36 and $4.58 per HFOGE, which are comparable to those with the similar technologies reported in the literature.[5,12,22,23] This analysis requires that the discount rate, depreciation method, income tax rates, plant life, and construction startup duration be specified. The fuel products (naphtha, jet, and marine/diesel blendstocks) are combined and referred to as a single-fuel product for simplicity. All MFSP calculations are performed and reported on a combined product basis. The cost contributions to the MFSP (Figure 1) are divided into capital charges and taxes, operating costs and coproduct credits, and feedstock costs (biomass, natural gas, and coal). The MFSPs for the evaluated scenarios for pathway 1 increase in the order: $2.36/HFOGE (GTL) <$2.87/HFOGE (GBTL) <$3.62/HFOGE (BTL) <$4.58/HFOGE (CBTL).

MFSPs for the two feed scenarios for pathway 2 are determined to be $4.23/HFOGE for the case with YG only and $3.57/HFOGE for the case with heavy oil cofeed (YG + HFO). These feedstock costs contribute about 70 and 68%, respectively, to the MFSPs. Under the *n*th plant assumptions, the MFSP for the fast pyrolysis marine bio-oil (pathway 3) is determined to be $2.36/HFOGE. Like pathway 2, feedstock cost is the most significant cost contributor to the MFSP (51% of the MFSP). Capital-related costs (capital depreciation, income tax, and return on investment) and operating costs (variable costs and labor) are about 32 and 17% of the MFSP, respectively.

GTL and fast pyrolysis show the lowest MFSPs ($2.36/HFOGE), attributing to a combination of favorable yields and lower operating costs for the former and high carbon-conversion efficiency (47%)[9] for the latter. Conversely, CBTL has the highest MFSP ($4.58/HFOGE) as a result of the higher capital expenditure associated with the air separation unit and the high-temperature slagging gasifier, as well as hydrogen cost. GBTL and YG + HFO exhibit relatively favorable MFSPs. TEA sensitivity study results are summarized in Figure S5, Supporting Information.

Cofeeding biomass with the fossil feedstock (natural gas or HFO) is a practical synergistic approach to improve liquid fuel yields while simultaneously lowering GHG emissions and suppressing cost. With this strategy, fuel producers could adjust the biomass and fossil feedstock shares over time to meet changing cost and greenhouse gas targets. Future study will explore opportunities to enable lower-cost biofuels, including using even lower-cost waste and low-quality feedstocks, intensifying process designs, and utilizing existing infrastructure to coprocess biomass or convert to biomass feedstocks altogether. While the costs for these processes could likely be improved through further integration and optimization, it is likely that environmental policy affecting the relative prices or ability to use HFO/very low-sulfur fuel oil would be the driver for the adoption of alternative energy sources for the maritime shipping industry.

3.5. Economic Viability of Biofuels for Marine Propulsion. The economic viability of biofuels for marine propulsion depends on several factors, primarily the price of low-sulfur HFO and other compliance costs with emission regulations. Fuel costs already represent more than 50% of total ship-operating expenses, even before complying with the IMO 2020, making it difficult for shippers to absorb additional expenses while remaining profitable.[1] The current TEA evaluations provide an important baseline analysis for establishing the feasibility of biofuel adoption for operating vessels. Figure 2 shows the rough projection of maximum biofuel prices as a function of the biofuel blending ratio, determined using the price premium for 0.5% sulfur bunker fuel over the current global standard of 3.5%, roughly $30/metric ton.[24] The isolines are for three HFO price scenarios: $1.50, $2.00, and $2.50 per gallon (1 gal HFO = 1 HFOGE). HFO accounts for more than 75% of the total marine shipping consumption[5] and is used for baseline comparison. The low-sulfur (0.5%) fuel price is the sum of the high-sulfur (3.5%) fuel price and the fuel premium (about $0.11/gal). For example, if the high-sulfur HFO price is $2.50/gal, the low-sulfur HFO price becomes $2.61/gal. The low-sulfur HFO prices would be the maximum threshold that shipowners are willing to pay for the biofuels for a one-to-one replacement; namely, the maximum acceptable biofuel prices would be the same as the low-sulfur fuel oil prices. However, as the biofuel blending ratio decreases, the biofuel price is allowed to increase and still meet the maximum price limit. For instance, the MFSP for GBTL is $2.87/HFOGE, and if the HFO (3.5% S) is $2.50/gal or $2.11/gal for the 0.5%S fuel, up to about 50% of the GBTL biofuel blendstock can be blended with the high-sulfur fossil HFO and still meet the $2.11/gal limit. Figure 2 also reveals the significant market penetration potential for marine biofuels. With the annual total marine fuel consumption of 400 million metric tons, even a 5% biofuel blending will translate to roughly 5.3 billion gallons of biofuel market. Here, we assume that biofuels contain virtually no sulfur and can be near-linearly blended with the sulfur components. FP, GTL, and GBTL appear to be economically viable as biofuel options for meeting the low-sulfur IMO regulation.

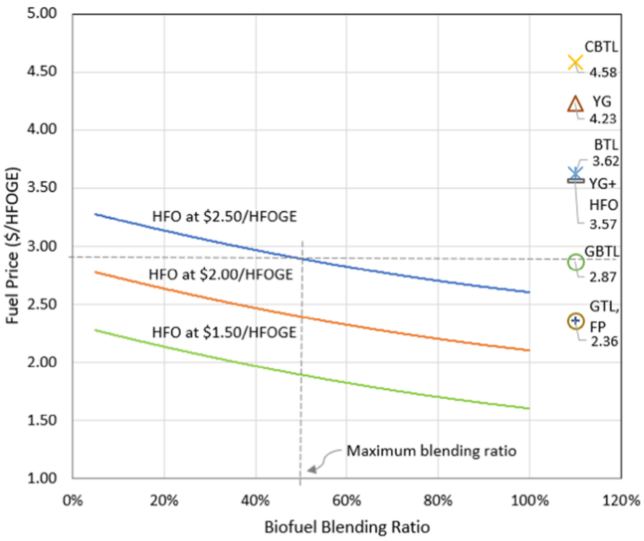

Figure 2. Estimated maximum allowable biofuel price for blending with conventional heavy oil (3.50% m/m S content) to achieve the 0.50% m/m sulfur target. Biofuel price estimates are based on a $30/metric ton premium of 0.50% over 3.50% m/m (1 metric ton = 285.05 HFOGE).

3.6. Life-Cycle GHG and Criteria Air Pollutant Emissions. Life-cycle GHG and CAP emissions for the 19 fuel pathways are presented in Figure 3 on a per-MJ basis. Results for each emission metric were normalized to the maximum to allow all metrics to be presented on the same axis. The figure's left side provides results for GHGs, with the first (top) figure showing total GHG emissions by substance and the next three showing contributions by the life-cycle stage for total GHGs, CO_2, and CH_4. For total GHGs and CO_2, carbon uptake during feedstock growth is shown as a negative contribution for these cases and the net result is shown as a black square in front of the stacked bar. Figures on the right show results for SO_x, $PM_{2.5}$, NO_x, and CO. The exact values are provided in the Supporting Information, Tables S9−S12. This per-MJ comparison is appropriate for comparing these fuel options as all are used in the same/similar internal combustion engine drivetrain. However, when comparing results, it is important to keep in mind that LNG and pyrolysis oil may be affected by other factors. The onboard storage and handling requirements for LNG differ significantly from those for the other fuels. For example, the requirement for LNG to be stored above deck could affect the container ships' cargo capacity. Pyrolysis oil is distinct among the fuel options in having a lower heating value of 15 MJ/kg compared with 37−49 MJ/kg for the other fuels (Table S4). Our preliminary analysis of the effect of this low energy density, based on the share of ship space devoted to fuel, suggests that while it may not have a large effect on cargo-carrying capacity or ship energy efficiency, it would affect a vessel's range for a given tank size.

The biofuel pathways generally offer improvements in GHG, SO_x, and PM over the petroleum-based fuels, while the trend for NO_x and CO is less clear with less variation across the pathways studied. Predictably, results across metrics are dominated by combustion emissions. Significant reductions in life-cycle GHG emissions for the biofuels are driven by carbon uptake during feedstock growth. Biofuels produced from 100% biomass have higher reductions in GHG emissions (67−93% compared with HFO 2.7%S) than those from the cofed pathways, CBTL and HFO + YG (reductions of 40−45%). Biofuels also offer low SO_x emissions due to their inherently low-sulfur contents. While life-cycle SO_x emissions for low-sulfur (0.1%S) HFO, MGO, and MDO are also quite low, ∼0.06 g/MJ, SO_x emissions for biofuel pathways are even lower, ranging from 0.005 to 0.03 g/MJ, the exception being the cofed pathways CBTL and HFO + YG. While combustion is the most significant source of SO_x emissions for the HFO, MGO, and MDO pathways, the conversion phase is a significant contributor to several biofuel pathways. The trend for PM emissions is like that for SO_x, although in the case of

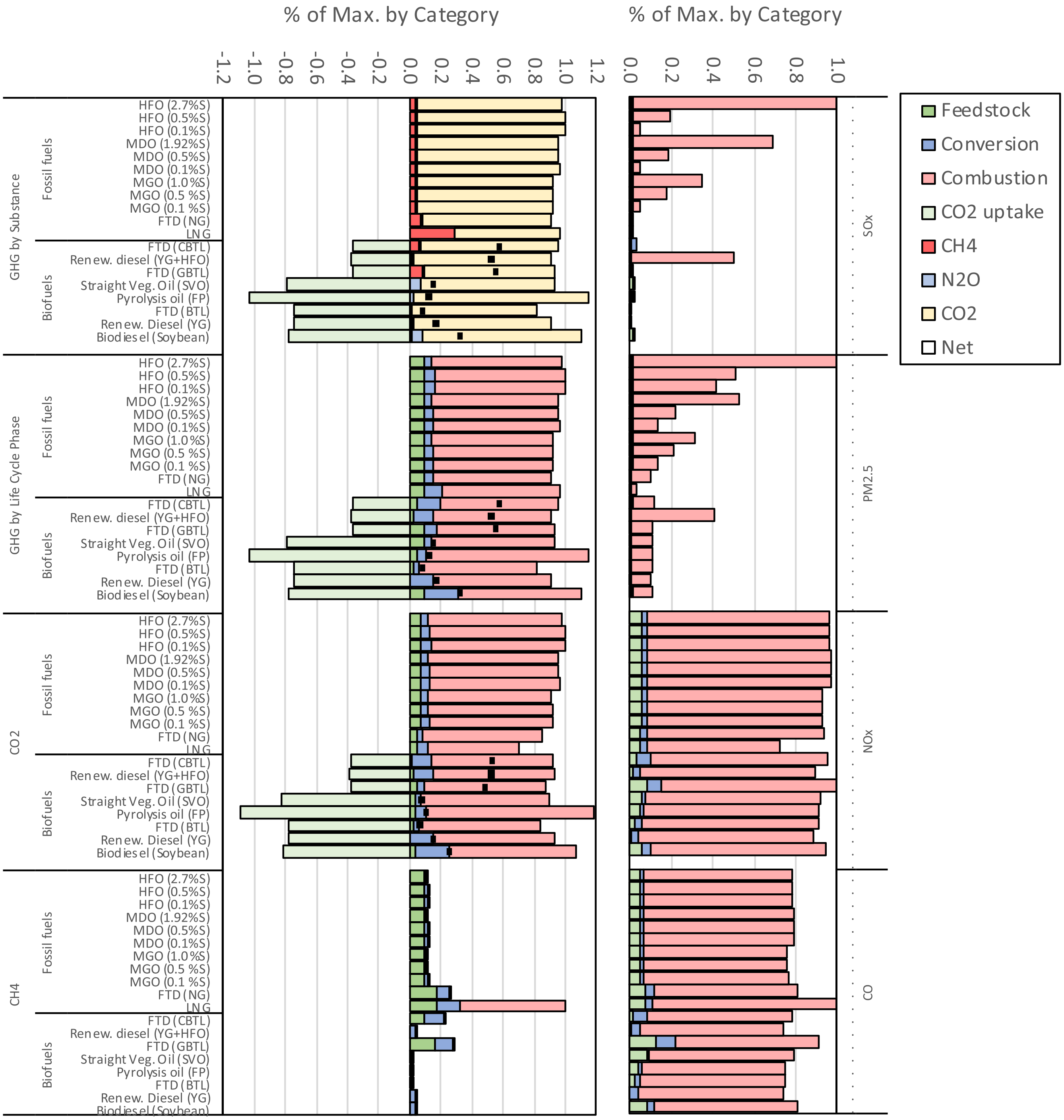

Figure 3. Emission results for marine fuel pathways. Bars are scaled to the maximum net result within each emission type. Medium-speed diesel engines under IMO emission tier 3 are assumed to be used. Results for greenhouse gas (GHG) emissions are shown both by greenhouse gas, uppermost left, and by life-cycle stage, second from top on left. Legend entries below the dashed line relate to the first, while those above relate to the second.

PM, the reduction in combustion emissions for biofuels is less dramatic and the differences compared with HFO, MGO, and MDO are less pronounced.

Low-sulfur HFO and MGO offer significant improvements in life-cycle SO_x emissions while incurring a relatively minor increase in GHG emissions. The life-cycle SO_x emissions of HFO 2.7%S and MGO 1.0%S are 1.35 and 0.47 g SO_x/MJ, respectively. These are reduced to 0.26, 0.064, 0.24, and 0.058 g SO_x/MJ through fuel desulfurization for HFO 0.5%S, HFO 0.1%S, MGO 0.5%S, and MGO 0.1%S, respectively. The life-cycle GHG emissions of HFO 2.7%S and MGO 1.0%S are 95 and 89 gCO_2e/MJ, respectively, with combustion contributing around 85%. Low-sulfur HFO 0.5%S and MGO 0.5%S have slightly higher life-cycle GHG emissions due to the additional hydrogen inputs estimated for reducing sulfur content by hydrotreating, 97 and 89 gCO_2e/MJ, respectively (i.e., an increase of 1.9 and 0.9%, respectively, for the sulfur removal).

LNG does not provide an appreciable GHG benefit. The life-cycle GHG emissions for the LNG pathway are virtually the same as those for HFO 2.7%S; we find a 1% decrease for

LNG, which is well within the margin of error for this analysis, attributing to significant methane emissions in both its supply chain and methane slip during combustion. Methane contributes 29% of the life-cycle GHG emissions for LNG, with roughly two-thirds of this from methane slip[25] and the remaining one-third from the supply chain.

LNG offers benefits for PM, SO_x, and NO_x emissions compared with HFO. Reductions in life-cycle SO_x and PM emissions for LNG are significantly reduced from HFO 2.7, 99, and 97%, respectively. LNG offers the lowest PM emissions across all of the pathways studied. LNG also stands out as offering a 26% reduction in NO_x compared with HFO 2.7%S.

Results for characteristic trips were also calculated for the various fuel options. The trends observed in these results on a per-trip basis (Figures S6−S9, Supporting Information) are similar to those presented in Figure 3.

These results corroborate other studies' findings while adding detail for specific biofuel pathways and providing a consistent basis for comparison across these fuels/pathways. The range of GHG reduction potential here, 67−93% relative to HFO 2.7%S, is on the high end of reductions for biofuel use reported by Bouman and colleagues,[26] who review quantitative life-cycle GHG results for seven studies of biofuels and find the range of GHG reductions to be 25−84%, which makes sense considering our selection of biofuel pathways with significant GHG reductions. The result for soybean oil biodiesel, 22 gCO_2e/MJ, matches the base-case value reported by the California version of the GREET model used to evaluate pathways for low carbon fuel standard credits, 28 gCO_2e/MJ. It is important to note that the results reported here are for U.S. production practices, reflecting an allocation of the soybean farming impacts to the soybean meal coproduct and U.S. transportation distances. Kesieme and colleagues report significantly higher results for biodiesel from soybean oil, ∼110 gCO_2e/MJ, due to higher impacts from soybean production in Argentina and transportation to the United Kingdom, and a higher allocation of impacts to the soy oil coproduct.[27] For this reason, it is important to carefully reflect the specific production system when considering biofuels produced via different pathways. Our LNG results correlate well with those reported by Thomson and colleagues,[25] who report results for a range of scenarios in the United States and Norway and find that the GHG impacts of LNG can range from an improvement to as much as a 12% increase compared with petroleum fuels. They also find that the GHG impacts of LNG are highly dependent on estimates of fugitive methane emissions along the supply chain and methane slip during use. Their results for SO_x, PM, and NO_x agree well with those presented here, finding that LNG can offer a significant improvement in SO_x and PM and comparable NO_x emissions relative to HFO.

3.7. Marginal Abatement Costs for SO_x and GHGs.

Marginal abatement costs for SO_x and GHGs for the seven alternative fuel pathways with TEAs are presented in Figure 4. Costs for GHGs are presented relative to HFO 0.5%S, as it is the business-as-usual fuel for the foreseeable future. In contrast, costs for SO_x are presented relative to HFO 2.7%S as HFO 0.5%S and these alternatives are competing options to meet the IMO 2020 sulfur limits. Recall that the difference in life-cycle GHGs for HFO 2.7%S versus HFO 0.5%S is only ∼2%. Costs are presented across a range of HFO price, as HFO prices have been volatile in recent months, with the price of HFO 0.5%S at the time of writing at roughly $1.30/gallon ($380/tonne), HFO380 as high as $1.78/gallon ($522/tonne) in the past year, and HFO 0.5%S at $2.34/gallon ($686/tonne) at its recent peak in January 2020. The alternative fuel prices used in the calculation are presented alongside the legend entries.

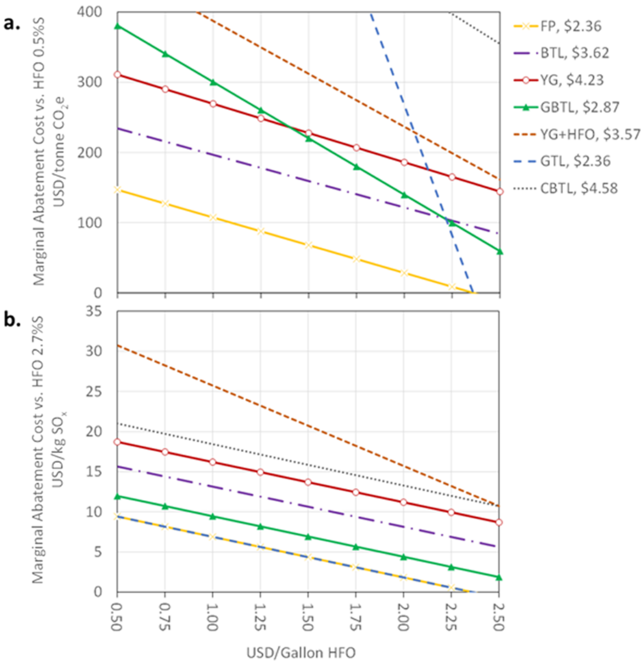

Figure 4. Marginal abatement costs for SO_x and GHGs for alternative fuel pathways relative to (a) HFO 0.5%S and (b) HFO 2.7%S. Results are shown for each of the biofuel pathways: pyrolysis oil (FP); Fischer−Tropsch diesel from biomass (BTL), natural gas and biomass (GBTL), coal and biomass (CBTL), and natural gas (GTL); and renewable diesel from hydroprocessing yellow grease (YG) and yellow grease with HFO (YG + HFO). Prices in the legend reflect the baseline price estimates for each in HFO gallon equivalents (HFOGE). Note that both fast pyrolysis and GTL have a price of $2.36/HFOGE and negligible sulfur content; thus, their results overlap in (b).

With HFO at $1.50/gallon, the MAC for fast pyrolysis oil via pathway 3 is $68/tonne CO_2e. This reduces to zero at higher HFO prices as it reaches cost parity with the fast pyrolysis oil. Similarly, HFO reaches cost parity with GTL within the range presented here; however, because the GHG reductions for the GTL pathway are much smaller than those for the biofuel pathways, the MAC increases sharply at lower HFO prices. MACs for FP, BTL, and YG have similar slopes due to their similar life-cycle GHG profiles and are separated by $60−100/tonne CO_2e and price differences. The MAC for the FP pathway 3 is consistently the lowest of the pathways considered, with MAC <$100/tonne CO_2e for HFO prices > $1.09/gallon ($320/tonne). The MACs for the BTL, GBTL, and GTL pathways all fall below $100/tonne CO_2e for HFO prices greater than roughly $2.25/gallon ($660/tonne). MAC below $100/tonne CO_2e for the biofuel options is promising, as decarbonizing the marine fuel sector is notoriously challenging due to the low cost of HFO and slim profit margins for ship operators. As a point of comparison, recent credit transfer rates for the California Low Carbon Fuel Standard have been around $200/tonne CO_2e. The marginal CO_2 abatement cost for pyrolysis oil falls below $200/tonne CO_2e regardless of the HFO price (down to zero), and BTL, GBTL, YG, GTL, and YG + HFO all fall below $200/tonne

CO_2e at HFO prices of around \$1.00, 1.60, 1.90, 2.10, and 2.25, respectively. These costs are in the range of those reported by Lindstad and colleagues,[28] who estimated ∼200 euros/tonne CO_2 reduction for marine biofuels in the European context. They could also be compared with the MAC for speed reduction and main engine retrofit, roughly \$55 and \$165 per tonne CO_2e, respectively, estimated by the International Council on Clean Transportation.[29] We find that the pathways combining fossil and biomass feedstocks do not perform as well as the pathways based solely on biomass feedstocks for the marginal CO_2 abatement cost, particularly at lower HFO prices.

The MAC for SO_x ranges from \$4 to \$21/kg SO_x for HFO at \$1.50/gallon. As most biofuel options have similarly very low life-cycle SO_x emissions, the slopes for the different pathways are nearly the same, while their y-intercepts are dependent on price. An exception is the YG + HFO pathway, which has higher SO_x emissions due to sulfur in the HFO portion of the feedstock. Note that GTL obscures the FP line as both have very similar prices and SO_x emissions. These costs could be compared with those for sulfur scrubbing presented by Jiang and colleagues (see Table S13, Supporting Information),[30] who report marginal SO_2 abatement costs of ∼\$2.40/kg SO_x for a new vessel and ∼\$3.30/kg SO_x for a retrofit based on an HFO price of ∼\$540/tonne or \$1.80/HFOGE. Jiang and colleagues also find that switching from HFO to MGO is cost-effective when the price difference is less than ∼\$279/tonne or \$0.95/HFOGE.

4. CONCLUSIONS

Biofuels could potentially be a cost-effective means of decreasing GHG, SO_x, and PM emissions from the marine shipping industry. This study evaluates a series of scenarios for three pathways for producing biofuels to offset the maritime use of HFO. Prices for the alternative fuels evaluated here range from 2.36 to 4.58 USD 2016/HFOGE and offer GHG reductions of 40−93% compared with HFO. While cofeeding natural gas and HFO improved the MFSP for the BTL and YG pathways, respectively, cofeeding coal did not improve the MFSP for the CBTL pathway. All of the alternative fuel pathways, except the one cofeeding HFO with yellow grease, offer significant SO_x and PM reductions, with SO_x reductions of 97−100% and PM reductions of 84−90%. Results for NO_x, CO, and VOC were mixed, with the biofuel pathways never exceeding 10% reductions and, in some cases, causing slight increases, up to 20%.

The pyrolysis oil, FT-diesel from biomass, and yellow grease hydroprocessing pathways all offer promising marginal CO_2 abatement costs below \$200/tonne CO_2e, with pyrolysis oil offering the lowest marginal CO_2 abatement cost with results falling below \$100/tonne CO_2e for HFO prices >\$1.09/HFOGE. Pathways combining fossil fuels and biomass do not perform as well for marginal CO_2 abatement costs, particularly at lower HFO prices. Similarly, the FP, GTL, GBTL, and BTL pathways offered the best marginal SO_x abatement costs, with pyrolysis oil and GTL offering marginal SO_x abatement costs < \$5/kg SO_x for HFO prices >\$1.38/HFOGE and GBTL dropping below \$5/kg SO_x for HFO prices >\$1.88/HFOGE.

Taken together, these results demonstrate the potential for biofuels to contribute to both decarbonization and SO_x mitigation objectives, with marginal CO_2 abatement costs comparable to those for other transportation fuels and in the range of current California Low Carbon Fuel Standard credit prices. The biomass and natural gas mixed feed pathways investigated here could potentially offer marine fuel producers a strategy to produce more cost-competitive fuels in the near term using abundant natural gas feedstocks and to dial down CO_2 emissions in the longer term by increasing the share of biomass feed. Meanwhile, the biofuels presented here could serve as drop-in alternatives, avoiding costly modifications to ships for sulfur scrubbing or LNG handling and use. Additional research is needed to better understand the effect of these new marine fuels on fuel handling equipment. This study is a step toward a better understanding of the potential for biofuels to contribute to the maritime transportation sector's sustainability.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c06141.

> Detailed description of conceptual process description, capital cost estimation, GREET modeling of the marine fuel production, and consumption and emissions by fuel and engine types (PDF)

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Notes

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■ ACRONYMS AND ABBREVIATIONS

BTL	biomass-to-liquid
CAP	criteria air pollutant
CBTL	biomass and coal-to-liquid
CO_2e	carbon dioxide equivalent
FT	Fischer−Tropsch
FP	fast pyrolysis
GBTL	biomass and natural gas-to-liquid
GHG	greenhouse gas
GTL	natural gas-to-liquid
GREET	Greenhouse gases, Regulated Emissions, and Energy use in Transportation
HFO	heavy fuel oil
HFOGE	heavy fuel oil gallon equivalent
LCA	life-cycle analysis
LNG	liquefied natural gas
MAC	marginal abatement cost
MDO	marine diesel oil
MFSP	minimum fuel selling price
MGO	marine gas oil
PM	particulate matter
SVO	straight vegetable oil
TEA	technoeconomic analysis
YG	yellow grease

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