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Comparison of Life Cycle Greenhouse Gases from Natural Gas Pathways for Light-Duty Vehicles

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Supporting Information

ABSTRACT: Low prices and abundant resources open new opportunities for using natural gas, one of which is the production of transportation fuels. In this study, we use a Monte Carlo analysis combined with a life cycle analysis framework to assess the greenhouse gas (GHG) implications of a transition to natural gas-powered vehicles. We consider six different natural gas fuel pathways in two representative light-duty vehicles: a passenger vehicle and a sport utility vehicle. We find that a battery electric vehicle (BEV) powered with natural gas-based electricity achieves around 40% life cycle emissions reductions when compared to conventional gasoline. Gaseous hydrogen fuel cell electric vehicles (FCEVs) and compressed natural gas (CNG) vehicles have comparable life cycle emissions with conventional gasoline, offering limited reductions with 100-year global warming potential (GWP) yet leading to increases with 20-year GWP. Other liquid fuel pathways (methanol, ethanol, and Fischer−Tropsch liquids) have larger GHG emissions than conventional gasoline even when carbon capture and storage technologies are available. Life cycle GHG emissions of natural gas pathways are sensitive to the vehicle fuel efficiency, to the methane leakage rates of natural gas systems, and to the GWP assumed. With the current vehicle technologies, the break-even methane leakage rates of CNG, gaseous hydrogen FCEV, and BEV are 0.9%/2.3%, 1.2%/2.8%, and 4.5%/10.8% (20-year GWP/100-year GWP). If the actual methane leakage rate is lower than the break-even rate of a specific natural gas pathway, that natural gas pathway reduces GHG emissions compared to conventional gasoline; otherwise, it leads to an increase in emissions.

INTRODUCTION

The past decade has seen a significant increase in U.S. natural gas production due to the technological success in extracting natural gas from unconventional resources. While in 2005 the United States (U.S.) shale gas production was negligible, by 2012 it reached 25.7 billion cubic feet per day (BCF/d),[1] and today it accounts for 40% of total dry natural gas production in the U.S.[2] The U.S. Energy Information Agency (EIA) forecasts that shale gas production will reach 45.8 BCF/d by 2040.[3] The rapid increase of natural gas supply has led to a large decrease in wellhead prices, which dropped from $7.97 per thousand cubic feet (Mcf) in 2008 to $2.66/Mcf in 2012.[4] As a result of the emergence of this domestic natural gas resource, there is a growing interest in using natural gas for electricity generation, for producing transportation fuels, for petrochemical manufacturing, and also for exports.[3]

Light duty vehicles (LDV) are the largest providers of mobility services to the U.S. population. More than 90% of U.S. families have at least one vehicle, and, on average, each household owns more than two vehicles.[5] Currently, there are more than 244 million LDVs in use in the U.S., and each year around 15 million new LDVs are sold.[5] In 2013, more than half (54%) of the new LDVs were gasoline-powered passenger vehicles, while the other half were gasoline-powered sport utility vehicles (SUVs) (32%), and pick-up trucks (11%).[5] By comparison, there are less than 1.2 million alternative fuel vehicles (AFVs) in use,[5] representing only 0.5% of the LDV fleet.

In the transportation sector, gasoline and distillate fuel from petroleum meet more than 90% of energy consumption.[5] The emergence of natural gas supply may open the opportunity for use of natural gas for transportation.[6−15] If so, several different pathways can be used. For example, natural gas could be used directly as a transportation fuel through compression or liquefaction, or it can be converted into other transportation fuels, such as hydrogen, electricity, and even gasoline and diesel via the Fischer−Tropsch process.

Life cycle analysis (LCA) is a widely used method to assess the environmental effects of a product or service from production to end of life.[16] There is an extensive body of research about the life cycle greenhouse (GHG) emissions of alternative transportation fuels, including hybrid electric vehicles (HEVs), plug-in hybrid electric vehicles (PHEVs), battery electric vehicles (BEVs), and hydrogen fuel cell electric vehicles (FCEVs).[17−40] Similarly, another large body of work has analyzed the life cycle GHG emissions of using natural gas to meet end uses (including transportation).[19,34,37−53] In 1999, Wang et al.[19] evaluated the life cycle GHG emissions of nine natural gas-based fuels, compressed natural gas (CNG), liquefied natural gas (LNG), liquid petroleum gas (LPG), electricity, methanol, gaseous hydrogen, liquid hydrogen, Fischer−Tropsch diesel, and dimethyl ether (DME), and they found that the "use of NG-based fuels can help reduce per-mile fossil energy use considerably and eliminate petroleum use in most cases; all [but near-term M85 FFVs] help reduce GHG emissions." More recently, Venkatesh et al.[34] used a Monte Carlo analysis to characterize the uncertainty of the life cycle GHG emissions of CNG and gasoline HEVs for passenger

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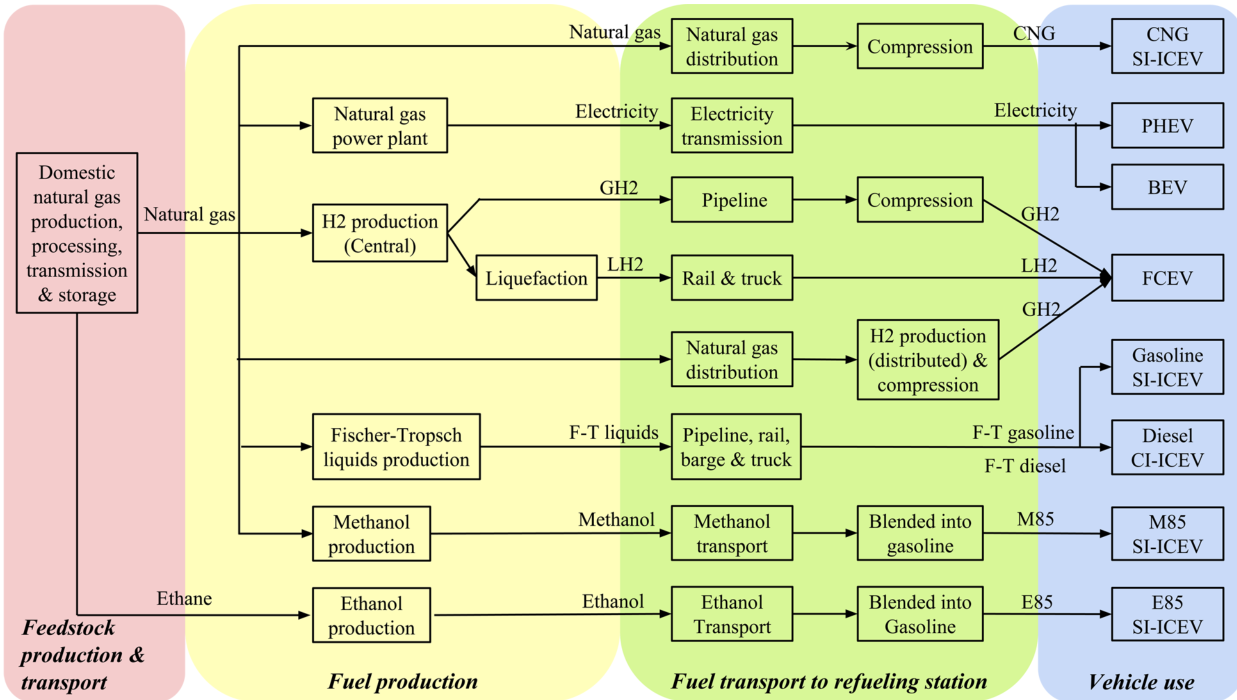

Figure 1. Study system boundary of natural gas pathways. Colored areas correspond to different life cycle stages: natural gas upstream (pink), fuel production (yellow), fuel transport (green), and vehicle operation (blue) (indicated by engine technologies). Both feedstock and energy carriers are marked along each pathway. CNG = compressed natural gas; H2 = hydrogen; GH2 = gaseous hydrogen; LH2 = liquid hydrogen; F-T = Fischer–Tropsch; M85 = a blend of methanol (85% by volume) and gasoline (15%); E85 = a blend of ethanol (85% by volume); ICEV = internal combustion engine vehicle; SI = sparking ignition; CI = compression ignition; PHEV = plug-in hybrid electric vehicle; BEV = battery electric vehicle.

vehicles. The authors found that both HEVs and CNG vehicles achieve emission reductions over conventional gasoline vehicles (on average 25% reduction for HEV and 5% reduction for CNG), but with some probabilities that either pathway is worse than conventional gasoline vehicles. Similarly, Curran et al.[38] used the GREET model (version 2012) to analyze the well-to-wheel energy use and GHG emissions from natural gas pathways. They specifically compared CNG vehicles and electric vehicles charged with natural gas-based electricity and found that the latter is better. Dai et al.[39] considered other environmental impacts (air pollutants, toxicity, land use, and water consumption) in addition to GHG emissions and found that BEVs and FCEVs with natural gas-based electricity and hydrogen reduce environmental impacts. Luk et al.[40] analyzed the life cycle GHG emissions and ownership costs of CNG and natural gas-derived electricity in BEVs. They found that CNG is more cost-effective in reducing GHG emissions than BEVs. While these studies reached similar conclusions, they also shared common limitations. Except those of Venkatesh et al.[34] and Luk et al.,[40] both of which focused only on CNG and electricity from natural gas, other studies reported point estimates and largely ignored the uncertainty and variability in the life cycle of natural gas pathways, especially the uncertainty in methane emissions from natural gas systems. In addition, they used outdated global warming potential (GWP) values that do not reflect the most recent Intergovernmental Panel on Climate Change (IPCC) estimates, and they did not include SUVs in the analysis.

In this paper, we address these shortcomings by performing an LCA coupled with a Monte Carlo analysis to estimate GHG emissions from a broad set of potential fuel pathways that use natural gas directly or indirectly to power passenger vehicles and SUVs. We use scenario analysis and break-even analysis to understand the implications of policy-relevant choices (such as the GWP time frame) and highly uncertain variables (such as methane emissions from natural gas systems).

METHODS AND DATA

System Boundary. Figure 1 illustrates the natural gas pathways and engine technologies considered in this study. The analysis includes six different types of transportation fuels: CNG, natural gas-based electricity, natural gas-based hydrogen, natural gas-based Fischer–Tropsch liquids (gasoline and diesel), natural gas-based methanol, and ethane-based ethanol. We evaluate six vehicle technologies: a sparking ignition internal combustion engine vehicle (SI-ICEV), a flex fuel vehicle (FFV), a compression ignition internal combustion engine vehicle (CI-ICEV), a hybrid electric vehicle (HEV), a plug-in hybrid electric vehicle (PHEV), a battery electric vehicle (BEV), and a fuel cell electric vehicle (FCEV). For both passenger vehicles and SUVs, the functional unit is one vehicle kilometer traveled.

We consider GHG emissions from the full life cycle for each pathway. For instance, GHGs result from the combustion of natural gas and other fossil fuels used to provide energy in the production and transport of natural gas, natural gas flaring, noncombusted emissions (such as vents and fugitive methane and CO_2 emissions), and land use (associated with well pad and well constructions). Downstream activities include production of transportation fuels from natural gas, transportation of final fuels from plants to fueling stations, and use in vehicles. In addition to these fuel-related GHG emissions, we also include emissions from vehicle manufacturing. We exclude emissions associated with building the infrastructure needed to deploy different fuel pathways. Recent literature suggests that they contribute to less than 1% of the cradle-to-gate (from extraction of feedstock to the finished product from the production facilities) GHG emissions in the case of oil and natural gas production as well as the production of electricity and hydrogen from natural gas.[54−56]

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We convert emissions of different GHGs into CO_2-equivalent emissions by multiplying the mass of emissions and their GWP. We consider fossil methane, and model the uncertainty in GWP using a normal distribution[57,58] (Table 1). We use the latest GWP values with inclusion of climate-carbon feedbacks reported in the Fifth Assessment Report of the IPCC[57] and assume a normal distribution. All else being equal, the choice of time horizon for GWP greatly changes the equivalent CO_2 emissions of methane, which has a much higher GWP over 20 years than over 100 years. While most life cycle studies used 100-year GWP, short-term implications of methane emissions are increasingly of interest.[48,59] We report GHG emissions using GWP with both 100-years and 20-years. There are limitations in using GWP, such as ignoring the timing of emissions,[35,57,60−63] but taking account of alternative climate metrics is beyond the scope of this paper.

Table 1. Global Warming Potential (GWP) Values (Climate-Carbon Feedbacks of Non-CO_2 Gases Are Considered)

Greenhouse Gas	100-yr	20-yr
CO_2	1	1
CH_4 (fossil)	Norm (36, 8.5)	Norm (87, 15.9)
N_2O	Norm (298, 52.5)	Norm (268, 34.2)

Natural Gas Upstream Emissions. U.S. production of natural gas in 2013 came from four sources, including conventional natural gas (38%), shale gas (40%), associated gas as a coproduct of crude oil (18%), and coal-bed methane (5%).[2] We focus on shale gas given its prevalence in the U.S.[3] Differences in life cycle GHG emissions from conventional natural gas and shale gas are small.[45,64]

We use natural gas upstream estimations estimates from Tong et al.,[52] which follows a bottom-up life cycle assessment framework shared by a number of existing studies.[34,42,45,48,65] In this previous work, we used a bottom-up framework where we divided the natural gas systems in five stages - preproduction, production, processing, transmission and storage, and distribution−following the U.S. Environmental Protection Agency (EPA)'s GHG emissions inventory.[66] For each stage, we modeled key emission sources individually, which required estimates of emission factors (emission per unit of activity) and corresponding activity data (total units of activity). We accounted for two types of emissions sources, combustion emissions and noncombustion (i.e., fugitive) emissions. Combustion sources include well drilling, transportation of hydraulic liquids and wastewater, lease fuel use, plant fuel, and pipeline fuel use. We relied on previous studies[34,42,66,67] to model these combustion sources. Noncombustion sources include intentional venting and nonintentional leakage. Sources of intentional venting include well completion, well workover, liquid unloading, and blowdowns and upsets. Nonintentional sources include leaks from production devices and pipelines. Existing studies usually rely on the U.S. EPA's GHG emissions inventory for emissions factors and activity data. However, since U.S. EPA still uses outdated emission factors originating from a field campaign in 1990s, recent research efforts have focused on updating these emission factors with on-site measurements.[68−70] In ref 52 we thus used recent field measurements (such as well completion, well work-over, and liquid unloading[68,69]). In addition, we relied on the U.S. EPA's GHG emissions inventory[66] for emissions sources that recent studies have not evaluated.

As in ref 52 we use Monte Carlo analysis to account for variability and uncertainty of emissions factors and activity data of emissions sources considered in the bottom-up framework. Some recent studies suggest that there is a 50% difference between top-down estimates and bottom-up estimates of methane emissions from natural gas systems.[46,47] Unfortunately, there is no sufficient information to develop meaningful probability distributions that account for bottom-up and top-down estimates. Instead, to account for potential bias in our baseline bottom-up estimate, we develop a pessimistic scenario of methane emissions. In this pessimistic scenario, we multiply the distribution of our bottom-up estimate by 1.5. We refer the readers interested in the technical details of the bottom-up framework and Monte Carlo analysis for the upstream natural gas emissions to ref 52.

Natural Gas-derived Fuel Production and Distribution Emissions. Dry natural gas and ethane (the feedstock for ethanol production) go through different conversion and distribution processes to produce transportation fuels. We assume that the first four upstream stages (from preproduction to transportation) are common to all fuels produced from dry natural gas, while the last stage (distribution) is only included in pathways in which fuel production takes place at refueling stations, such as CNG and distributed gaseous hydrogen (see Figure 1). Ethane, which is used to produce ethanol, shares the first three upstream stages with dry natural gas.

Electricity. Although it is difficult to identify individual power plants for specific electricity consumptions, it is likely that increasing electricity demand from electric vehicles, as well as stringent regulations on new coal power plants,[71,72] will drive further deployment of natural gas combined cycle (NGCC) plants. In fact, nearly half of the new power plant capacity in U.S. in 2013 was NGCC.[73] In this study, we assume that natural gas-based electricity powers centrally located fuel production plants (hydrogen, ethanol, methanol, and Fischer−Tropsch liquids) as well as the charging of BEVs and PHEVs. Other, smaller, electricity consumers (such as fuel production at refueling stations) rely on the U.S. grid (with an emission factor of 612 gCO_2-equiv/MJ_{LHV}[74]). We use NETL (2013)[75] to model NGCC power plants without and with carbon capture and sequestration (CCS) technologies (energy efficiencies in lower heating value are 55% and 47.5% and carbon capture rates are 0% and 88.2%, respectively). We assume transmission and distribution losses to be 6.5% of generated electricity.[76] We also assume that the charging efficiency of BEVs and PHEVs follows a uniform distribution of 85%−88%.[26,36,76]

CNG. The CNG pathway relies on natural gas pipelines to deliver natural gas to refueling stations, where compression occurs to "produce" CNG. There are two types of compressors: electric compressors and natural gas-fueled compressors, with electric compressors being the prevalent choice.[76] We thus only consider

Table 2. Hydrogen Production Profile for One MJ of Hydrogen Produced[55,76,77]

Key parameters	Central hydrogen plant (without CCS)		Central hydrogen plant (with CCS)[77]		Hydrogen production at refueling stations	
	Distribution	Distribution parameters	Distribution	Distribution parameters	Distribution	Distribution parameters
Energy efficiency*	triangular**	(0.72, 0.74, 0.79)	triangular	(0.71, 0.73, 0.78)	triangular	(0.71, 0.72, 0.74)
Electricity share of all inputs	triangular	(0.007, 0.012, 0.044)	triangular	(0.018, 0.026, 0.03)	triangular	(0.024, 0.050, 0.083)
Natural gas share of all inputs	1-(electricity share)		1-(electricity share)		1-(electricity share)	
Process GHG emission factor (gCO_2-equiv/MJ of H_2)+	uniform++	77−79	uniform	7.7−7.9	uniform	77−77.2

*The energy efficiency is the ratio of the energy contents of all outputs to those of all inputs (natural gas as feedstock, natural gas as fuel, and electric power). +Process GHG emission factor includes GHG emissions within the hydrogen production plant but does not include emissions embodied in electricity inputs and upstream emissions of natural gas inputs. **Parameters of the triangular distribution are lower limit, mode, and upper limit. ++Parameters of the uniform distribution are lower limit and upper limit.

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electric compressors, which have an energy efficiency that follows a uniform distribution of 0.94 to 0.98.[34]

Hydrogen (H_2). Current industry practice uses steam methane reforming technology to produce gaseous hydrogen from natural gas. We consider three configurations of a hydrogen supply chain: centrally produced gaseous hydrogen (GH_2); centrally produced hydrogen used in the liquid phase (LH_2); and distributed production of GH_2. We also consider CCS technologies in central hydrogen production plants. Table 2 summarizes hydrogen plant assumptions.[55,76,77] Liquid hydrogen has a higher energy density than gaseous hydrogen but requires an energy-intensive liquefaction process and suffers from boil-off leakage. We assume loss factors of 0.3%, 0.16% and 0.5% at liquefaction plant, transport and distribution, and storage of LH_2 after accounting for an 80% rate of capture and reuse of boil-off gas.[76]

Fischer–Tropsch Liquids. In the Fischer–Tropsch liquid production process, natural gas delivered through the transmission system undergoes thermo-chemical transformation to produce liquid fuels (gasoline and diesel) similar to those produced in an oil refinery. We use the process-level data of a Fischer–Tropsch plant (see ref 78 for details) and perform an energy-based emissions allocation. Upon production, Fischer–Tropsch liquids are transported in existing petroleum-product infrastructure and used in current petroleum ICEVs.

Methanol. The late 1990s and early 2000s saw a growing interest in using methanol as an alternative fuel in the U.S. and Canada.[79,80] While there are currently no methanol-fueled vehicles in the market, high performance of methanol attracts some niche markets.[79] We model a M85 (85% methanol and 15% gasoline in volume) pathway since pure methanol suffers from cold start issue and has safety concerns (such as invisible flame and erosion of mechanical systems).[79] Methanol can be produced from natural gas in a centralized methanol production plant using steam reforming technologies. It is a two-step process, where the first step is to produce synthesis gas from natural gas, and the second step is the catalytic synthesis of methanol from the synthesis gas.[79] Table 3 summarizes the assumptions in our M85 model.[25,76,81,82]

Table 3. Methanol Production Plant Profile for One MJ of Methanol Produced[25,76,81,82]

Key parameter	Distribution type	Distribution parameters
Energy efficiency*	Triangular+	(0.41, 0.57, 0.68)
Natural gas share of all inputs	Triangular+	(0.994, 0.999, 1.000)
Electricity share of all inputs	1-(natural gas share of all inputs)	
Feedstock share of natural gas inputs	Triangular+	(0.63, 0.78, 0.88)
Fuel share of natural gas inputs	1-(feedstock share of natural gas inputs)	

*The energy efficiency is the ratio of the energy contents of all outputs to those of all inputs (natural gas as feedstock, natural gas as fuel, and electric power). +Parameters of the triangular distribution are lower limit, mode, and upper limit.

Ethanol. While the production of ethanol has already transitioned to biomass-based pathways (such as corn grain, sugar cane, and cellulosic biomass[83]), ethanol can be produced from fossil fuel-based naphtha and ethane, a coproduct of methane. There are two steps to produce ethanol from ethane—the first step is to produce ethylene through ethane cracking[50] and the second step is to produce ethanol using catalytic ethylene hydration.[84] The Supporting Information provides specific details about the technical specifications of this two-step process.

Petroleum Upstream and Combustion Emissions. The baseline fuel for LDVs is conventional gasoline in the U.S. We rely on existing studies[76,85–87] to model GHG emissions from its life cycle, which includes oil production, oil transport, oil refining, gasoline transport, and combustion during vehicle use (Table 4). We also include conventional diesel and one specific type of unconventional oil,[88] Canadian oil sand-derived crude, which accounts for the largest fraction of imported oil in the U.S.[89]

Vehicle Specifications. We model new vehicles available in the market instead of the existing fleet. We use functionally equivalent vehicles across fuel pathways - compact passenger vehicles and compact SUVs - to eliminate the bias of vehicle choices.[90] There is at least one vehicle model currently offered in the market for all natural gas and petroleum pathways except for M85. The fuel economy assumptions of these vehicles are from the U.S. Department of Energy (DOE) and the U.S. EPA[91] (see Table 5). We rely on the literature for fuel economy assumptions of M85 vehicles and PHEVs.[29,36,76] We further rely on the National Household Travel Survey (NHTS)[92] to estimate the fraction of electric and gasoline driving for any ride of PHEVs (PHEV30 and PHEV60 with 30 km and 60 km electric-only range, respectively). On-road fuel economy may be different from measurements due to factors such as speed, weight, age, road gradient, and ambient temperate.[93,94] While it is beyond our scope to consider these factors, they should be carefully studied in future studies that focus on regional variations. The Supporting Information includes the technical specifications of the vehicles and assumptions for M85 vehicles and PHEVs.

Vehicle tailpipe emissions include CO_2, CH_4, and N_2O emissions. We calculate tailpipe CO_2 emissions using the fuel economy of the vehicle and combustion emissions of the fuel (based on the fuel's carbon content). We model CH_4 and N_2O emissions using emission factors from the GREET model.[76] We assume that electric vehicles (both BEVs and FCEVs) have zero GHG emissions at the tailpipe. As a result of incomplete combustion of natural gas, CNG vehicles have much higher CH_4 emission factors than conventional gasoline vehicles.

We include emissions from vehicle manufacturing. We assume all ICEVs have similar vehicle manufacturing emissions.[76] We rely on the GREET model[76] for manufacturing emissions of ICEVs and FCEVs. For HEVs, PHEVs, and BEVs, we assume they have incremental emissions associated with battery manufacturing, which we calculate using emission factors[95] and activity data (battery size and number of batters per vehicle lifetime), compared to the ICEVs (see the Supporting Information for details).

RESULTS

Comparisons between Natural Gas Pathways and Conventional Gasoline. Figure 2 shows the main results: the life cycle GHG emissions of natural gas pathways for passenger vehicles and SUVs in gCO_2-equiv/km and the associated uncertainty and variability in the results. For light-duty vehicles, the median results suggest that BEVs powered by electricity generated by a natural gas plant provide the lowest GHG emissions across all technologies and pathways considered. This is due to the fact that the high efficiency of BEVs outweighed the emission penalty of electricity generation and battery manufacturing. PHEVs, either with a 30- or 60-km range, when powered by natural gas electricity, have the second lowest average emissions. Both BEVs and PHEVs provide large (more than 20%) emissions reductions compared to conventional gasoline, but none of them is a dominant strategy when compared to gasoline HEVs. Gaseous hydrogen FCEVs and CNG vehicles have comparable life cycle emissions with conventional gasoline, offering limited reductions with 100-year GWP yet leading to increases with 20-year GWP. All other fuel pathways (E85, M85, and Fischer–Tropsch liquids) have larger GHG emissions than conventional gasoline.

Compared to passenger vehicles, SUVs have larger life cycle GHG emissions for all natural gas pathways. For example, a hydrogen-powered fuel-cell SUV has life cycle GHG emissions that are at least 19% higher than those of a fuel-cell passenger vehicle, while a battery electric SUV has life cycle GHG emissions that are 41% higher than those of a battery electric

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Table 4. Life Cycle GHG Emissions of Gasoline and Diesel (Unit: gCO_2-equiv/MJ of fuel delivered)[a]

Fuel	Stage	Distribution	Mean	S.D.+	95% C.I.+
Conventional gasoline	Upstream	Venkatesh et al. (2011)[86]*	18.6	4.0	12.6–28.0
	Combustion	Triangular (71.0, 72.7, 74.9)[76,85–87]	72.9	0.8	71.4–74.4
	Life cycle	Upstream plus combustion	91.5	4.1	85.2–101.0
Conventional diesel	Upstream	Venkatesh et al. (2011)[86]**	17.5	4.3	11.5–27.7
	Combustion	Triangular (72.6, 74.1, 75.2)[76,85–87]	74.0	0.5	72.9–74.9
	Life cycle	Upstream plus combustion	91.5	4.3	85.3–101.7
Oil sand-derived gasoline/diesel	Life cycle	Uniform (103,118)[88]	110.5	4.3	103.4–111.6

[a]Notes: We assume that upstream emissions of conventional gasoline and conventional diesel follow the same distributions as in Venkatesh et al. (2011). *Conventional gasoline upstream emissions follow the difference between a shifted log–logistic distribution (mu = 2.2, alpha = 0.2, delta = 80) and a triangle distribution (68.2, 70.2, 74.6). **Conventional diesel upstream emissions follow the difference between a shifted log–logistic distribution (mu = 2.3, alpha = 0.2, delta = 82) and a triangle distribution (73.6, 75.3, 76.6). +SD stands for standard deviation; C.I. stands for confidence interval.

Table 5. Fuel Economy Assumptions (Unit: Miles Per Gallon of gasoline equivalent)

Pathway		Passenger Vehicle	SUV	Source
Gasoline (baseline)		33	25	fueleconomy.gov[91]
Diesel		32.3	26.2	fueleconomy.gov[91]
Gasoline HEV		45	33	fueleconomy.gov[91]
PHEV30	CS*	43.8		Karabasoglu et al. (2013)[56]
	CD*	112		Karabasoglu et al. (2013)[56]
PHEV60	CS*	42.4		Karabasoglu et al. (2013)[56]
	CD*	105		Karabasoglu et al. (2013)[56]
BEV		110	76	fueleconomy.gov[91]
CNG dedicated		31		
M85 dedicated		35.3		GREET 2014[76]
E85 flex fuel vehicle		31.6	24.7	fueleconomy.gov[91]
Hydrogen fuel cell vehicle		59	49	fueleconomy.gov[91]

*CS is short for Charging Sustaining, and CD is short for Charging Depleting.

passenger vehicle. While SUVs provide advantages such as larger cargo space and better road accessibility, their functions are not significantly different from those of passenger vehicles in most applications but they have larger carbon footprints.

When we look at emissions changes between natural gas pathways and conventional gasoline, we find that all but two natural gas pathways have the same sign (emission increase or emission reduction) under all scenarios considered (i.e., 20- and 100-year GWP; baseline and pessimistic methane emissions). The exceptions are gaseous hydrogen and CNG, for which the time horizon of GWP determines whether they reduce or increase emissions when compared to gasoline. Still, GWP has non-negligible effects on the absolute levels of GHG emissions. Using the 20-year GWP, the life cycle GHG emissions of natural gas pathways increase by 6–17% compared to emission estimates with 100-year GWP.

We find that the benefits of natural gas pathways largely depend on two factors, vehicle fuel efficiency and carbon intensity of the fuel (cradle-to-gate GHG emissions per MJ of fuel delivered; see the Supporting Information for our estimates). Pathways that run on electric vehicles (except liquid hydrogen) have smaller emissions than pathways that run on ICEVs. Within each vehicle technology group, if vehicles' fuel efficiencies are comparable, pathways with higher supply chain efficiency emit less than pathways with lower supply chain efficiency (e.g., CNG vs methanol or gaseous hydrogen vs liquid hydrogen). To assess the contribution of these two effects, we developed a break-even analysis that shows the trade-offs between vehicle fuel efficiency and methane leakage rate that influence the carbon intensity of a natural gas-based fuel.

Break-even Methane Leakage Rates. We perform a break-even analysis for three pathways, CNG, GH_2 FCEV, and BEV, as their emissions are highly dependent on fugitive methane emissions. Further, these pathways are the closest to commercial deployment. We provide a parametric analysis where we determine the methane leakage rate at which life cycle GHG emissions from each of these natural gas pathways equal that of conventional gasoline, hereafter called the *break-even* rate. (For technical details regarding the break-even analysis, we refer the reader to the Supporting Information.) If the actual methane leakage rate from the natural gas systems is lower than the calculated break-even rate of a specific pathway, that pathway has lower life cycle emissions than conventional gasoline. Further, the higher the break-even rates, the larger the emissions reduction coming from that pathway.

Our analysis shows that the break-even rate depends on vehicle fuel efficiency (expressed as the ratio of the gasoline equivalent fuel economy of a natural gas-powered vehicle relative to a conventional gasoline vehicle, hereafter denoted as the *energy economy ratio*, EER) and GWP (see Figure 3). There is a linear relationship between the break-even rate and the vehicle's EER; that is, all else being equal, the break-even rate increases with a higher EER. See, for instance, the relationship between the break-even rate and the EER of the CNG pathway using a 100-year GWP: increasing the EER of the CNG vehicle from 100% (i.e., the same fuel efficiency as its gasoline counterpart) to 110% (i.e., 10% more efficient than its gasoline counterpart) allows for an 1.4 percent point increase in methane leakage rate (from 3.1% to 4.5%).

Figure 3 suggests that current CNG vehicles offer emissions reductions if the life cycle methane leakage rate is lower than 2.3% (using the 100-year GWP) or 0.9% (using the 20-year GWP). A shorter time horizon (such as 20 years), which considers a higher warming potential of methane, requires a lower break-even rate than a longer time horizon (such as 100 years). Current FCEVs offer emission reductions if the life cycle methane leakage rate is lower than 2.8% (100 years) or 1.2% (20 years). Of the three pathways considered, BEVs have largest break-even rates with current vehicle technologies,

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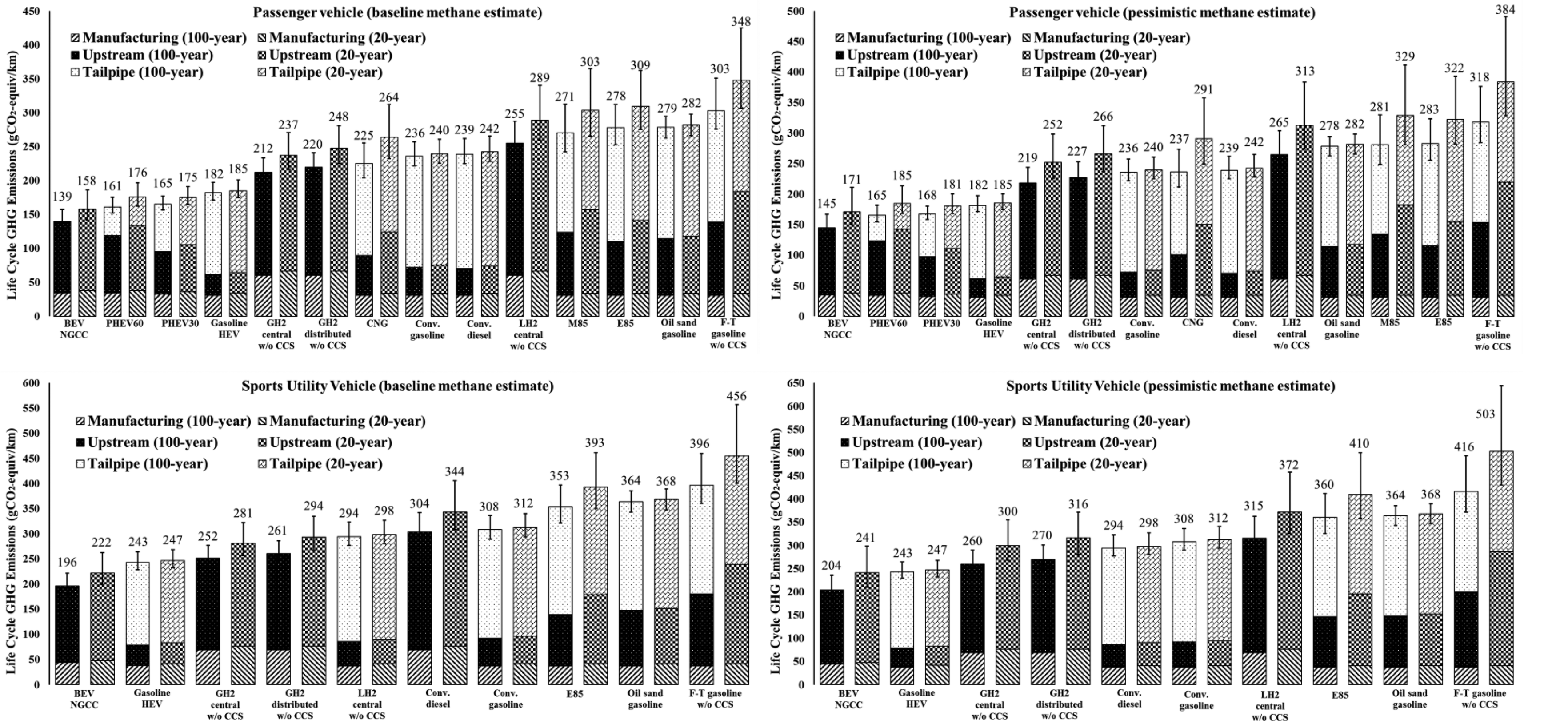

Figure 2. Life cycle GHG emissions of natural gas pathways for passenger vehicles (top panels) and SUVs (bottom panels) in gCO_2-equiv/km. Here we assume both baseline (left panels) and pessimistic estimates (right panels) of methane emissions from natural gas systems. Error bars represent the 95% confidence interval of life cycle GHG emissions, which comprise three sources: vehicle manufacturing, upstream (well-to-pump), and tailpipe (pump-to-well) emissions. Upstream emissions include all use-related emissions from primary energy extraction to dispensing the fuel into vehicles. Tailpipe emissions include all use-related emissions from vehicle operation. Estimates using both 100-year GWPs (left bars) and 20-year GWPs (right bars) are presented side by side for each pathway. Pathways are sorted based on life cycle emissions with 100-year GWP. Data labels represent mean life cycle GHG emissions.

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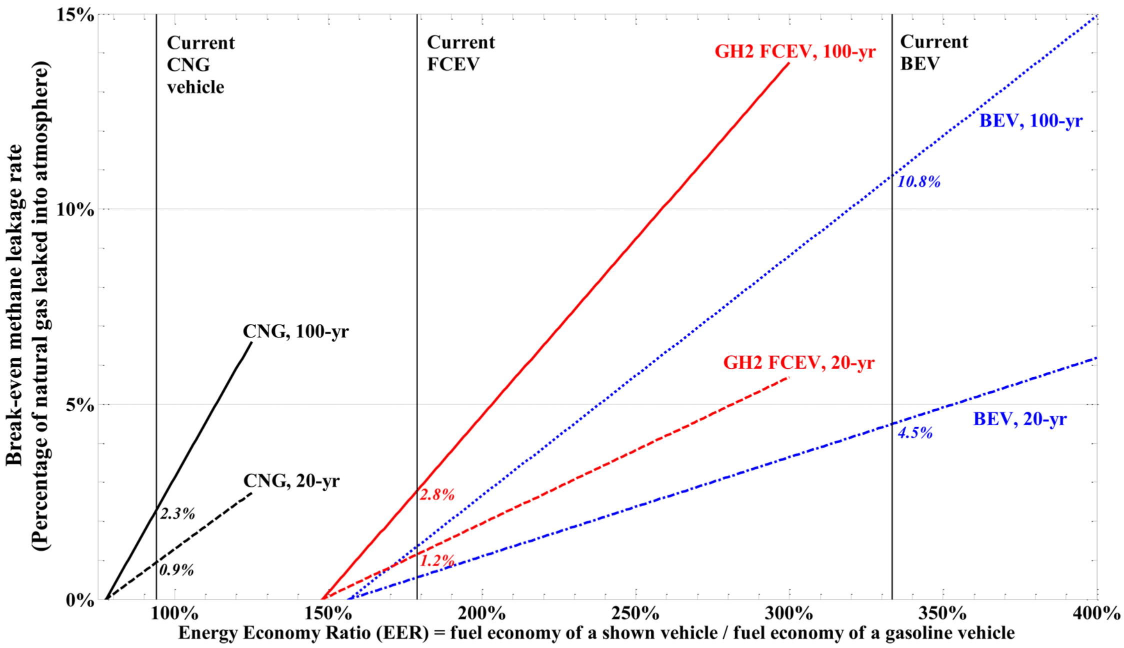

Figure 3. Break-even methane leakage rates of CNG, distributed gaseous hydrogen FCEV, and BEV pathways. At the break-even rate (defined as the volumetric percentage of natural gas that leaks directly into the atmosphere in the life cycle), life cycle GHG emissions from these natural gas pathways are comparable with conventional gasoline. If the actual methane leakage rate from the natural gas systems is lower than the calculated break-even rate of a specific pathway, that pathway has lower life cycle emissions than conventional gasoline. The current EER values for these vehicles are marked with vertical lines.

10.8% (100 years) and 4.5% (20 years). This is consistent with the previous findings that BEVs achieve the largest emissions reductions (see Figure 2). For comparison, our baseline estimate of methane leakage is 1.3%, and our pessimistic estimate is 2.0%, which means that CNG vehicles, FCEVs, and BEVs could achieve emissions reductions with 100-year GWP but not with 20-year GWP, as we have shown in Figure 2.

Other Important Factors. While vehicle manufacturing emissions account for less than 30% of life cycle GHG emissions across all vehicles, manufacturing emissions from FCEVs, which are almost double the manufacturing emissions of ICEVs, merit further discussion. In the sections presented above, we assumed that fuel cells work for the entire lifetime of the vehicle. However, if consumers need to replace fuel cells during vehicle lifetime, increased manufacturing emissions of FCEVs would cause hydrogen pathways to have larger life cycle GHG emissions than gasoline vehicles. Although there is not enough data to quantify the actual lifetime of fuel cells, as they are still in the demonstration phrase, the U.S. DOE has pushed for increasing the durability and reliability of the fuel cell system.[96]

CCS Technology. One potential way to further reduce GHG emissions from natural gas pathways is to capture the carbon emitted from centralized fuel production facilities and store it in geological structures. We consider a scenario in which carbon CCS is available at centralized hydrogen production facilities, natural gas combine cycle power plants, and Fischer−Tropsch plants. CCS technologies reduce the cradle-to-gate emissions of these fuels significantly (37% for Fischer−Tropsch liquids, 38−46% for hydrogen, and 64% for NGCC electricity, compared to the same fuel pathway without CCS technologies). Compared to conventional gasoline passenger vehicles, when CCS technologies are available at fuel production facilities, BEVs and FCEVs have much lower emissions than conventional gasoline (71%, 47%, and 29% reductions for BEVs, gaseous hydrogen, and liquid hydrogen, respectively), but Fischer−Tropsch liquids still have higher emissions than gasoline vehicles.

DISCUSSION

We find that the use of natural gas to produce electricity to then charge BEVs has the lowest life cycle GHG emissions of all natural gas-based fuels considered in this study, and it achieves large emission reductions compared to conventional gasoline. On the other extreme, E85, M85, and Fischer−Tropsch liquids, which have low requirements for new infrastructure, are most likely to lead to increases in GHG emissions. Hydrogen and CNG pathways have the ability to reduce life cycle GHG emissions in LDVs, but such reductions require that methane leakage rates decrease from their current levels.

We find larger uncertainty and variability in life cycle GHG emissions of natural gas pathways than those of conventional gasoline. There is stochastic dominance among natural gas pathways (see Supporting Information), so we simplified the discussions by referring to mean emissions. However, it is important that policy-makers consider uncertainty and variability when they set policy goals based on relative or absolute emissions.[97]

While this paper focuses on GHG emissions, natural gas-based fuels may provide other environmental benefits, such as the reduction of other air pollutants. On the other hand, the adoption of natural gas has to include consumers as part of the equation, namely in what concerns costs, fueling convenience, performance, and safety. These issues are outside the scope of this work and will be addressed in future research.

Increasing vehicle fuel efficiency and reducing methane emissions from the natural gas system are promising strategies to reduce GHG emissions from using natural gas for road transportation. For the same mobility service, a higher vehicle

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fuel efficiency leads to lower life cycle emissions or translates into a higher allowable break-even methane leakage rate. Recent studies find evidence of "super emitters" in natural gas systems: a small number of emission sources that lead to a significant share of methane emissions.[47,69,70,98−101] There are cost-effective technologies to reduce these emissions[102−105] that would reduce the methane leakage rate and provide better opportunities for reducing GHG emissions from light-duty vehicles by using natural gas as a transportation fuel.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.energyfuels.5b01063.

> The Supporting Information provides further discussions on the Monte Carlo model, such as fuel properties and combustion emission factors, assumptions related to ethanol production, and vehicle assumptions; comparisons with existing studies (the GREET model) and discussions of data quality; and additional figures, such as bar plots of natural gas-derived fuels' carbon intensity and cumulative density plots of vehicle life cycle GHG emissions (PDF) as well as numerical values behind these figures (XLSX)

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Notes

The authors declare no competing financial interest.

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■ REFERENCES

(1) U.S. Energy Information Agency (EIA). North America leads the world in production of shale gas. 2014; http://www.eia.gov/todayinenergy/detail.cfm?id=13491 (accessed Oct 23, 2013).

(2) U.S. Energy Information Agency (EIA). Natural gas gross withdrawals and production. http://www.eia.gov/dnav/ng/ng_prod_sum_dcu_NUS_m.htm (accessed Feb 20, 2015).

(3) *Annual Energy Outlook, 2013*; U.S. Energy Information Agency (EIA): Washington, DC, U.S., 2013; http://www.eia.gov/forecasts/aeo/.

(4) U.S. Energy Information Agency (EIA). Natural gas prices; http://www.eia.gov/dnav/ng/ng_pri_sum_dcu_nus_a.htm (accessed Dec 20, 2013).

(5) Davis, S. C.; Diegel, S. W.; Boundy, R. G. *Transportation Energy Data Book*; Oak Ridge National Laboratory (ORNL): Oak Ridge, TN, U.S., 2013; http://cta.ornl.gov/data/index.shtml.

(6) Tezak, C. (BAIRD) Energy Policy - Clean & Green: NGVs: Progress on an Uphill Road. *Energy & Environmental Policy Research*; September 28, 2009. https://baird.bluematrix.com/docs/pdf/e8bd177a-87a5-43cd-8c7f-d8fc23bde70f.pdf.

(7) Marbek. *Study of Opportunities for Natural Gas in the Transportation Sector*; Natural Resources Canada: Ottawa, Ontario, Canada, 2010; http://www.xebecinc.com/pdf/Marbek-NGV-Final-Report-April-2010.pdf.

(8) Werpy, M. R.; Santini, D.; Burnham, A.; Mintz, M. *Natural Gas Vehicles: Status, Barriers, and Opportunities*; Argonne National Laboratory: Lemont, IL, U.S., 2010; http://www.afdc.energy.gov/pdfs/anl_esd_10-4.pdf.

(9) Whyatt, G. *Issues Affecting Adoption of Natural Gas Fuel in Light- and Heavy-Duty Vehicles*; Pacific Northwest National Laboratory: Richland, WA, U.S., 2010; http://s3.amazonaws.com/zanran_storage/www.pnl.gov/ContentPages/184758856.pdf.

(10) Krupnick, A. J. *Will Natural Gas Vehicles Be in Our Future?*; Resources for the Future: Washington, DC, U.S., 2011; http://www.rff.org/Publications/Resources/Pages/181-Natural-Gas-Vehicles.aspx.

(11) Encouraging Natural Gas Vehicles in Pennsylvania. *Allegheny Conference on Community Development (ACCD)*: Pittsburgh, PA, U.S., 2012; http://www.alleghenyconference.org/NaturalGasVehicles/Resources/ACCDNGVehiclesAnalysis.pdf.

(12) Knittel, C. R. *Leveling the Playing Field for Natural Gas in Transportation*; Brookings Institution: Washington, DC, U.S., 2012; http://www.brookings.edu/~/media/research/files/papers/2012/6/13%20transportation%20knittel/06_transportation_knittel.pdf.

(13) Lee, A.; Zinaman, O.; Logan, J. *Opportunities for Synergy Between Natural Gas and Renewable Energy in the Electric Power and Transportation Sectors*; National Renewable Energy Laboratory (NREL): Golden, CO, U.S., 2012; http://www.nrel.gov/docs/fy13osti/56324.pdf.

(14) *Advancing Technology for America's Transportation Future*; National Petroleum Council (NPC): Washington, DC, U.S., 2012; https://www.npc.org/FTF-80112.html.

(15) Morse, E. L.; Yuen, A.; Kleinman, S. M.; Wetherbee, C.; Thein, T.; Michaeli, I.; Cunningham, G.; Smith, R. P. *Energy 2020: Trucks Trains & Automobiles*; Citi Global Perspectives & Solutions (Citi GPS): 2013; https://ir.citi.com/ROzgbJktqIDIbvbbj8Kae%2BI%2F1te4V9Hh91ex5wFyG1wuStLT5EL8L%2BP7DiZLBZC%2FZIYJ%2BHi13iU%3D.

(16) Guinée, J. B.; Heijungs, R.; Huppes, G.; Zamagni, A.; Masoni, P.; Buonamici, R.; Ekvall, T.; Rydberg, T. Life cycle assessment: past, present, and future. *Environ. Sci. Technol.* **2011**, *45*, 90−96.

(17) Delucchi, M. A. *Emissions of greenhouse gases from the use of transportation fuels and electricity*; Argonne National Laboratory: Lemont, IL, U.S., 1991; http://www.osti.gov/scitech/servlets/purl/5457338/.

(18) Delucchi, M. *Emissions of criteria pollutants, toxic air pollutants, and greenhouse gases, from the use of alternative transportation modes and fuels*; Institute of Transportation Studies, University of California, Davis: Davis, CA, U.S., 1996; http://www.its.ucdavis.edu/wp-content/themes/ucdavis/pubs/download_pdf.php?id=617.

(19) Wang, M. Q.; Huang, H. S. *A Full Fuel-Cycle Analysis of Energy and Emissions Impacts of Transportation Fuels Produced from Natural Gas*; Argonne National Laboratory: Lemont, IL, U.S., 1999; http://www.transportation.anl.gov/pdfs/TA/13.pdf.

(20) *Well-to-Wheel Energy Use and Greenhouse Gas Emissions of Advanced Fuel/Vehicle Systems - North American Analysis*; General Motors Corporation, Argonne National Laboratory, BP, ExxonMobil, and Shell: 2001; http://www.transportation.anl.gov/pdfs/TA/163.pdf.

(21) Nylund, N.; Laurikko, J.; Ikonen, M. *Pathways for natural gas into advanced vehicles Part A: Technology and Fuels for New Generation Vehicles*; International Association for Natural Gas Vehicles (INAGV): Auckland, New Zealand, 2002; http://www.fischer-tropsch.org/DOE/DOE_reports/Pathways_Part_A/Pathways_Part_A.pdf.

(22) Delucchi, M. A. *A Lifecycle Emissions Model (LEM): Lifecycle Emissions from Transportation Fuels, Motor Vehicles, Transportation Modes, Electricity Use, Heating and Cooking Fuels, and Materials*; Institute of Transportation Studies, University of California, Davis:

DOI: 10.1021/acs.energyfuels.5b01063
Energy Fuels 2015, 29, 6008−6018

Davis, CA, U.S., 2003; http://www.its.ucdavis.edu/wp-content/themes/ucdavis/pubs/download_pdf.php?id=273.
(23) Hekkert, M. P.; Hendriks, F. H. J. F.; Faaij, A. P. C.; Neelis, M. L. Natural gas as an alternative to crude oil in automotive fuel chains well-to-wheel analysis and transition strategy development. *Energy Policy* **2005**, *33*, 579−594.
(24) Brinkman, N.; Wang, M.; Weber, T.; Darlington, T. *Well-to-Wheels Analysis of Advanced Fuel/Vehicle Systems - A North American Study of Energy Use, Greenhouse Gas Emissions, and Criteria Pollutant Emissions*; Argonne National Laboratory: Lemont, IL, U.S., 2005; http://www.transportation.anl.gov/pdfs/TA/339.pdf.
(25) TIAX. *Full Fuel Cycle Assessment: Well-To-Wheels Energy Inputs, Emissions, and Water Impacts*; California Energy Commission: Sacramento, CA, U.S., 2007; http://www.energy.ca.gov/2007publications/CEC-600-2007-004/CEC-600-2007-004-F.PDF.
(26) Samaras, C.; Meisterling, K. Life cycle assessment of greenhouse gas emissions from plug-in hybrid vehicles: implications for policy. *Environ. Sci. Technol.* **2008**, *42*, 3170−3176.
(27) Stephan, C. H.; Sullivan, J. Environmental and energy implications of plug-in hybrid-electric vehicles. *Environ. Sci. Technol.* **2008**, *42*, 1185−1190.
(28) Jaramillo, P.; Samaras, C.; Wakeley, H.; Meisterling, K. Greenhouse gas implications of using coal for transportation: Life cycle assessment of coal-to-liquids, plug-in hybrids, and hydrogen pathways. *Energy Policy* **2009**, *37*, 2689−2695.
(29) Shiau, C.-S. N.; Samaras, C.; Hauffe, R.; Michalek, J. J. Impact of battery weight and charging patterns on the economic and environmental benefits of plug-in hybrid vehicles. *Energy Policy* **2009**, *37*, 2653−2663.
(30) Elgowainy, A.; Han, J.; Poch, L.; Wang, M.; Vyas, A.; Mahalik, M.; Rousseau, A. *Well-to-Wheels Analysis of Energy Use and Greenhouse Gas Emissions of Plug-In Hybrid Electric Vehicles*; Argonne National Laboratory: Lemont, IL, U.S., 2010; http://www.afdc.energy.gov/pdfs/argonne_phev_evaluation_report.pdf.
(31) U.S. National Research Council (NRC). *Hidden Costs of Energy: Unpriced Consequences of Energy Production and Use*; The National Academies Press: Wahington, DC, U.S., 2010.
(32) Lipman, T.; Delucchi, M. A. Comparing Greenhouse Gas Emissions. In *Sustainable Transportation Energy Pathways: A Research Summary for Decision Makers*; Ogden, J., Anderson, L., Eds.; Institute of Transportation Studies, University of California, Davis: Davis, CA, U.S., 2011; http://steps.ucdavis.edu/files/09-06-2013-STEPS-Book-A-Research-Summary-for-Decision-Makers-Sept-2011.pdf.
(33) Michalek, J. J.; Chester, M.; Jaramillo, P.; Samaras, C.; Shiau, C.-S. N.; Lave, L. B. Valuation of plug-in vehicle life-cycle air emissions and oil displacement benefits. *Proc. Natl. Acad. Sci. U. S. A.* **2011**, *108*, 16554−16558.
(34) Venkatesh, A.; Jaramillo, P. Uncertainty in life cycle greenhouse gas emissions from United States natural gas end-uses and its effects on policy. *Environ. Sci. Technol.* **2011**, *45*, 8182−8189.
(35) 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. U. S. A.* **2012**, *109*, 6435−6440.
(36) Karabasoglu, O.; Michalek, J. Influence of driving patterns on life cycle cost and emissions of hybrid and plug-in electric vehicle powertrains. *Energy Policy* **2013**, *60*, 445−461.
(37) Wang, M. Key Issues for GREET Transportation Fuel LCA (presentation slides); *CRC Workshop on Life Cycle Analysis of Transportation Fuels*, Argonne, IL, Oct. 16−17, 2013; Argonne National Laboratory: Lemont, IL, U.S., 2013; http://www.crcao.org/workshops/LCA%20October%202013/Session%202/Wang,%20Michael%202013.11.20.pdf.
(38) Curran, S. J.; Wagner, R. M.; Graves, R. L.; Keller, M.; Green, J. B. Well-to-wheel analysis of direct and indirect use of natural gas in passenger vehicles. *Energy* **2014**, *75*, 194−203.
(39) Dai, Q.; Lastoskie, C. M. Life Cycle Assessment of Natural Gas-Powered Personal Mobility Options. *Energy Fuels* **2014**, *28*, 5988−5997.
(40) Luk, J. M.; Saville, B. A.; MacLean, H. L. Life Cycle Air Emissions Impacts and Ownership Costs of Light-Duty Vehicles Using Natural Gas As a Primary Energy Source. *Environ. Sci. Technol.* **2015**, *49*, 5151−5160.
(41) Jaramillo, P.; Griffin, W. M.; Matthews, H. S. Comparative life-cycle air emissions of coal, domestic natural gas, LNG, and SNG for electricity generation. *Environ. Sci. Technol.* **2007**, *41*, 6290−6296.
(42) Jiang, M.; Michael Griffin, W.; Hendrickson, C.; Jaramillo, P.; VanBriesen, J.; Venkatesh, A. Life cycle greenhouse gas emissions of Marcellus shale gas. *Environ. Res. Lett.* **2011**, *6*, 034014.
(43) Burnham, A.; Han, J.; Clark, C. E.; Wang, M.; Dunn, J. B.; Palou, I. Life-Cycle Greenhouse Gas Emissions of Shale Gas, Natural Gas, Coal, and Petroleum. *Environ. Sci. Technol.* **2011**, *46*, 619−627.
(44) Howarth, R.; Shindell, D.; Santoro, R.; Ingraffea, A.; Phillips, N.; Townsend-Small, A. *Methane Emissions from Natural Gas Systems*; Background Paper Prepared for the National Climate Assessment, Reference number 2011-0003; Department of Ecology and Evolutionary Biology, Cornell University: Ithaca, NY, U.S., 2012; http://www.eeb.cornell.edu/howarth/publications/Howarth_et_al_2012_National_Climate_Assessment.pdf.
(45) Weber, C. L.; Clavin, C. Life cycle carbon footprint of shale gas: review of evidence and implications. *Environ. Sci. Technol.* **2012**, *46*, 5688−5695.
(46) Miller, S. M.; Wofsy, S. C.; Michalak, A. M.; Kort, E. A.; Andrews, A. E.; Biraud, S. C.; Dlugokencky, E. J.; Eluszkiewicz, J.; Fischer, M. L.; Janssens-Maenhout, G.; et al. Anthropogenic emissions of methane in the United States. *Proc. Natl. Acad. Sci. U. S. A.* **2013**, *110*, 20018−20022.
(47) Brandt, A. R.; Heath, G. A.; Kort, E. A.; O'Sullivan, F.; Pétron, G.; Jordaan, S. M.; Tans, P.; Wilcox, J.; Gopstein, A. M.; Arent, D.; et al. Methane leaks from North American natural gas systems. *Science* **2014**, *343*, 733−735.
(48) Howarth, R. W. A bridge to nowhere: methane emissions and the greenhouse gas footprint of natural gas. *Energy Sci. Eng.* **2014**, *2*, 47−60.
(49) Schwietzke, S.; Griffin, W. M.; Matthews, H. S.; Bruhwiler, L. M. P. Natural gas fugitive emissions rates constrained by global atmospheric methane and ethane. *Environ. Sci. Technol.* **2014**, *48*, 7714−7722.
(50) Posen, I. D.; Griffin, W. M.; Matthews, H. S.; Azevedo, I. L. Changing the Renewable Fuel Standard to a Renewable Material Standard: Bioethylene Case Study. *Environ. Sci. Technol.* **2015**, *49*, 93−102.
(51) Abrahams, L. S.; Samaras, C.; Griffin, W. M.; Matthews, H. S. Life Cycle Greenhouse Gas Emissions From U.S. Liquefied Natural Gas Exports: Implications for End Uses. *Environ. Sci. Technol.* **2015**, *49*, 3237−3245.
(52) Tong, F.; Jaramillo, P.; Azevedo, I. Comparison of Life Cycle Greenhouse Gases from Natural Gas Pathways for Medium and Heavy-Duty Vehicles. *Environ. Sci. Technol.* **2015**, *49*, 7123−7133.
(53) Ou, X.; Zhang, X.; Zhang, X.; Zhang, Q. Life Cycle GHG of NG-Based Fuel and Electric Vehicle in China. *Energies* **2013**, *6*, 2644.
(54) Spath, P. L.; Mann, M. K. *Life Cycle Assessment of a Natural Gas Combined - Cycle Power Generation System*; National Renewable Energy Laboratory (NREL): Golden, CO, U.S., 2000; http://library.umac.mo/ebooks/b28020261.pdf.
(55) Spath, P. L.; Mann, M. K. *Life-Cycle Assessment of Hydrogen Production via Natural Gas Steam Reforming*; National Renewable Energy Laboratory (NREL): Golden, CO, U.S., 2011; http://www1.eere.energy.gov/hydrogenandfuelcells/pdfs/27637.pdf.
(56) Beath, J.; Black, N.; Boone, M.; Roberts, G.; Rutledge, B.; Elgowainy, A.; Wang, M.; Kelly, J. *Contribution of Infrastructure to Oil and Gas Production and Processing Carbon Footprint*; Argonne National Laboratory: Lemont, IL, U.S., 2014; https://greet.es.anl.gov/publication-oil-gas-prod-infra.
(57) Intergovernmental Panel on Climate Change (IPCC). *Climate Change 2013: The Physical Science Basis. Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*; Stocker, T. F., Qin, D., Plattner, G., Tignor, M. M. B., Allen,

DOI: 10.1021/acs.energyfuels.5b01063
Energy Fuels 2015, 29, 6008−6018

S., Boschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P. M., Eds.; Cambridge University Press: Cambridge, United Kingdom, and New York, NY, USA, 2014.

(58) Shindell, D. T.; Faluvegi, G.; Koch, D. M.; Schmidt, G. A.; Unger, N.; Bauer, S. E. Improved attribution of climate forcing to emissions. *Science* **2009**, *326*, 716−718.

(59) Shindell, D.; Kuylenstierna, J. C. I.; Vignati, E.; van Dingenen, R.; Amann, M.; Klimont, Z.; Anenberg, S. C.; Muller, N.; Janssens-Maenhout, G.; Raes, F.; et al. Simultaneously mitigating near-term climate change and improving human health and food security. *Science* **2012**, *335*, 183−189.

(60) Shine, K. P.; Fuglestvedt, J. S.; Hailemariam, K.; Stuber, N. Alternatives to the Global Warming Potential for comparing climate impacts of emissions of greenhouse gases. *Clim. Change* **2005**, *68*, 281−302.

(61) Peters, G. P.; Aamaas, B.; Lund, M. T.; Solli, C.; Fuglestvedt, J. S. Alternative "global warming" metrics in life cycle assessment: A case study with existing transportation data. *Environ. Sci. Technol.* **2011**, *45*, 8633−8641.

(62) Kendall, A. Time-adjusted global warming potentials for LCA and carbon footprints. *Int. J. Life Cycle Assess.* **2012**, *17*, 1042−1049.

(63) Edwards, M. R.; Trancik, J. E. Climate impacts of energy technologies depend on emissions timing. *Nat. Clim. Change* **2014**, *4*, 347−352.

(64) Stephenson, T.; Valle, J. E.; Riera-Palou, X. Modeling the relative GHG emissions of conventional and shale gas production. *Environ. Sci. Technol.* **2011**, *45*, 10757−10764.

(65) Bradbury, J.; Obeiter, M.; Draucker, L.; Wang, W.; Stevens, A. *Clearing the Air: Reducing Upstream Greenhouse Gas Emissions from U.S. Natural Gas Systems*; World Resources Institute: Washington, DC, 2013.

(66) *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990−2012*; U.S. Environmental Protection Agency (EPA): Washington, DC, U.S., 2014; http://epa.gov/climatechange/ghgemissions/usinventoryreport.html.

(67) U.S. Energy Information Agency (EIA). *Natural gas consumption by end use.* http://www.eia.gov/dnav/ng/ng_cons_sum_dcu_nus_a.htm.

(68) Shires, T.; Lev-on, M. *Characterizing Pivotal Sources of Methane Emissions from Natural Gas Production: Summary and Analysis of API and ANGA Survey Responses*; American Petroleum Institute (API): Washington, DC, U.S., 2012; http://www.api.org/~/media/Files/News/2012/12-October/API-ANGA-Survey-Report.pdf.

(69) 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. U. S. A.* **2013**, *110*, 17768−17773.

(70) U.S. Environmental Protection Agency (EPA). *Greenhouse Gas Reporting Program (GHGRP): 2011 Data Sets.* http://www.epa.gov/ghgreporting/ghgdata/reported-2011/index.html (accessed Nov 10, 2013).

(71) Saha, A. *Review of Coal Retirements*; MJB&A: Concord, MA, U.S., 2013; http://www.mjbradley.com/sites/default/files/Coal_Plant_Retirement_Review_Apr2013_0.pdf.

(72) U.S. Energy Information Agency (EIA). *AEO2014 projects more coal-fired power plant retirements by 2016 than have been scheduled.* http://www.eia.gov/todayinenergy/detail.cfm?id=15031 (accessed Feb 14, 2014).

(73) U.S. Energy Information Agency (EIA). *Half of power plant capacity additions in 2013 came from natural gas.* http://www.eia.gov/todayinenergy/detail.cfm?id=15751 (accessed Apr 8, 2015).

(74) Cai, H.; Wang, M.; Elgowainy, A.; Han, J. *Updated Greenhouse Gas and Criteria Air Pollutant Emission Factors of the U. S. Electric Generating Units in 2010*; Argonne National Laboratory: Lemont, IL, U.S., 2013; https://greet.es.anl.gov/files/electricity-13.

(75) Haslbeck, J. L.; Kuehn, N. J.; Lewis, E. G.; Pinkerton, L. L.; Simpson, J.; Turner, M. J., Varghese, E.; Woods, M. C.; *Cost and performance baseline for fossil energy plants. Vol. 1: Bituminous coal and natural gas to electricity final report*; National Energy Technology Laboratory (NETL): 2013; http://www.netl.doe.gov/File%20Library/Research/Energy%20Analysis/OE/BitBase_FinRep_Rev2a-3_20130919_1.pdf.

(76) *The GREET (Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation) Model*; Argonne National Laboratory: Lemont, IL, U.S., 2015; https://greet.es.anl.gov/.

(77) U.S. Department of Energy (DOE). *H2A Analysis.* http://www.hydrogen.energy.gov/h2a_analysis.html.

(78) Jaramillo, P.; Griffin, W. M.; Matthews, H. S. Comparative analysis of the production costs and life-cycle GHG emissions of FT liquid fuels from coal and natural gas. *Environ. Sci. Technol.* **2008**, *42*, 7559−7565.

(79) Bromberg, L.; Cheng, W. K. *Methanol as an alternative transportation fuel in the U.S.: Options for sustainable and energy-secure transportation*; Massachusetts Institute of Technology: Cambridge, MA, U.S., 2010; http://www.afdc.energy.gov/pdfs/mit_methanol_white_paper.pdf.

(80) Kramer, U.; Anderson, J. E. *Prospects for Flexible- and Bi-Fuel Light Duty Vehicles: Consumer Choice and Public Attitudes*; Massachusetts Institute of Technology: Cambridge, MA, U.S., 2012; http://mitei.mit.edu/system/files/Alt_Fuel_Vehicles.pdf.

(81) *Cradle-to-Gate Life Cycle Inventory of Nine Plastic Resins and Two Polyurethane Precursors*; American Chemistry Council (ACC): Washington, DC, U.S., 2007.

(82) *Cradle-to-Gate Life Cycle Inventory of Nine Plastic Resins and Four Polyurethane Precursors*; American Chemistry Council (ACC): Washington, DC, U.S., 2011; http://plastics.americanchemistry.com/LifeCycle-Inventory-of-9-Plastics-Resins-and-4-Polyurethane-Precursors-Rpt-Only.

(83) Parker, A.; Jenkins, B.; Dempster, P.; Higgins, B.; Ogden, J. The Biofuels Pathway. In *Sustainable Transportation Energy Pathways: A Research Summary for Decision Makers*; Ogden, J., Anderson, L., Eds.; Institute of Transportation Studies, University of California Davis, Davis, CA, U.S., 2011; http://steps.ucdavis.edu/files/09-06-2013-STEPS-Book-A-Research-Summary-for-Decision-Makers-Sept-2011.pdf.

(84) Frischknecht, R.; Jungbluth, N.; Althaus, H.-J.; Doka, G.; Dones, R.; Heck, T.; Hellweg, S.; Hischier, R.; Nemecek, T.; Rebitzer, G.; Spielmann, M.; The ecoinvent database: Overview and methodological framework. *Int. J. Life Cycle Assess..* **2005**, *10*, 3−9 (Ecoinvent version 2.0; http://www.ecoinvent.org/).10.1065/lca2004.10.181.1

(85) Gerdes, K.; Skone, T. J. *Development of Baseline Data and Analysis of Life Cycle Greenhouse Gas Emissions of Petroleum-Based Fuels*; National Energy Technology Laboratory (NETL): 2008; http://www.netl.doe.gov/research/energy-analysis/publications/details?pub=39a03247-40ab-441d-8f32-1654e76e2ee4.

(86) Venkatesh, A.; Jaramillo, P.; Griffin, W. M.; Matthews, H. S. Uncertainty analysis of life cycle greenhouse gas emissions from petroleum-based fuels and impacts on low carbon fuel policies. *Environ. Sci. Technol.* **2011**, *45*, 125−131.

(87) Boland, S.; Unnasch, S. *Carbon Intensity of Marginal Petroleum and Corn Ethanol Fuels*; Life Cycle Associates: Portola Valley, CA, U.S., 2014; http://ethanolrfa.3cdn.net/8f1f5d7e868d849da0_gam6b4eab.pdf.

(88) Englander, J. G.; Bharadwaj, S.; Brandt, A. R. Historical trends in greenhouse gas emissions of the Alberta oil sands (1970−2010). *Environ. Res. Lett.* **2013**, *8*, 044036.

(89) U.S. Energy Information Agency (EIA). *U.S. crude oil imports fall, but share of top three suppliers highest in four decades*; 2014; http://www.eia.gov/todayinenergy/detail.cfm?id=15711 (accessed April 4, 2014).

(90) MacLean, H. L.; Lave, L. B. Evaluating automobile fuel/propulsion system technologies. *Prog. Energy Combust. Sci.* **2003**, *29*, 1−69.

(91) U.S. Department of Energy (DOE) and U.S. Environmental Protection Agency (EPA). *Model Year 2015 Fuel Economy Guide*; Washington, DC, U.S., 2015; https://www.fueleconomy.gov/feg/pdfs/guides/FEG2015.pdf.

DOI: 10.1021/acs.energyfuels.5b01063
Energy Fuels 2015, 29, 6008−6018

(92) *National Household Travel Survey 2009*; Federal Highway Administration (FHWA): Washington, DC, U.S., 2011; http://nhts.ornl.gov/.
(93) Reyna, J. L.; Chester, M. V.; Ahn, S.; Fraser, A. M. Improving the Accuracy of Vehicle Emissions Profiles for Urban Transportation Greenhouse Gas and Air Pollution Inventories. *Environ. Sci. Technol.* **2015**, *49*, 369−376.
(94) Yuksel, T.; Michalek, J. J. Effects of Regional Temperature on Electric Vehicle Efficiency, Range, and Emissions in the United States. *Environ. Sci. Technol.* **2015**, *49*, 3974−3980.
(95) Dunn, J. B.; Gaines, L.; Sullivan, J.; Wang, M. Q. Impact of recycling on cradle-to-gate energy consumption and greenhouse gas emissions of automotive lithium-ion batteries. *Environ. Sci. Technol.* **2012**, *46*, 12704−12710.
(96) Eudy, L.; Gikakis, C. *Fuel Cell Buses in U.S. Transit Fleets: Current Status 2014*; National Renewable Energy Laboratory (NREL): Golden, CO, U.S., 2014; http://www.afdc.energy.gov/uploads/publication/fc_buses_2014_status.pdf.
(97) Kocoloski, M.; Mullins, K. A.; Venkatesh, A.; Michael Griffin, W. Addressing uncertainty in life-cycle carbon intensity in a national low-carbon fuel standard. *Energy Policy* **2013**, *56*, 41−50.
(98) Allen, D. T.; Pacsi, A. P.; Sullivan, D. W.; Zavala-Araiza, D.; Harrison, M.; Keen, K.; Fraser, M. P.; Daniel Hill, A.; 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.
(99) 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.
(100) Subramanian, R.; Williams, L. L.; Vaughn, T. L.; Zimmerle, D.; Roscioli, J. R.; Herndon, S. C.; Yacovitch, T. I.; Floerchinger, C.; Tkacik, D. S.; Mitchell, A. L.; et al. Methane Emissions from Natural Gas Compressor Stations in the Transmission and Storage Sector: Measurements and Comparisons with the EPA Greenhouse Gas Reporting Program Protocol. *Environ. Sci. Technol.* **2015**, *49*, 3252−3261.
(101) Mitchell, A. L.; Tkacik, D. S.; Roscioli, J. R.; Herndon, S. C.; Yacovitch, T. I.; Martinez, D. M.; Vaughn, T. L.; Williams, L. L.; Sullivan, M. R.; Floerchinger, C.; et al. Measurements of Methane Emissions from Natural Gas Gathering Facilities and Processing Plants: Measurement Results. *Environ. Sci. Technol.* **2015**, *49*, 3219−3227.
(102) Harvey Consulting LLC. *Leaking Profits: The U.S. Oil and Gas Industry Can. Reduce Pollution, Conserve Resources, and Make Money by Preventing Methane Waste*; Natural Resources Defense Council (NRDC): New York, NY, U.S., 2012; http://www.nrdc.org/energy/files/Leaking-Profits-Report.pdf.
(103) ICF International. *Economic Analysis of Methane Emission Reduction Opportunities in the U.S. Onshore Oil and Natural Ga Industries*; Environmental Defense Fund (EDF): New York, NY, U.S., 2014; http://www.edf.org/sites/default/files/icf_methane.pdf.
(104) Hein, J. F. *Capturing Value: Science and Strategies to Curb Methane Emissions from the Oil and Natural Gas Sector*; Institute for Policy Integrity, New York Unviersity School of Law: New York, NY, U.S., 2014; http://policyintegrity.org/files/publications/Capturing_Value_-_Methane_Policy_Brief.pdf.
(105) McCabe, D.; Geertsma, M.; Matthews, N.; Fleischman, L.; Schroeder, D.. *Waste Not Common Sense Ways to Reduce Methane Pollution from the Oil and Natural Gas Industry*; Clean Air Task Force; Natural Resources Defense Council; Sierra Club: 2014; http://docs.nrdc.org/energy/files/ene_14111901b.pdf.

DOI: 10.1021/acs.energyfuels.5b01063
Energy Fuels 2015, 29, 6008−6018