Environmental Science & Technology

POLICY ANALYSIS

pubs.acs.org/est

Uncertainty in Life Cycle Greenhouse Gas Emissions from United States Natural Gas End-Uses and its Effects on Policy

Aranya Venkatesh,[*,†] Paulina Jaramillo,[‡] W. Michael Griffin,[‡,§] and H. Scott Matthews[†,‡]

[†]Civil and Environmental Engineering Department, [‡]Department of Engineering and Public Policy, and [§]Tepper School of Business, Carnegie Mellon University, 5000 Forbes Avenue, Pittsburgh, Pennsylvania 15213-3890, United States

Supporting Information

ABSTRACT: Increasing concerns about greenhouse gas (GHG) emissions in the United States have spurred interest in alternate low carbon fuel sources, such as natural gas. Life cycle assessment (LCA) methods can be used to estimate potential emissions reductions through the use of such fuels. Some recent policies have used the results of LCAs to encourage the use of low carbon fuels to meet future energy demands in the U.S., without, however, acknowledging and addressing the uncertainty and variability prevalent in LCA. Natural gas is a particularly interesting fuel since it can be used to meet various energy demands, for example, as a transportation fuel or in power generation. Estimating the magnitudes and likelihoods of achieving emissions reductions from competing end-uses of natural gas using LCA offers one way to examine optimal strategies of natural gas resource allocation, given that its availability is likely to be limited in the future.

In this study, the uncertainty in life cycle GHG emissions of natural gas (domestic and imported) consumed in the U.S. was estimated using probabilistic modeling methods. Monte Carlo simulations are performed to obtain sample distributions representing life cycle GHG emissions from the use of 1 MJ of domestic natural gas and imported LNG. Life cycle GHG emissions per energy unit of average natural gas consumed in the U.S were found to range between −8 and 9% of the mean value of 66 g CO_2e/MJ. The probabilities of achieving emissions reductions by using natural gas for transportation and power generation, as a substitute for incumbent fuels such as gasoline, diesel, and coal were estimated. The use of natural gas for power generation instead of coal was found to have the highest and most likely emissions reductions (almost a 100% probability of achieving reductions of 60 g CO_2e/MJ of natural gas used), while there is a 10−35% probability of the emissions from natural gas being higher than the incumbent if it were used as a transportation fuel. This likelihood of an increase in GHG emissions is indicative of the potential failure of a climate policy targeting reductions in GHG emissions.

INTRODUCTION

In an effort to reduce greenhouse gas (GHG) emissions from energy consumption and promote energy security within the United States, alternatives to conventional fuels such as gasoline and diesel are being considered. Natural gas has been considered a promising low carbon alternative as a fuel source. The early release of the Annual Energy Outlook 2011 (Reference case) by the Energy Information Administration (EIA)[1] predicts the increased long-term use of natural gas in various sectors, driven by forecasts of increased shale gas production and low prices. In the EIA report, natural gas consumption is projected to increase from 23.9 trillion cubic feet (tcf) in 2010 (680 billion m^3) to 25.7 tcf (730 billion m^3) in 2030, with the largest growth seen in the transportation sector (an increase of 400% from 0.03 tcf or 0.8 billion m^3 to 0.13 tcf or 3.7 billion m^3).[2] In a recent paper, Yeh and Sperling[3] suggest that compressed natural gas (CNG) is likely to be considered an alternative transportation fuel, especially for heavy-duty vehicles, within national and regional policy frameworks aimed at increasing the use of low carbon fuels. Newcomer and Apt[4] examine alternatives to meet future electricity demand in the face of increasing restrictions on the construction of new coal-fired power plants in the U.S. In all the scenarios they consider, they conclude that there is likely to be a large push toward building natural gas combined cycle (NGCC) plants.

A number of life cycle assessment (LCA) studies have been published on the subject of GHG emissions from natural gas, for example, the environmental impacts of using natural gas for electricity generation in the U.S.,[5] of natural gas substituted as transportation fuel in Europe,[6] of domestic natural gas and liquefied

Received: March 19, 2011
Accepted: August 16, 2011
Revised: June 17, 2011
Published: August 16, 2011

natural gas (LNG) consumed in the U.S.,[7] and of LNG used in Japan,[8,9] among others.[10−12] While comprehensive in scope, these studies do not include a rigorous treatment of the uncertainty and variability in estimating life cycle environmental impacts.

Recently, some policies aiming to mitigate climate change impacts, such as the Energy Independence and Security Act[13] and the California Low Carbon Fuel Standard (LCFS),[14] have made use of LCA methods as tools to compare GHG emissions of competing fuels, and to estimate emissions reductions possible by the use of low carbon fuels. However, these policies have been informed by the results of analyses based only on point estimates that ignored the uncertainty and variability prevalent in LCA data and methods. For instance, the California LCFS uses a modified version of the comprehensive Greenhouse Gases, Regulated Emissions and Energy Use in Transportation model[15] to obtain single-point estimates of GHG emissions from compressed natural gas (CNG) as a potential transportation fuel.[16] Using single point-estimates in policy designed to reduce GHG emissions, however, may not provide reliable estimates of emissions reductions possible with the use of low carbon fuels, as demonstrated by Mullins et al.[17] and Venkatesh et al.[18] As concluded by Plevin et al.,[19] the robust design of any such policies requires that the risks associated with uncertainties in estimates of life cycle impacts be acknowledged.

Natural gas can be used for different energy demands as a low carbon alternative to achieve emissions reductions. For example, it can be used as a transportation fuel (as CNG) instead of gasoline or diesel in vehicles, or in power generation plants. While considering optimal ways of allocating natural gas to competing end-uses, estimating the likelihoods of achieving emissions reductions in these scenarios is important. Even though natural gas production is projected to increase in the future,[1] multiple new energy demands that use large amounts of gas will likely be constrained by available supply.

Motivated by the forecast of increased natural gas consumption, this study quantifies the uncertainty in life cycle GHG emissions from natural gas consumed in the U.S. It builds upon the deterministic natural gas life cycle framework developed by Jaramillo et al.,[5] using analyses similar to those performed by Mullins et al.[17] and Venkatesh et al.[18] Included are probability distributions representing the uncertainty and/or variability in model input parameters, obtained from different data sources. The differences between uncertainty and variability relevant to this approach are discussed. Monte Carlo simulations are performed to obtain sample distributions representing life cycle GHG emissions from the use of 1 MJ of domestic natural gas and imported LNG. A probability mixture model combines the emissions from domestic natural gas and LNG imports to obtain a representative range of life cycle emissions from natural gas used in the U.S. Emissions from natural gas consumed for different end-uses are stochastically compared to emissions from incumbent fuels to examine the magnitudes and likelihoods of achieving emissions reductions.

■ MATERIALS AND METHODS

The functional unit of this study is 1 MJ (higher heating value) of natural gas consumed in the U.S. Data applicable to the year 2008 are used wherever possible unless otherwise specified, as this is the most recent year for which data were available. Publicly available data or data from peer-reviewed literature were used in this analysis. U.S. specific data were used to estimate domestic natural gas life cycle emissions. Continuous probability distributions that best fit the available data were used to represent the various model input parameters. When the quality and/or the quantity of data were not sufficient to estimate best-fitting distributions that satisfied standard goodness-of-fit tests, uniform distributions (when two data points were available) and triangular distributions (when three or more data points were available) were used. Discrete distributions were used when data was found to take on a few discrete random values. Further details on fitting distributions to data along with examples are presented in the Supporting Information. Canadian and Mexican natural gas imports (unliquefied) are assumed to be transported by pipeline and to have the same life cycle GHG emissions as domestic natural gas. This is considered a reasonable assumption in the absence of better data given the small percentage of imports from Mexico (<1%), the similarity of natural gas technology for the U.S. and operators in Canada (contributing to 15% of U.S. imports) and the large range of upstream emissions modeled. For CH_4 and N_2O emissions, 100-year global warming potentials (GWP) were used, as suggested by the 2007 IPCC Fourth Assessment Report.[20]

Domestic Natural Gas. In 2008, U.S. domestic natural gas was produced from a number of sources including conventional gas wells (62%), gas associated oil wells (23%), and unconventional sources such as shale gas (8%) and coalbed methane (8%).[21,22] In this study, the production, processing, transmission, distribution and combustion of domestic natural gas were considered. The GHG emissions from each of these stages were assumed to be similar for natural gas produced from different sources, whether conventional or otherwise. The impacts from preproduction activities such as well drilling and completion, which are considered to be higher for unconventional natural gas sources compared to conventional, were not considered in the system boundary. These impacts are normalized over the quantity of natural gas produced over the well lifetimes and are likely to be negligible, as demonstrated by Jiang et al.[23]

Producing natural gas contributes to GHG emissions via a number of activities incuding lease fuel combustion (fuel at natural gas wells and fields for activities such as drilling operations[24]), flaring and/or venting of gas, and other fugitive emissions. A triangular distribution was used to model emissions from the combustion of lease fuel. Uniform distributions were used to represent flared and vented emissions based on state-level data from the U.S. Energy Information Administration (EIA),[25] and fugitive CO_2 emissions based on data from the 2006 IPCC Guidelines report.[26] A discrete distribution was used to represent CH_4 fugitive emissions, based on data from Environmental Protection Agency's (EPA) 2010 Inventory of U.S. Greenhouse Gas Emissions and Sinks.[27]

GHG emissions from processing natural gas are due to fuel combustion at processing plants, fugitive emissions and from CO_2 that is separated from the gas processed and vented. A log-normal distribution was found to fit state-level plant fuel consumption data obtained from the U.S. EIA.[25] Uniform distributions were used to represent CO_2 and N_2O emissions from fugitive and flared sources, using data from the 2006 IPCC Guidelines report.[26] Fugitive CH_4 emissions were estimated using data from the U.S. EIA[28] and EPA's State Greenhouse Gas Inventory tool.[29] The GHG emissions from plant fuel combustion, fugitives and flaring (based on the total quantity of natural gas processed in the U.S) thus estimated were

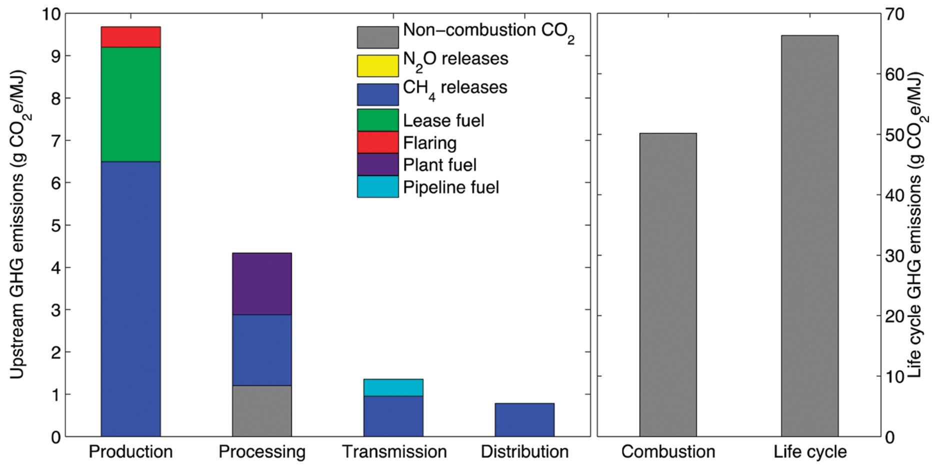

Figure 1. Mean GHG emissions from domestic natural gas life cycle activities and stages.

multiplied by a factor of 0.76, since only 76% of the total quantity of natural gas produced required processing in 2008.[22,30] If this factor were increased to 89%, the highest observed since the year 2000, this would affect the total life cycle estimate by less than 1%, and hence no sensitivity analysis was considered for this parameter. The amount of CO_2 vented at processing plants was modeled as a discrete distribution using data from the EPA's 2010 Inventory of U.S. Greenhouse Gas Emissions and Sinks.[27]

Sources of GHG emissions from the transmission and distribution of natural gas include the combustion of pipeline fuel in these systems, and fugitive and vented CH_4 and CO_2. A generalized extreme value distribution was found to fit state-level data from the U.S. EIA,[25,31] representing emissions from pipeline fuel use. Data from the 2006 IPCC Guidelines report[26] were used to fit uniform distributions to represent fugitive and vented CO_2 emissions. Fugitive CH_4 emissions were estimated using data from EPA's State Inventory Tool,[29] Interstate Natural Gas Infrastructure Map Book[32] and the Gas Facts Data 2007 report.[33]

The 2006 IPCC Guidelines Report[26] suggests a minimum, maximum, and an average value for GHG emissions per TJ of natural gas combusted. The minimum and maximum, as percentages of the average, were used in conjunction with the average combustion emissions factor for natural gas reported in Jaramillo et al.,[5] to estimate the parameters of a triangular distribution, representing combustion emissions per MJ of natural gas consumed. Note that, while the emissions per MJ of natural gas remain unchanged, the actual emissions from a particular end-use depend on the efficiency of the vehicles/equipment that consume natural gas as as fuel source.

Liquefied Natural Gas. The U.S. imports LNG from a number of countries such as Trinidad and Tobago, Egypt, Nigeria and Norway.[34] Emissions from the production, liquefaction, shipping, regasification, transmission, distribution and combustion of LNG were considered, consistent with the life cycle stages considered in previous LNG studies.[5,6,8,9,35] Relatively few data sources report GHG emissions from natural gas production in countries other than the U.S. Some of these include the ETH-ESU library (database) in the Simapro life cycle analysis software[36] and Arteconi et al.,[6] who in turn summarized data from an ARI and ICF study[7] and Okamura et al.[8] These data and those developed in this study for domestic natural gas were used to estimate the parameters of a triangular distribution representing emissions from producing natural gas that is imported to the U.S.

In the liquefaction stage, natural gas is processed to remove water and contaminants, and liquefied for transport to the U.S. Using data from Tamura et al.,[9] the ARI and ICF study,[7] Okamura et al.,[8] a study by the Climate Mitigation Services for the Environmental Defense Center[37] and the ETH-ESU and Ecoinvent libraries in Simapro[36] a triangular distribution was developed to represent emissions from liquefaction.

To estimate emissions associated with LNG transport, the distance that a unit volume of LNG is shipped from an exporting country to reach the U.S. was modeled as a discrete distribution, based on port-to-port distance data obtained from online distance calculators,[38,39] along with import volumes reported by EIA[40] that were used as probabilities of the distribution. The shipping emissions factors due to ocean tankers were estimated from Jaramillo et al.,[5] the Ecoinvent database in Simapro[36] and Okamura et al.[8]

Imported LNG is regasified before entering the U.S. natural gas transmission and distribution systems, through which it is delivered to end-users for combustion. A uniform distribution representing regasification emissions was developed using data from Tamura et al.[9] and Ruether et al.[35] The emissions from transmission, distribution and combustion of regasified LNG and domestic natural gas are assumed to be the same.

The probability distributions described were used to represent the uncertainty and variability in GHG emissions from various activities in the life cycle of domestic natural gas and LNG. A summary of modeling parameters and the distributions used is presented in the Supporting Information. These distributions were used as input to Monte Carlo simulations of 10 000 runs to obtain output sample distributions for life cycle GHG emissions per MJ of domestic natural gas and LNG.

Compressed Natural Gas. When natural gas is compressed for use as a transportation fuel, this additional life cycle stage needs to be included in order to facilitate life cycle comparisons with other transportation fuels. The study on life cycle GHG emissions from North American CNG conducted for the California LCFS[16] estimates an electric compressor efficiency of 98%. A similar study by Wang et al.[41] estimates an average electric compressor efficiency of 96% with a lower bound of 94%. Using a range of 94−98%, GHG emissions due to the use of average electricity (from the grid) for compression were estimated and added to natural gas life cycle emissions. This is in accordance with Weber et al.[42] who recommend using the national grid average for products that are spatially dispersed over the U.S, in the absence of better data.

■ RESULTS

Model Results. The mean GHG emissions from the upstream life cycle stages and constituent activities of domestic natural gas are presented in Figure 1, represented by the primary *y*-axis on the left. The two bars to the right represent the combustion and total life cycle GHG emissions, which are scaled using the secondary *y*-axis. Note that the scale of the secondary *y*-axis is an order of maginitude higher than the primary. The height of the life cycle GHG emissions bar represents the sum of the heights of the combustion and upstream GHG emissions bars. Emissions from natural gas production, processing, and transport account for 25% of life cycle emissions. As expected for the natural gas system, emissions from CH_4 release dominate the upstream stages. The average methane emissions rate was found to be about 2.2% by volume of the natural gas produced, with a range between 1.5 and 3.2%. The mean life cycle GHG emissions were estimated to be 66 g CO_2e/MJ, 6% higher than average emissions obtained by Jaramillo et al.[5] This is primarily due to the use of updated CH_4 emissions factors from production, emissions from CO_2 venting that were not included in the previous study and the use of disaggregated state level data rather than aggregated national data.

The 90% confidence intervals for the GHG emissions from the domestic natural gas and LNG life cycle stages are presented in the Supporting Information. The interval of the total life cycle estimate for domestic natural gas was found to span between 61 and 72 g CO_2e/MJ, a range equal to nearly 17% (−8% to 9%) of the mean value. A log-normal distribution (μ = 4.19, σ = 0.05) was found to best fit the sample output. The interval of the total life cycle estimate for LNG was found to span between 64 and 77 g CO_2e/MJ, nearly 18% of 70 g CO_2e/MJ, the mean value. A log-normal distribution (μ = 4.24, σ = 0.06) was found to best fit the sample output. Histograms representing life cycle emissions from domestic natural gas and LNG and the results of an uncertainty importance analysis are presented in the Supporting Information.

The U.S. EIA reports that about 1.5% of the total natural gas consumed in the U.S. was imported as LNG [25,40] in 2008. A probability mixture model (presented in the Supporting Information) using the percentages of domestic natural gas and LNG consumed in 2008 was used to estimate the life cycle emissions representing average natural gas consumed in the U.S. The distribution used to represent this average, as expected, was almost identical to the one for domestic natural gas, since LNG currently contributes to a small percentage of total natural gas consumed. Increasing the LNG fraction to 10% was found to increase life cycle results by less than 2%. However, EIA's Annual Energy Outlook 2011[1] predicts that this percentage is likely to decrease in the future, due to increases in domestic shale gas production. Thus the GHG impact that LNG will have on average natural gas consumed in the U.S. is not expected to change dramatically.

The emissions from compressing natural gas ranged between 4 and 11 g CO_2e/MJ, with a mean of 8 g CO_2e/MJ. The life cycle emissions of CNG used as transportation fuel were thus estimated to be 74 g CO_2e/MJ on average.

Natural Gas End-Use Scenarios. Mullins et al.[17] quantify GHG emissions reductions achievable by the use of biomass derived transportation fuels as alternatives to gasoline, the incumbent fuel. They develop curves representing the relationship between emission reductions targets based on the incumbent and the corresponding probabilities that biofuels use will meet the target. Similarly, in this study, the use of natural gas as an alternate fuel is examined, although the scope of comparison with the incumbent fuel is broadened. To obtain emissions reductions through natural gas use, it can be consumed as a gasoline or diesel substitute in light duty vehicles, as a diesel substitute in buses, or in power plants instead of coal. The potential emissions reductions from these scenarios, for every MJ of natural gas consumed, were quantified and compared. The probabilities of achieving such reductions were also quantified.

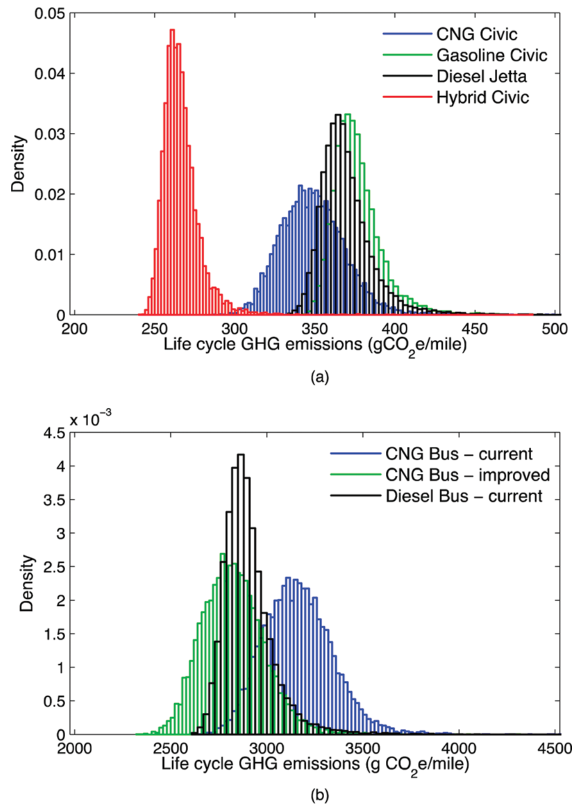

Figure 2. Comparison of the sample output probability distributions representing GHG emissions per mile driven in (a) small cars using CNG, gasoline, diesel and gasoline hybrids and (b) in buses using CNG and diesel.

Similar to this analysis, Venkatesh et al.[18] also used life cycle and probabilistic modeling methods to develop sample output probability distributions representing GHG emissions from petroleum-based fuels such as gasoline, diesel and kerosene. The study also fitted distributions to available data to represent life cycle model inputs and used a Monte Carlo simulation to obtain sample outputs. Life cycle GHG emissions from petroleum-based fuels, obtained from Venkatesh et al.,[18] and those for CNG, from this study, were used to estimate emissions per mile driven in comparable CNG, gasoline and diesel powered light-duty vehicles. The fuel economy of CNG passenger vehicles is reported in miles per gasoline equivalent gallon, where an equivalent gallon of gasoline equals 121.5 cubic feet of CNG.[43] The U.S. Department of Energy Fuel Economy web site[44] currently reports the fuel economy of only one CNG light duty

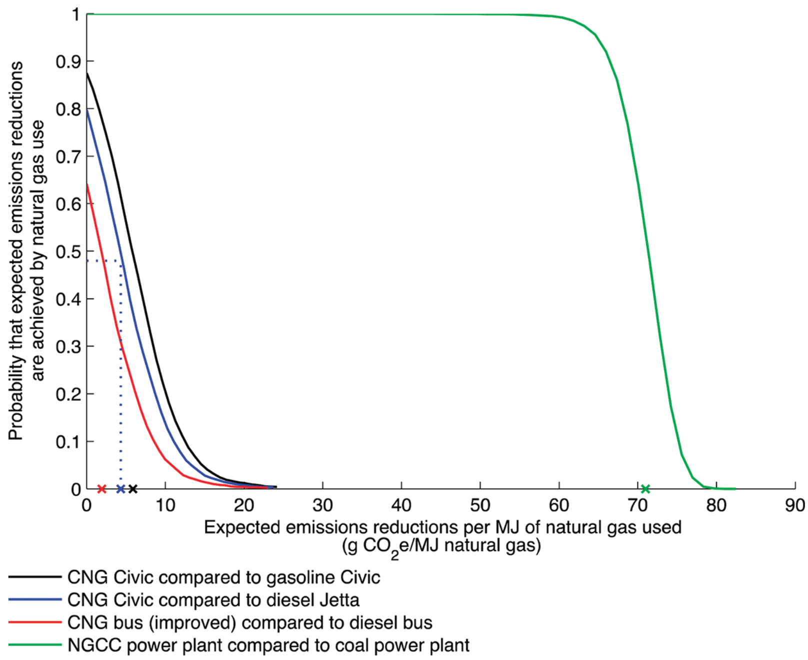

Figure 3. Trade-off between emissions reductions per MJ of natural gas consumed and the probabilities of achieving these reductions. The cross markers on the *x*-axis indicate the estimated mean emissions reductions per MJ of natural gas used as a substitute for the incumbent. The blue dotted line illustrates the probability of achieving the expected mean emisisons reductions from using a CNG Civic instead of a diesel Jetta.

vehicle in 2011—the Honda Civic (4 cylinder, 1.8 L, automatic, 5-speed) at 28 mpg (combined city and highway). The corresponding gasoline Honda Civic compares at 29 mpg, while the gasoline Honda Civic hybrid is at 41 mpg. Since a 2011 diesel Honda Civic does not exist, a diesel Volkswagen Jetta (4 cylinder, 2 L, Automatic, S6) with a fuel economy of 34 mpg was selected for this comparison. Using these fuel economies, the sample output distributions of GHG emissions per mile driven by the four different vehicle types were estimated and are presented in Figure 2(a). The hybrid vehicle dominates the other vehicle considered. The CNG Civic, however, seems to perform better than both the diesel Jetta (with 6% lower GHG emissions) and the gasoline Civic (with 7% lower GHG emissions), per mile driven by each vehicle on average, although there is a significant overlap between the distributions.

A study by the National Renewable Energy Laboratory (NREL)[43] reports that nearly 25% of U.S. transit buses purchased over the past decade and a half were CNG powered. Considering that these buses are likely to continue being used, examining the emissions from the use of CNG and diesel (the incumbent) in these buses makes for an interesting comparison. The NREL report,[43] containing survey findings of 10 transit agencies in the U. S. and around 4000 buses, documents the average fuel efficiency of a 40-foot CNG bus to be 3.5 miles per diesel equivalent gallon (where one diesel equivalent gallon equals 137 cubic feet of CNG), 20% lower than a comparable diesel powered bus with an average fuel efficiency of 4.3 mpg. The report also indicates that new CNG buses are expected to have improved efficiencies of 3.9 mpg, which is still 10% lower than diesel buses. Note that this comparison focuses on decreases in GHG per mile driven in these buses only, and does not consider any other benefits such as potential reductions in PM and NO_X emissions.

The sample output distributions of GHG emissions per mile driven corresponding to each of these cases ("CNG Bus – current", "Diesel Bus – current", and "CNG Bus – improved") are presented in Figure 2(b). These results indicate that the current diesel driven buses fare better than their CNG counterparts on average. However, the mean emissions from CNG buses with improved fuel efficiencies were about 3% lower than the diesel emissions with a significant overlap of the two distributions.

For the final scenario, emissions reductions from substituting an NGCC plant for a coal plant, as suggested by Newcomer and Apt,[4] were estimated. Comparing single-cycle natural gas plants (inefficient) with base-load coal plants is inappropriate since the most inefficient natural gas plants are only used for load-following, a service that coal plants cannot provide. Natural gas can only compete with coal plants when the most efficient power plants are used for base-load. Based on this understanding, an average coal plant was compared with an efficient natural gas (NGCC) plant. Mean life cycle GHG emissions from coal (point estimate) and an average coal power plant efficiency of 34%, were obtained from Jaramillo et al.[5] to estimate emissions per kWh of power generated using coal. On average, life cycle GHG emissions from the NGCC plant (with 50% efficiency, from Jaramillo et al.[5]) were about 50% lower than from the coal plant, per kWh of power generated. It is to be noted that this last comparison between electricity produced from coal and natural gas is intended to be for illustrative purposes only and to match the context of the other parts of this study considering the uses of natural gas that would lead to the most reductions. It is acknowledged that many other factors are likely to affect to the use of these fuels for power generation, such as the relative prices of the fuels, and whether they are to be used to meet baseline, seasonal or peak demands.

For the four scenarios where mean emissions reductions were observed by the use of natural gas (i.e., as a substitute for gasoline and diesel in small cars, for diesel in buses and for coal in power plants), a range of possible emissions reductions compared to the incumbent were specified. In the case of the CNG Civic, emissions reductions of 0−100 g CO_2e/mile from the baseline of 380 g CO_2e/mile (mean value of the emissions from the gasoline Civic) were considered. The interval was chosen such that it included the mean emissions reductions for the scenario, for instance, the CNG Civic achieves mean emissions reductions of 30 g CO_2e/mile, compared to the gasoline-powered Civic. For each value in this range, the probability of achieving the emissions reductions was estimated. This was done for all four scenarios and the emissions reductions along with their corresponding probabilities were compared to one another. However, the emissions reductions are based on the mean emissions from their respective incumbents, which differ in all four scenarios. For instance, in the scenario comparing the gasoline and CNG Civic cars, emissions reductions from the incumbent (gasoline Civic) are quantified on a per mile basis, while for the natural gas and coal power plant scenario, the emissions reductions from the incumbent (coal power plant) are quantified on a per kWh basis. These scenarios cannot be compared directly in this form, but could be compared if the quantities of natural gas used in each were the same. Therefore the range of emissions reductions were converted to emissions reductions per MJ of natural gas used, for consistency.

A set of curves was developed for all scenarios, representing a range of emissions reductions per MJ of natural gas used as a substitute fuel, and the corresponding probabilities of actually achieving those reductions. These curves are presented in Figure 3, along with mean emissions reductions for each scenario represented by crosses on the *x*-axis. If a CNG Civic were used in place of a diesel Jetta, the emissions reductions expected by comparing only the mean values of the emissions from the two cars is represented by the blue cross, at about 4 g CO_2e/MJ. Reading up from the cross to the blue curve applicable to this scenario (represented by the blue dotted line, to illustrate this scenario), the probability of actually achieving these emissions reductions is only about 50%. Similarly, the probability of achieving 0 g CO_2e/MJ of emissions reductions using the CNG bus (i.e., emissions from the CNG bus being unchanged versus the diesel bus) is about 65%, as indicated by the red curve, which implies that emissions from CNG buses have a 35% probability of being higher than the diesel bus. Such outcomes with low probabilities of achiving emissions reductions, or worse, likely increases in emissions, indicate a risk of failure in climate policy. In comparison, the quantities of emissions reductions possible as well as the probabilities of achieving them are higher if natural gas were used in place of coal at power plants. For instance the green curve shows about a 100% probability of achieving emissions reductions of up to 60 g CO_2e/MJ.

To summarize, the results in Figure 3 indicate that using CNG in small cars has very low probabilities of achieving any significant emissions reductions, with even lower probabilities for buses that are represented by the red curve. In fact, there is 10−35% probability that the emissions from using CNG as a transportation fuel might cause higher emissions than the incumbent fuels, as indicated by the points where these curves meet the *y*-axis at 0 g CO_2e/MJ reductions. On the other hand, NGCC plants have the potential for much more likely as well as more significant emissions reductions per MJ of natural gas consumed, indicating that using it for power generation is probably a better allocation of natural gas as a resource, to achieve GHG emissions reductions.

Sensitivity Analysis. The choice of GWP value could have serious impacts on the actual and relative results of this study. The analysis presented here uses a 100-year GWP value of 25 for methane.[20] Shindell et al.[45] recently modeled the effects of gas-aerosol interactions in the atmosphere and reported that the most-likely 100-year GWP estimate for methane is 33 and has a potential range from 25 to 41. Using a GWP value of 33 increases the life cycle emissions of natural gas from 66 g CO_2e/MJ to 70 g CO_2e/MJ (ranging between 64 and 77 g CO_2e/MJ), an increase of about 6% from the baseline estimates. The lower bound on the GWP estimate in Shindell's study[45] is the same as the baseline value used for this analysis. Using the upper bound on the GWP increases the value to 73 g CO_2e/MJ. The increase does not affect the qualitative conclusions of the study. Using the most-likely value in fact increases the probability of emissions from CNG being higher than incumbent transportation fuels (gasoline and diesel) from 10 to 35% originally, to 35−60%.

Another source of uncertainty is the assumption that all natural gas reported by EIA to be flared and vented (which was not disaggregated) is flared (a lower bound). If all of the natural gas is assumed to be vented (as an upper bound), the mean emissions increase from 66 g CO_2e/MJ to 70 g CO_2e/MJ (ranging between 63 and 81 g CO_2e/MJ). This also increases the probability of CNG emissions being higher than the incumbent transportation fuels from 10 to 35% orginially, to 35−60%. The probability of achieving emissions reductions of 60 g CO_2e/MJ of natural gas used in NGCC plants instead of coal is reduced to about 90% from being almost a 100% originally.

If a two-way sensitivity analysis were carried out, using the highest possible GWP value for methane reported by Shindell et al.[45] and assuming all of the natural gas reported by EIA to be flared and vented[25] was vented as methane, the mean value of life cycle GHG emissions increases from 66 g CO_2e/MJ to 79 g CO_2e/MJ. This increases the probability of emissions from CNG being higher than the incumbent fuels from 10 to 35% originally, to 80−100%. This also reduces the probability of achieving emissions reductions of 60 g CO_2e/MJ of natural gas used in NGCC plants instead of coal to about 50% from being almost a 100% originally. However, there is still almost a 100% probability of achieving emissions reductions of 30 g CO_2e/MJ of natural gas used at NGCC plants.

DISCUSSION

Results obtained from this study indicate that the uncertainty and variability in the life cycle GHG emissions associated with a unit volume of natural gas consumed in the U.S. is, at the least, 17% of the mean value. This range can be considered significant when compared to the actual differences in emissions between natural gas and other similar fuel types such as gasoline. For instance, the mean GHG emissions reductions benefit of using natural gas instead of gasoline in comparable light-duty vehicles was estimated to be 7% by this study. Therefore, the development of life cycle emissions estimates based on probabilistic modeling methods is recommended in addition to (or instead of) conventional deterministic approaches, especially when done in support of policy designed to mitigate climate impacts.

Uncertainty and variability have been analyzed together in this study, although these are inherently different. For instance, using state level data to estimate a probability distribution representing emissions at the national level attempts to quantify the spatial variability in these emissions. But as suggested in Venkatesh et al.,[18] this approach is appropriate since consumers and policy-makers have no way of differentiating between uncertainty and variability when considering a random unit volume of natural gas, given the level of aggregated data publicly available at present. However, although any variability could be considered as uncertainty, using disaggregated data when available would certainly improve the uncertainty estimates. It should also be pointed out that the uncertainty ranges derived in this study are likely to be underestimates, since literature and databases (such as those referred to in this study) usually report only average data without reporting variances.[18,46]

Although emissions reductions due to the use of CNG in vehicles are possible, comparing only mean values were found to somewhat overestimate the emissions reductions potential. Plots like Figure 3 could be used to decide when to promote specific fuels over others to achieve emissions reductions, and minimize the risk of policy failure. The plot could also be used to understand how the end-uses of a particular fuel type compare to each other, in order to determine the optimal uses of the fuel. Using natural gas for power generation in NGCC plants, as an alternative to coal, is likely to achieve higher and more certain GHG emissions reductions, since the differences between life cycle GHG emissions from the use of coal and natural gas at power plants almost certainly overcome any life cycle estimate uncertainties. Understanding the relative emissions reductions potential associated with different end-uses of natural gas is important considering that the quantity of natural gas available in the future will be constrained.

In addition, multiobjective optimization approaches can be applied when dealing with fuel types that might offer potential benefits in some areas, and drawbacks in others. Even if little or no reductions in GHG emissions are obtained, using CNG as a substitute for diesel could reduce non-GHG emissions such as particulate matter, CO and NO_X, as well as promote energy security within the U.S. by reducing dependence on petroleum imports. By using methods such as assigning subjective weights to the multiple objectives, Pareto fronts describing the optimal solution space can be developed, which can be used to identify the existing trade-offs and make better-informed choices. Further research is needed to expand this knowledge base.

■ ASSOCIATED CONTENT

S Supporting Information. Detailed explanation of methods and modeling parameters employed, additional results and references. This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author

*Phone:(412) 268-2940; fax:(412) 268-7813; e-mail: aranya@cmu.edu.

■ ACKNOWLEDGMENT

This material is based upon work supported by the U.S. National Science Foundation (grant number 0628084), the Energy Foundation, and the Steinbrenner Graduate Fellowship Program. Any opinions, findings, and conclusions expressed in this material are those of the authors and do not necessarily reflect the views of these organizations.

■ REFERENCES

(1) *Annual Energy Outlook 2011—Early Release*; U.S. Energy Information Administration:Washington, D.C., 2010; p 12.

(2) U.S. Energy Information Administration Annual Energy Outlook 2011, Reference Case Tables 2010; http://www.eia.doe.gov/oiaf/aeo/tablebrowser/#release=AEO2011&subject=15-AEO2011&table=50-AEO2011&region=0-0&cases=ref2011-d120810c.

(3) Yeh, S.; Sperling, D. Low carbon fuel standards: Implementation scenarios and challenges. *Energy Policy* **2010**, *38*, 6955–6965.

(4) Newcomer, A.; Apt, J. Near-term implications of a ban on new coal-fired power plants in the United States. *Environ. Sci. Technol.* **2009**, *43*, 3995–4001.

(5) 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.

(6) Arteconi, A.; Brandoni, C.; Evangelista, D.; Polonara, F. Life-cycle greenhouse gas analysis of LNG as a heavy vehicle fuel in Europe. *Appl. Energy* **2010**, *87*, 2005–2013.

(7) *Life-Cycle Emissions Study: Fuel Life-Cycle of U.S. Natural Gas Supplies and International LNG*; Advanced Resources International Inc., 2008.

(8) Okamura, T.; Furukawa, M.; Ishitani, H. Future forecast for life-cycle greenhouse gas emissions of LNG and city gas 13A. *Appl. Energy* **2007**, *84*, 1136–1149.

(9) Tamura, I.; Tanaka, T.; Kagjao, T.; Kuwabara, S.; Yoshioka, T.; Nagata, T.; Kurahashi, K.; Ishitani, H. M. S. Life cycle CO_2 analysis of LNG and city gas. *Appl. Energy* **2001**, *68*, 301–319.

(10) Odeh, N. a; Cockerill, T. T. Life cycle GHG assessment of fossil fuel power plants with carbon capture and storage. *Energy Policy* **2008**, *36*, 367–380.

(11) Ally, J.; Pryor, T. Life-cycle assessment of diesel, natural gas and hydrogen fuel cell bus transportation systems. *J. Power Sources* **2007**, *170*, 401–411.

(12) Kim, S.; Dale, B. E. LCA case studies life cycle inventory information of the United States electricity system. *Int. J. Life Cycle Assess.* **2005**, *10*, 294–304.

(13) *Energy Independence and Security Act of 2007*, Public Law 110-140, 2007.

(14) Schwarzenegger, A. *Executive Order S-01-07: Low Carbon Fuel Standard*; Sacramento, CA, 2007.

(15) Wang, M. *The Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET) Model*; Illinois, 2009.

(16) *Detailed California-Modified GREET Pathway for Compressed Natural Gas (CNG) from North American Natural Gas*, Version 2.1; California Environmental Protection Agency Air Resources Board, 2009.

(17) Mullins, K.; Griffin, W. M.; Matthews, H. S. Policy impacts of uncertainty in modeling life-cycle greenhouse gas emissions of biofuels. *Environ. Sci. Technol.* **2010**, *45*, 132–138.

(18) 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.* **2010**, *45*, 125–131.

(19) Plevin, R. J.; O'Hare, M.; Jones, A. D.; Torn, M. S.; Gibbs, H. K. Greenhouse gas emissions from biofuels... indirect land use change are uncertain but may be much greater than previously estimated. *Environ. Sci. Technol.* **2010**, *44*, 8015–21.

(20) Forster, P.; Ramaswamy, V.; Artaxo, P.; Berntsen, T.; Betts, R.; Fahey, D. W.; Haywood, J.; Lean, J.; Lowe, D. C.; Myhre, G.; Nganga, J.; Prinn, R.; Raga, G.; Schulz, M.; Dorland, R. V. Changes in atmospheric constituents and in radiative forcing. In *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*; Cambridge, United Kingdom, and New York, NY, 2007.

(21) *Natural Gas Navigator: Shale Gas Production*; U.S. Energy Information Administration, 2010; http://www.eia.gov/dnav/ng/ng_prod_shalegas_s1_a.htm.

(22) *Natural Gas Navigator: U.S. Natural Gas Gross Withdrawals and Production*; U.S. Energy Information Administration, 2010; http://www.eia.gov/dnav/ng/ng_prod_sum_dcu_NUS_a.htm.

(23) Jiang, M.; Griffin, W. M.; Hendrickson, C.; Jaramillo, P.; VanBriesen, J.; Venkatesh, A. Life Cycle Greenhouse Gas Emissions of Marcellus Shale Gas. *Environ. Res. Lett.* **2011**, *6*, 034014.

(24) *Natural Gas Navigator: Definitions, Sources and Explanatory Notes*; U.S. Energy Information Administration, http://www.eia.doe.gov/dnav/ng/TblDefs/ng_cons_sum_tbldef2.asp.

(25) *Natural Gas Navigator: U.S. Natural Gas Consumption by End Use*; U.S. Energy Information Administration, 2010; http://www.eia.gov/dnav/ng/ng_cons_sum_dcu_nus_m.htm.

(26) *2006 IPCC Guidelines for National Greenhouse Gas Inventories*; Intergovernmental Panel on Climate Change: Japan, 2006.

(27) *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2008*; U.S. Environmental Protection Agency Washington, DC.

(28) *Natural Gas Processing: The Crucial Link Between Natural Gas Production and Its Transportation to Market*; U.S. Energy Information Administration: Washington, D.C., 2006; p 11.

(29) U.S. Environmental Protection Agency State Inventory and Projection Tool http://securestaging.icfconsulting.com/sit/.

(30) *Natural Gas Navigator: Natural Gas Processed*; U.S. Energy Information Administration, 2010; http://www.eia.gov/dnav/ng/ng_prod_pp_a_EPG0_ygp_mmcf_a.htm.

(31) *State to State Natural Gas Pipeline Capacities and Flows*; U.S. Energy Information Administration, 2009, http://www.eia.doe.gov/pub/oil_gas/natural_gas/analysis_publications/ngpipeline/usage.html.

(32) *Interstate Natural Gas Infrastructure Map Book: 2009–2010*; Rextag Strategies, 2010; p. 350.

(33) Wilkinson, P.; Pierson, P. *Gas Facts 2007 Data*; Washington DC, 2009.

(34) U.S. Energy Information Administration Natural Gas Navigator: U.S. Natural Gas Imports by Country 2010, http://www.eia.gov/dnav/ng/ng_move_impc_s1_a.htm.

(35) Ruether, J.; Grol, E.; Ramezan, M. In *Second International Conference on Clean Coal Technologies for our Future*; Sardinia, Italy, 2005.

(36) *SimaPro Version 7.1*; PRé Consultants: Amersfoort, The Netherlands, 2007; http://www.pre.nl/.

(37) Heede, R. *LNG Supply Chain Greenhouse Gas Emissions for the Cabrillo Deepwater Port: Natural Gas from Australia to California*; Snowmass, CO, 2006.

(38) World Ports Distances http://distances.com/.

(39) PortWorld Distance Calculator—Ship Voyage Distance Calculator http://www.portworld.com/map/.

(40) *U.S. Liquefied Natural Gas Imports by Point of Entry*; U.S. Energy Information Administration, http://www.eia.gov/dnav/ng/ng_move_poe1_a_EPG0_IML_Mmcf_a.htm.

(41) Wang, M. Q.; Huang, H.-S. *A Full Fuel-Cycle Analysis of Energy and Emissions Impacts of Transportation Fuels Produced from Natural Gas*; Argonne, IL, 1999.

(42) Weber, C. L.; Jaramillo, P.; Marriott, J.; Samaras, C. Life cycle assessment and grid electricity: What do we know and what can we know?. *Environ. Sci. Technol.* **2010**, *44*, 1895–1901.

(43) Adams, R.; Horne, D. B. *Compressed Natural Gas (CNG) Transit Bus Experience Survey April 2009—April 2010*; National Renewable Energy Laboratory: Golden, CO, 2010.

(44) U.S. Department of Energy-Energy Efficiency & Renewable Energy http://www.fueleconomy.gov/feg/bymanu.htm.

(45) 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.

(46) Lloyd, S. M.; Ries, R. Characterizing, propagating, and analyzing uncertainty in life-cycle assessment. *J. Ind. Ecol.* **2007**, *11*, 161–179.