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Life Cycle Greenhouse Gas Emissions and Freshwater Consumption of Marcellus Shale Gas

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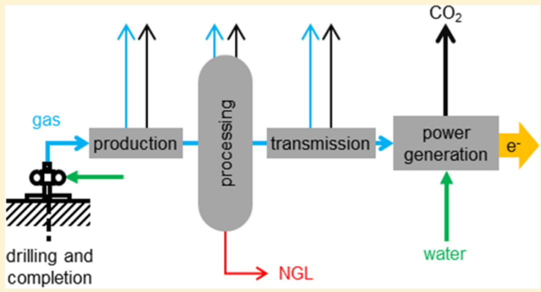
Supporting Information

ABSTRACT: We present results of a life cycle assessment (LCA) of Marcellus shale gas used for power generation. The analysis employs the most extensive data set of any LCA of shale gas to date, encompassing data from actual gas production and power generation operations. Results indicate that a typical Marcellus gas life cycle yields 466 kg CO_2eq/MWh (80% confidence interval: 450−567 kg CO_2eq/MWh) of greenhouse gas (GHG) emissions and 224 gal/MWh (80% CI: 185−305 gal/MWh) of freshwater consumption. Operations associated with hydraulic fracturing constitute only 1.2% of the life cycle GHG emissions, and 6.2% of the life cycle freshwater consumption. These results are influenced most strongly by the estimated ultimate recovery (EUR) of the well and the power plant efficiency: increase in either quantity will reduce both life cycle freshwater consumption and GHG emissions relative to power generated at the plant. We conclude by comparing the life cycle impacts of Marcellus gas and U.S. coal: The carbon footprint of Marcellus gas is 53% (80% CI: 44−61%) lower than coal, and its freshwater consumption is about 50% of coal. We conclude that substantial GHG reductions and freshwater savings may result from the replacement of coal-fired power generation with gas-fired power generation.



1. INTRODUCTION

In recent years "shale gas" has become an increasingly important energy resource. Like natural gas produced from other geological formations, it is composed of methane, a variety of other hydrocarbons, and various contaminants such as carbon dioxide and nitrogen. Unlike other sources of natural gas, shale gas is produced from low-permeability deep shale formations. In recent years, production of gas from such "tight" reservoirs was made possible by advances in horizontal drilling and improvements in hydraulic fracturing. These innovations have been employed to produce gas from the Barnett, Haynesville, Fayetteville, and Marcellus shales in the U.S., among others.

As a consequence of these technological developments, the U.S. EIA forecasted that natural gas production in the U.S. would increase 29% between 2010 and 2035.[1] However, some regions have had substantially higher increases in production already. For example, between 2009 until 2011, Marcellus shale development resulted in a 4-fold increase in gas production in Pennsylvania alone.[2] The increased availability of natural gas to consumers has resulted in a decrease in the price of gas for all consumers, including electrical utilities. As a consequence, the fraction of electricity generated from gas has increased substantially.[3]

Interest in the environmental impacts of shale gas has included greenhouse gas (GHG) impact and water use associated with horizontal drilling and completion practices, which include hydraulic fracturing. Comprehensive assessment of the environmental impact of shale gas requires consideration of its entire life cycle, including drilling, well completion, production, processing, pipeline transmission, and the end use of the processed gas by consumers.

These complete impacts may be estimated via life cycle assessment (LCA). When conducted in accordance with ISO 14040 and 14044 guidelines, of which the definition of scope and "functional unit" are paramount, an LCA will yield a "cradle to grave" estimate of the environmental impact of a product or process that can meaningfully distinguish alternatives in terms of their function or use.[4,5]

Howarth and co-workers were the first to publish a study evaluating GHG emissions from shale gas in the peer-reviewed academic literature. They claimed that methane emissions from shale gas are 30−100% higher than conventional gas.[6] They also claimed that the carbon footprint of shale gas (GHG emissions per low heating value of fuel) is 20−100% higher than the footprint of coal on the 20 year time horizon, and that the carbon footprints of shale gas and coal are comparable on the 100 year time horizon.[6]

Shortly thereafter, several LCAs of shale gas were published,[7−10] each of which used the ISO guidelines to evaluate the carbon footprint of shale gas from the well to a combined cycle gas turbine power plant. We refer the interested reader to the recent meta-analysis of these studies

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by Weber and Clavin, which comprehensively reviews the data sources, assumptions, and results of these LCAs.[11] The consensus of these studies was that life cycle GHG emissions from gas are about half of those from coal when the fuels are utilized for power generation (their only common use). However, as Weber and Clavin have noted, all of these studies made extensive use of assumptions or EPA emission estimates in the absence of data or measurements, for example, utilization of the EPA estimate for gas emitted to the atmosphere during the "flowback" phase of well completion.[12] Indeed, previous studies, including that of Howarth and co-workers, stressed the need for "better data" and measurements to quantify GHG emissions over the shale gas life cycle.

In this work, we present the results of a life cycle assessment of Marcellus gas based on ExxonMobil *field data* for drilling, completion, production and power plant operations, focusing on the carbon and water footprints of Marcellus gas from "well to wire" (i.e., drilling the well to generation of electricity at a power plant). We begin by defining the goal, scope and boundaries of our study in accordance with ISO guidelines. Next, we define our methodology and present key findings, including robust statistical metrics for the upper and lower limits of the GHG and water footprints. We also present sensitivity analyses of our results, identifying uncertain variables or features of the Marcellus gas LCA that are the most likely to have a significant effect on the total GHG emissions. Finally, we compare our results for the Marcellus gas footprints with results of other studies of coal, conventional gas and shale gas. We conclude with a discussion regarding the longer-term environmental consequences of power generation from Marcellus shale gas.

2. MATERIALS AND METHODS

2.1. Goal and Scope. The primary goal of this study was to assess GHG emissions over the life cycle of Marcellus shale gas from the well pad to generation of electricity at a combined cycle gas turbine (CCGT) power plant. We limited the assessment of GHG to CO_2, CH_4 and N_2O. Emissions were assessed in units of CO_2-equivalents as specified by the Fourth IPCC Annual Report (AR4).[13] CO_2-equivalency is a metric that compares the radiative forcing associated with a GHG relative to that of CO_2. Because GHGs have different atmospheric lifetimes, the IPCC reports "global warming potentials" (GWP) for each GHG for three time-horizons: 20 year, 100 year, and 500 year. In our study, we utilized 100 year GWPs, that is, 25 kg CO_2eq/kg CH_4 and 298 kg CO_2eq/kg N_2O, following the precedent of previously published LCAs of fossil fuels[7−11,14−16,28,30] and guided by Decision 2/CP.3 of the Kyoto protocol.[17]

The secondary goal of the study was to assess the life cycle freshwater consumption associated with shale gas. This includes water consumed for (1) hydraulic fracturing and (2) evaporative cooling at the power plant where the gas is used. Freshwater is also consumed indirectly via the life cycles of diesel and gasoline, which are used in various phases of the shale gas life cycle (e.g., well pad use and transportation). Although some processes in the shale gas life cycle result in the generation of water by way of combustion, we excluded this water from the scope of our assessment.

2.2. System Boundaries. The boundaries of the shale gas life cycle include drilling, well completion, wastewater disposal, production (i.e., delivery of gas from the well via gathering pipelines), treatment and processing, transmission (i.e., transportation of "pipeline quality gas"), and power generation at a natural gas combined cycle gas turbine (CCGT) power plant (Supporting Information (SI) Figure S1). We collectively denote as "Upstream" all phases of the gas life cycle preceding the power plant. Gas distribution networks, which deliver gas from transmission pipelines to nonutility customers (e.g., for home heating) were excluded from the assessment because power plants withdraw gas directly from transmission pipelines. Likewise, we excluded operations associated with the fractionation and use of natural gas liquids (NGLs) that are separated from raw natural gas at the processing plant: Such operations are best considered in LCAs of products manufactured from NGLs, which may include polyethylene, polypropylene, and liquefied petroleum gas (LPG, i.e. heating propane), among others.

2.3. Functional Unit. ISO specifies that LCAs must report environmental impacts in terms of a "functional unit", that is, impacts must be reported relative to the function of the assessed product. This permits comparisons of the impacts associated with alternative products, for example, gas electricity and coal electricity. In our LCA, Marcellus gas is a fuel for power generation. Therefore, we employed a functional unit of one MWh of power generated at the plant, and report GHG emissions and water consumption as kg CO_2eq/MWh and gal/MWh, respectively.

2.4. Information Sources. Most of the information used for the modeling of the drilling, completion, and production phases of the LCA was obtained from XTO Energy, a subsidiary of ExxonMobil (XTO). To our knowledge, this kind of information for shale gas has not appeared in the public domain. The composition of Marcellus gas employed in our study is reported in SI Table S1, and is typical of gas produced from the Marcellus shale in Southwestern Pennsylvania.[18] To evaluate the effect of compositional variability upon the carbon footprint, we also performed an analysis using a Marcellus gas composition typical of Northeastern Pennsylvania[18] (SI Table S8). The amounts of steel and cement required for the casing and isolation of the well were estimated from the design of CNX No. 3 in Greene County, Pennsylvania;[19] our analysis assumes complete cementing of the production casing. Some data associated with the hydraulic fracturing step of the gas life cycle are reported in Figure 1. Other data are included in SI Figure S2. CO_2 emissions from engine exhaust were modeled using an emission factor derived from XTO engine emissions and gross XTO production in 2011 (SI, Section 4).

To perform a material balance on methane from well to wire, one requires the gross production of gas from the well over its lifetime. "Ultimate recovery" is not known with absolute certainty prior to gas production. However, it may be estimated from production time series. To this end, we applied the method of Ilk and co-workers[20] to monthly production data for 222 Marcellus wells in Pennsylvania and West Virginia, which include 54 wells drilled and completed by XTO. The resulting "estimated ultimate recoveries" (EURs, Bcf) are reported in Figure 1C. Production data from other companies were obtained from the IHS Energy Well Production Database, which contains production data reported to states by operators.[21]

We did not have a sufficient amount of data to model certain production-phase GHG emissions. For these sources, we utilized EPA emission factors (EFs)[22] or regulatory emission limits[23] (SI Tables S4 and S5). Methane and noncombustive CO_2 emissions were estimated using an approach similar to the

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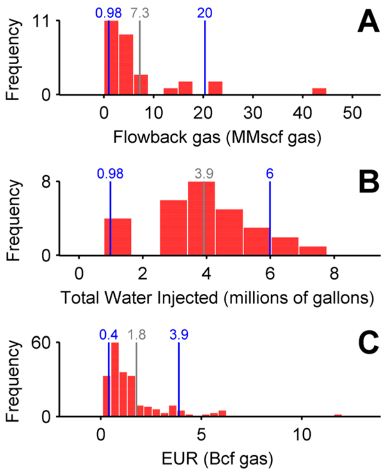

Figure 1. Key data employed in the assessment of the drilling and completion phases of the Marcellus shale gas life cycle. Amounts of flowback gas, water injected and EUR are reported on a "per well" basis,that is, the average EUR of a Marcellus shale gas well is 1.8 Bcf. Blue lines indicate the 10th and 90th percentiles of the data, and gray lines indicate average values. Note that the average amount of gas flared during flowback is 7.25×10^6 scf due to the influence of a single well; the median amount of flared gas is 3.88×10^6 scf. EPA[12], by contrast, assumes that 9.18×10^6 scf of gas are *vented* during flowback.

one employed by EPA for its annually published greenhouse gas inventory.[24] However, there were key differences, for example, adjustments to the EPA emission factors to account for the composition of raw Marcellus gas. Additional details are provided in the SI.

We modeled the processing and transmission stages of the gas life cycle using a combination of publicly available data and process simulation. Details are discussed in SI Section 5.

For our base case assessment of the power generation stage, we adopted the NETL model of a CCGT plant, which features an efficiency of 50.2% (HHV basis);[25] similar efficiencies are used in other LCAs of gas-fired power generation. We employed the EIA-923 power generation data file from 2010[26] to estimate the efficiencies of U.S. CCGT plants for our LCA of Marcellus gas used in the U.S. We adopted plant cooling water requirements from NREL[27] and NETL of 130–300 gal/MWh, with a median of 198 gal/MWh.

2.5. Material Balance and Allocation. The LCA was calculated by way of a material balance for the flow of raw natural gas from the source (the well) to various sinks, including noncombustive and combustive atmospheric emissions as well as the processing plant, where NGLs depart the system boundary. Fifty-four collections of data and randomly distributed parameters were included in the analysis. Gas reaching the power plant was assumed to be completely combusted, and the corresponding heat generation was converted to electrical output via the power plant efficiency. Impacts associated with flow dependent activities (e.g., trucking of materials used at or removed from the pad) were calculated separately. Total GHG emissions (kg CO_2eq) and freshwater consumption (gal) for each stage were then calculated from the material balances.

Since NGLs are constituents of raw gas, we accounted for their impacts via allocation. Fractions of the GHG and freshwater impacts associated with drilling, completion and gas processing were allocated to the NGLs and subtracted from the life cycle impacts of electricity generated from Marcellus gas. Following ISO guidelines,[5] we allocated impacts associated with pipeline quality gas and NGLs according to their energy content (HHV), that is, emissions and freshwater consumption associated with all stages up to and including processing were decreased by 19.7% (SI Table S1). However, we also investigated the effect of mole- and mass-based allocations upon our life cycle estimates (SI Section 7).

After allocation, the adjusted life cycle GHG emissions and freshwater consumption were normalized by the amount of electricity generated at the plant. We have grouped emission sources and freshwater consumers into thirty-seven activities or processes.

2.6. Calculation Approaches. We employed two types of calculations. In the first type, we employed the average values of all process data sets (e.g., flowback gas) and distributed variables (e.g., EPA emission factors) to conduct the LCA. The resulting life cycle GHG emission (kg CO_2eq/MWh) and freshwater consumption (gal freshwater/MWh) represent the

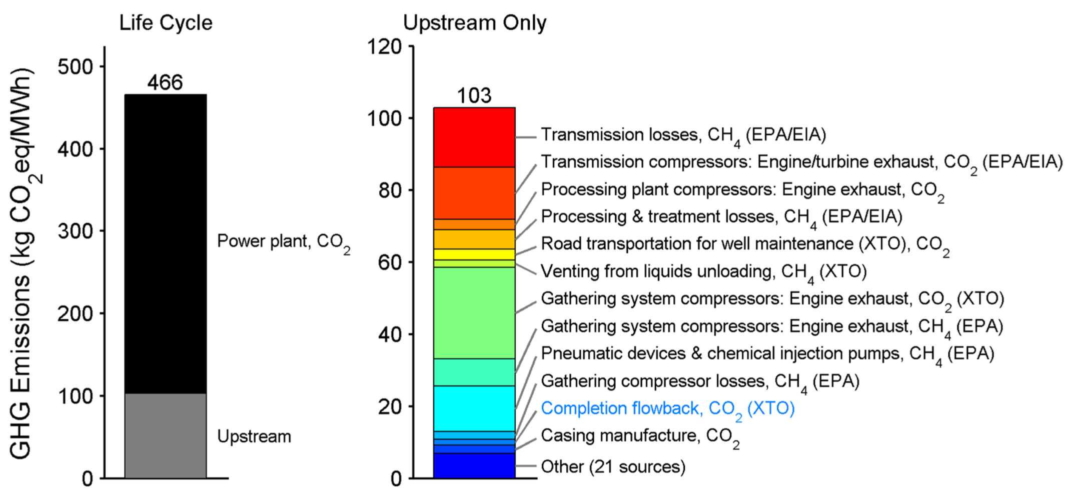

Figure 2. Breakdown of GHG emissions over a typical life cycle of Marcellus gas. The primary source of GHG emissions is the power plant, where most of the gas is burned. Gathering systems are the largest source of upstream GHG emissions, primarily due to the use of gas as fuel for compressor engines. Gas flared following well completion constitutes 1.9 kg CO_2eq/MWh, or 0.42% of the life cycle GHG emissions ("Completion flowback"). Emissions are reported in terms of 100 year GWPs[13].

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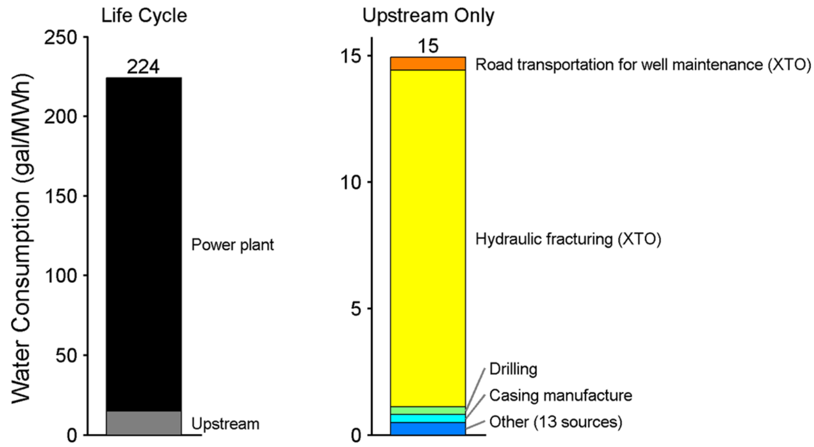

Figure 3. Freshwater consumption over the life cycle of Marcellus gas. The primary freshwater consumer is the cooling water system for the power plant, which withdraws freshwater and rejects it to the atmosphere as steam. Freshwater consumption due to hydraulic fracturing constitutes 6.2% of the freshwater consumption over the gas life cycle.

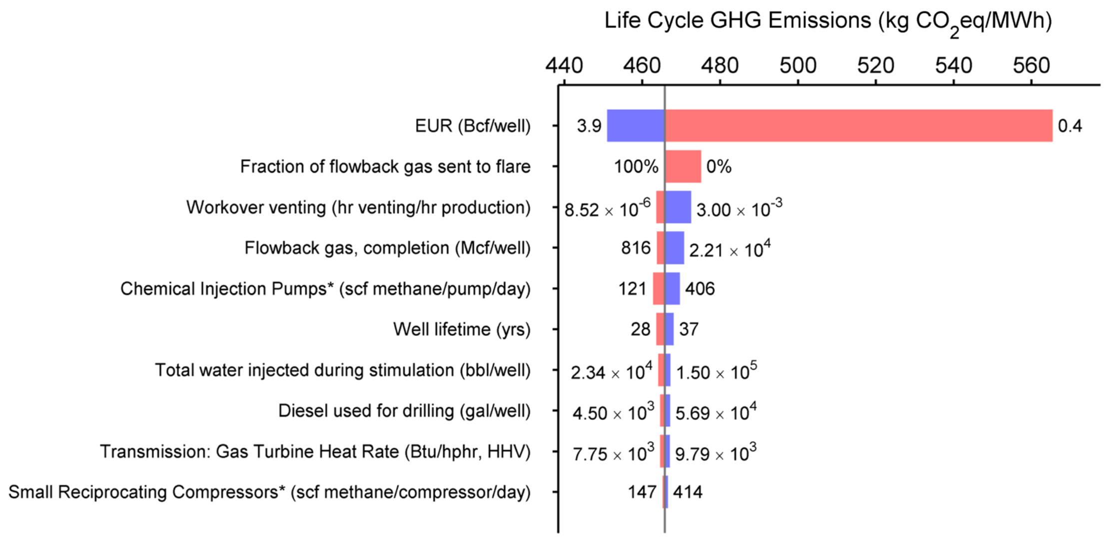

Figure 4. Sensitivity of the life cycle GHG emissions to key data and model parameters. Input parameters or data were varied between their 10th and 90th percentiles. Parameters marked with asterisks are EPA emission factors. Emissions are reported in terms of 100 year GWPs[13].

impacts associated with an "average" Marcellus gas life cycle from well to wire.

We also conducted analyses of all possible environmental impacts of power generation fueled by Marcellus shale gas using Monte Carlo simulation. For each "MC run", each input variable was selected *at random* from its distribution (e.g., EPA EFs) or data set (e.g., EUR). Life cycle emissions and freshwater consumption were then calculated using these values. The process was repeated 10 000 times using Crystal Ball (version 11), yielding distributions of life cycle emissions and freshwater consumption from which statistics (e.g., percentiles) may be estimated to sufficient precision. These distributions quantify the uncertainties associated with our life cycle estimates of GHG emissions and water consumption.

3. RESULTS

3.1. Base Case. Results of the "base case" LCA are reported in Figures 2 and 3. Key results are as follows:

- Life cycle GHG emissions are 466 kg CO_2eq/MWh generated (100 year GWPs, "Methods").
- Life cycle freshwater consumption is 224 gal/MWh.
- 77.9% of GHG emissions (363 kg CO_2eq/MWh) occur at the power plant.
- 93.3% of freshwater consumption (209 gal/MWh) occurs at the power plant.
- Hydraulic fracturing and its associated operations account for 1.17% of the life cycle GHG emissions and 6.15% of the life cycle freshwater consumption.

Nearly all upstream freshwater consumption is associated with hydraulic fracturing (13.7 gal/MWh). Approximately 1 gal/MWh of this is consumed in association with the life cycles of diesel and gasoline used for power generation at the pad or for transportation.

Gas engines used to drive gathering system compressors are the next largest source of GHG emissions, after the power plant. Gathering engine exhaust results in the emission of 25.3 kg CO_2/MWh, as well as 7.7 kg CO_2eq/MWh of methane. This methane emission results from incomplete combustion, and was calculated via an EPA EF. This EF was obtained from measurements of gas engine exhaust in 1992. If this EF is accurate, then gas engine exhaust is the second largest source of methane emissions over the gas life cycle: Of every 24

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molecules of methane emitted from "well to wire", four are emitted in the exhaust of gas engines.

3.1.1. Sensitivity and Representativeness. To evaluate the sensitivity of the LCA results to the life cycle inventory data, we independently varied each parameter within its 10th and 90th percentiles. The results of this analysis are reported in Figure 4. The life cycle impacts were most sensitive to the parameters at the top of the chart; only the top 10 most LCA-sensitive parameters are reported. The analysis clearly shows that the life cycle GHG emissions are most sensitive to EUR, in accordance with the results of sensitivity analyses of the Argonne and Shell LCAs.[8,9] With the exception of EUR, these key parameters are all specific to the pad phase of the gas life cycle.

Our analysis also revealed that the life cycle GHG emissions are sensitive to well lifetime, confirming the trends observed by Jiang and co-workers.[7] This results from the use of EPA EFs to model blowdowns and upsets associated with the production phase of the gas life cycle: Emissions were estimated by multiplying the EFs by a well lifetime.

3.1.2. Effect of EUR Upon Life Cycle GHG Emissions. The life cycle GHG emissions are sensitive to several of the key parameters due to the relationship between GHG emissions and the EUR. Absolute GHG emissions (kg CO_2eq) associated with drilling and completion occur before a well produces gas; they are *not related* to the EUR. By contrast, absolute GHG emissions from other phases of the life cycle (kg CO_2eq) and the power output from the plant (MWh) are *proportional* to the EUR. The normalization step in LCA entails division of emissions by the functional unit, that is, power generation. Thus, the normalized GHG emissions associated with drilling and completion (kg CO_2eq/MWh) are *inversely proportional* to EUR, whereas the normalized GHG for other stages of the life cycle are *independent* of EUR. For this reason the relative GHG emissions from the drilling and completion stages of the gas life cycle LCA will diminish with respect to the total amount of gas that a well produces.

We illustrate this dependency in Figure 5. Each point represents the result of a Monte Carlo simulation corresponding to a particular well EUR. The predicted life cycle GHG emissions are inversely related to EUR: The asymptotic limit of the life cycle GHG emission is 443 kg CO_2eq/MWh (80% CI: 440–450 CO_2eq/MWh).

3.1.3. Effect of Power Plant Efficiency Upon Life Cycle GHG Emissions. The results illustrated in Figure 4 were calculated using a constant power plant efficiency of 50.2% (HHV basis). In SI Figure S7, we report a sensitivity analysis that varies the efficiency between the 10th and 90th percentiles of U.S. efficiencies in 2010; in that year, the average efficiency was 48% (HHV basis). The results indicate that the LCA is more sensitive to power plant efficiency than any other factor other than EUR. This is a consequence of the functional unit of the study: power generation is proportional to efficiency. Therefore, life cycle emissions are inversely proportional to the power plant efficiency.

3.1.4. Effect of Raw Gas Composition and Allocation. In SI Figure S8, we report the effects of raw gas composition and method of allocation of coproducts upon the carbon footprint of Marcellus shale gas. If impacts associated with drilling, completion, production, and processing are allocated to pipeline quality gas and NGLs in proportion to the mass flow rates of the streams departing the processing plant, then the life cycle GHG emission decreases to 465 kg CO_2eq/MWh; allocation by mole yields a life cycle GHG emission of 473 kg CO_2eq/MWh. The apparent increase in life cycle GHG emissions due to molar allocation is a consequence of the higher molecular weights of ethane and propane relative to methane: allocation by mass or energy directs more of the GHG emissions to the NGL product.

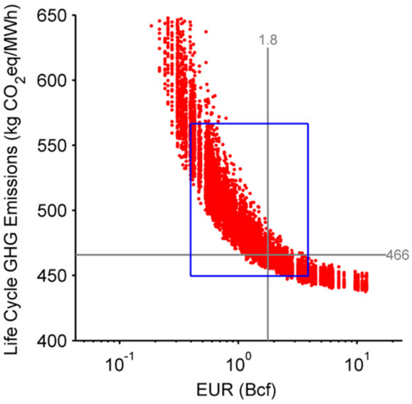

Figure 5. EUR-dependence of the life cycle GHG emissions from Marcellus gas used for power generation (50.2% efficiency, HHV basis). The results of 10 000 Monte Carlo simulations are illustrated (Materials and Methods); each point represents the results of an LCA using randomly selected inputs, including EURs. The blue box indicates the 10th and 90th percentiles of Marcellus EURs (cf. Figure 1) and the 10th and 90th percentiles of the life cycle emissions; gray lines indicate the averages. The life cycle GHG emissions decrease inversely with respect to the EUR of the gas well. Emissions are reported in terms of 100 year GWPs[13].

Marcellus shale gas produced from Northeastern Pennsylvania meets the HHV and inert specifications for its injection into a transmission pipeline. Therefore, no processing is needed and there is no NGL coproduct. As a result, there is no cryogenic separation process, and no need to use pipeline quality gas to fuel a booster compressor (SI Figure S1). Therefore, more gas proceeds to the power plant, and there are fewer GHG emissions. The net result is a decrease in the life cycle GHG emissions from 465 to 463 kg CO_2eq/MWh.

3.1.5. Comparison with Other Life Cycle Studies. Activities associated with hydraulic fracturing include flowback flaring, the operation of diesel-fueled frac pumps, and mining and transport of sand, which is the largest nonwater constituent of fluids injected into the well. They also include road transport of wastewater to disposal sites. Insofar as these activities comprise only 1.26% of the life cycle GHG emissions, we expect that the carbon footprints of Marcellus shale gas and many "conventional" gases produced from onshore wells will be essentially identical, provided that the wells have equivalent recoveries, compositions, completions (e.g., the flowback gas of both wells is flared), etc.

Indeed, our results are statistically consistent with results of most of the published LCAs of conventional gas and shale gas (SI Figure S11). However, the results of LCA of "Onshore Domestic Gas" published by NETL[28] do not fall within our 80% confidence interval for Marcellus gas. NETL reports a life cycle emission of 467 kg CO_2eq/MWh of *delivered* electricity of which 3.28 kg CO_2eq/MWh results from electricity distribution. Correcting for a 7% loss of electricity from plant to consumer, one obtains a "well to wire" GHG emission of 433 kg CO_2eq/MWh. The major difference between our results and those of NETL appear to originate in the modeling of the well pad and production stages: NETL utilizes a constant rate of gas

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production throughout the well life, whereas our analysis is based upon the EUR. In failing to account for the decline of production over the well lifetime, NETL may be effectively increasing the EUR to higher levels, thereby reducing the life cycle emissions (cf. Figure 5).

The aforementioned work of Howarth and co-workers[6] reports GHG emissions with respect to the lower heating value (LHV) of gas and uses non-IPCC GWPs[29] to express emissions of methane and CO_2 on a common basis. Using a 20 year time horizon, they predict a range of GHG emissions of 35−60 g C/MJ LHV; Using a 100 year time horizon, their range is 23−32 g C/MJ LHV. These may be converted to an electrical basis by dividing by the LHV efficiency of a CCGT plant (55% LHV, equivalent to 50% HHV). Expressed in terms of IPCC GWPs and a basis of electrical generation, Howarth's results are 830−1430 kg CO_2eq/MWh for a 20 year horizon, and 550−770 kg CO_2eq/MWh for a 100 year horizon. These are approximately two times larger than the GHG emissions reported by other peer-reviewed LCAs (SI Figure S11), including ours. In SI Figure S12, we express the results of our LCA using Howarth's basis of MJ LHV.

3.2. Marcellus Gas in the U.S. CCGT Fleet. Thus far we have only considered power plants operating at 50.2% efficiency (HHV basis). In practice, CCGT plants may operate over a range of efficiencies: Plants constructed in the early 1990s tend to generate power with lower efficiency, whereas newer plants will operate at efficiencies equal or greater to that used in our base case. In SI Figure S13, we report the distribution of CCGT plant efficiencies in 2010: Due to the persistence of older CCGT plants, 80% of U.S. plants operate in the range of 43−48% net efficiency (HHV basis). Newer plants operate at efficiencies higher than our base case.

In Figure 6 we contrast the distributions of GHG emissions associated with (1) a life cycle terminating at a plant with the base case efficiency of 50.2% HHV and (2) a life cycle terminating at a plant featuring an efficiency representative of the current U.S. CCGT fleet. The major consequences of replacing the base case plant efficiency with the efficiencies of the U.S. fleet are (1) an upward shift of emissions, concomitant with a lower average efficiency for the fleet and (2) an increase in the width of the distribution due to the introduction of plant-to-plant variability.

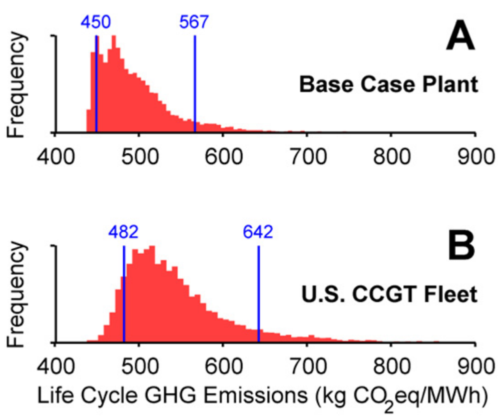

Figure 6. Effect of power plant efficiency upon the life cycle GHG emissions of Marcellus gas. Blue lines indicate the 10th and 90th percentiles of the data of emissions. (A) Monte Carlo simulations of the Marcellus gas life cycle terminating at a typical new base load power plant with an efficiency of 50.2% (HHV). (B) Monte Carlo simulations of the Marcellus gas life cycle terminating at any of the U.S. combined cycle power plants (cf. SI Figure S13). Emissions are reported in terms of 100 year GWPs[13].

The LCA results utilizing actual U.S. efficiencies may be utilized to statistically resolve the controversies regarding the life cycle GHG emissions from coal- and gas-fired power generation. To do so, we compared the distribution of life cycle GHG emissions from the U.S. CCGT fleet in Figure 6B with a distribution of the life cycle GHG emissions from coal-fired power generation. Recently, Venkatesh and co-workers[30] reported a distribution of life cycle emissions of U.S. coal in terms of kg CO_2eq/MJ heat (HHV). Like the results published by Howarth and co-workers, this does not reflect the functional use of coal, that is, power generation. Therefore, we integrated the results of Venkatesh and co-workers with the distribution of coal power plant efficiencies in 2010, as calculated from data in the EIA 923 Data File for that year. Integration was conducted via Monte Carlo (SI Figure S6).

In Figure 7 we report the distributions of life cycle GHG emissions for coal and Marcellus shale gas. The distributions

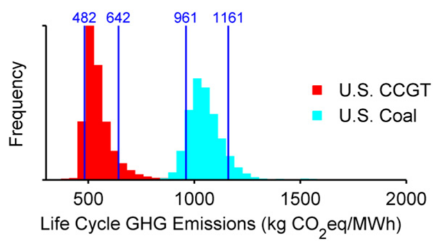

Figure 7. Comparison of base load Marcellus gas-fired power generation with base load coal-fired power generation. Coal emissions are reported for all actual coal-to-plant life cycles, calculated via integration of the results of Venkatesh and co-workers[30] with the 2010 efficiencies of coal plants as calculated from the EIA 923 Data File.[26] Gas emissions are reported for the use of Marcellus shale gas in every base load combined cycle plant in the continental U.S. (cf. plant efficiencies in SI Figure S13). Blue lines indicate the 10th and 90th percentiles of the emissions. Emissions are reported in terms of 100 year GWPs[13].

show no overlap: The highest possible life cycle GHG emission for Marcellus gas, corresponding to the lowest observed EUR and power plant efficiency, is lower than the lowest possible life cycle GHG emission for coal. Moreover, the key statistics for the distribution accord with the trends of previously published LCAs of coal and gas (SI Figure S11): The 10th and 90th percentiles (450 and 567 kg CO_2eq/MWh, respectively) for the Marcellus gas life cycle GHG emissions are approximately half those of coal. Additional statistical analysis reveals that the carbon footprint of Marcellus shale gas is 53% (80% CI: 44−61%) lower than power generated from U.S. coal when a 100 year time horizon is used. If one uses a 20 year time horizon to quantify GHG emissions from the coal and gas life cycles, one obtains similar results (SI Table S9 and Figure S8).

4. DISCUSSION

Absence of data is a key challenge in LCAs, potentially introducing uncertainty and bias. In this LCA, we have utilized large sets of production, emission and fuel use data from Marcellus gas wells spanning West Virginia and Pennsylvania. Although other LCAs of shale gas have appeared in the peer-reviewed literature, only ours has utilized actual production data for the key factors that affect the life cycle GHG emissions, for example, EUR, gas engine emission data, and flowback flaring. Moreover, our study is the first to explicitly consider the role of compositional variation in the shale gas life cycle.

Our results suggest that GHG emissions associated with natural gas may be reduced most directly by increasing the efficiencies of CCGT power plants or increasing the recovery of gas (EUR) from a well. The results in Figure 6 imply that nominal improvements in the operational efficiencies of CCGTs may decrease the median GHG emissions by as much as 50 kg CO_2eq/MWh, which is far larger than any single upstream source of emissions (Figure 2). Moreover, water consumption may be reduced most directly via reduction of the cooling water requirements at the power plant.

In the assessment of freshwater consumption, we have not attempted to differentiate water used in different watersheds (e.g., water used at the well pad for hydraulic fracturing vs cooling water at the power plant) or the degradation of water quality (e.g., discharge of nonconsumed water that is "blown down" at a power plant). Unlike atmospheric emissions of GHGs, freshwater consumption or changes to freshwater quality may have different impacts at the local level. Assessment of total freshwater consumption neglects consideration of water stress or scarcity: Availability of freshwater in the vicinity of the pad may be completely unrelated to availability of freshwater at the power plant, which may be hundreds of miles away. Furthermore, hydraulic fracturing and deep underground wastewater disposal (SI) permanently remove freshwater from the global ecosystem, whereas freshwater evaporatively consumed at the power plant may return to the surface freshwater supply via precipitation.

In our LCA, we have not considered water vapor generated via combustion of gas. This process yields 9.3 million gallons of water (if condensed to liquid form) over the life cycle of a well. By contrast, 3.6×10^6 gal/well is consumed on average for Marcellus wells due to hydraulic fracturing and wastewater disposal. Although the difference implies a net global ecosystem gain of 5.7×10^6 gal/well, this water may not precipitate in the same region from which freshwater was withdrawn. We view the water consumption and water quality areas as topics for additional research to better define appropriate metrics and potential impacts.

One of the key findings of our study is that only 1.17% of the total GHG emissions are specific to Marcellus shale gas production and processing. This is well within the range of uncertainty associated with the life cycle emissions. Therefore, for all practical purposes, the life cycle GHG emissions from shale gas and conventional gas are statistically indistinguishable. That said, wells are defined as "conventional" if the permeability of the gas-yielding formation exceeds a certain number (e.g., 0.1 millidarcy[31]). This definition neglects geological and chemical properties of the reservoir that in turn may affect the EUR. Considering the strong effect of EUR (Figure 5) upon life cycle emissions, we conclude that there may be differences between the life cycle GHG emissions of "conventional" gas and Marcellus shale gas if there are significant differences in the EURs of their wells.

Our analysis made use of some external information. Most notably, we made use of EPA EFs[22] to model fugitive emissions from equipment that is common to all natural gas operations, regardless of whether they are associated with shale gas or conventional gas wells. Emission factors describing new technology may be considerably lower than those utilized by EPA. For example, two recent studies called key EPA EFs into question: El Paso Energy Corporation presented measurement-based emission factors for the processing and transmission phases of the gas life cycle.[32] These emission factors, which ostensibly represent U.S. midstream operations in 2011, are an order of magnitude lower than the EPA EFs. Therefore, it is possible that we have overestimated methane emissions from the processing and transmission phases of the gas life cycle. Similarly, API and ANGA[33] recently presented new EFs for liquids unloading, calculated from data provided by API members. EPA will employ these improved EFs in its forthcoming Inventory.[34] As noted by Burnham and co-workers,[9] such data-based EFs may play a key role in distinguishing the carbon footprints of certain "conventional" gases (i.e., those obtained from wells requiring liquids unloading) and shale gas.

The major emissions sources highlighted in Figure 2 and sensitivities of our LCA to key variables and assumptions reported in Figure 4 (and SI Figures S9 and S10) suggest priorities for future work. First and foremost, we recommend that gas EFs should be updated to reflect current operations, particularly those for chemical injection pumps and gathering compressors (cf. Figure 2). This will soon be possible: EPA is now receiving data from U.S. gas producers as a result of new GHG emission reporting rules ("Subpart W"); these data may in turn be utilized to update the EPA EFs. Another possible area of improvement is the characterization of well lifetime. As the Marcellus shale gas play matures and more production data become available (e.g., production curves), better predictions of well lifetime may be estimated. Finally, more accurate information regarding freshwater consumption at U.S. CCGT power plants will help improve estimates of water consumption over the shale gas life cycle from well to wire.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Figures S1−S13, Tables S1−S10, and text. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

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■ ABBREVIATIONS

EUR expected ultimate recovery
EPA Environmental Protection Agency
EIA Energy Information Administration
GHG greenhouse gas
CCGT combined cycle gas turbine
NGCC natural gas combined cycle

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HHV higher heating value
LHV lower heating value
BD blowdown
NREL National Renewable Energy Laboratory
UMD University of Maryland
CMU Carnegie Mellon University
NETL National Energy Technology Laboratory
GWP global warming potential
IPCC Intergovernmental Panel on Climate Change
GRI Gas Research Institute
LCA life cycle assessment
EUR estimated ultimate recovery
AR4 Fourth Annual Report of the IPCC
LPG liquefied petroleum gas
NGL natural gas liquids

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