Supporting Information for

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

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This document includes 38 pages, 11 figures, and 22 tables.

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S1. Assumptions - Fuel Properties and Emission Factors from Combustion

Table S1 summarizes properties (e.g., energy density and mass density) and emissions factors of energy carriers used in this study. There are two reported values for energy contents, LHV (lower heating value) and HHV (higher heating value). MacLean and Lave[(1)] suggested using LHV for mobile use (such as in vehicles) and HHV for stationary use (such as in power plants and fuel production plants). To be consistent, we use LHV for all energy sources in this study. Furthermore, we rely on the GREET model[(2)] for all properties of energy carriers to maintain consistency, even though there is a noticeable difference in the energy content (HHV) for dry natural gas used in existing studies: 1,089 BTU/ft^3 in the GREET model[(2)]; 1,030 BTU/ ft^3 in Jaramillo et al. (2007)[(3)], Venkatesh et al. (2001)[(4)], and Jiang et al. (2011)[(5)].

We calculate the combustion emission factor of natural gas based on Venkatesh et al. (2011)[(4)]. The combustion emission factor of natural gas reported in Venkatesh et al. (2011)[(4)] follows a normal distribution with a mean of 50 gCO_2-equiv/MJ_{HHV} and a standard deviation of 0.7 gCO_2-equiv/MJ_{HHV}. To convert it to a LHV-basis, we used the ratio of natural gas HHV (1,089 BTU/cf) and LHV (983 BTU/cf)[(2)].

Table S1. Energy content and emissions factors for different energy carriers.

Fuel	Energy density (Unit: BTU/cubic foot or BTU/gallon)	Mass density (Unit: gram/cubic foot or gram/gallon)	Combustion Emissions factor (Unit: gCO_2-equiv /MJ_{LHV})
Liquid Fuels (at 32F and 1atm)			
Conventional Gasoline	112,194	-	See main text
Oil sand gasoline		-	
Conventional Diesel	128,450	-	
Oil sand diesel		-	
Methanol	57,250	3,006	68.4[b]
Ethanol	76,330	2,988	70.9[b]
LPG	84,950	1,923	64.5[b]
Ethane	20,295 (Btu/lb)[a]	-	-
Butane	94,970	2,213	-
n-Hexane	105,125	2,479	-
Gaseous Fuels (at 32F and 1atm)			
Natural gas	983	22.0	Normal dist. (50, 0.7)/983*1089[c]
Pure methane	962	20.3	-
Gaseous hydrogen	290	2.6	-

Note: a. The energy density of ethane is used in modeling the ethane steam cracking process. And the source of the value is http://www.engineeringtoolbox.com/heating-values-fuel-gases-d_823.html. b. The combustion emission factor of methanol, ethanol, and LPG are calculated using energy density, mass density, and carbon weight ratio from the GREET model[(2)]. c. The HHV of natural gas is 1,089 BTU/cubic foot[(2)].

Generally speaking, as natural gas flows from the well site to end users, its methane composition increases due to various processing and purification processes. **Table S2** summarizes methane composition of four types of natural gas. We note, however, that the methane composition of natural gas varies by region[6], so a region-specific analysis may have slightly different results to those presented in this paper.

Table S2. Methane composition of natural gas.

Fuel	**Methane composition** (volume)	**Reference**
Natural gas (production)	0.894	U.S. EPA (2014)[6]
Natural gas (pipeline)	0.934	U.S. EPA (2014)[6]
CNG (Compressed Natural Gas)	0.934	Assumed to be the same as pipeline-quality natural gas
LNG (Liquefied Natural Gas)	0.95	Foss (2007)[7]

S2. Assumptions - Natural Gas Upstream and Fuel Productions

We discussed key assumptions related to natural gas upstream activities, fuel production and transport in the main text. Here we provide additional details that do not fit in the main text due to page limits.

Ethanol Production

While the production of ethanol has already transitioned to biomass-based pathways (such as corn grain, sugarcane, and cellulosic biomass[8]), ethanol was historically produced from fossil fuel-based naphtha and ethane. We consider ethane produced along with natural gas, and we assume that separation of ethane and other natural gas liquids takes place at natural gas processing plants. We perform an energy-based allocation to assign greenhouse gas (GHG) emission associated with natural gas preproduction, production, and processing to ethane.

There is a two-step process to produce ethanol from ethane, and both steps are well studied. The first step is ethane cracking, from which ethylene is produced[9]. We rely on Posen et al.[10] to model the ethane cracking process, as detailed in **Table S3**. The second step is catalytic ethylene hydration, where a mixture of gaseous steam water and gaseous ethylene react over phosphoric acid catalysts[11–13]. The conversion rate of the process is very low (around 4%) so the unreacted feedstock is recycled until the overall conversion rate is economically favorable (usually more than 95%)[14]. We model this ethylene hydration process using data from the Ecoinvent database[14], as shown in **Table S4**. According to the Ecoinvent database[14], the intermediate co-products (butane, and acetaldehyde) are burned as fuel and the final outputs of this process include ethanol and diethyl ether. The Ecoinvent database also suggests a mass-based emission allocation for these co-products (99.2% ethanol and 0.8% diethyl ether). The inputs of the process include ethane as a feedstock, as well as dry natural gas to provide steam and electricity. We assume that the steam boiler has an energy efficiency of 80%[2]. We also calculate carbon dioxide emissions from the process by performing the carbon balance between the inputs and outputs. The ethanol produced is anhydrous ethanol[14], so we do not account for a dehydration process of ethanol. After production, ethanol is transported to refueling stations. We assume that natural gas-based ethanol and methanol have the same transportation emission factor (per one unit energy transported), as listed in **Table S5**, because these two pathways are likely to operate with similar infrastructure.

Table S3. Ethylene steam cracking (ethylene production) profile[10].

Key parameter	Distribution parameters	Units
Specific Energy Required	Uniform (15,25)	GJ/ metric ton ethylene
Ethylene Produced	Triangular (764, 803, 840)	kg/metric ton ethane
Propylene Produced	Triangular (14.1, 16, 29.9)	
Butadiene Produced	Triangular (17.4, 19.9, 23)	
Aromatics Produced	Uniform (0, 19.9)	
Hydrogen Produced	Triangular (57.9, 60, 89.7)	
Methane Produced	Triangular (58.8, 61, 70.1)	
C4 Components Produced	Triangular (0, 6, 8.1)	
C5 and C6 Components Produced	Uniform (0, 26)	
Product Losses	Uniform (3, 20)	
Methane leakage	Triangular (5.45,6,6.6)	

Table S4. Ethylene hydration profile for one energy unit (MJ) of ethanol[14].

Key parameter	Distribution type	Distribution parameters
Molecular conversion rate	Uniform	(0.968, 0.971)
Total energy demand (MJ/kg ethanol produced)	Uniform	(2.85, 2.98)
Steam share	Uniform	(0.88, 0.98)
Electricity share	1 – share of steam as energy input	
Co-product: Ethylene	99.2% (by weight)	
Co-product: Diethyl ether	0.8% (by weight)	

Transport and Distribution of Liquid Fuels

All the natural gas pathways fall into two groups in terms of where the fuel is produced and where the fuel is pumped into vehicles (i.e. fueling stations): (1) distributed production pathways where alternative fuels are produced at distributed refueling stations (CNG and gaseous hydrogen from distributed productions (GH_2d)), and (2) centralized production pathways where alternative fuels are produced at centralized locations and then transported to refueling stations (E85, M85, Fischer-Tropsch liquids, GH_2 central, LH_2 central, and electricity).

For distributed pathways, emissions related to delivering natural gas to refueling stations are included in the natural gas upstream emissions. For centralized pathways (as well as gasoline pathways), the final fuels are transported either as electricity or liquids. We assumed a 6.5% loss[(2)] in electricity transmission. For other fuel pathways, we rely on the emission factors reported in the GREET model (version 2013)[(2,15)]. We summarize transportation emission factors in **Table S5**. The GREET model [(2)] does not have a natural gas-based ethanol pathway or a Fischer-Tropsch gasoline pathway, so we made two further assumptions: (1) the ethanol pathway has the same transportation emission factor as methanol; (2) Fischer-Tropsch gasoline has the same transportation emission factor as Fischer-Tropsch diesel.

Table S5. Emission factors of fuel transport[(2)] (Unit: gram/MJ_{LHV}).

Fuel pathways	Fischer-Tropsch Gasoline/Diesel	GH_2 central	LH_2 central	Methanol/Ethanol
CO_2	1.0	4.8	0.6	1.7
CH_4	0.002	0.014	0.001	0.003
N_2O	1×10^{-5}	6×10^{-5}	9×10^{-5}	3×10^{-5}

S3. Assumptions: Vehicles

Fuel Economy

For this analysis, we model new vehicles available on the market rather than existing fleets. We use functionally-equivalent vehicles for different fuel pathways for two vehicle types, a compact passenger vehicle and a compact Sports Utility Vehicle (SUV), to eliminate bias[(1)]. We use the official fuel economy estimates published by the U.S. Department of Energy (DOE) and the U.S. Environmental Protection Agency (EPA)[(16)], who test all vehicles on the same duty cycle. There are currently no methanol vehicles in the market, so we rely on the literature for fuel economy estimates. We also rely on the literature to model “standardized” plug-in hybrid vehicle (PHEVs) - PHEV30 and PHEV60 that have 30 or 60 km of all electric range (AER) - since PHEVs offered by vehicle manufacturers differ in their AERs. **Table S6** and **Table S7** summarize fuel economy assumptions, and representative vehicle models as well as their specifications (engine, transmission, range, and weight) for passenger vehicles and SUVs included in this analysis.

Table S6. Passenger Vehicle Fuel Economy Assumptions.

Pathway		Representative vehicle	Source	MPGge[1]	Range (km)
Gasoline vehicle (baseline)		2015 Honda Civic (1.8 L, 4 cyl, Automatic (variable gear ratios))	fueleconomy.gov(16)	33	702
Diesel vehicle		2015 BMW 328d (A-S8, 2.0 L, 4cyl)	fueleconomy.gov(16)	32.3[2]	840
Gasoline hybrid electric vehicle (HEV)		2015 Honda Civic Hybrid (1.5 L, 4 cyl, Automatic (variable gear ratios))	fueleconomy.gov(16)	45	956
PHEV30	CS*	Karabasoglu et al. (2013)(17)	Karabasoglu et al. (2013)(17)	43.8	-
	CD*			112	30
PHEV60	CS*		Karabasoglu et al. (2013)(17); weight-fuel economy relationship from Shiau et al. (2009)(18).	42.4	-
	CD*			105	60
Battery Electric Vehicle (BEV) 130[3]		2015 Ford Focus Electric	fueleconomy.gov(16)	105	122
		2015 Nissan Leaf	fueleconomy.gov(16)	114	135
		Our assumptions	-	110	-
CNG dedicated		2015 Honda Civic Natural Gas (1.8 L, 4 cyl, Automatic 5-spd)	fueleconomy.gov(16)	31	311
M85 dedicated		N/A	GREET 2013(2) (7% more energy efficient than conventional gasoline vehicle)	35.3	-
E85 flex fuel vehicle (FFV)		2015 Ford Focus FWD FFV (2.0 L, 4 cyl, Auto (AM6))	fueleconomy.gov(16)	31.6[4]	459
Hydrogen fuel cell electric vehicle (FCEV)		2014 Honda Clarity FCX	fueleconomy.gov(16)	59	372

* CS stands for Charge-Sustaining, and CD stands for Charge-Deleting.

[1] MPGge, miles per gallon of gasoline equivalent.
[2] The fuel economy of BMW 328d is 37 MPG, which is equivalent to 32.3 MPGge using the energy intensity of diesel and gasoline.
[3] Since Honda Civic does not have an all-electric version, we use a comparable BEVs from other manufacturers. The Nissan Leaf has an AER of 84 miles with a 24 kWh battery and the Ford Focus Electric has an AER of 76 miles and a 23kWh battery (from fueleconomy.gov). While the Nissan Leaf and the Ford Focus have roughly the same all-electric range, their equivalent fuel economy differs by 10%. To be more conservative, we assume that the representative BEV has the average parameters of Nissan Leaf and Ford Focus Electric, i.e. 100 MPG with an AER of 130 km (80 miles) and a 24 kWh battery.
[4] The fuel economy of Ford Focus FFV is 23 MPG, which is equivalent to 31.6 MPGge using the energy intensity of E85 and gasoline. Note that the GREET model (versions 2013)(2) achieves that FFV has the same MPGge as conventional gasoline vehicle.

Table S7. Sports Utility Vehicle Assumptions. All vehicle economy and range estimates are taken from fueleconomy.gov[16].

Pathway		Representative vehicle	Specifications	MPGge[5]	Range (km)	GVW (lbs.)
Gasoline vehicle (baseline)	Marketed vehicles	2015 Hyundai Tuscon 2WD, 2015 BMW X3 xDrive28i, 2015 Toyota RAV4 2015 Lexus NX 200t	2.0 L, 4 cyl, Automatic 6-spd 2.0 L, 4 cyl, Automatic (S8), Turbo 2.5 L, 4 cyl, Automatic (S6) 2.0 L, 4 cyl, Automatic (S6), Turbo	25 24 26 25	615 665 684 641	3294 4150 3700 3940
	Our Assumption	N/A	N/A	25	N/A	N/A
Diesel vehicle		2015 BMW X3 xDrive 28d	2.0 L, 4 cyl, Automatic (S8), Turbo	26.2[6]	855	4230
Gasoline HEV		2015 Lexus NX 300h	2.5 L, 4 cyl, Automatic (S6)	33.0	785	4055
E85 flex fuel vehicle		2015 Chevrolet Captiva FWD	2.4 L, 4 cyl, Automatic 6-spd	24.7[7]	563	3801
Hydrogen fuel cell vehicle		2015 Hyundai Tucson Fuel Cell	Fuel cell power (max): 100 kW	49.0	426	-
BEV165		2014 Toyota RAV4 EV	Automatic (variable gear ratios)	76.0	166	4032

Tailpipe Methane and N_2O Emissions

Table S8 summarizes tailpipe CH_4 and N_2O emissions from different vehicle technologies. The main text explains key assumptions and data sources.

Table S8: Tailpipe methane emissions of light-duty vehicles (LDV)[2].

GHG	Absolute values (Unit: g/km)		Relative values (percentage as gasoline pathway emissions)				
	Gasoline	Diesel	CNG	M85	HEV	PHEV	BEV / FECV
CH_4	0.014	0.006	1,000%	100%	47%	47%	0%
N_2O	0.007	0.007	100%	100%	100%	100%	0%

PHEV-specific Assumptions

This analysis includes two types of PHEVs for passenger vehicles: PHEV30 and PHEV60 with an AER of 30 and 60 kilometers, respectively. The operation of PHEVs is categorized into two modes depending on the battery state of charge (SOC): charge-depleting (CD) mode, in which the vehicle receives some or

[5] MPGge, miles per gallon of gasoline equivalent.
[6] The fuel economy of 2015 BMW X3 xDrive 28d given at fueleconomy.gov is 30.0 MPG. We have converted the number to MPGge using the energy intensity of diesel and gasoline.
[7] The fuel economy of 2015 Chevrolet Captiva FWD given at fueleconomy.gov is 18.0 MPG. We have converted the number to MPGge using the energy intensity of E85 and gasoline.

all of its propulsion energy from the battery; and charge-sustaining (CS) mode, in which gasoline provides all propulsion energy[17]. There are two control strategies for the CD mode, all-electric control (or extended-range) and blended control, which differ if the CD mode uses any non-electric energy sources (such as gasoline)[17–20]. For simplicity, we assume an all-electric control strategy for the CD mode, which is used first until the battery is depleted to predefined SOC.

Following Samaras et al.[19], we approximate the fractions of vehicle trips powered by electricity and gasoline using National Household Travel Survey (NHTS) 2009[21]. We assume that the fraction of a PHEV (with an AER X km)'s electric drive equals the probability of daily vehicle kilometer traveled less than X km. Furthermore, we assume PHEVs are charged once every day (in the night). **Figure S1** shows the calculated cumulative distribution, while **Table S9** shows the selected fractions of electric drive for different types of PHEVs. For PHEV30, this electric range share is 0.44, while for PHEV60 it is 0.68 (**Table S9**).

We use a probability mixture model to combine the GHG emissions from CD and CS mode (i.e. electric and gasoline drives). We rely on Karabasoglu et al.[17] for the technical specifications of the PHEVs examined. Shiau et al.[18] reported that the additional battery weight associated with increasing the All Electric Range (AER) by 10 mile reduces CD-mode and CS-mode efficiencies by 0.10 mile/kWh and 0.68 MPGge (mile per gallon gasoline equivalent), respectively. Thus, we adjust the fuel economy of the PHEV60, and assume the battery weight effects are accounted for in the fuel economy estimates of HEV and PHEV30.

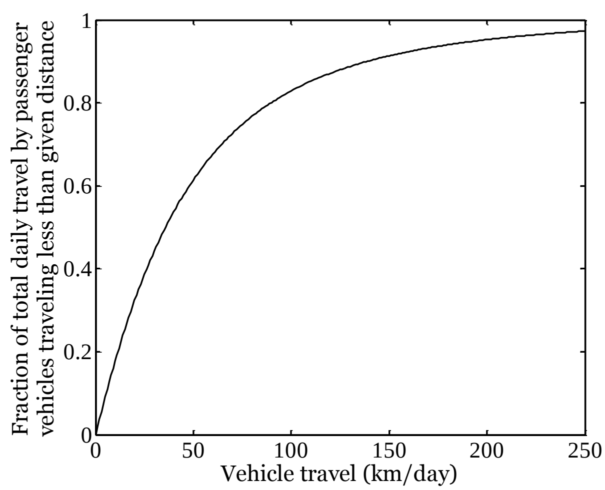

Figure S1. Cumulative distribution of daily passenger vehicle travel (km/day). The distribution is constructed with data from the National Household Travel Survey 2009[21].

Table S9. Fraction of vehicle kilometers powered by electricity in PHEVs if PHEVs are charged overnight (based on Figure S1).

Range	(mile)				(km)			
	20	30	40	60	20	30	60	90
Probability	0.47	0.60	0.70	0.82	0.32	0.44	0.68	0.80

Vehicle Manufacturing Emissions

We include cradle-to-gate vehicle manufacturing emissions in the system boundary. **Table S10** summarizes relevant assumptions used to calculate a vehicle's emissions. The main text explains key assumptions and data sources.

Table S10. Vehicle manufacturing emissions.

Application	Technology	Battery/Fuel cell power plant (FCPP) type and size	Numbers of batteries/FCPP per vehicle lifetime
Hybrid and plug-in electric vehicles			
Passenger Vehicle	Gasoline HEV	1.3 kWh Li-Ion battery	1[2,17,19]
	PHEV30	9.9 kWh Li-Ion battery	
	PHEV60	19.9 kWh Li-Ion battery	
	BEV130	24.0 kWh Li-Ion battery	
SUV	Gasoline HEV	1.6 kWh Ni-Mh battery	
	BEV165	41.8 kWh Li-Ion battery	
Fuel cell electric vehicles (FCEV)			
Passenger vehicle	FCEV	100 kW fuel cell + Li-ion battery[22]	1[2]
SUV	FCEV	100 kW fuel cell	
Vehicle lifetime travel distance			
LDV (passenger vehicles & SUVs)		150,000 miles, or 240,000 km[17,19]	
Vehicle manufacturing emission factors			
Passenger vehicle	Internal Combustion Engine Vehicle (ICEV)	[CO_2, CH_4, N_2O] = [45.2, 0.1, 0.011] gram/mile[2]	
	PHEV/BEV	Calculated as the sum of vehicle manufacturing emissions (same as the ICEV) and additional battery manufacturing emissions	
	FCEV	[CO_2, CH_4, N_2O] = [89.3, 0.2, 0.002] gram/mile[2]	
SUV	ICEV	[CO_2, CH_4, N_2O] = [55.4, 0.2, 0.001] gram/mile[2]	
	PHEV/BEV	Calculated as the sum of vehicle manufacturing emissions (same as the ICEV) and additional battery manufacturing emissions	
	FCEV	[CO_2, CH_4, N_2O] = [102.2, 0.2, 0.002] gram/mile[2]	
Battery manufacturing emission factors			
Battery manufacturing emission factors		5.1 kg CO_2-equiv/kg[23] (Li-Ion battery)	
Battery specific energy	HEV	0.11 kWh/kg of battery[23]	
	BEV	0.13 kWh/kg of battery[23]	

S4. Break-Even Methane Leakage Rate Analysis

We derive the closed-form formulas to estimate the break-even life cycle methane leakage rate with respect to relative vehicle fuel economy (normalized to that of a conventional gasoline vehicle), global warming potential (GWP), and the baseline fuel choice (conventional gasoline). We estimate the break-even rate for three pathways: CNG, distributed gaseous hydrogen FCEV, and NGCC (Natural Gas Combined Cycle Power Plants) electricity BEV pathways. The system boundary is the same for the break-even analysis and the Monte-Carlo simulations.

$$\begin{aligned}&\textit{gasoline life cycle emissions}\left[gCO_2e/\mathrm{km}\right]\\&=\frac{\textit{gasoline life cycle GHG emissions}\left[gCO_2e/MJ\right]}{\textit{energy efficiency}_{\textit{gasoline vehicle}}\ [\mathrm{km}/MJ]}+\textit{vehicle manufacturing}_{\text{gasoline vehicle}}\left[gCO_2e/\mathrm{km}\right]\end{aligned}\qquad(1)$$

Equation (1) calculates the life cycle emissions for conventional gasoline vehicles. Here the life cycle include upstream, combustion, and manufacturing emissions but exclude tailpipe methane and N_2O emissions, both of which contribute little to the life cycle emissions. We include the unit in the bracket in equations to help readers understand the unit conversions.

$$\begin{aligned}&\textit{CNG life cycle emissions}\\&=\frac{\textit{CNG life cycle } CO_2 \textit{ emissions}}{\textit{efficiency}_{CNGV}}+\frac{\textit{CNG life cycle methane emissions}}{\textit{efficiency}_{CNGV}}\times GWP_{methane}+\textit{vehicle manufacturing}_{CNGV}\end{aligned}\qquad(2)$$

Equation (2) calculates the life cycle emissions for the CNG pathway (with a dedicated CNG vehicle). Here we split the use-related life cycle emissions into two parts, CO_2 emissions and methane emissions. We the calculate CO_2 emissions and methane emissions as sum of emissions from natural gas upstream, compression, and vehicle tailpipe. Later we will represent methane emissions as a function of leakage rate (percentage of natural gas lost into the atmosphere) and then back calculate the break-even methane leakage rate.

$$\begin{aligned}&\textit{BEV life cycle GHG emissions}\\&=\textit{BEV life cycle } CO_2 \textit{ emissions}+\textit{BEV life cycle methane emissions}+\textit{vehicle manufacturing}_{BEV}\\&=\frac{\textit{Natural gas upstream } CO_2 \textit{ emissions}+\textit{Natural gas combustion } CO_2 \textit{ emissions}}{\textit{efficiency}_{power\ plant}\times\left(1-loss_{transmission\ line}\right)\times \textit{efficiency}_{charging}\times \textit{efficiency}_{BEV}}+\\&\quad\frac{\textit{Natural gas life cycle } CH_4 \textit{ emissions}}{\textit{efficiency}_{power\ plant}\times\left(1-loss_{transmission\ line}\right)\times \textit{efficiency}_{charging}\times \textit{efficiency}_{BEV}}\times GWP_{methane}+\textit{vehicle manufacturing}_{BEV}\end{aligned}\qquad(3)$$

Equation (3) calculates the life cycle emissions for NGCC-electricity-BEV pathway. Again, we split the use-related life cycle emissions into CO_2 and methane emissions. The pathway of generating electricity through NGCC power plants is quite straightforward; natural gas is produced and transported to NGCC power plants (natural gas upstream emissions), then combusted to generate electricity, then transmitted to charging stations where electricity is used to power a vehicle.

$$
\begin{aligned}
& Gaseous\ hydrogen\ FCEV\ life\ cycle\ GHG\ emissions \\
&= GH_2d\ life\ cycle\ emissions\ (except\ natural\ gas\ feedstock) + \\
&\quad Natural\ gas\ feedstock\ for\ hydrogen\ production\ emissions + vehicle\ manufacturing_{FCEV} \\
&= \frac{GH_2d\ life\ cycle\ emissions\ (except\ natural\ gas\ feedstock)}{efficiency_{FCEV}} + \\
&\quad \left(\frac{Natural\ gas\ upstream\ CO_2\ emissions}{efficiency_{FCEV}} + \frac{Natural\ gas\ upstream\ methane\ emissions}{efficiency_{FCEV}}\right) \times Hydrogen\ plant\ feedstock_{natural\ gas} \\
&\quad + vehicle\ manufacturing_{FCEV} \\
&= \frac{GH_2d\ life\ cycle\ emissions\ (except\ natural\ gas\ feedstock)}{efficiency_{FCEV}} + \\
&\quad \frac{Natural\ gas\ upstream\ CO_2\ emissions}{efficiency_{FCEV}} \times Hydrogen\ plant\ feedstock_{natural\ gas} + \\
&\quad \frac{life\ cycle\ methane\ emissions}{efficiency_{FCEV}} \times Hydrogen\ plant\ feedstock_{natural\ gas} + vehicle\ manufacturing_{FCEV}
\end{aligned}
\tag{4}
$$

Equation (4) calculates the life cycle emissions for a gaseous hydrogen distributed FCEV pathway. Because hydrogen production plants involve multiple inputs and outputs, we split the hydrogen life cycle emissions in a different way. The life cycle emissions of the gaseous hydrogen pathway include emissions associated with the natural gas feedstock (only natural gas upstream emissions), emissions associated with electricity input (assumed to be grid-average electricity), hydrogen production process emissions, and hydrogen compression emissions.

In Equations (1) through (4), we can further represent vehicle energy efficiency and methane emissions using Equation (5) and (6), respectively. In Equation (5), fuel economy is in mile per gallon of gasoline equivalent (MPGge), as shown in **Table S6** and **Table S7**. In Equation (6), methane emissions are calculated as a function of methane leakage rate (the percentage of produced natural gas leaked into the atmosphere).

$$
energy\ efficiency_{vehicle}\left[{km}/{MJ}\right] = \frac{fuel\ economy_{vehicle}\left[{mile}/{gge}\right]}{energy\ intensity_{gasoline}\left[{MJ}/{gallon}\right] \times conversion\left[{mile}/{km}\right]} \tag{5}
$$

$$
\begin{aligned}
& methane\ emission\left[{gCO_2e}/{MJ}\right] \\
&= \frac{methane\ leakage\ rate \times natural\ gas\ composition_{methane} \times methane\ density\left[{gram}/{cf}\right] \times GWP_{methane}\left[{gCO_2e}/{gram}\right]}{energy\ intensity_{Natural\ gas}\left[{MJ}/{cf}\right]}
\end{aligned}
\tag{6}
$$

The break-even methane leakage rate is defined as the methane leakage rate at which natural gas-based fuels' life cycle emissions equal conventional gasoline's life cycle emissions. To calculate the break-even methane leakage rate, we equaled Equation (1) with Equation (2-4), re-arranged terms, and reached the following formulas (7-9) for the three natural gas pathways:

$$\begin{aligned}&\textit{Break-even methane leakage rate}_{CNG}\\&=\left(EER_{CNG\ vehicle}\times \textit{gasoline life cycle GHG emissions} - \textit{CNG life cycle } CO_2 \textit{ emissions}\right)\times\frac{\textit{energy density}_{methane}}{GWP_{methane}}\end{aligned} \tag{7}$$

$$\begin{aligned}&\textit{Break-even methane leakage rate}_{NGCC-BEV}\\&=\begin{bmatrix}EER_{BEV}\times EF_{natural\ gas\text{-}electricity}\times\left(\textit{gasoline life cycle GHG emissions} - \Delta \textit{vehicle manufacturing}_{BEV}\times \textit{efficiency}_{gasoline\ vehicle}\right)\\ -\left(\textit{Natural gas upstream } CO_2 \textit{ emissions} + \textit{Natural gas combustion } CO_2 \textit{ emissions}\right)\end{bmatrix}\\&\times\frac{\textit{energy density}_{methane}}{GWP_{methane}}\\&\textit{where } EF_{natural\ gas\text{-}electricity} \doteq \textit{efficiency}_{power\ plant}\times\left(1-loss_{transmission\ line}\right)\times \textit{efficiency}_{charging}\end{aligned} \tag{8}$$

$$\begin{aligned}&\textit{Break-even methane leakage rate}_{GH_2d\text{-}FCEV}\\&=\begin{bmatrix}\frac{EF_{natural\ gas\text{-}electricity}}{\textit{Hydrogen plant feedstock}_{natural\ gas}}\times\left(\textit{gasoline life cycle GHG emissions} - \Delta \textit{vehicle manufacturing}_{BEV}\times \textit{efficiency}_{gasoline\ vehicle}\right)\\ -\frac{1}{\textit{Hydrogen plant feedstock}_{natural\ gas}}\times GH_2d \textit{ life cycle emissions (except natural gas feedstock)} - \textit{Natural gas upstream } CO_2 \textit{ emissions}\end{bmatrix}\\&\times\frac{\textit{energy density}_{methane}}{GWP_{methane}}\end{aligned} \tag{9}$$

Here we define the energy economy ratio (EER) as the ratio between a vehicle's gasoline-equivalent fuel economy to another vehicle's gasoline-equivalent fuel economy

$$\textit{Energy Economy Ratio }(EER)_{vehicle\ A} = \frac{\textit{fuel economy}_{vehicle\ A,\ gasoline\ equivalent}}{\textit{fuel economy}_{gasoline\ vehicle}} \tag{10}$$

and the energy density of methane can be calculated as

$$\textit{energy density}_{methane}\left[{MJ}/{gram}\right] = \frac{\textit{energy intensity}_{Natural\ gas}\left[{MJ}/{cf}\right]}{\textit{natural gas composition}_{methane}\times \textit{methane density}\left[{gram}/{cf}\right]} \tag{11}$$

Equations (7)-(11) give the exact formulas to calculate the break-even methane leakage rates for the selected natural gas pathways. All the inputs used to solve these equations are from the Monte-Carlo simulation model but we use the average estimates instead of distributions. Some of the key inputs are as follows.

- Methane density is 20.3 g/ft^3, and a cubic foot (ft^3) of natural gas has 93.4% methane on average. The energy density of natural gas is 1.037 MJ/ ft^3 (or 983 BTU/ ft^3). The energy intensity of gasoline is 118.4 MJ_{LHV}/gallon or 112,194 BTU/gallon.

- The conventional ICEV vehicle has a fuel economy of 33 MPG and an energy efficiency of 0.28. Conventional gasoline has life cycle GHG emissions of 91.5 gCO_2-equiv/MJ_{LHV} (for both 20-year and 100-year time periods).
- CO_2 emissions from natural gas upstream and combustion are 7.99 gCO_2-equiv/MJ_{LHV} and 55.39 gCO_2-equiv/MJ_{LHV}.
- The energy efficiency of NGCC power plants is 55.7% (LHV basis), the average transmission and distribution line loss is 6.5%, and the average BEV charging efficiency is 86.5%. In other words, the overall energy efficiency from the delivery of natural gas at the front gate of the NGCC power plant to the electricity in the BEV is 45%.
- The input of natural gas feedstock for a hydrogen production plant is 1.3 MJ per MJ of hydrogen produced. The life cycle emissions of hydrogen include the cradle-to-bus-bar emissions of the natural gas input and electricity input (13.2 gCO_2-equiv/MJ_{LHV}), emission factors of hydrogen production plant (77.1 gCO_2-equiv/MJ_{LHV}), and hydrogen compression emissions (14.8 gCO_2-equiv/MJ_{LHV}), which brings the subtotal to be 127.6 gCO_2-equiv/MJ_{LHV}.
- While all the above assumptions are fuel-specific and are transparent to vehicle types, vehicle manufacturing emissions are different for passenger vehicles and SUVs. For passenger vehicles, compared to conventional ICEVs, the additional vehicle manufacturing emissions of a BEV (with a battery size of 24 kWh) are 6.3 gCO_2-equiv/mile and the additional vehicle manufacturing emissions of a FCEV are 48.1 gCO_2-equiv/mile.

With all these assumptions and values, the break-even formulas can be simplified to the following expressions, where the break-even methane leakage rates are only dependent on the EER and the GWP.

$$\begin{aligned}
&Break\text{-}even\ methane\ leakage\ rate_{CNGV} = \left(91.5 \times EER_{CNGV} - 71.0\right) \times \frac{0.055}{GWP_{methane}} \\
&Break\text{-}even\ methane\ leakage\ rate_{NGCC-BEV} = \left(40.4 \times EER_{BEV} - 63.4\right) \times \frac{0.055}{GWP_{methane}} \\
&Break\text{-}even\ methane\ leakage\ rate_{GH_2d\text{-}FCEV} = \left(59.3 \times EER_{FCEV} - 87.5\right) \times \frac{0.055}{GWP_{methane}}
\end{aligned} \tag{12}$$

We plot the break-even methane leakage rates with regard to EERs in the main text, and calculate the break-even methane leakage rates for selected EERs in the **Table S11**. Note that current vehicle technologies have an EER of 94%, 179%, and 333% for a CNGV, a FCEV, and a BEV, respectively.

Table S11. Break-even methane leakage rates for natural gas pathways (the baseline fuel pathway is conventional gasoline).

EER	CNG		NGCC-BEV		GH_2d-FCEV	
	100-year GWP	20-year GWP	100-year GWP	20-year GWP	100-year GWP	20-year GWP
90%	1.7%	0.7%	-	-	-	-
95%	2.4%	1.0%	-	-	-	-
100%	3.1%	1.3%	-	-	-	-
110%	4.5%	1.9%	-	-	-	-
120%	5.9%	2.4%	-	-	-	-
150%	-	-	-	-	0.2%	0.1%
175%	-	-	1.1%	0.5%	2.5%	1.0%
200%	-	-	2.6%	1.1%	4.7%	2.0%
225%	-	-	4.2%	1.7%	7.0%	2.9%
250%	-	-	5.7%	2.4%	9.2%	3.8%
275%	-	-	7.2%	3.0%	11.5%	4.8%
300%	-	-	8.8%	3.6%	13.7%	5.7%
325%	-	-	10.3%	4.3%	-	-
350%	-	-	11.9%	4.9%	-	-
375%	-	-	13.4%	5.5%	-	-
400%	-	-	14.9%	6.2%	-	-

S5. Additional Results

Natural Gas Upstream GHG Emissions

Here we provide detailed results with breakdowns by GHGs and by processes in each upstream stage (**Table S12**, **Table S13**, **Figure S2** and **Figure S3**). In accordance with the main text, we consider four scenarios: baseline methane estimate with 100-year GWP; baseline methane estimate with 20-year GWP; pessimistic methane estimate with 100-year GWP; and pessimistic methane estimate with 20-year GWP. Our estimates show that there is a very large uncertainty range of natural gas upstream GHG emissions. The mean upstream total GHG emission is 17.4 gCO_2-equiv/MJ_{LHV} and the 95% confidence interval is 10.3-29.5 gCO_2-equiv/MJ_{LHV}. We find that the distribution of total upstream GHG emissions is highly asymmetrical. In addition, we fit distributions to the GHG emissions from the natural gas system (the functional unit is one mega joule of natural gas delivered at the end of distribution pipelines) in **Table S12.** We chose the fitted distribution[8] based on maximizing the negative of the log likelihood, Bayesian information criterion (BIC), and Akaike information criterion (AIC).

In the breakdown of upstream stages, well-site production (including both preproduction and production) and pipeline transportation (including transmission and distribution) contribute most to GHG emissions

[8] Candidate distributions include Beta, Birnbaum-Saunders, Exponential, Extreme Value, Gamma, Generalized Extreme Value, Generalized Pareto, Inverse Gaussian, Logistic, Log Logistic, Lognormal, Nakagami, Normal, Rayleigh, Rician, t location-scale, and, Weibull.

and all these stages have large methane emissions. By comparison, natural gas processing is responsible for a much lower share of GHG emissions and only 79% of produced natural gas has to be processed at processing plant[24,25].

We find that the relative contribution from methane is higher than carbon dioxide (**Figure S2** and **Figure S3**). For the baseline scenario, methane contributes slightly more than carbon dioxide. For the 20-year GWP scenario, the contribution of methane is nearly two times more than carbon dioxide. While carbon dioxide is quite evenly distributed across natural gas upstream stages (mostly as result of fuel combustion), methane emissions are more concentrated. Most methane emissions occur either at the well site (liquid unloading, well completion and well workover), or in the pipeline system (fugitive emissions). In addition, there is evidence that a small share of super-emitters is responsible for a larger share of GHG emissions (as summarized in Brandt et al.[26] and reflected in the right-skewed distribution shown in this study).

Existing studies of natural gas GHG emissions span a wide range with a 95% uncertainty range as of 11.0-21.0 gCO_2-equiv/MJ_{LHV} (compiled using estimates from six individual bottom-up studies)[27]. While our baseline estimate (mean value) aligns well with existing studies, there are two differences. First, we find smaller emissions from natural gas preproduction, production, and processing stages[9] while larger emissions from natural gas pipeline systems compared to most existing studies (Howarth et al.[28], is an exception). Second, we find a lower methane leakage rate from natural gas systems and higher carbon dioxide emissions. We think these two observations are partially the results of reduced methane emissions in well completions and well workovers that have been the result of better industry practices and stringent regulation on well completions.

[9] See Table SI-5 in the Supporting Information of Weber et al. (2012)[27] for a summary of GHG emissions from preproduction, production & processing, and transmission stages.

Table S12. Natural gas upstream emissions with breakdown of upstream stages and GHGs. Both 100-year and 20-year GWP estimation results are shown as well as the pessimistic case (methane emissions are multiplied by 1.5). Mean estimate and 95% confidence interval (in parenthesis) are shown in table entries.

Stage	GHG emissions breakdown		Total GHG emissions			
	CO_2 (baseline)	CH_4 (baseline)	100-year GWP (baseline)	100-year GWP (pessimistic)	20-year GWP (baseline)	20-year GWP (pessimistic)
Unit	gram/MJ_{LHV}		gCO$_2$-equiv/MJ_{LHV}			
Pre-production	1.3 (0.5-3.1)	0.006 (0-0.05)	1.5 (0.6-4.2)	1.7 (0.6-4.9)	1.9 (0.6-6.3)	2.2 (0.6-8.2)
Production	2.6 (0.7-7.3)	0.09 (0.06-0.23)	6.0 (2.8-13.3)	7.7 (3.7-17.1)	10.7 (5.7-24.1)	14.8 (7.9-34.2)
Processing	2.2 (0.3-8.6)	0.008 (0-0.05)	2.5 (0.4-9.2)	2.7 (0.4-9.6)	2.9 (0.5-10.5)	3.3 (0.5-12)
Transmission	1.8 (0.4-6.1)	0.09 (0.08-0.11)	5.2 (2.7-9.6)	6.9 (3.7-11.7)	9.9 (6.2-15.2)	14 (8.8-20.7)
Distribution	0.002 (0.001-0.002)	0.06 (0.05-0.07)	2.0 (1.1-3.1)	3.0 (1.6-4.7)	4.9 (3.0-7.1)	7.3 (4.5-10.7)
Upstream total	8.0 (3.6-16.9)	0.26 (0.20-0.43)	17.2 (10.2-29.3)	22.0 (12.9-36.7)	30.3 (19.3-49.7)	41.7 (26.3-68.8)
Fitted distribution for upstream total	Generalized extreme value (‘shape’=0.13, ‘scale’=2.28, ‘location’ = 6.33)	Generalized extreme value (‘shape’=0.30, ‘scale’=0.024, ‘location’ = 0.23)	Log logistic (‘log location’ = 2.80, ‘log scale’ = ‘0.15’)	Log logistic (‘log location’ = 3.37, ‘log scale’ = ‘0.13’)	Log logistic (‘log location’ = 3.05, ‘log scale’ = ‘0.14’)	Log logistic (‘log location’ = 3.69, ‘log scale’ = ‘0.13’)

Table S13. Natural gas upstream emissions with the breakdown of processes in each upstream stage (Unit: gCO_2-equiv/MJ_{LHV}). Both 100-year and 20-year GWP estimation results are shown as well as the pessimistic case (methane emissions are multiplied by 1.5). Mean estimate and 95% confidence interval (in parenthesis) are shown in table entries.

Stage	Process	100-year GWP (baseline)	100-year GWP (pessimistic)	20-year GWP (baseline)	20-year GWP (pessimistic)
Pre-production	Wellpad Construction	0.2 (0.1-0.6)			
	Well Drilling	0.4 (0.1-1.0)			
	Hydraulic Fracturing	0.6 (0.2-1.5)			
	Well Completion	0.3 (0.0-2.4)	0.4 (0.0-3.2)	0.7 (0.0-4.8)	0.9 (0-6.8)
Production	Lease Fuel Use	2.0 (0.4-6.2)			
	Flaring	0.6 (0.0-2.8)	0.7 (0.1-3.1)	0.8 (0.1-3.7)	0.9 (0.1-4.4)
	Liquid Unloading	0.7 (0.0-4.7)	1.0 (0.0-7.0)	1.6 (0.0-11.2)	2.4 (0-16.9)
	Well Workover	0.3 (0.0-2.5)	0.4 (0.0-3.4)	0.7 (0.0-5.0)	0.9 (0-7.1)
	Other Fugitive Emissions	2.4 (1.2-3.6)	3.5 (1.8-5.5)	5.7 (3.5-8.2)	8.6 (5.3-12.4)
Processing	CO_2	2.2 (0.3-8.6)			
	CH_4	0.3 (0.0-1.8)	0.4 (0.0-2.7)	0.7 (0.0-4.3)	1.1 (0-6.4)
	N_2O	< 0.01			
Transmission	Fuel Use – Natural gas	1.7 (0.3-6.0)			
	Fuel Use – Electricity	0.1 (0.1-0.1)			
	Fugitive Emissions	3.4 (1.7-5.2)	5.1 (2.6-7.8)	8.1 (5.0-11.8)	12.2 (7.5-17.7)
Distribution	Fugitive Emissions	2.0 (1.1-3.1)	3.0 (1.6-4.7)	4.9 (3.0-7.1)	7.3 (4.5-10.7)
Upstream total emissions		17.4 (10.3-29.5)	22.0 (12.9-36.7)	30.3 (19.3-49.7)	41.7 (26.3-68.8)
Implicit methane leakage rate		1.3% (1.0%-2.2%)		2.0% (1.6%-3.3%)	

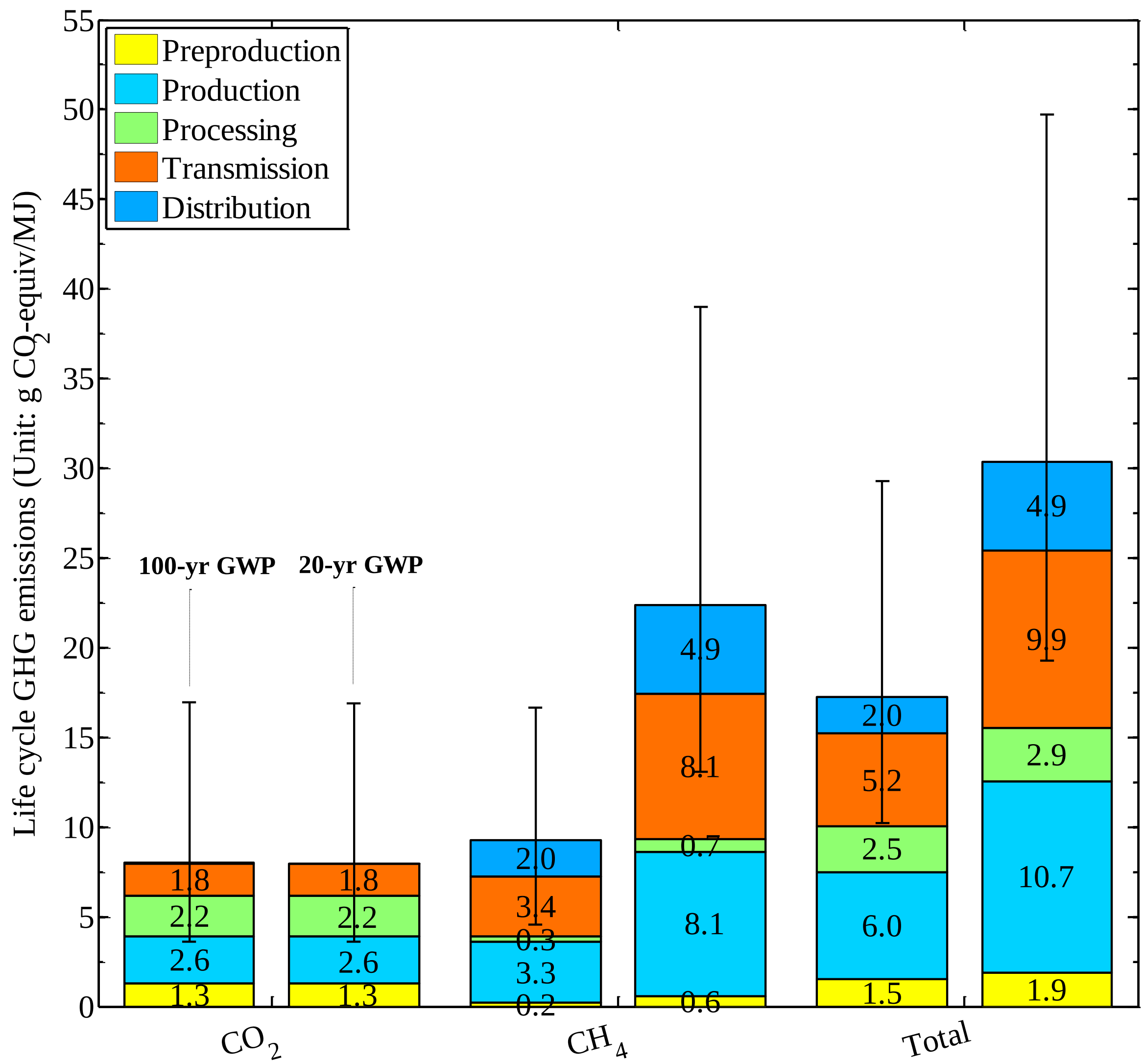

Figure S2. Breakdown of natural gas upstream GHG emissions by greenhouse gas and by upstream stages. Error bar are based on the 95 percent confidence interval of the total emissions for each GHG. Estimation results with 100-year GWP (left bars) and 20-year GWP (right bars) are shown side by side.

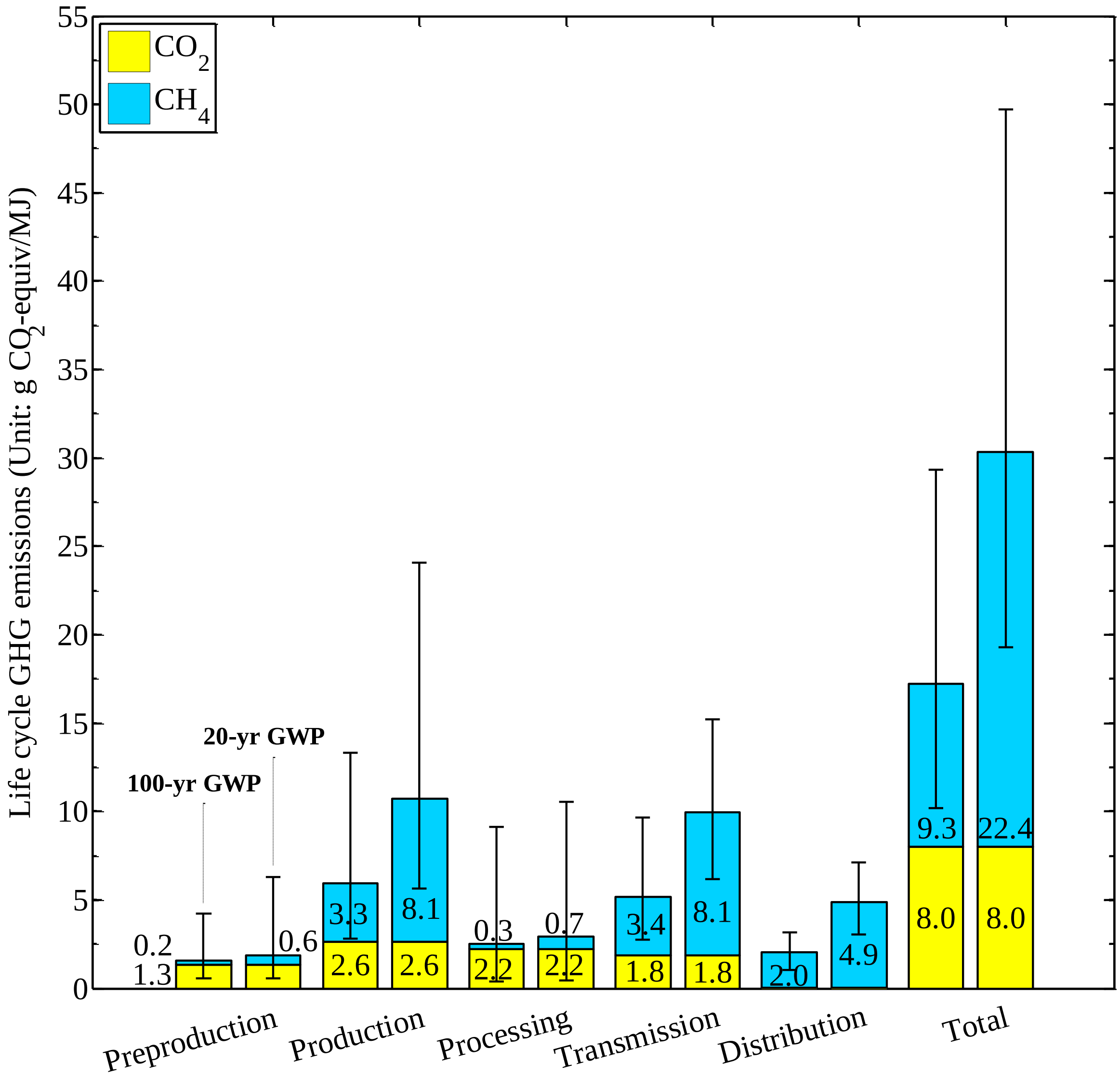

Figure S3. Breakdown of natural gas upstream GHG emissions by upstream stages and by greenhouse gases. Error bars are based on the 95 percent confidence interval of the total emissions for each GHG. Estimation results with 100-year GWP (left bars) and 20-year GWP (right bars) are shown side by side.

Carbon Intensity of Fuel Pathways

Figure S4 and **Figure S5** shows the carbon intensity of all fuel pathways considered in this study. The figure shows the breakdown of GHG emissions from upstream activities (well-to-pump) and fuel combustion. **Table S14** summarizes the carbon intensity of power generation in a different unit. A spreadsheet Supplemental Data file is available online and includes numbers behind the figures. While fuel carbon intensity is of interest to policymakers (for instance, California sets the Low Carbon Fuel Standards for transportation fuels), we caution that fuel carbon intensity should not be directly compared unless the efficiency of end use technology is considered. These results, however, may be useful to other researchers who wish to compare our estimates with other sources or wish to evaluate a wide range of end-use technologies beyond those included in this paper.

Table S14. GHG emissions for electricity generation (Unit: gCO_2-equiv/kWh). Shown in the table are mean estimates and 95% confidence interval (in parenthesis) from this study.

	NGCC without CCS	NGCC with CCS	Grid average (Year 2010)[29]
Upstream	98 (57, 174)	115 (67, 204)	48
Combustion	358 (348, 368)	50 (48, 51)	564
Total (at power plant gate)	456 (413, 533)	165 (116, 254)	612

* Here we report energy efficiency in lower heating value.

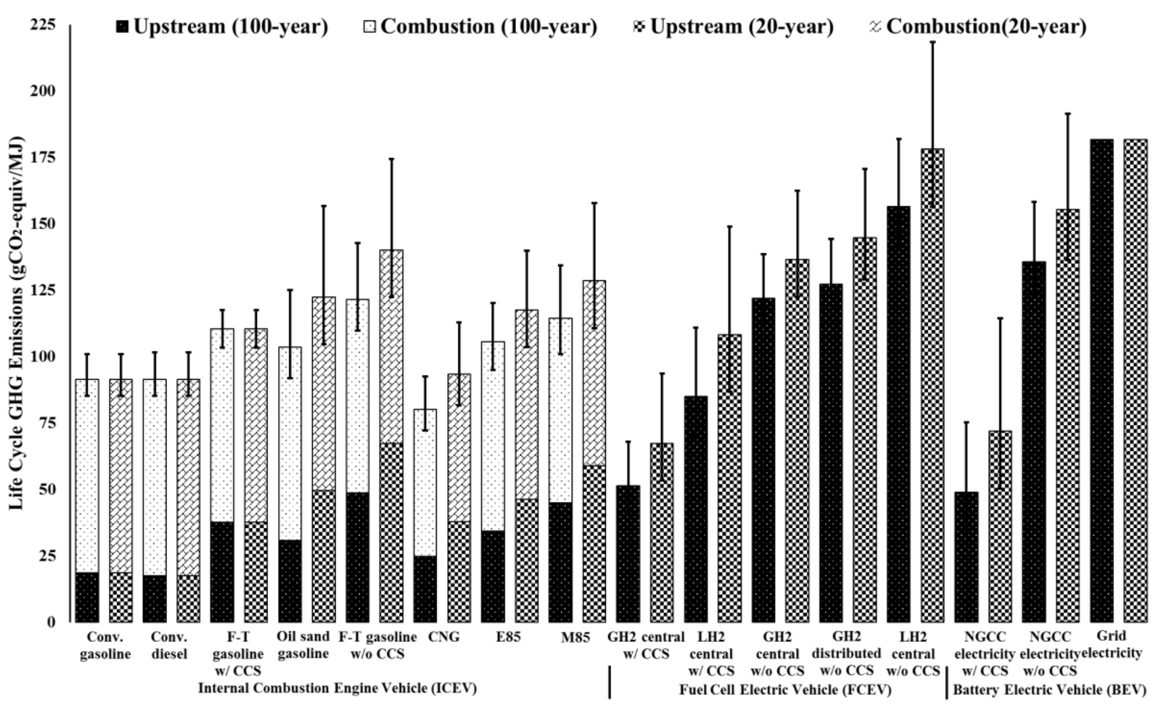

Figure S4. Life cycle GHG emissions ('carbon intensity') of natural gas-derived fuels and petroleum fuels with 100-year and 20-year GWP. Baseline methane emissions estimates are assumed for natural gas pathways. The functional unit is 1 MJ (lower heating value) of fuel

delivered to end use. These estimates do not account for vehicle fuel efficiencies or tailpipe CH_4 and N_2O emissions, as they depend on vehicle technologies. Error bars represents the 95% confidence interval of the life cycle GHG emissions. See the spreadsheet Supplementary Data file for numbers.

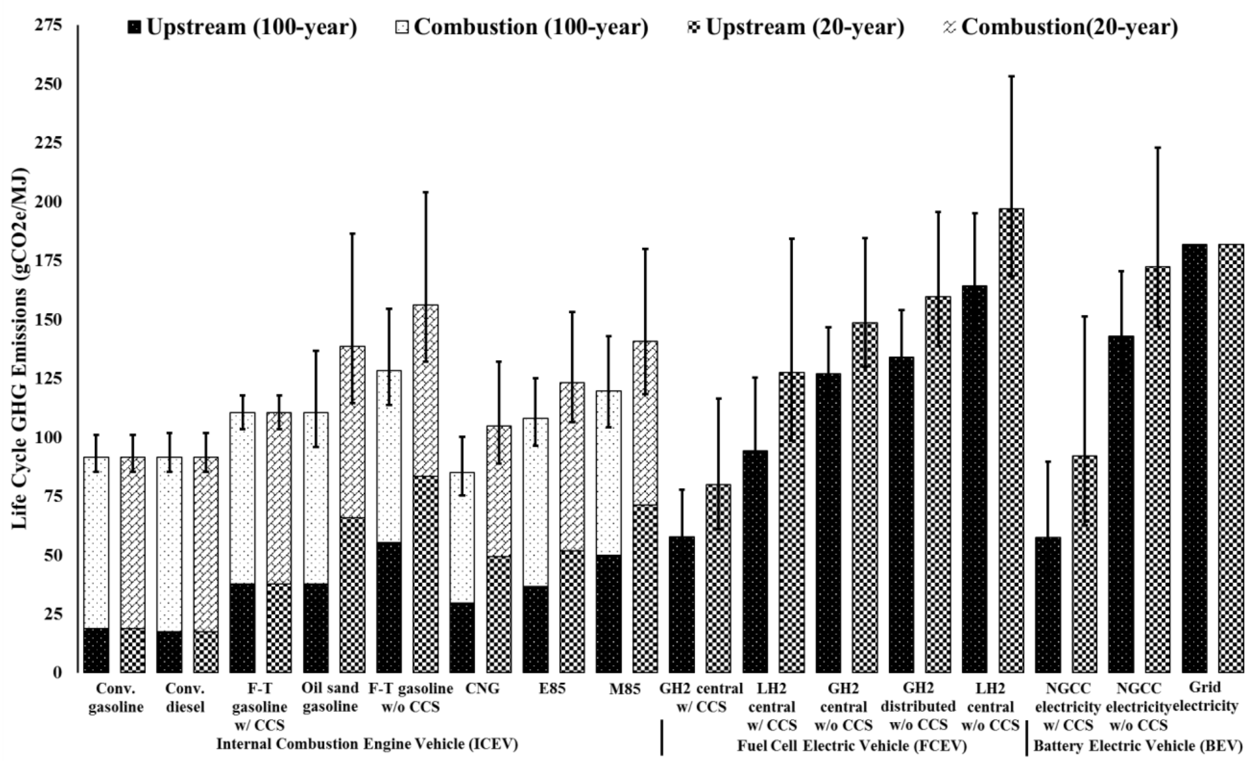

Figure S5. Life cycle GHG emissions ('carbon intensity') of natural gas-derived fuels and petroleum fuels with 100-year and 20-year GWP. Pessimistic methane emissions estimates are assumed for natural gas pathways. The functional unit is 1 MJ (lower heating value) of fuel delivered to end use. These estimates do not account for vehicle fuel efficiencies or tailpipe CH_4 and N_2O emissions, as they depend on vehicle technologies. Error bars represents the 95% confidence interval of the life cycle GHG emissions. See the spreadsheet Supplementary Data file for numbers.

Additional Results from the Monte-Carlo Simulations

Figure S6-Figure S9 shows the cumulative distribution function of the relative changes between life cycle GHG emissions of natural gas pathways and that of conventional gasoline. These figures highlight that natural gas pathways have larger uncertainty and variability than the conventional gasoline pathway. We also find strict stochastic dominance among natural gas pathways, which allows us to use the average GHG emissions to determine the ranks of natural gas pathways. Note that a spreadsheet Supplemental Data file is available online and includes numbers behind the figures.

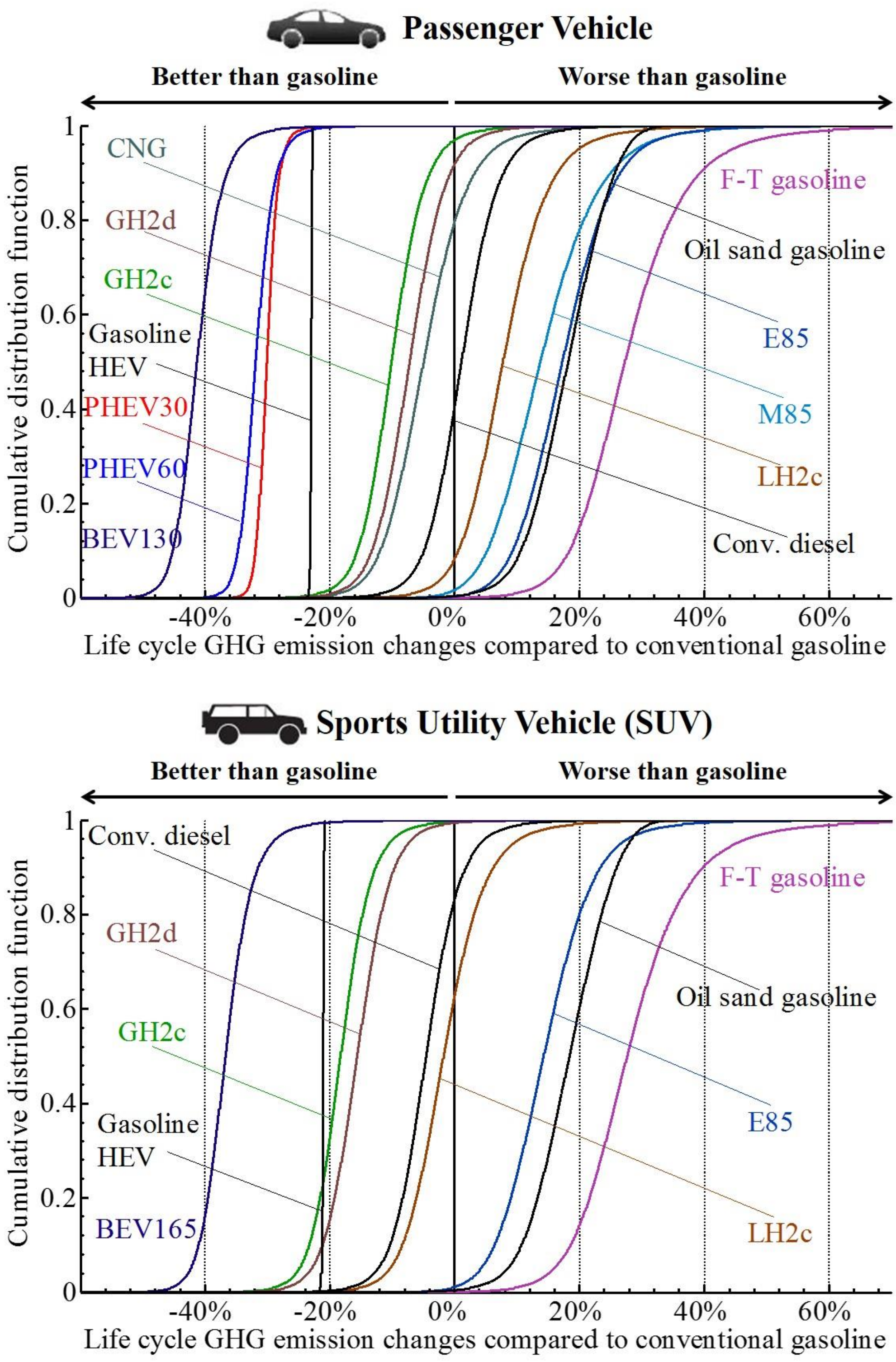

Figure S6. Cumulative probability distributions (CDF) of life cycle GHG emission changes compared to the baseline gasoline pathways for LDVs with 100-year GWPs and baseline methane emissions estimate. (The cartoon icons are from the Alternative Fuels Data Center; http://www.afdc.energy.gov/.)

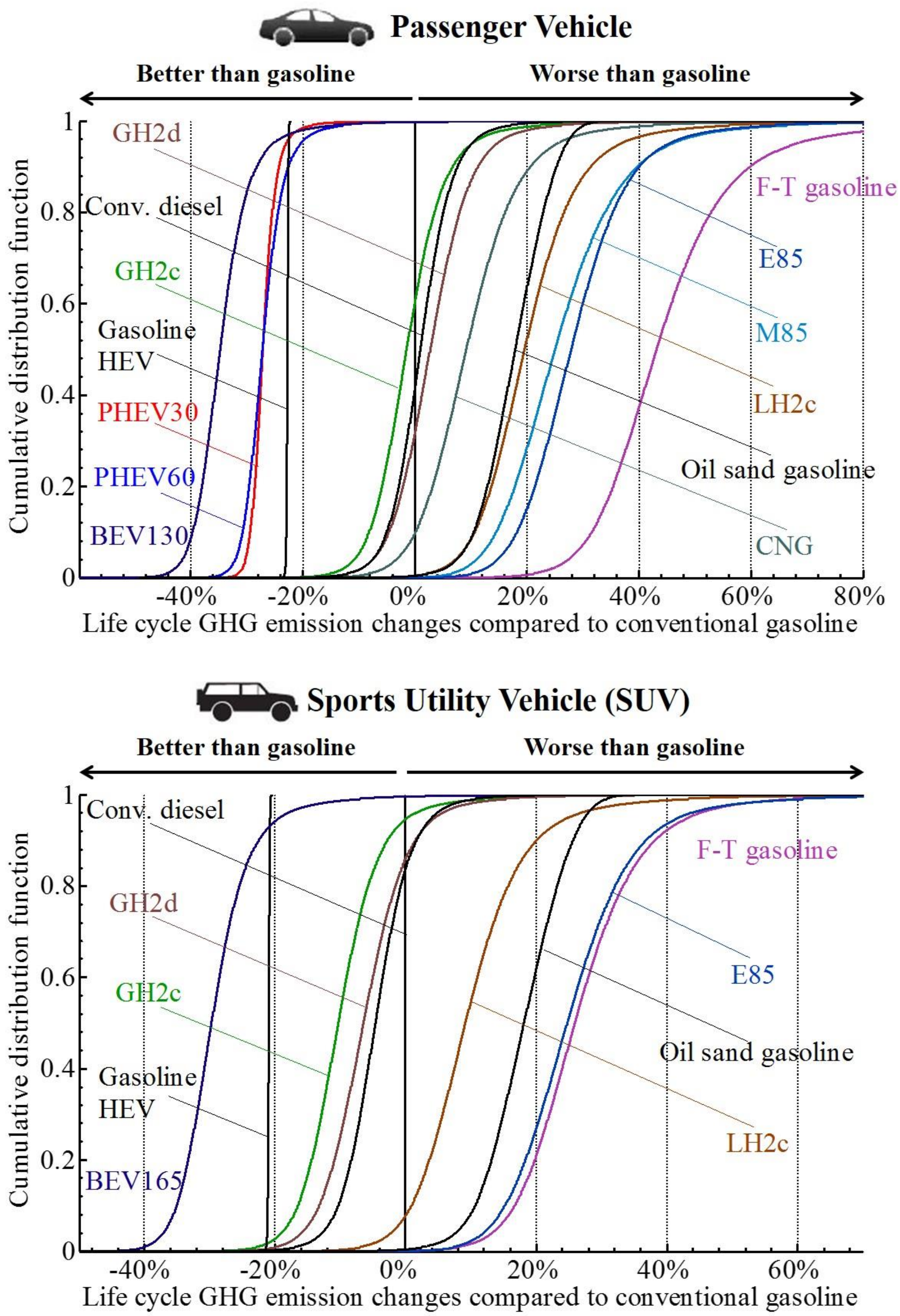

Figure S7. Cumulative probability distributions (CDF) of life cycle GHG emission changes compared to the baseline gasoline pathways for LDVs with 20-year GWPs and baseline methane emissions estimate. (The cartoon icons are from the Alternative Fuels Data Center; http://www.afdc.energy.gov/.)

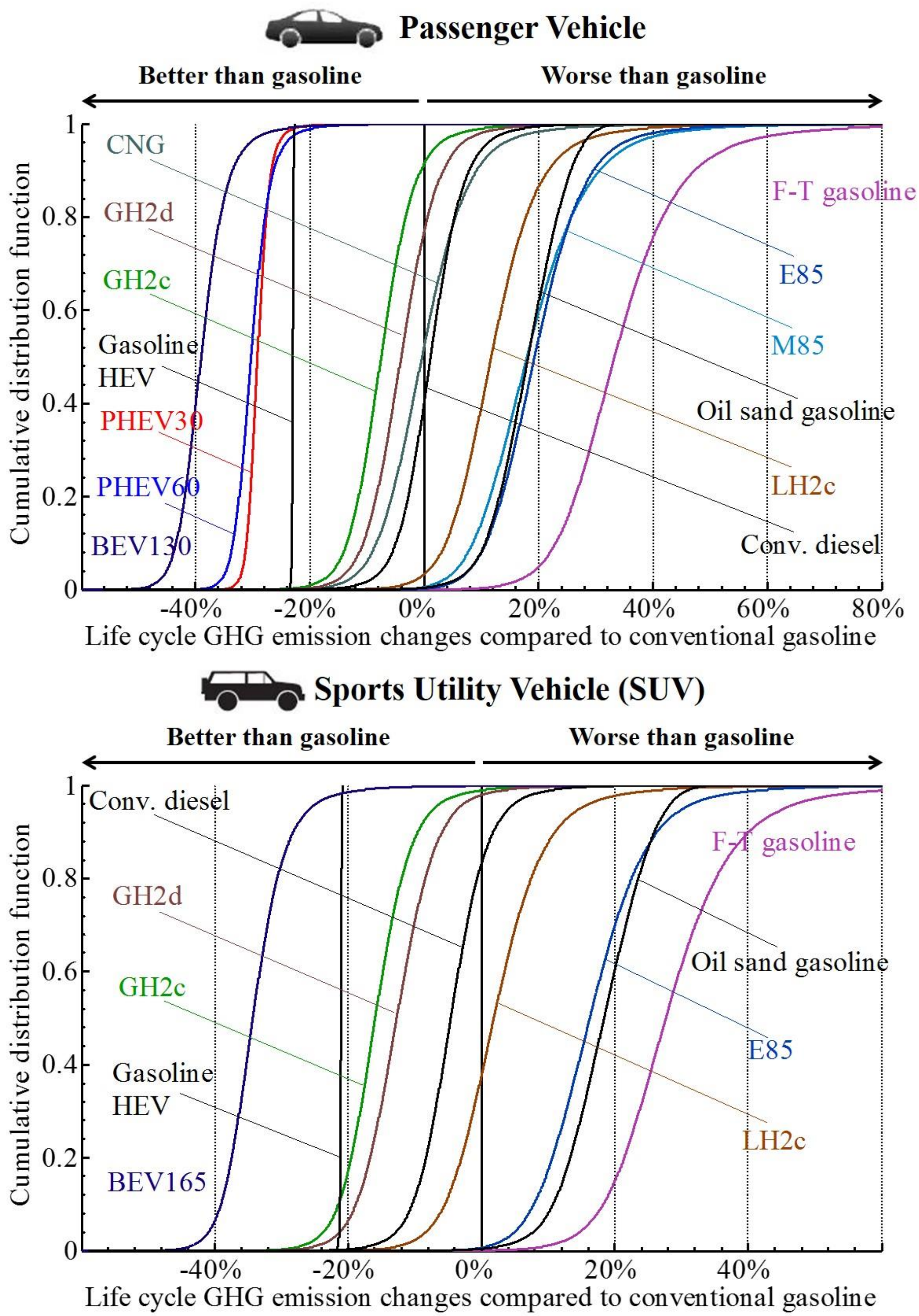

Figure S8. Cumulative probability distributions (CDF) of life cycle GHG emission changes compared to the baseline gasoline pathways for LDVs with 100-year GWPs and pessimistic methane emissions estimate. (The cartoon icons are from the Alternative Fuels Data Center; http://www.afdc.energy.gov/.)

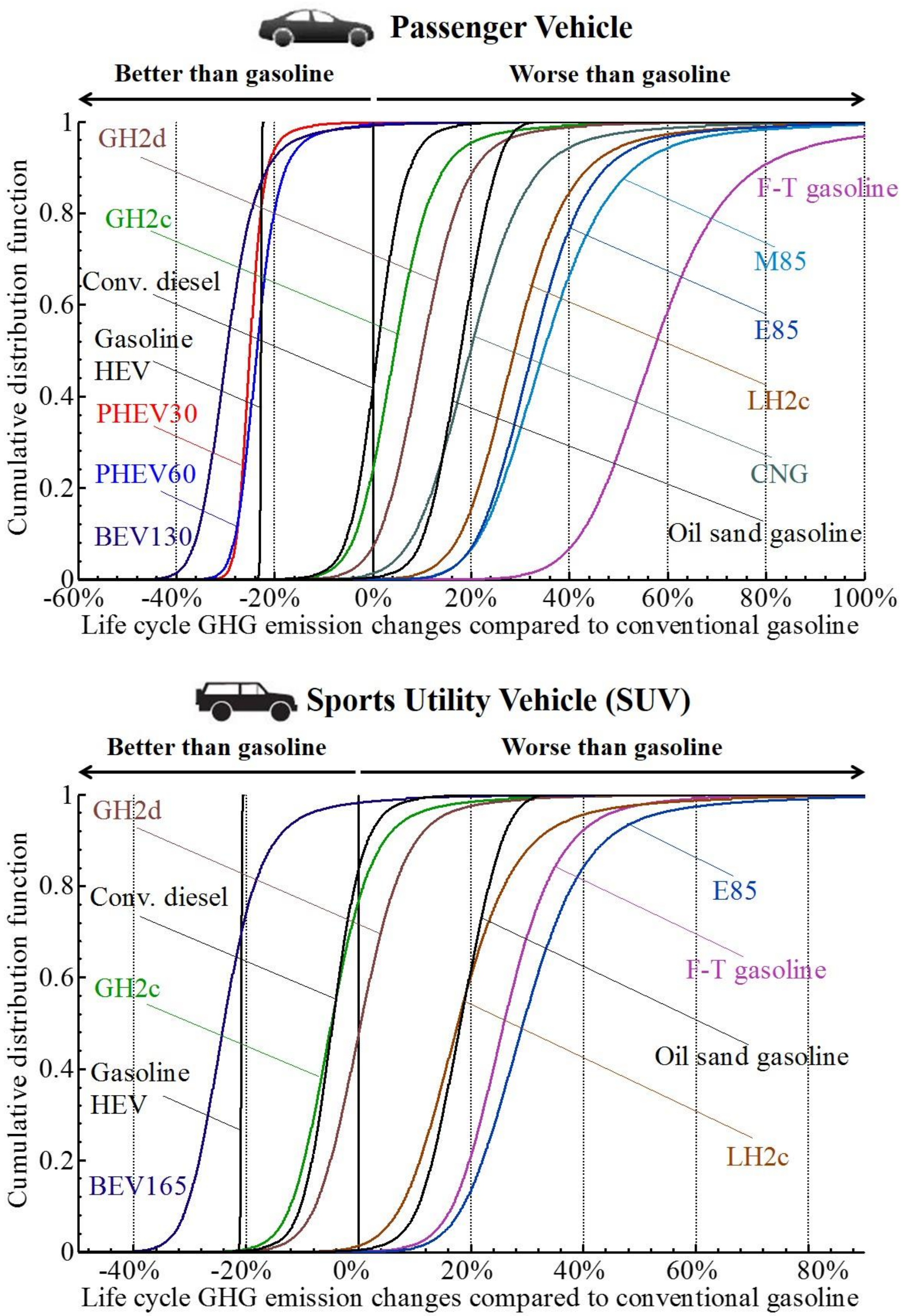

Figure S9. Cumulative probability distributions (CDF) of life cycle GHG emission changes compared to the baseline gasoline pathways for LDVs with 20-year GWPs and pessimistic methane emissions estimate. (The cartoon icons are from the Alternative Fuels Data Center; http://www.afdc.energy.gov/.)

Impact of the Carbon Intensity of Electricity on GHG emissions of Electric Vehicles

Figure S10 shows that life cycle GHG emissions from HEVs, PHEVs, and BEVs are linear functions of the carbon intensity of electricity sources. Here, we use the same system boundary and formulas with the Monte-Carlo simulation model except that we treat electricity input as a parameter. We use the 100-year GWP[(30)]. We find that with current U.S. grid, BEV130 and PHEV30 are slightly better than gasoline HEVs in terms of life cycle GHG emissions, while PHEV60 is worse. If carbon capture and storage (CCS) technology is available at NGCC power plants, BEVs reduce GHG emissions by nearly 75% compared to gasoline HEVs. With NGCC electricity, BEV130 is much better than gasoline HEVs as we have shown in the main text.

Table S15 summarizes the tipping points of carbon intensity of electricity inputs where these electric pathways have the same emissions. Our estimates of the tipping points are different from Samaras et al. (2008)[(19)] in that we account for incremental vehicle manufacturing emissions and include non-CO_2 GHG emissions from the vehicle tailpipe. In addition, we use updated assumptions on fuel economy of electric vehicles and battery manufacturing emissions. Samaras et al. (2008)[(19)] report a tipping point of roughly 690 gCO_2-equiv/kWh, below which PHEV60 emits less GHGs than PHEV30. We find that a much cleaner electricity source is needed[10] (520 gCO_2-equiv/kWh) in order for PHEV60 to emit less than PHEV30.

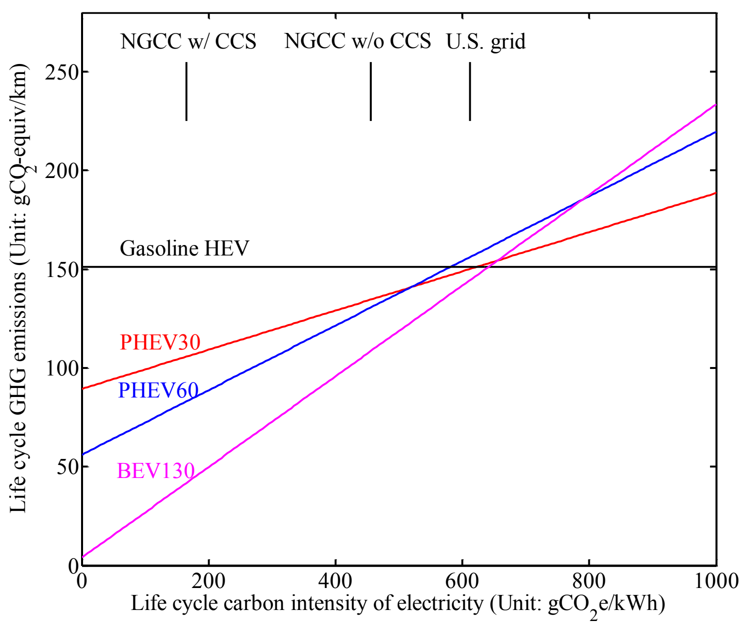

Figure S10. Life cycle GHG emissions from HEV, PHEV, and BEV as a function of life cycle carbon intensity of electricity generation.

[10] An interesting fact in our study is that the tipping point between PHEV30 and PHEV60 is around 510 gCO_2-equiv/kWh which is also the mean life cycle GHG emissions of NGCC electricity.

Table S15. Break-even carbon intensity of electricity inputs for plug-in electric vehicles.

Index	Break-even between		Break-even carbon intensity of electricity (Unit: gCO_2-equiv/kWh)
1	PHEV30	BEV130	790
2	HEV	BEV130	640
3	HEV	PHEV30	625
4	HEV	PHEV60	580
5	PHEV30	PHEV60	520

S6. Comparison with Existing Studies

We compare our results with the GREET model[(2)] which has been widely used in the literature [(31–38)]. We choose two recent versions of the GREET model, version 2013 and version 2014, which differ primarily in updated assumptions on fugitive methane emissions from natural gas systems and GWP values. While the version 2013 is not most up-to-date, it provides fuel production and transport assumptions used in this study. The GREET model by default focuses on existing LDVs and assumes the U.S. grid as the electricity source. We thus modified the GREET model settings to use fuel economy assumptions in this paper (**Table S6**) and to use NGCC electricity for plug-in electric vehicles.

Figure S11 shows the comparisons for life cycle GHG emissions for natural gas pathways in passenger vehicles. We find that our results are comparable with those from the GREET models. The point estimates of most natural gas pathways (except the M85 and E85 pathways) from the GREET models fall in the 95% confidence intervals of life cycle emissions from our model. In fact, for most of these pathways, our average estimates are comparable with the point estimates in the GREET model. We find smaller differences between our average emission estimates with the GREET model Version 2014 than with Version 2013. Version 2014 updated the fugitive methane emissions from natural gas systems and used the new GWPs in the IPCC AR5 report[(30)], thus having higher emission estimates for all natural gas pathways.

There are differences between our model and the GREET model for M85 and E85 pathways. For the M85 pathway, the differences are solely from different estimates in upstream (well-to-pump) emissions of M85: 34.9 gCO_2-equiv/MJ in our model compared to 17.5 gCO_2-equiv/MJ. Our assumptions of methanol production are less optimistic than those used in the GREET model. If we use GREET's assumptions of energy efficiency and input shares, our upstream emissions of M85 change to 27.9 gCO_2-equiv/MJ, bringing the differences in life cycle emissions to less than 6%. In addition, our natural gas upstream emissions are slightly higher than the GREET model. The differences for the E85 pathway are much easier to explain, as the GREET model does not a natural gas-based E85 pathways but we include the corn-based E85 for comparison.

As we have discussed in the main text, although existing studies used the GREET model to estimate the emission reduction potentials and the cost-effectiveness of natural gas pathways compared to petroleum fuels, they failed to include a comprehensive set of pathways, used outdated data with regard to natural gas upstream emissions and GWP, and largely ignored uncertainty and variability, especially those related to fugitive methane emissions from natural gas systems. Our study addresses these limitations and provides an independent emission inventory in addition to the GREET model.

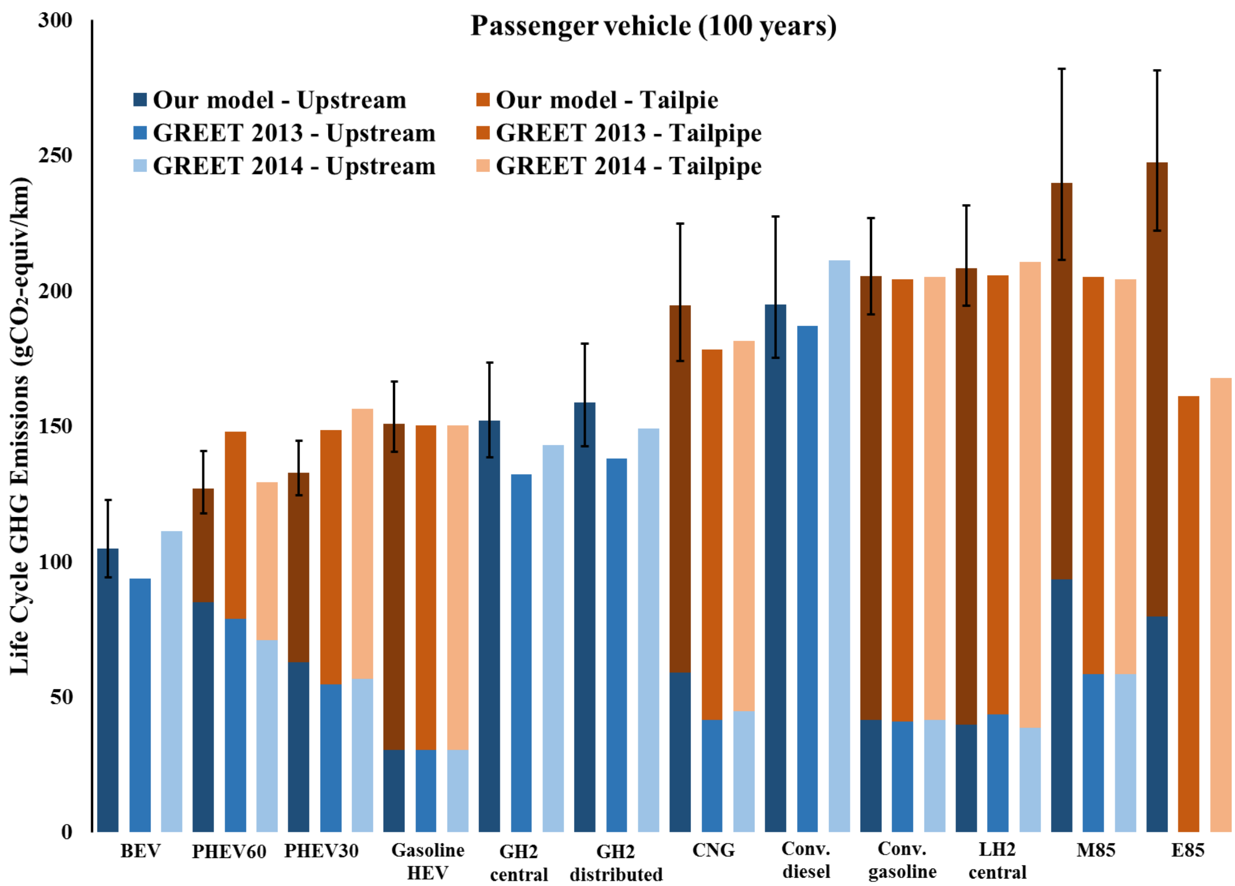

Figure S11. Comparison of life cycle GHG emissions of natural gas pathways for passenger vehicles from this study, the GREET model (version 2013), and the GREET model (version 2014). Results from this study are from the baseline scenario (baseline methane estimate and 100-year GWPs). Results from the GREET model (version 2013 and 2014) use the same fuel economy assumptions and assume NGCC electricity for plug-in electric pathways. Error bars represent the 95% confidence interval of life cycle GHG emissions. The E85 pathway in the GREET model assumes corn as the feedstock input.

S7. Data Quality

In this paper, as with other LCA work, we relied on many data sources. In the main text and this document (Supporting Information), we discussed how we used these data sources and what our model leads to. Here, we discuss the data quality of these sources. Specifically we discuss the type of source and where we used it in our model. We divide data sources into the following categories: peer-reviewed journal papers (including peer-reviewed conference proceedings), thesis, conference presentations, academic working papers, government sources (including those authored or contracted by national laboratories), vehicle manufacturer specifications, and industry consulting reports. For several model assumptions, peer-reviewed data sources do not exist, in which case we relied on alternative data sources. In addition to data sources, we also include the information regarding the nature of data for natural gas upstream emissions and vehicle assumptions. For instance, are they based on actual emission measurements or vehicle tests? Direct data sources, such as emission measurements or vehicle tests are preferred over indirect sources.

Units and Metrics

We summarize data sources in **Table S16**. Here we used the authoritative sources wherever possible. We use the fuel properties and combustion emission factors from the GREET model(2) because it is the mostly-widely used LCA model in the transportation sector and some of their periodic updates are peer-reviewed.

Table S16. Review of data sources related to units, metrics and fuel properties.

Type	Data Source	Type of source
Global warming potential	IPCC AR5(30)	Peer-reviewed inter-government report.
Fuel properties and combustion emission factors (except for natural gas)	GREET(2)	Government or national laboratory data source (LCA model).
Methane composition in natural gas	EPA (2014)(6)	Government or national laboratory report.
	Foss (2007)(7)	Non peer-reviewed academic institute source.

Gasoline and Diesel

We rely on peer-reviewed journal papers for upstream and combustion emissions of conventional and oil-sand derived gasoline and diesel, as summarized in **Table S17**.

Table S17. Review of data sources used to estimate GHG emissions of gasoline and diesel.

Type	Data Source	Type of source
Upstream emissions for conventional gasoline and diesel	Venkatesh et al. (2011)(39)	Peer-reviewed journal paper.
Combustion emissions for gasoline and diesel	(2,39–41)	Two peer-reviewed journal papers, and one government or national laboratory report.
Oil sand-derived gasoline/diesel	Englander et al.(42)	Peer-reviewed journal paper.

Fuel Production

We rely on peer-reviewed journal papers and reports from the national laboratories to model fuel production profiles (**Table S18**). We use multiple data sources for validation purposes as well as for constructing distributions for the Monte Carlo model.

Table S18. Review of data sources for fuel production assumptions.

Type	Data Source	Type of source
Electricity		
NGCC Energy efficiency	NETL (2013)[43]	Government or national laboratory report.
U.S. grid average electricity	Cai et al. (2013)[2][29]	Government or national laboratory report.
Compressed Natural Gas (CNG)		
Energy efficiency of electric compressors	Venkatesh et al. (2011)[4]	Peer reviewed journal paper.
Natural Gas-Based Hydrogen (H2)		
Central hydrogen plant profile (w/o CCS)	[2,44,45]	Government or national laboratory reports.
Central hydrogen plant profile (w/ CCS)	H2A 3.0[45]	Government or national laboratory report.
Distributed hydrogen plant profile (w/o CCS)	[2,45]	Government or national laboratory reports.
Boil-Off Effects of Liquid H_2	GREET[2]	Government or national laboratory report.
Fischer-Tropsch Liquids		
Centralized Fischer-Tropsch liquids production plant	Jaramillo et al. (2008)[46]	Peer-reviewed journal paper.
Methanol		
Methanol production profile	[2,34,47,48]	One government or national laboratory data source (LCA model), two scientific society reports, and one consulting report
Ethanol		
Ethane steam cracking production profile	(10)	Peer-reviewed journal paper.
Ethylene hydration production profile	(14)	A LCA database provided by Universities and national laboratories in Switzerland

Fuel Transport

We rely on the GREET model[2] for fuel transport assumptions for liquid fuels (**Table S19**).The GREET model assumes a national-average transportation profile in terms of transportation modes, energy intensities, and distances. While regional variations exist in terms of transportation profiles, the shares of transportation emissions in life cycle GHG emissions are very small.

Table S19. Review of data sources used for fuel transportation assumptions.

Type	Data Source	Type of source
GHG emission factors of fuel transport for F-T liquids, GH_2 (central), LH_2 (central), methanol and ethanol.	GREET[2]	Government or national laboratory data source (LCA model).

Vehicle

We summarize data sources on vehicle fuel economy assumptions in **Table S20**. Most natural gas pathways have new vehicles offered by original equipment manufacturers (OEM). The U.S. EPA regulates, monitors and publishes fuel economy information through standardized vehicle tests.

Table S20. Review of data sources for vehicle fuel economy assumptions.

Type	Data Source	Type of source
Passenger vehicle		
Gasoline vehicle, diesel vehicle, gasoline HEV, BEV, CNG vehicle, E85 FFV, Hydrogen FFV	fueleconomy.gov[16]	Government or national laboratory data source (vehicle tests).
PHEV	(17,18)	Peer-reviewed journal papers.
M85	GREET[2]	Government or national laboratory data source (LCA model).
Sports Utility vehicle (SUV)		
All pathways	fueleconomy.gov[16]	Government or national laboratory data source (vehicle tests)

We summarize data sources used for calculating vehicle manufacturing emissions in **Table S21**. Emission factors of battery and fuel cell manufacturing are from peer-reviewed journal papers and the GREET model[2]. Assumptions related to the battery and fuel cell sizes are collected from vehicle specifications of literature if actual vehicles do not exist. We note that emissions from FCEVs vehicle manufacturing are significantly larger than those of conventional ICEVs. In the main text, we mentioned that if the fuel cells have to be replaced even once during the life of the vehicle, all hydrogen pathways would increase life cycle GHG emissions compared to the conventional gasoline pathway. Given that current FCEVs have only been available in certain regions for less than a decade, it is still early to say much about the life time of fuel cells.

Table S21. Review of data sources for battery and fuel cell manufacturing emissions.

Type	Data Source	Type of source
Emission factors		
Battery manufacturing emissions	[23]	Peer-reviewed journal paper.
Battery specific energy	[23]	Peer-reviewed journal paper.
Vehicle manufacturing emissions	GREET[2]	Government or national laboratory data source (LCA model).
PHEV/BEV charging energy efficiency	(2,17,19)	Two peer-reviewed journal papers and one government or national laboratory report.
PHEV use patterns		
Household travel survey	(21)	Government or national laboratory report (national survey).
LDV lifetime travel distance		
LDV lifetime travel distance	(17,19)	Two peer-reviewed journal papers.
EV battery and fuel cell size and replacement		
gasoline HEV and BEV battery size	fueleconomy.gov[16]	Government or national laboratory data source (vehicle tests)
PHEV battery size	(17,18)	Peer-reviewed journal papers.

FCEV fuel cell size	fueleconomy.gov[16]	Government or national laboratory data source (vehicle tests)
Battery replacement	(2,17,19)	Two peer-reviewed journal papers, and one government or national laboratory data source (LCA).
Fuel cell replacement	GREET[2]	Government or national laboratory data source (LCA model).

We summarize data sources for tailpipe methane and N_2O emissions in **Table S22**. Tailpipe emissions are relatively small and comparable across natural gas pathways. The only exception is CNG, which has a tailpipe methane emission factor that is 10 times higher than the conventional ICEVs. However, tailpipe emissions only represent 2% of life cycle emissions of the CNG pathway.

Table S22. Review of data sources for tailpipe methane and N_2O emissions.

Type	Data Source	Type of source
Tailpipe methane and N_2O emissions	GREET[2]	Government or national laboratory data source (LCA model).

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