sustainability

MDPI

Article

Pollution Tradeoffs for Conventional and Natural Gas-Based Marine Fuels

James J. Winebrake [1,*], James J. Corbett [2], Fatima Umar [1] and Daniel Yuska [3]

[1] Rochester Institute of Technology, Rochester, NY 14623, USA; fu1476@rit.edu
[2] Marine Policy Program, University of Delaware, Newark, DE 19711, USA; jcorbett@udel.edu
[3] U.S. Department of Transportation, Maritime Administration, Washington, DC 20590, USA; daniel.yuska@dot.gov
* Correspondence: jjwgpt@rit.edu

Received: 4 March 2019; Accepted: 6 April 2019; Published: 13 April 2019

Abstract: This paper presents a life-cycle emissions analysis of conventional and natural gas-based marine transportation in the United States. We apply a total fuel cycle—or "well-to-propeller"—analysis that evaluates emissions along the fuel production and delivery pathway, including feedstock extraction, processing, distribution, and use. We compare emissions profiles for methanol, liquefied natural gas, and low sulfur marine fuel in our analysis, with a focus on exploring tradeoffs across the following pollutants: greenhouse gases, particulate matter, sulfur oxides, and nitrogen oxides. For our greenhouse gas analysis, we apply global warming potentials that consider both near-term (20-year) and long-term (100-year) climate forcing impacts. We also conduct uncertainty analysis to evaluate the impacts of methane leakage within the natural gas recovery, processing, and distribution stages of its fuel cycle. Our results indicate that natural-gas based marine fuels can provide significant local environmental benefits compared to distillate fuel; however, these benefits come with a near-term—and possibly long-term—global warming penalty, unless such natural gas-based fuels are derived from renewable feedstock, such as biomass. These results point to the importance of controlling for methane leaks along the natural gas production process and the important role that renewable natural gas can play in the shipping sector. Decision-makers can use these results to inform decisions related to increasing the use of alternative fuels in short sea and coast-wise marine transportation systems.

Keywords: marine shipping; emissions; alternative fuels; methanol; natural gas

1. Introduction

Marine transportation is an important source of regional air pollution in many parts of the world and represents 15% and 13% of the global emissions burden for nitrogen oxides (NO_x) and sulfur oxides (SO_x), respectively [1]. These pollutants pose significant health risks to exposed populations and are responsible for hundreds of thousands of premature deaths and millions of respiratory illnesses worldwide annually [2–4]. Due to human health and other ecological risks, some nations have imposed emissions control areas (ECAs) that restrict emissions from vessels operating within defined coastal boundaries, and the International Maritime Organization (IMO) has adopted new fuel standards aimed at significantly reducing the sulfur content in marine fuels by 2020 [2–6].

The shipping sector is considering several approaches to meet ECA and IMO regulations, including the installation of on-board pollution control equipment and the switch from petroleum-based fuels to cleaner fuels. In this latter category, two fuels that have gained attention due to their fuel properties are liquefied natural gas (LNG) and methanol (MeOH) [7–10]. These fuels are derived from natural gas (primarily methane (CH_4)) and are expected to reduce NO_x, particulate matter (PM), and SO_x at the vessel stack compared to petroleum-based fuels [11].

However, LNG and MeOH have the potential to increase greenhouse gases (GHG). Ocean-going vessels contribute about 2.2% of total GHG emissions globally, mostly in the form of carbon dioxide (CO_2) [1]. The movement towards LNG and MeOH in the marine sector may increase CH_4 emissions (an important GHG), especially during fuel production and distribution [12].

This paper explores pollution tradeoffs among marine distillate oil (MDO), LNG, and MeOH in the shipping sector. Using recent research on LNG and MeOH production, we apply a total fuel cycle analysis (TFCA) methodology to evaluate "well-to-propeller" (W2P) emissions for various marine fuels. We evaluate emissions along the entire fuel pathway, including feedstock extraction and fuel processing, distribution, and use in vessels.

We characterize the following pollutant emissions for these fuels: GHGs (CO_2, CH_4), NO_x, $PM_{2.5}$, and SO_x. We also present a GHG comparison using two different global warming potentials (GWP) representing a 20-year and 100-year GWP (GWP_{20} and GWP_{100}, respectively). The choice of GWP is important in evaluating near-term vs. long-term climate forcing impacts [13]. All results are presented in mass per energy units (e.g., grams per mega-Joule, or g/MJ) so results can be applied across different vessel configurations. We demonstrate our results for a short sea shipping case study along the east coast of the United States (US).

Section 2 presents background literature and motivation for considering natural gas-based fuel as an alternative to current petroleum marine fuels. Section 3 describes the methodology and data used, with particular attention paid to CH_4 leakage along the fuel production supply chain. Section 4 presents the key findings of the analysis. Finally, Section 5 presents overall conclusions and describes areas of further research.

2. Background and Motivation

2.1. Emissions and International Shipping

The International Convention for the Prevention of Pollution from Ships (MARPOL) was adopted by the IMO in 1973 to address the issue of pollution from ships [14]. IMO has amended MARPOL several times in response to new information about the causes, effects, and extent of such pollution. For example, subsequent changes have decreased the allowed emissions from vessel exhaust stacks and have allowed for the creation of ECAs with strict emissions requirements.

Recently, IMO approved new regulations that will impose a global sulfur cap of 5000 parts per million (ppm) sulfur (i.e., 0.5% S) on marine fuels used in ships operating in global waters, and 1000 ppm S for fuels used on vessels operating in designated ECAs [5]. Ships operating in ECAs will have the option to (1) switch to low sulfur marine distillates (<1000 ppm S); (2) continue using high sulfur fuel, but install after-treatment desulfurization technology (such as scrubbers) to remove sulfur from the exhaust stream; or (3) switch to alternative fuels—such as LNG and MeOH—that have little to no sulfur content. The choice that ship operators make will depend on costs, fuel availability, refueling infrastructure, safety, and operational factors, among others [8].

The literature suggests that LNG and MeOH fuel can be effective at meeting these new regulations, as these fuels have been shown to reduce certain local pollutants from vessel operations [8,10–12,15]. However, LNG and MeOH fuel production pathways are relatively energy-intensive compared to petroleum pathways, and fugitive emissions of CH_4 that accompany natural gas extraction and distribution may increase GHG impacts [13,16–24]. Since the IMO has committed to reductions in both local pollution (PM, SO_x, and NO_x) and GHG emissions, decision-makers ought to look at the life-cycle GHG emissions generated by NG-based fuels compared to traditional marine bunkers before committing to one fuel over another.

2.2. Natural Gas-Based Fuels in the Shipping Sector

As of May 2018, there were ~120 vessels operating on LNG internationally (excluding LNG carriers), with another 135 under construction or on order [25]. At the same time, there are only about seven internationally-registered vessels operating on MeOH [26]—but interest is growing, as demonstrated by the IMO Marine Safety Committee's invitation to the International Organization for Standardization (ISO) to consider new standards for MeOH use and refueling processes [27].

Attributes supporting interest in LNG and MeOH as a marine fuel are documented in previous work [8,12]. With respect to LNG, Thomson et al. (2015) showed that environmental and economic advantages make LNG a competitive fuel, and there is extensive experience on the use of LNG in the marine sector. The greatest drawbacks of LNG are its energy-intensive production processes, its relatively low energy density, and its distribution and refueling network, which remains more complicated than conventional oil.

With regards to MeOH, fuel distribution is not as much of a concern compared to LNG, and MeOH can be blended or used directly in marine engine systems with modest modifications [28]. Marine engines designed for MeOH combustion can achieve engine efficiencies similar to diesel combustion, and emissions from such engines are reported to meet or exceed current pollutant emission regulations [29]. Unplanned spills also biodegrade more quickly than traditional petroleum fuels. However, MeOH is an expensive fuel vis-à-vis conventional oil. February 2019 prices in North America for both MeOH and heavy fuel oil are in the range of ~$430 per metric ton (mt), while lighter marine gas oil is ~$650/mt [30,31]. But due to its lower energy content (which is about 50% less than the energy content of petroleum fuels), the cost of MeOH on a per energy basis is about twice as costly as heavy fuel oil and 20–30% higher than MDO. This cost differential may be a major barrier to MeOH commercialization in the shipping sector.

Despite these challenges, the application of LNG and MeOH in shipping operations—and in particular, short sea or coast-wise shipping—remains an interesting opportunity. Because short sea shipping is near-coast, the local emissions benefits associated with these NG-based fuels are readily achieved on land. Short sea shipping also allows for greater refueling opportunities and predictability compared to trans-oceanic shipping, thereby addressing one of the major barriers for NG-based fuels. Interest in short sea shipping has been increasing in the European Union [32], the United States (US) [33], and Southeast Asia [34]; and it shows promise as a growing part of goods movement elsewhere, as well [35,36]. Our goal in this paper is to explore the emissions tradeoffs associated with NG-based fuels vs. conventional fuels, particularly in a short sea shipping context.

3. Methodology

3.1. Total Fuel-Cycle Analysis Modeling

We apply a *total fuel-cycle analysis* (TFCA) methodology in this paper. A TFCA is a type of life-cycle analysis modeling approach that enables calculation of the total emissions profile associated with the use of a given fuel in a marine vessel [24,37,38]. These analyses account for emissions along the entire "fuel cycle", which includes the following stages, also depicted in Figure 1:

- *Feedstock-related stages*—encompassing the extraction of fuel feedstock through the delivery of that feedstock to the refinery or processing facility;
- *Fuel-related stages*—encompassing the processing of feedstock into usable fuel through the delivery of that fuel from to the bunkering location; and,
- *Vessel operation*—encompassing the refueling of the vessel and use of the fuel in the vessel itself.

We refer to the feedstock-related and fuel-related stages as "upstream" stages—or "well-to-tank" processes; and the last stage (vessel refueling and operation) as the "downstream" stage, consistent with the TFCA literature [7,9].

Total fuel-cycle analysis gained prominence in the life-cycle analysis literature in the 1990s, when alternative fuels for personal transportation (e.g., automobiles using natural gas, electricity, propane, ethanol, etc.) received much attention due to their energy independence and environmental attributes [39]. In the US, TFCA became even more critical with the emergence of low-carbon fuel standards in California [40], renewable fuel standards at the national level [41], and the expansion of certain feedstock extraction methods, such as hydraulic fracturing in the natural gas sector [24,42]. The first peer-reviewed application of TFCA to the marine sector was published in 2007 [7]. The literature still remains limited in this regard, with only a handful of papers published since that time.

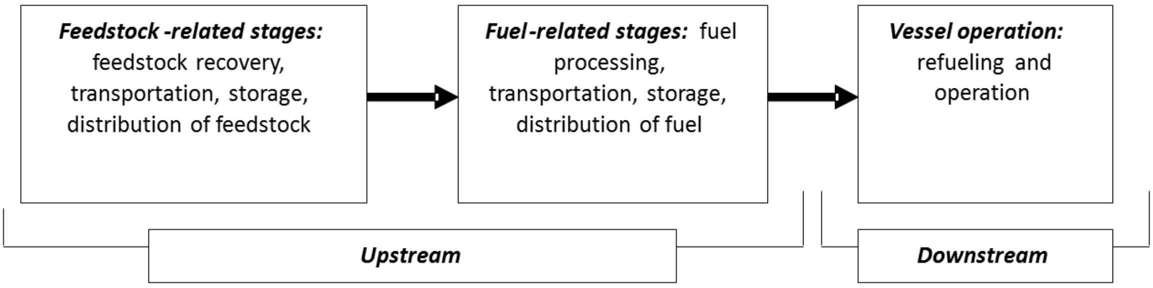

Figure 1. Components of a total fuel cycle analysis showing upstream ("well-to-tank") and downstream ("tank-to-propeller") activities [7].

We employ two well-regarded, peer-reviewed models to conduct our TFCA. For our upstream analysis we employ the Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET) model developed by Argonne National Laboratory (ANL), Version 2018 [43]. GREET allows researchers to evaluate fuel production emissions for a wide variety of fuels obtained using over 100 different pathways and has been used extensively in previous work to evaluate alternative fuels for transportation technologies [7,37,39,44–49]. For downstream emissions analysis we use the Total Energy and Environmental Analysis for Marine System (TEAMS) model developed by a subset of these authors with support from the US Maritime Administration. TEAMS links a vessel operation model with GREET to estimate emissions along simulated vessel voyages and using specific vessel characteristics. More information about TEAMS is available in Supplementary Materials and the peer-reviewed literature [7,9]. The advantage of TEAMS is that it marries the upstream modeling capacity of GREET with a flexible and integrated downstream emissions model for marine vessels.

3.2. Production Pathways and Assumptions

3.2.1. Fuel Pathways

Natural gas-based marine fuels can be produced in many different ways. The production pathways we evaluate in this paper are listed below, with details about the inputs for each pathway provided in Tables 1 and 2. Each pathway is constructed using assumptions consistent with fuel production in the US and vessel operations and bunkering at a US port.

- *Pathway I: MeOH from US Natural Gas.* This pathway represents a traditional pathway for the production of MeOH in the US. The feedstock is natural gas from US wells (USNG). Because we are not modeling production from a *particular* natural gas field, we use an average feedstock mix of ~50% from shale production and ~50% from conventional recovery practices (shale production represented ~62% of total US production in 2017 [50]). We assume that gas is transported to MeOH production facilities via pipeline and that the resulting MeOH is transported to bulk terminals using a mix of pipeline, rail, and transport options. We assume transport from bulk terminals to refueling sites occur via a diesel tanker truck.
- *Pathway II: MeOH from Non-North American Natural Gas.* This pathway addresses MeOH produced outside of North America from non-North American Natural Gas (NNANG). The pathway

assumes that natural gas travels a short distance to the MeOH production facility via pipeline and then the MeOH is shipped via ocean tanker to a US port. The MeOH is then moved to bulk terminals using barge, rail, and truck. The product is then moved to refueling facilities via diesel tanker truck.

- *Pathway III: MeOH from Biomass.* This pathway relies on forest residue as a MeOH feedstock and demonstrates the potential to gasify biomass and convert to MeOH. We assume biomass is transported to the MeOH production facility via diesel truck, and that MeOH is produced using gasification processes as outlined in GREET 2018. The MeOH is then moved to bulk terminals using barge, rail, and truck; and then to refueling sites via tanker truck.
- *Pathway IV: LNG from US Natural Gas.* This pathway represents production of LNG from an average blend of US natural gas sources. The natural gas feedstock travels via pipeline to a liquefaction plant, and the resulting LNG is delivered to bulk terminals via barge and rail. Final distribution to the refueling location is conducted by truck.
- *Pathway V: Low Sulfur (US ECA-Compliant) Marine Distillate.* This pathway models the production of low sulfur marine distillate fuel (1000 ppm, or 0.1% S) that complies with US ECA regulations. We assume the feedstock crude comes from an average blend representing the lower 48 states of the US. That crude is shipped via pipeline and barge to a refinery, and the distillate is moved from the refinery to bulk terminals via barge, pipeline, and rail. Final delivery to the refueling location is via truck.

Table 1. Overview of production and distribution pathways with key variables identified.

Fuel Pathway	Feedstock	Transportation of Feedstock to Production Facility (Miles)	Transportation to Bulk Terminal	Transportation to Refueling Facility (Miles)
I: MeOH from USNG	U.S. Natural Gas	Pipeline (100 mi)	To Terminal: Mix (Barge—10%/520 mi; Pipeline—20%/550 mi; Rail—20%/650 mi; Truck—50%/80 mi.)	Truck (100%/30 mi)
II: MeOH from NNANG	Non-North American Conventional Recovery	Pipeline (10 mi)	To US: Ocean Tanker—100%/3000 mi To Terminal: Mix (Barge—40%/520 mi; Rail—20%/700 mi; Truck—40%/80 mi)	Truck (100%/30 mi)
III: MeOH from Biomass	Biomass (Forest Residue)	Truck (100%/90 mi)	Mix (Barge—40%/520 mi; Rail—20%/700 mi; Truck—40%/80 mi.)	Truck (100%/30 mi)
IV: LNG from USNG	U.S. Natural Gas	Pipeline (50 mi)	To Bulk Terminal: Mix (Barge—50%/520 mi; Rail—50%/800 mi)	Truck (100%/30 mi)
V: US ECA-Compliant Distillate Oil (MDO)	Crude Oil (Average from US Conventional Sources)	Mix (Pipeline—76%/420 mi; Barge—24%/750 mi)	To Bulk Terminal: Mix (Barge—49%/200 mi; Pipeline—46%/110 mi; Rail—5%/490)	Truck (100%/30 mi)

Table 2. Feedstock recovery, processing, and production efficiencies (MJ/MJ).

Feedstock and Fuel Production Process	Process Efficiency
Natural Gas Recovery from US Wells	97.5%
Natural Gas Processing for Use as Feedstock	97.4%
Natural Gas to LNG Production	91.0%
Natural Gas to Methanol Production	70.0%
Biomass to Methanol Production	58.0%
Crude Oil Recovery from US Wells	98.0%
Crude Oil to Marine Distillate Oil Refining	90.9%

3.2.2. Natural Gas Recovery and Leakage Factors

An important assumption for natural gas pathways is the fugitive CH_4 emissions that occur during feedstock extraction, fuel processing, and fuel distribution. Recent literature demonstrates the importance of these emissions with respect to climate change, since CH_4 has significantly higher climate forcing potential than the more ubiquitous CO_2. For example, a kilogram of CH_4 released to the atmosphere has a global warming potential (GWP) that is 28 and 84 times greater than a kilogram of CO_2 over a 100-year and 20-year period, respectively [51]. Due to increasing concerns over near-term climate change, researchers have concentrated efforts to obtain accurate estimates of CH_4 releases during natural gas recovery and processing. New studies have shown that existing emissions factors used by the US government in its inventory calculations under-estimate actual emissions [19].

Methane emissions that occur during natural gas recovery and distribution processes can be of three types: unintended leaks (e.g., fugitive emissions from leaky equipment or pipelines); intended releases (e.g., vented emissions by design in equipment, such as pneumatic devices, vents, or chemical injection pumps); or combustion-related (e.g., from the burning of fossil fuels during the extraction and production process) [17,21]. There is no "single" emission factor to apply to CH_4 along these processes. Instead, what has emerged in the literature is a collection of emissions factors informed by ongoing research, testing, and demonstration projects.

Alvarez et al. [19] and Littlefield et al. [52] synthesized a set of source-specific and site-specific analyses to derive emissions factors for certain parts of the natural gas supply chain. Littlefield et al. [52] synthesized component-based data related to well completion, pumps, and equipment leaks [17], pneumatic controllers [53], liquids unloading [54], general production [55], gathering and processing [56], transmission and storage [20], and local distribution systems [57]. Alvarez et al. (2018) provide the most comprehensive assessment to date of CH_4 emissions from the natural gas supply chain, demonstrating that site-based analyses show CH_4 emissions levels that are 1.2–2 times higher than the EPA's estimates. We conduct our analysis using both EPA estimates and EDF estimates to create a range of results. The EPA and EDF emissions factors are shown in Table 3 for conventional gas and shale gas recovery.

Table 3. Emissions factors for methane leakage and venting ($mgCH_4/MJ$) related to natural gas feedstock recovery, processing, and distribution showing values attributed to the US Environmental Protection Agency (EPA) and values attributed to the Environmental Defense Fund (EDF) [58].

Upstream Stage of Natural Gas Fuel Production	EPA ($mgCH_4/MJ$)		EDF ($mgCH_4/MJ$)	
	Conventional	Shale	Conventional	Shale
Recovery [a]	129.9	133.3	203.1	203.1
Processing	5.6	5.6	9.0	9.0
Transmission and Storage [b]	37.7	37.7	52.3	52.3
Distribution	18.4	18.4	18.4	18.4

[a] Recovery includes CH_4 leakage and venting related to well completion, workover, liquid unloading, and well equipment venting and leakage. [b] Units for transmission and storage are per 1000 km of transmission as calculated by the authors.

3.2.3. Fuel Properties

An important factor associated with emissions at the vessel is the fuel characteristics, particularly energy content of the fuel, its density, its carbon ratio (affecting CO_2 emissions), and its sulfur content (affecting SO_x emissions). For the fuels studied for this paper, we assume a set of fuel properties shown in Table 4.

Table 4. Fuel properties for fuels evaluated in this study, showing low heating value (LHV), fuel density, carbon ratio by weight, and sulfur ratio by weight.

Fuel	Low Heating Value (MJ/Gallon)	Density (Grams/Gallon)	Carbon Ratio (% by Weight)	Sulfur Ratio (ppm_w and %)
Methanol (MeOH)	60.45	3000	37.5%	0
Liquefied Natural Gas (LNG)	78.89	1620	75.0%	0
Marine Distillate Oil (MDO)	135.62	3167	86.5%	1000 ppm (0.1%)

4. Results

The upstream "well-to-use" (WTU) results from our emissions analysis for each pathway are shown in Table 5. Separate results are shown using EPA and EDF assumptions (which we denote as "low" and "high", respectively) regarding CH_4 leakage in the natural gas-based fuel production pathway. Results show that the NG-based fuels generate a high level of GHGs on a per energy delivered basis. This is due to both high CO_2 emissions (in the case of MeOH) and high CH_4 emissions. The CH_4 emissions are also shown to drive GHG emissions factors. When using a GWP_{100}, CH_4 emissions make up about one-third of the GHG emissions factors for pathways I, II, and V—and more than half for pathway IV; but when using a GWP_{20}, the CH_4 emissions make up a majority of the GHG_{20} emissions factors for pathways I, II, and IV—and almost half for pathway V. It is only in pathway III (MeOH from biomass) that we see GHG factors little affected by CH_4 emissions, due to the fact that there is essentially no CH_4 "leakage" in the collection and delivery of biomass feedstock.

In terms of upstream local pollution (NO_x, $PM_{2.5}$, and SO_x), results vary. For example, MeOH from non-North American Natural Gas (pathway II) generates higher emissions of these pollutants—but much of that is due to the delivery of MeOH via ocean tanker and is spread over a large geographic area. The greater concerns with regard to these local pollutants—especially in a short sea context—occur in the downstream stages of the fuel cycle, discussed below.

Table 5. Well-to-use (WTU) results for each production pathway showing emissions per MJ fuel throughput using EDF (high) and EPA (low) methane leakage factors for natural gas production and distribution; and showing GHG values using a GWP_{20} (GHG_{20}) and GWP_{100} (GHG_{100}).

Pathway		CO_2 (g/MJ)	CH_4 (g/MJ)	GHG_{20} (g/MJ)	GHG_{100} (g/MJ)	NOx (mg/MJ)	$PM_{2.5}$ (mg/MJ)	SOx (mg/MJ)
I: MeOH from USNG	High	21.9	0.347	51.02	31.60	34.2	1.9	24.1
	Low		0.231	41.29	28.35			
	Mid		0.289	46.16	29.98			
II: MeOH from NNANG	High	23.1	0.342	51.79	32.63	66.4	4.3	43.4
	Low		0.225	41.98	29.36			
	Mid		0.284	46.88	31.00			
III: MeOH from Biomass	High	4.3	0.006	4.80	4.45	21.6	1.0	0.7
	Low		0.006	4.77	4.44			
	Mid		0.006	4.79	4.45			
IV: LNG from USNG	High	11.1	0.380	43.06	21.76	27.6	0.9	12.7
	Low		0.291	35.55	19.25			
	Mid		0.336	39.31	20.50			
V: US ECA-Compliant MDO	High	10.5	0.109	19.59	13.50	19.4	0.9	8.3
	Low		0.100	18.90	13.27			
	Mid		0.105	19.24	13.38			

Downstream emissions factors for methanol-fueled, natural gas-fueled, and petroleum-fueled marine engines were compiled from various literature sources [8,11,12,29,59,60]. For gas engines, the type of engine used by the shipowner can have an impact on vessel emissions. We consider both lean-burn/spark-ignited (LBSI) and low-pressure/dual-fuel (LPDF) engines, since these dominate current markets [11]. Both of these types of engines are evaluated in the downstream analysis by indicating two sub-pathways for Pathway IV shown in the results below.

Table 6 presents our assumed vessel emissions factors by type of marine engine. Combustion conditions and fuel properties matter to these estimates. For example, spark-ignited (Otto cycle) natural gas engines typically have higher methane emissions (methane slip) than auto-ignited (Diesel cycle) engines. Table 6 associates lower NO_x and higher $PM_{2.5}$ with spark-ignited engines, and different sulfur emissions according to the fuel sulfur contents; in the absence of published test data for some criteria pollutants, this work assigns similar values across engine types while acknowledging these may, in fact, differ. We also assume for the natural gas-based fuels a 3% diesel fuel injection on average with a sulfur content of 1000 ppm.

Combining the upstream (well-to-tank) emissions for each pathway with downstream (tank-to-propeller) emissions factors provides us with the results shown in Table 7. These results are also depicted in Figures 2 and 3.

Table 6. Summary of vessel combustion emission factors for fuels on a per MJ fuel throughput.

Fuel/Engine	Natural Gas/LPDF	Natural Gas/LBSI	Marine Distillate Oil	Methanol/LBSI
CO_2 (g/MJ)	55.9	55.5	73.2	65.8
CH_4 (g/MJ)	0.087	0.625	0.005	0.002 *
GHG_{20} (g/MJ)	63.76	108.47	74.1	65.8
GHG_{100} (g/MJ)	58.9	73.5	73.8	65.8
NOx (mg/MJ)	2400	230	2400	300
$PM_{2.5}$ (mg/MJ)	0.7	4.7	69.2	4.3
SO_x (mg/MJ)	0.28	0.28	9.48	0.28

* The literature provides insufficient data regarding CH_4 emissions from MeOH marine engines. We assume these emissions to be equivalent to CH_4 emissions for MeOH spark ignition engines found in GREET for methanol fueled spark ignition engines.

Table 7. Summary of results showing upstream and downstream emissions for each pollutant studied, as well as low and high values for CH_4 and GHG emissions based on EPA (low) and EDF (high) emissions factors.

Pathway/Fuel	Upstream (Low, High)	Downstream	Total
	CO_2 (g/MJ)		
I—MeOH	21.89	65.83	87.72
II—MeOH	23.05	65.83	88.88
III—MeOH	4.28	65.83	70.11
IV—LNG/LPDF	11.10	55.93	67.04
IV—LNG/LBSI	11.10	55.49	66.59
V—MDO	10.46	73.20	83.66
	CH_4 (g/MJ)		
I—MeOH	0.289 (0.231, 0.347)	0.002	0.291
II—MeOH	0.284 (0.225, 0.342)	0.002	0.286
III—MeOH	0.006 (0.006, 0.006)	0.002	0.008
IV—LNG/LPDF	0.336 (0.291, 0.380)	0.087	0.423
IV—LNG/LBSI	0.336 (0.291, 0.380)	0.625	0.960
V—MDO	0.105 (0.100, 0.109)	0.005	0.109
	GHG-20 (g/MJ)		
I—MeOH	46.16 (41.29, 51.02)	66.00	112.16
II—MeOH	46.88 (41.98, 51.79)	66.00	112.89
III—MeOH	4.79 (4.77, 4.80)	66.00	70.79
IV—LNG/LPDF	39.31 (35.55, 43.06)	63.26	102.56
IV—LNG/LBSI	39.31 (35.55, 43.06)	107.97	147.27
V—MDO	19.24 (18.90, 19.59)	73.60	92.84
	GHG-100 (g/MJ)		
I—MeOH	29.98 (28.35, 31.60)	65.89	95.87
II—MeOH	31.00 (29.36, 32.63)	65.89	96.89
III—MeOH	4.45 (4.44, 4.45)	65.89	70.34
IV—LNG/LPDF	20.50 (19.25, 21.76)	58.37	78.88
IV—LNG/LBSI	20.50 (19.25, 21.76)	72.98	93.49
V—MDO	13.38 (3.27, 13.50)	73.34	86.72

Table 7. *Cont.*

Pathway/Fuel	Upstream (Low, High)	Downstream	Total
	NO_x (mg/MJ)		
I—MeOH	34.24	342.55	376.79
II—MeOH	66.35	342.55	408.90
III—MeOH	21.64	342.54	364.18
IV—LNG/LPDF	27.62	2351.99	2379.61
IV—LNG/LBSI	27.62	224.68	252.30
V—MDO	19.44	2351.04	2370.48
	$PM_{2.5}$ (mg/MJ)		
I—MeOH	1.89	4.30	6.19
II—MeOH	4.34	4.30	8.64
III—MeOH	0.96	4.30	5.26
IV—LNG/LPDF	0.86	0.66	1.52
IV—LNG/LBSI	0.86	4.74	5.60
V—MDO	0.92	69.20	70.13
	SO_x (mg/MJ)		
I—MeOH	24.13	0.28	24.41
II—MeOH	43.41	0.28	43.70
III—MeOH	0.71	0.28	0.99
IV—LNG/LPDF	12.74	0.28	13.03
IV—LNG/LBSI	12.74	0.28	13.03
V—MDO	8.28	9.48	17.76

We apply the results above to a short sea case study previously published in the peer-reviewed literature [8]. The case study involves shipping along the east coast of the US, from the Port of Jacksonville, Florida to the Port of New York/New Jersey. The details of the vessel and route attributes are shown in Tables 8 and 9. We evaluate this case using the emissions factors from above for MDO, LNG, and MeOH along pathways I–IV.

Our results are shown in Table 10. These results clearly show some of the tradeoffs that are apparent when trying to balance both local and global emissions reductions. Although the NG-based fuels perform relatively well with respect to local pollutants (NO_x, $PM_{2.5}$, and SO_x) compared to MDO, the same cannot be said for their climate change impacts. Values for near-term (GHG_{20}) and long-term (GHG_{100}) emissions demonstrate the potentially negative impacts on climate change, especially in the near term. These types of tradeoffs must be carefully considered by decision makers looking to promote shifts from conventional to alternative fuels in the shipping sector.

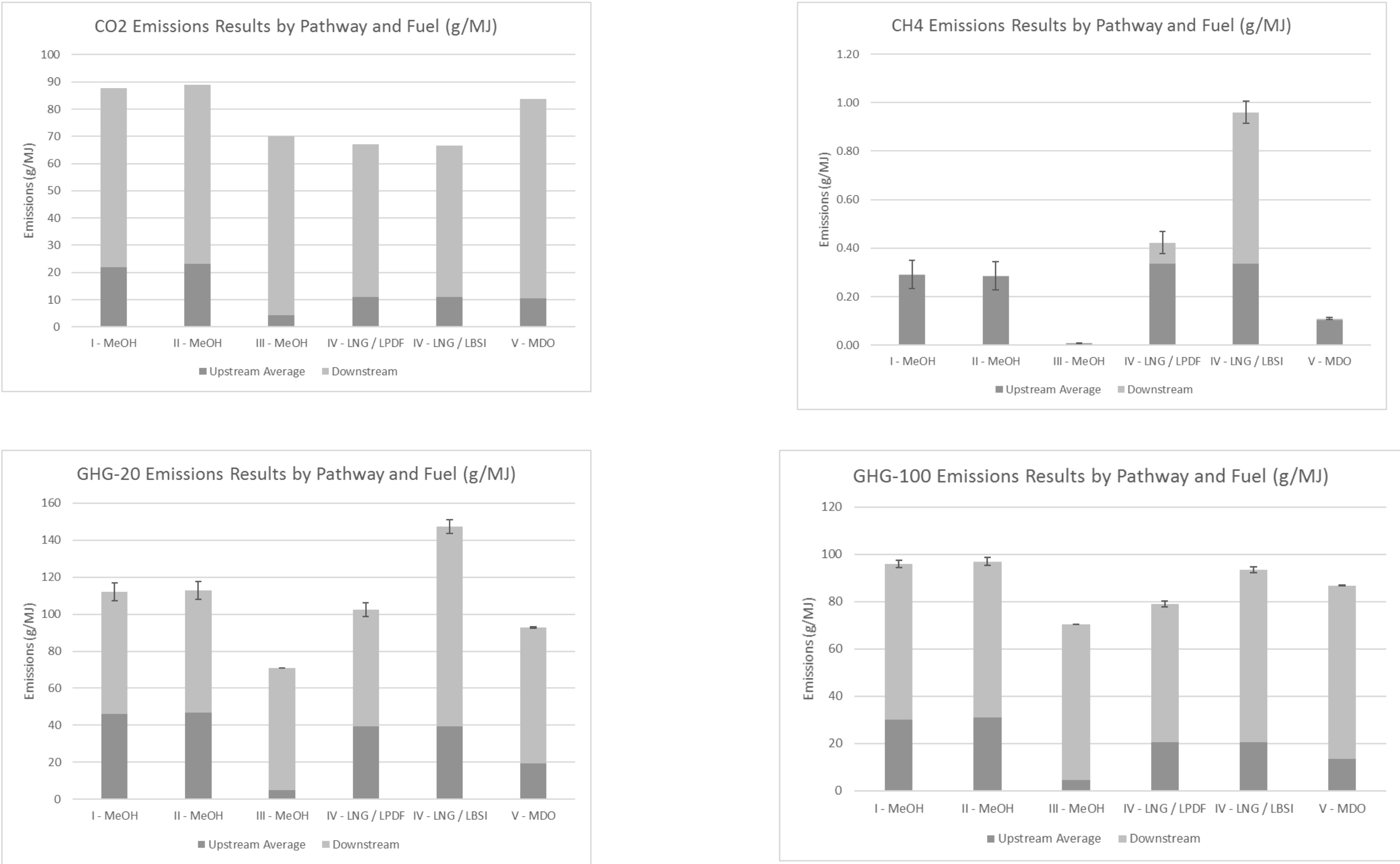

Figure 2. *Cont.*

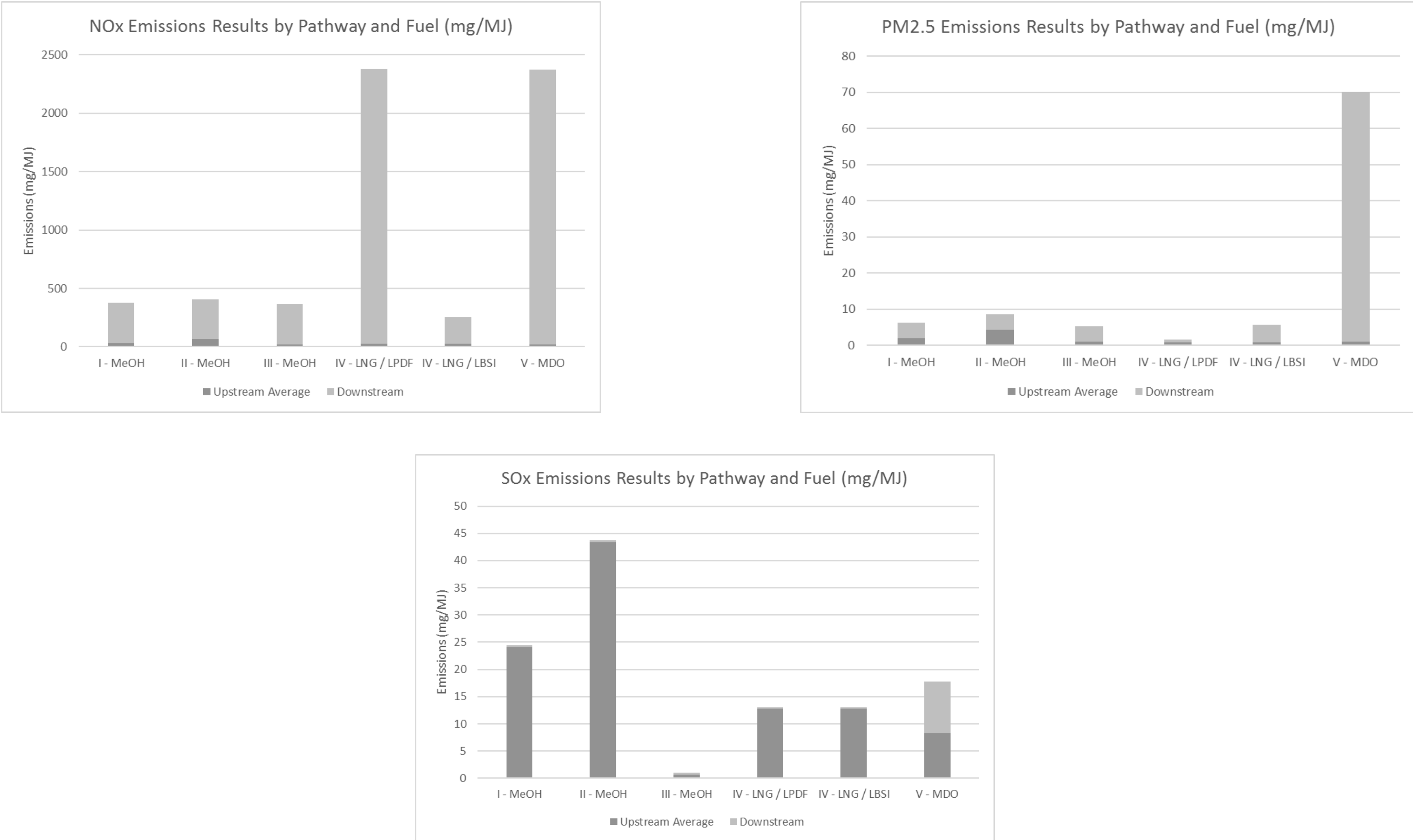

Figure 2. Summary of well-to-propeller results with error bars showing emissions ranges due to different assumption of methane leakage in natural gas production and distribution systems.

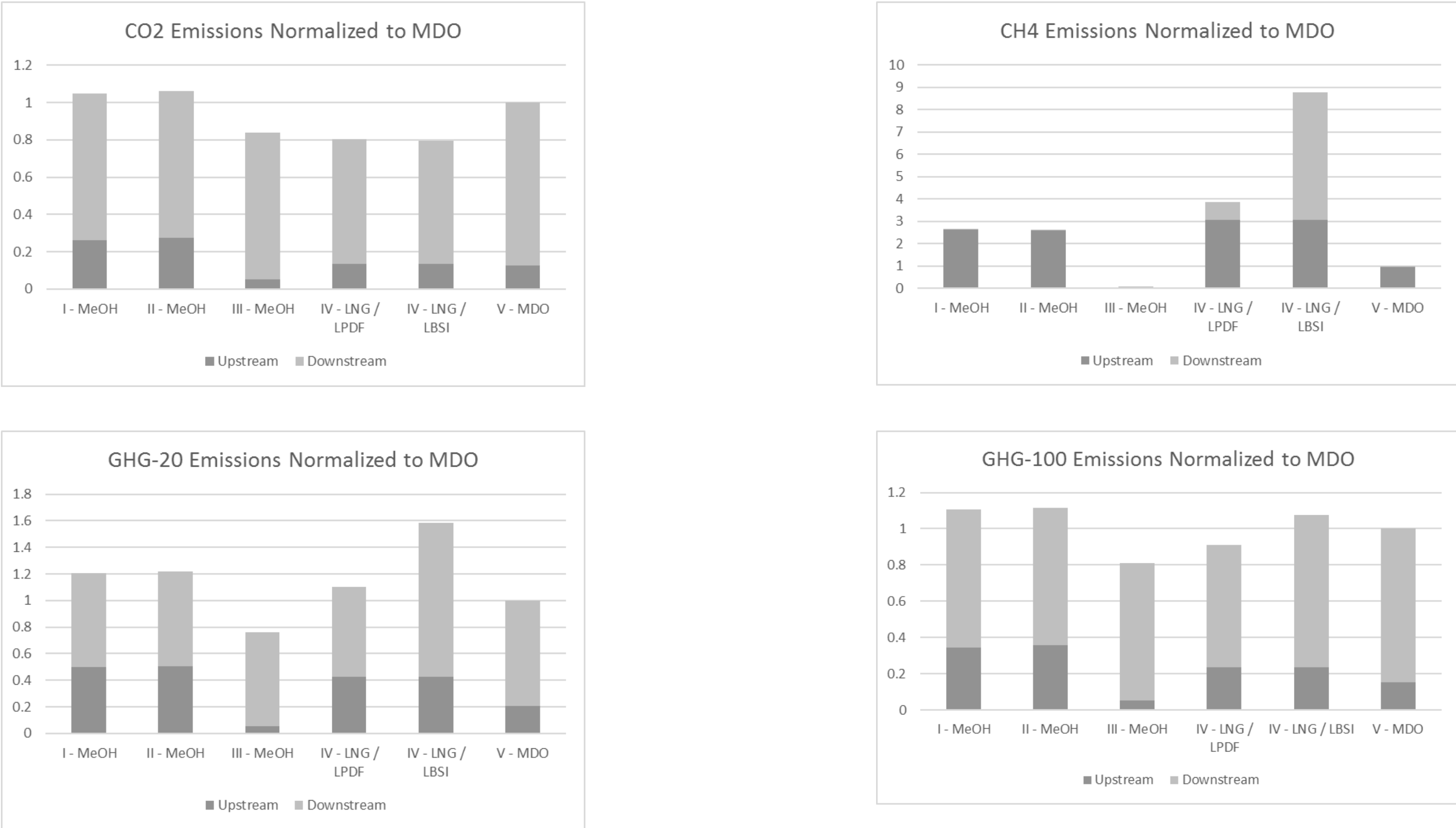

Figure 3. *Cont.*

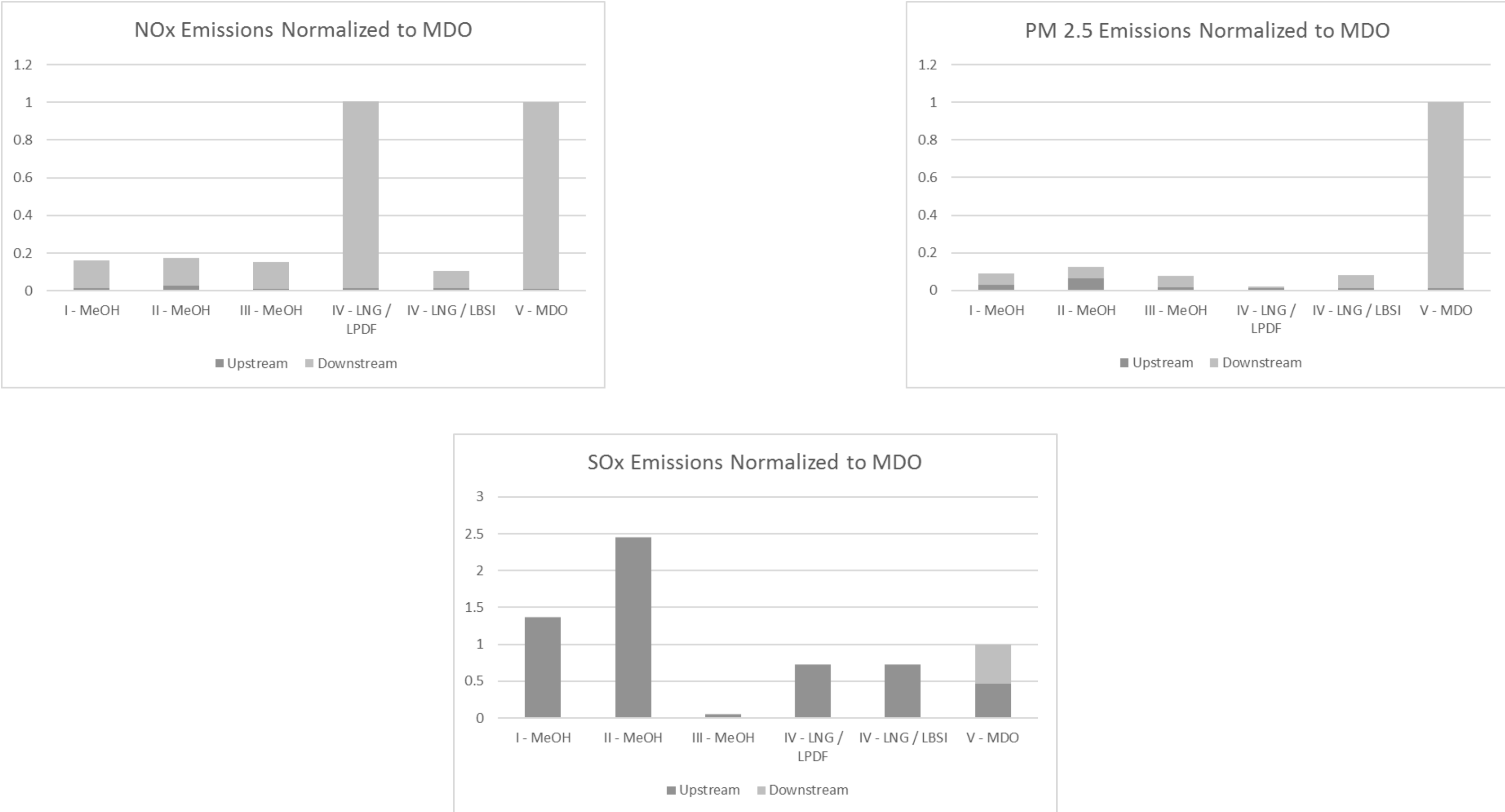

Figure 3. Summary of well-to-propeller results normalized to the MDO pathway.

Table 8. Vessel attributes of US east coast short sea (coast-wise) shipping case study [8].

Vessel Characteristic	Value
Vessel Type	Container
Average DWT	37,300
Rated Power (kW)	22,000
Distance (miles)	828
Rated Speed (knots)	22
Time for one-way trip (hours)	40
Engine Efficiency (%)	45

Table 9. Time spent in each operating stage as a percentage of total trip time (%) for the US east coast short sea (coast-wise) shipping case study [8].

Mode Type	Load (%)	Time in Mode (%)
Idle	2%	1.25
Maneuvering	8%	1.75
Precautionary	12%	5.00
Slow Cruise	50%	7.00
Full Cruise	95%	85.00

Table 10. Results of emissions from US east coast case study.

Emissions (kg/Trip)/Pathway		I MeOH	II MeOH	III MeOH	IV LNG/LPDF	IV LNG/LBSI	V MDO
CO_2 (000)	Upstream	94.1	99.1	18.4	47.7	47.7	44.9
	Downstream	282.9	282.9	282.9	240.4	238.5	315.3
	Total	377.0	382.0	301.3	288.1	286.2	360.2
CH_4 (000)	Upstream	1.2	1.2	0.0	1.4	1.4	0.4
	Downstream	0.0	0.0	0.0	0.4	2.7	0.0
	Total	1.3	1.2	0.0	1.8	4.1	0.5
GHG_{20} (000)	Upstream	198.4	201.5	20.6	168.9	168.9	82.7
	Downstream	283.7	283.7	283.7	271.9	464.0	317.0
	Total	482.0	485.2	304.2	440.8	633.0	399.7
GHG_{100} (000)	Upstream	128.8	133.2	19.1	88.1	88.1	57.5
	Downstream	283.2	283.2	283.2	250.9	313.7	315.9
	Total	412.0	416.4	302.3	339.0	401.8	373.4
NO_x (000)	Upstream	0.1	0.3	0.1	0.1	0.1	0.1
	Downstream	1.5	1.5	1.5	10.1	1.0	10.1
	Total	1.6	1.8	1.6	10.2	1.1	10.2
$PM_{2.5}$	Upstream	8.1	18.6	4.1	3.7	3.7	4.0
	Downstream	18.5	18.5	18.5	2.9	20.4	283.2
	Total	26.6	37.1	22.6	6.5	24.1	287.1
SO_x	Upstream	103.7	186.6	3.0	54.8	54.8	35.6
	Downstream	1.2	1.2	1.2	1.2	1.2	40.8
	Total	104.9	187.8	4.3	56.0	56.0	76.4

5. Discussion and Conclusions

In this paper, we conduct a TFCA for NG-based fuels for the marine sector and compare results to low-sulfur MDO. A novel contribution of the paper is the application of a range of CH_4 emissions factors for the upstream stages of the fuel cycle. These emissions factors are derived from new research indicating that fugitive CH_4 emissions in natural gas extraction, processing, and distribution activities are much higher than previously thought. Combined with new emphasis on near-term climate forcing impacts (evaluated using a GWP_{20} climate forcing factor), we show these upstream CH_4 emissions to have considerable impact on the overall climate change impacts of switching from MDO to NG-based fuels, except in cases where the natural gas is derived from renewable sources, such as biomass.

In particular, MeOH derived from natural gas from conventional wells and shale formations have a near-term and long-term GHG footprint that is ~20% and ~10% higher, respectively, than MDO. Methanol performs well when derived from renewable biomass, exhibiting ~20–25% reductions in GHGs compared to MDO. Methanol also performs extremely well vis-à-vis MDO with respect to $PM_{2.5}$ and NO_x. With regards to SO_x, MeOH demonstrates emissions comparable to MDO in the domestic case (from US natural gas) and much higher in the international transport case (from non-North American natural gas); these emissions are almost entirely due to transportation aspects of feedstock and fuel using petroleum-based fuels that are relatively high in sulfur content. If a low-sulfur fuel is used for such transport, these upstream SO_x emissions will decrease.

Liquefied natural gas performs worse than MDO on a near-term GHG basis, and significantly worse when this LNG is used in a spark-ignited engine (due to higher levels of CH_4 emissions in such systems). On a longer term GHG basis, the performance of LNG is slightly better (with dual-fuel, compression ignition engines) and slightly worse (with spark ignition engines) when compared to MDO. Similar to MeOH, LNG fares well compared to MDO with respect to $PM_{2.5}$ and SO_x, and equal or better with respect to NO_x (depending on type of LNG engine).

These results are demonstrated in absolute terms in the East Coast short sea shipping case study. That case study shows excellent performance for MeOH and LNG on local pollutants (NO_x, $PM_{2.5}$, and SO_x) compared to MDO—especially when considering only downstream emissions. Specifically, emissions reductions of up to 90% can be achieved depending on fuel choice. However, in terms of GHG emissions, the alternative fuels do not perform as well, particularly when considering near-term climate effects. For example, one trip from Jacksonville, FL to the Port of NY/NJ leads to ~480 metric tonnes of GHG_{20} emissions, while the same trip using MDO emits about 20% less, or ~400 metric tonnes of GHG_{20}.

These results present challenges to decision makers aiming to balance improvements in local and regional air quality and climate change. The results imply that decision makers need to consider additional policies that would complement incentives for natural gas-based fuel use in the marine sector. Such policies might include, for example, tighter standards on fugitive emissions control in natural gas production system, or more energy efficiency vessel designs that reduce overall energy consumption onboard ships.

Supplementary Materials: The following are available online at http://www.mdpi.com/2071-1050/11/8/2235/s1.

Author Contributions: Conceptualization: J.J.W., J.J.C., and D.Y.; methodology: J.J.W. and J.J.C.; formal analysis: J.J.W., J.J.C., and F.U.; writing—original draft, J.J.W., J.J.C., and D.Y.; writing—review and editing: J.J.W., J.J.C., F.U., and D.Y.

Funding: This research was partly funded by the U.S. Department of Transportation Maritime Administration grant number DTMA91H150002, and the APC was funded by the Rochester Institute of Technology.

Acknowledgments: The authors thank the US Maritime Administration Maritime Environmental Technical Assistance (META) Program for its financial support and the helpful comments of several anonymous reviewers.

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

Glossary of Terms

ANL	Argonne National Laboratory
CH_4	Methane
CO_2	Carbon dioxide
ECA	Emissions Control Area
EDF	Environmental Defense Fund
EPA	U.S. Environmental Protection Agency
GHG	Greenhouse gas
GHG_{20}	Greenhouse gas based on 20-year global warming potential
GHG_{100}	Greenhouse gas based on 100-year global warming potential
GREET	Greenhouse gases, Regulated Emissions, and Energy Use in Transportation Model
GWP	Global warming potential

GWP_{20}	Global warming potential over a 20-year time horizon
GWP_{100}	Global warming potential over a 100-year time horizon
IMO	International Maritime Organization
LBSI	Lean-burn/spark-ignited
LHV	Lower heating value
LNG	Liquefied natural gas
LPDF	Low-pressure/dual-fuel
MARPOL	International Convention for the Prevention of Pollution from Ships
MDO	Marine distillate oil
MeOH	Methanol
MJ	Mega-Joule
NNANG	Non-North American natural gas
NO_x	Nitrogen oxides
$PM_{2.5}$	Particulate matter with aerodynamic diameters less than 2.5 micrometers
SO_x	Sulfur oxides
TEAMS	Total Energy and Environmental Analysis of Marine Systems
TFCA	Total fuel cycle analysis
USNG	United States natural gas
WTU	Well-to-Use

References

1. Smith, T.W.P.; Jalkanen, J.P.; Anderson, B.A.; Corbett, J.J.; Faber, J.; Hanayama, S.; O'Keeffe, E.; Parker, S.; Johansson, L.; Aldous, L.; et al. *Third IMO Greenhouse Gas Study 2014*; International Maritime Organization: London, UK, 2015.
2. Corbett, J.J.; Winebrake, J.J.; Green, E.H.; Kasibhatla, P.; Eyring, V.; Lauer, A. Mortality from ship emissions: A global assessment. *Environ. Sci. Technol.* **2007**, *41*, 8512–8518. [CrossRef] [PubMed]
3. Sofiev, M.; Winebrake, J.J.; Johansson, L.; Carr, E.W.; Prank, M.; Soares, J.; Vira, J.; Kouznetsov, R.; Jalkanen, J.P.; Corbett, J.J. Cleaner fuels for ships provide public health benefits with climate tradeoffs. *Nat. Commun.* **2018**, *9*, 406.
4. Winebrake, J.J.; Corbett, J.J.; Green, E.H.; Lauer, A.; Eyring, V. Mitigating the health impacts of pollution from oceangoing shipping: An assessment of low-sulfur fuel mandates. *Environ. Sci. Technol.* **2009**, *43*, 4776–4782. [CrossRef] [PubMed]
5. International Maritime Organization. In Proceedings of the International Maritime Organization Marine Environment Protection Committee, Meeting Summary 2018, 72nd Session, Brussels, Belgium, 9–13 April 2018.
6. Corbett, J.J.; Winebrake, J.J. Environmental issues in international trade and transportation. In *Handbook of International Trade and Transportation*; Edward Elgar Publishing Limited: Cheltenham, UK, 2018; pp. 275–296.
7. Winebrake, J.J.; Corbett, J.J.; Meyer, P.E. Energy Use and Emissions from Marine Vessels: A Total Fuel Life Cycle Approach. *J. Air Waste Manag. Assoc.* **2007**, *57*, 102–110. [CrossRef] [PubMed]
8. Thomson, H.; Corbett, J.J.; Winebrake, J.J. Natural gas as a marine fuel. *Energy Policy* **2015**, *87*. [CrossRef]
9. Corbett, J.J.; Winebrake, J.J. Emissions tradeoffs among alternative marine fuels: Total fuel cycle analysis of residual oil, marine gas oil, and marine diesel oil. *J. Air Waste Manag. Assoc.* **2008**, *58*, 538–542. [CrossRef]
10. Deniz, C.; Zincir, B. Environmental and economical assessment of alternative marine fuels. *J. Clean. Prod.* **2016**, *113*, 438–449. [CrossRef]
11. Ushakov, S.; Stenersen, D.; Einang, P.M. Methane slip from gas fuelled ships: A comprehensive summary based on measurement data. *J. Mar. Sci. Technol.* **2019**, 1–18. [CrossRef]
12. Brynolf, S.; Fridell, E.; Andersson, K. Environmental assessment of marine fuels: Liquefied natural gas, liquefied biogas, methanol and bio-methanol. *J. Clean. Prod.* **2014**, *74*, 86–95. [CrossRef]
13. Alvarez, R.A.; Pacala, S.W.; Winebrake, J.J.; Chameides, W.L.; Hamburg, S.P. Greater focus needed on methane leakage from natural gas infrastructure. *Proc. Natl. Acad. Sci. USA* **2012**, *109*, 6435–6440. [CrossRef]
14. International Maritime Organization. *MARPOL 1973—Final Act and Convention*; International Maritime Organization: London, UK, 1973.

15. Burel, F.; Taccani, R.; Zuliani, N. Improving sustainability of maritime transport through utilization of Liquefied Natural Gas (LNG) for propulsion. *Energy* **2013**, *57*, 412–420. [CrossRef]
16. Zavala-Araiza, D.; Alvarez, R.A.; Lyon, D.R.; Allen, D.T.; Marchese, A.J.; Zimmerle, D.J.; Hamburg, S.P. Super-emitters in natural gas infrastructure are caused by abnormal process conditions. *Nat. Commun.* **2017**, *8*, 14012. [CrossRef] [PubMed]
17. Allen, D.T.; Torres, V.M.; Thomas, J.; Sullivan, D.W.; Harrison, M.; Hendler, A.; Herndon, S.C.; Kolb, C.E.; Fraser, M.P.; Hill, A.D.; et al. Measurements of methane emissions at natural gas production sites in the United States. *Proc. Natl. Acad. Sci. USA* **2013**, *110*, 17768–17773. [CrossRef] [PubMed]
18. Omara, M.; Sullivan, M.R.; Li, X.; Subramian, R.; Robinson, A.L.; Presto, A.A. Methane Emissions from Conventional and Unconventional Natural Gas Production Sites in the Marcellus Shale Basin. *Environ. Sci. Technol.* **2016**, *50*, 2099–2107. [CrossRef] [PubMed]
19. Alvarez, R.A.; Zavala-araiza, D.; Lyon, D.R.; Allen, D.T.; Barkley, Z.R.; Adam, R.; Davis, K.J.; Herndon, S.C.; Jacob, D.J.; Karion, A.; et al. Assessment of methane emissions from the US oil and gas supply chain. *Science* **2018**, *361*, 186–188. [PubMed]
20. Zimmerle, D.J.; Williams, L.L.; Vaughn, T.L.; Quinn, C.; Subramanian, R.; Duggan, G.P.; Willson, B.; Opsomer, J.D.; Marchese, A.J.; Martinez, D.M.; et al. Methane Emissions from the Natural Gas Transmission and Storage System in the United States. *Environ. Sci. Technol.* **2015**, *49*, 9374–9383. [CrossRef] [PubMed]
21. Allen, D.T. Emissions from oil and gas operations in the United States and their air quality implications. *J. Air Waste Manag. Assoc.* **2016**, *66*, 549–575. [CrossRef] [PubMed]
22. Howarth, R.W.; Santoro, R.; Ingraffea, A. Methane and the greenhouse-gas footprint of natural gas from shale formations. *Clim. Chang.* **2011**, *106*, 679–690. [CrossRef]
23. Howarth, R.W. A bridge to nowhere: Methane emissions and the greenhouse gas footprint of natural gas. *Energy Sci. Eng.* **2014**, *2*, 47–60. [CrossRef]
24. Burnham, A.; Han, J.; Clark, C.E.; Wang, M.; Dunn, J.B.; Palou-Rivera, I. Life-Cycle Greenhouse Gas Emissions of Shale Gas, Natural Gas, Coal, and Petroleum. *Environ. Sci. Technol.* **2012**, *46*, 619–627. [CrossRef]
25. Nilsen, O.V. LNG Regulatory Update. Available online: http://www.golng.eu/files/Main/20180417/2.OleVidarNilsen-DNVGL.pdf (accessed on 24 February 2019).
26. Methanex About Methanol. Available online: https://www.methanex.com/about-methanol/methanol-marine-fuel (accessed on 5 February 2018).
27. International Maritime Organization. *Report of the Maritime Safety Committee on Its 99th Session*; International Maritime Organization: London, UK, 2018.
28. Bromberg, L.; Cheng, W.K. *Methanol As an Alternative Transport Fuel*; Massachusetts Institute of Technology: Cambridge, MA, USA, 2010.
29. Ellis, J.; Ramne, B.; Bomanson, J.; Molander, P.; Tunér, M.; Aakko-Saksa, P.; Svanberg, M.; Rydbergh, T.; Berneblad, B. *SUMMETH—Sustainable Marine Methanol Deliverable D6.2 Final Report—Summary of the SUMMETH Project Activities and Results*; VIV Verbrandingsmotor: Rotterdam, The Netherlands, 2018.
30. Methanex Methanex Posts Regional Contract Methanol Prices for North America, Europe and Asia. Available online: https://www.methanex.com/our-business/pricing (accessed on 2 March 2019).
31. Ship and Bunker World Bunker Prices. Available online: https://shipandbunker.com/prices (accessed on 2 March 2019).
32. van den Bos, G.; Wiegmans, B. Short sea shipping: A statistical analysis of influencing factors on SSS in European countries. *J. Shipp. Trade* **2018**, *3*, 6. [CrossRef]
33. Perakis, A.N.; Denisis, A. A survey of short sea shipping and its prospects in the USA. *Marit. Policy Manag.* **2008**, *35*, 591–614. [CrossRef]
34. Arof, A.M. Decision Making Model for Ro-Ro Short Sea Shipping Operations in Archipelagic Southeast Asia. *Asian J. Shipp. Logist.* **2018**, *34*, 33–42. [CrossRef]
35. Medda, F.; Trujillo, L. Short-sea shipping: An analysis of its determinants. *Marit. Policy Manag.* **2010**, *37*, 285–303. [CrossRef]
36. Koliousis, I.G. *The Dynamics of Short Sea Shipping New Practices and Trends*; Springer: Berlin, Germany, 1842; ISBN 9783319980430.
37. Winebrake, J.J.; Wang, M.Q.; He, D. Toxic emissions from mobile sources: A total fuel-cycle analysis for conventional and alternative fuel vehicles. *J. Air Waste Manag. Assoc. 1995* **2001**, *51*, 1073–1086. [CrossRef]

38. Wang, M.; Wu, M.; Hong, H. Life-cycle energy and greenhouse gas emissions impacts of different corn ethanol plant types. *Environ. Res. Lett.* **2007**, *2*, 024001. [CrossRef]
39. Wang, M.Q. *Development and Use of the GREET Model to Estimate Fuel-Cycle Energy Use and Emissions of Various Transportation Technologies and Fuels*; Center for Transportation Research, Energy Systems Division: Argonne, IL, USA, 1996.
40. Yeh, S.; Sperling, D. Low carbon fuel standards: Implementation scenarios and challenges. *Energy Policy* **2010**, *38*, 6955–6965. [CrossRef]
41. Schnepf, R.; Yacobucci, B.D. *Renewable Fuel Standard (RFS): Overview and Issues*; Congressional Research Service: Washington, DC, USA, 2013.
42. Balcombe, P.; Anderson, K.; Speirs, J.; Brandon, N.; Hawkes, A. The Natural Gas Supply Chain: The Importance of Methane and Carbon Dioxide Emissions. *ACS Sustain. Chem. Eng.* **2017**, *5*, 3–20. [CrossRef]
43. Argonne National Lab GREET Model. Available online: https://greet.es.anl.gov (accessed on 21 January 2019).
44. Askin, A.C.; Barter, G.E.; West, T.H.; Manley, D.K. The heavy-duty vehicle future in the United States: A parametric analysis of technology and policy tradeoffs. *Energy Policy* **2015**, *81*, 1–13. [CrossRef]
45. Demir, E.; Bektaş, T.; Laporte, G. A review of recent research on green road freight transportation. *Eur. J. Oper. Res.* **2014**, *237*, 775–793. [CrossRef]
46. Nealer, R.; Matthews, H.S.; Hendrickson, C. Assessing the energy and greenhouse gas emissions mitigation effectiveness of potential US modal freight policies. *Transp. Res. Part A Policy Prct.* **2012**, *46*, 588–601. [CrossRef]
47. Plevin, R.J. Modeling corn ethanol and climate: A critical comparison of the BESS and GREET models. *J. Ind. Ecol.* **2009**, *13*, 495–507. [CrossRef]
48. Wang, M.Q.; Wu, M.; Huo, H.; Liu, J. Life-Cycle Energy Use and Greenhouse Gas Emission Implications of Brazilian Sugarcane Ethanol Simulated with the GREET Model. *Int. Sugar J.* **2008**, *110*.
49. Meyer, P.E.; Green, E.H.; Corbett, J.J.; Mas, C.; Winebrake, J.J. Total fuel-cycle analysis of heavy-duty vehicles using biofuels and natural gas-based alternative fuels. *J. Air Waste Manag. Assoc.* **2011**, *61*, 285–294. [CrossRef]
50. U.S. Energy Information Administration. *Annual Energy Outlook 2018*; U.S. Energy Information Administration: Washington, DC, USA, 2018.
51. IPCC. *Climate Change 2014 AR5 Synthesis Report*; IPCC: Geneva, Switzerland, 2014.
52. Littlefield, J.A.; Marriott, J.; Schivley, G.A.; Skone, T.J. Synthesis of recent ground-level methane emission measurements from the U.S. natural gas supply chain. *J. Clean. Prod.* **2017**, *148*, 118–126. [CrossRef]
53. Allen, D.T.; Pacsi, A.P.; Sullivan, D.W.; Zavala-Araiza, D.; Harrison, M.; Keen, K.; Fraser, M.P.; Hill, A.D.; Sawyer, R.F.; Seinfeld, J.H. Methane emissions from process equipment at natural gas production sites in the United States: Pneumatic controllers. *Environ. Sci. Technol.* **2015**, *49*, 633–640. [CrossRef]
54. Allen, D.T.; Sullivan, D.W.; Zavala-Araiza, D.; Pacsi, A.P.; Harrison, M.; Keen, K.; Fraser, M.P.; Daniel Hill, A.; Lamb, B.K.; Sawyer, R.F.; et al. Methane Emissions from Process Equipment at Natural Gas Production Sites in the United States: Liquid Unloadings. *Environ. Sci. Technol.* **2015**, *49*, 641–648. [CrossRef]
55. Zavala-Araiza, D.; Lyon, D.R.; Alvarez, R.A.; Davis, K.J.; Harriss, R.; Herndon, S.C.; Karion, A.; Kort, E.A.; Lamb, B.K.; Lan, X.; et al. Reconciling divergent estimates of oil and gas methane emissions. *Proc. Natl. Acad. Sci. USA* **2015**, *112*, 201522126. [CrossRef]
56. Marchese, A.J.; Vaughn, T.L.; Zimmerle, D.J.; Martinez, D.M.; Williams, L.L.; Robinson, A.L.; Mitchell, A.L.; Subramanian, R.; Tkacik, D.S.; Roscioli, J.R.; et al. Methane Emissions from United States Natural Gas Gathering and Processing. *Environ. Sci. Technol.* **2015**, *49*, 10718–10727. [CrossRef]
57. Lamb, B.K.; Edburg, S.L.; Ferrara, T.W.; Howard, T.; Harrison, M.R.; Kolb, C.E.; Townsend-Small, A.; Dyck, W.; Possolo, A.; Whetstone, J.R. Direct measurements show decreasing methane emissions from natural gas local distribution systems in the United States. *Environ. Sci. Technol.* **2015**, *49*, 5161–5169. [CrossRef]
58. Burnham, A. *Updated Natural Gas Pathways in the GREET1_2018 Model*; Argonne National Lab.(ANL): Argonne, IL, USA, 2018.

59. Bouman, E.A.; Lindstad, E.; Rialland, A.I.; Strømman, A.H. State-of-the-art technologies, measures, and potential for reducing GHG emissions from shipping—A review. *Transp. Res. Part D Transp. Environ.* **2017**, *52*, 408–421. [CrossRef]
60. Gilbert, P.; Walsh, C.; Traut, M.; Kesieme, U.; Pazouki, K.; Murphy, A. Assessment of full life-cycle air emissions of alternative shipping fuels. *J. Clean. Prod.* **2018**, *172*, 855–866. [CrossRef]