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Allocating Methane Emissions to Natural Gas and Oil Production from Shale Formations

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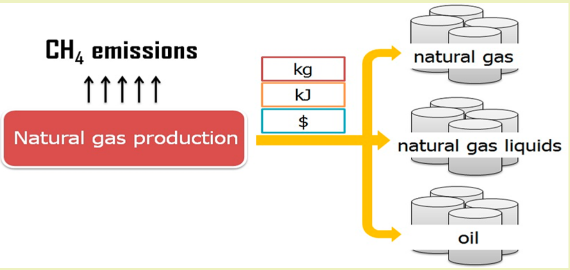
S *Supporting Information*

ABSTRACT: The natural gas supply chain includes production, processing, and transmission of natural gas, which originates from conventional, shale, coal bed, and other reservoirs. Because the hydrocarbon products and the emissions associated with extraction from different reservoir types can differ, when expressing methane emissions from the natural gas supply chain, it is important to allocate emissions to particular hydrocarbon products and reservoir types. In this work, life cycle allocation methods have been used to assign methane emissions from production wells operating in shale formations to oil, condensate, and gas products from the wells. The emission allocations are based on a data set of 489 gas wells in routine operation and 19 well completion events. The methane emissions allocated to natural gas production are approximately 85% of total emissions (mass based allocation), but there is regional variability in the data and therefore this work demonstrates the need to track natural gas sources by both formation type and production region. Methane emissions allocated to salable natural gas production from shale formations, based on this work, are a factor of 2 to 7 lower than those reported in commonly used life cycle data sets.



KEYWORDS: *Natural gas, shale gas, oil production, allocation of emissions, methane, GHG*

■ INTRODUCTION

A number of studies have examined greenhouse gas footprints of natural gas supply and use chains, and many of these studies have compared natural gas footprints to the greenhouse gas footprints for the production and use of other fuels.[1−6] A key factor in assessing the greenhouse gas footprint of natural gas systems is the quantification of methane emissions. Reported estimates of methane emissions from natural gas systems are characterized by a wide range of values and a significant degree of uncertainty,[7−9] and contributing to that variability and uncertainty are the methods used in attributing methane emissions from oil and gas systems to natural gas. In performing comparisons between natural gas and other fuel systems, it is important to recognize that parts of the natural gas supply chain, particularly production operations, produce both natural gas and liquid products. Coproduction of natural gas and liquid products is particularly common in some of the most rapidly growing shale gas production regions, such as the Eagle Ford region in Texas.[10] Associated gas from oil production is also extensive. In 2011, over 20% of the gross natural gas production was "associated gas" from oil wells.[11] A summary of natural gas flow from the well-head to end-users for 2011 is summarized in Figure S1-1 in the Supporting Information. Overall, the multiple and variable products associated with various types of natural gas production activities, and the diversity of natural gas sources, make it important to differentiate between sources of natural gas when assigning greenhouse gas emissions to the natural gas supply chain.

Recently, Allen et al.[12,13] have reported measurements of methane emissions from natural gas production sites, including wells that produced only gas and wells that produced both gas and liquids. All of these wells were classified by their operators as gas wells, all were in shale formations, and all were hydraulically fractured. Methane was the focus of these measurements because emissions of methane, a potent greenhouse gas, can significantly impact the supply chain greenhouse gas footprints of natural gas, relative to other fuels. Activities sampled included well completion flowbacks, liquid unloadings, pneumatic device operation, and equipment leaks from wells in routine operation. These data can be used to develop methane emission footprints of the shale gas production portion of the natural gas supply chain, with emissions allocated to the multiple products from the wells. For each of the wells sampled by Allen et al.,[12,13] gas composition,

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Table 1. Ratio of Methane Emissions Allocated to Each Product (Natural Gas, Natural Gas Liquids, and Oil) Divided by Its Respective Production, Based on a Mass Allocation, Energy Allocation, and Economic Value Allocation of Methane Emissions[a]

	pneumatic controllers	pneumatic pumps	equipment leaks	total
mass allocation				
emitted methane (scf)/dry natural gas (scf)[b] (%)	0.08	0.02	0.02	0.11
emitted methane (Mg) /produced natural gas liquids (Mg)[c] (%)	0.14	0.002	0.01	0.16
emitted methane (Mg) /produced oil (Mg) (%)	0.11	0.0002	0.01	0.12
energy allocation				
emitted methane (scf)/dry natural gas (scf)[b] (%)	0.08	0.02	0.02	0.12
emitted methane (Mg)/produced natural gas liquids (Mg)[c] (%)	0.13	0.002	0.01	0.15
emitted methane (Mg)/produced oil (Mg) (%)	0.10	0.0002	0.01	0.12
economic value allocation				
emitted methane (scf)/dry natural gas (scf)[b] (%)	0.07	0.02	0.02	0.11
emitted methane (Mg)/produced natural gas liquids (Mg)[c] (%)	0.17	0.005	0.02	0.19
emitted methane (Mg)/produced oil (Mg) (%)	0.16	0.0004	0.02%	0.17

[a]The values represent the ratio of the sum of emissions divided by the sum of production across all sites where each specific product is produced. From a total of 150 production sites, 144 sites reported natural gas production (for 3 sites production data was reported as "not available"; three sites reported zero gas production), 39 sites reported oil production, and 51 reported natural gas liquids production. [b]Dry natural gas refers to salable natural gas (also known as dry natural gas, referring to the remaining gas once the liquefiable hydrocarbon portion, propane and heavier, has been removed,[17] with the remaining nonmethane hydrocarbon gases,[14] being allocated to natural gas liquids. [c]Produced natural gas liquids, NGL refers to the remaining nonmethane hydrocarbon gases that are not part of salable natural gas. (For an expanded explanation of how nonmethane hydrocarbon gases are split between salable natural gas and natural gas liquids see the Supporting Information, Section S5.)

gas production and liquid production data are available.[13] The wells generally had high gas to oil production rate ratios (>12 500 standard cubic feet (scf) of gas per barrel of oil). This work reports allocations of methane emissions to natural gas and other gas and liquid products, based on a data set of 489 gas wells in routine operation and 19 well completion events.

■ METHODS

Allocation methods are commonly used in the life cycle analysis of emissions from supply chains when processes produce multiple products. For the case of a gas well that produces hydrocarbons that will eventually be separated into pipeline quality natural gas, natural gas liquids, and liquid hydrocarbon products, emissions from devices that handle all the products (e.g., a separator), should be allocated among multiple products. The most commonly used allocation methods are based on energy, mass, and economic value.[14−16]

In an energy based allocation, a hypothetical well that produces 6000 standard cubic feet (scf) of gas for every barrel (bbl) of hydrocarbon liquid would generate equal amounts of energy as gas and liquid products, assuming a heating value of 1000 BTU/scf for gas and 6 million BTU/bbl for hydrocarbon liquid. For this simple example, if an emission allocation is based on energy, half of the emissions from the well would be assigned to the gas and half to the liquid product. In contrast, a mass based allocation, for the same simple example, would allocate 60% of the emissions to the gas and 40% to the oil, based on a gas density of 25 g/scf and an oil density of 100 kg/bbl. An economic value based allocation, based on prices of \$3.50 per thousand scf (Mscf) for gas and \$90 per bbl for hydrocarbon liquid, would assign 19% of the emissions to gas and 81% to the liquid.

The gas leaving a well site will typically contain quantities of ethane, propane, butane, and heavier hydrocarbons and nonhydrocarbons. These products are largely removed from the methane in the gas before the product is supplied as "salable" or "dry" natural gas. In this work, emissions from well sites will initially be allocated to liquid products and to specific gas phase molecular species (methane, ethane, propane, butanes and pentane, and heavier hydrocarbons). The emissions allocated to the gases will then be attributed to three main products: (1) salable natural gas (also known as dry natural gas, referring to the remaining gas once the liquefiable hydrocarbon portion has been removed);[17] the composition of salable gas is based on commonly used life cycle analysis datasets for natural gas: 92.8% methane, and up to 5.54% nonmethane hydrocarbon gases (by mass%, the rest is N_2, CO_2, H_2S, and H_2O),[18] (2) natural gas liquids, which will be assumed to be the remainder of the hydrocarbon gas leaving the well, and (3) hydrocarbon liquids (oil) for which methane emissions will be assigned as a mixture.

Based on the assumed composition for salable natural gas, if the gas produced at a site has a nonmethane hydrocarbon content of up to 5.54% nonmethane hydrocarbon (mass), all nonmethane hydrocarbons are considered as part of salable natural gas (for that site there would be no natural gas liquids production). On the other hand, if the gas produced at each site is wetter (>5.54% nonmethane hydrocarbon gases), nonmethane hydrocarbon gases are assigned to salable natural gas, starting with ethane and then adding subsequent heavier hydrocarbons, until the 5.54% threshold is reached, the remaining portion of the gas leaving the well is considered natural gas liquids.

Properties and assumed economic values for each of these materials are provided in the Supporting Information (Table S2-1); the Supporting Information also includes an expanded explanation of how nonmethane hydrocarbon gases are split between salable natural gas and natural gas liquids (Supporting Information, Section S5). Calculated across all sites, the weighted average composition (% mass) of salable natural gas is 97.3% methane, 2.61% ethane, 0.120% propane, 0.007 butanes, and 0.003% pentane and heavier hydrocarbons. For natural gas liquids, the weighted average composition is 38.6% ethane, 27.8% propane, 18.8% butanes, and 14.8% pentane and heavier hydrocarbons (% mass).

For the case of the energy based allocation, a single heating value of 1027 BTU/scf (higher heating value) was assigned to salable natural gas (Table S2-1, Supporting Information); this heating value is commonly assumed in life cycle datasets.[5,18,20] A similar value is obtained when the mass weighted average heating value of salable natural gas across all sites is calculated (1022 BTU/scf), based on the gas composition. For natural gas liquids, an average heating value is derived for each site, based on its particular natural gas liquids composition. (mass weighted average heating value of natural gas liquids across all sites is 2349 BTU/scf (higher heating value)) (see the Supporting Information, Table S6-1).

Emissions for each product were allocated based on mass, energy and economic value, for each product (salable natural gas, natural gas liquids, and oil), for each of the individual sampling sites that were reported by Allen et al.[12,13] Average normalized emissions for each product are reported as the sum of emissions over all sites attributed to a particular product, divided by the sum of production of that product, over all sites. So, for example, the emissions attributed to natural gas liquids would be summed over all sites then divided by the production of natural gas liquids, summed over all sites.

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Table 2. Ratio of Methane Emissions from Completion Flowbacks Allocated to Specific Gas Phase Molecular Species (Methane, Ethane, Propane, Butanes and Pentane and Heavier Hydrocarbons) and to Each Product (Natural Gas, Natural Gas Liquids, and Oil) Divided by Its Respective Production[a]

	mass allocation	energy allocation	economic value allocation
emitted methane (scf)/dry natural gas (scf) (%)	0.002	0.002	0.001
emitted methane (Mg)/produced natural gas liquids (Mg) (%)	0.003	0.003	0.004
emitted methane (Mg)/produced oil (Mg) (%)	0.002	0.002	0.003

[a]Results are shown in mass, energy, and economic value basis.

Table 3. Ratio of Total Methane Emissions from Completion Flowbacks, Pneumatic Devices and Leaks and Routine Emissions) Allocated to Each Component (Natural Gas, Natural Gas Liquids, and Oil) Divided by Its Respective Production[a]

	mass allocation	energy allocation	economic value allocation
emitted methane (scf)/dry natural gas (scf) (%)	0.12	0.12	0.11
emitted methane (Mg)/produced natural gas liquids (Mg) (%)	0.16	0.15	0.20
emitted methane (Mg)/produced oil (Mg) (%)	0.12	0.12	0.18

[a]Results are shown on mass, energy, and economic value bases.

Production for the sites can be reported either as instantaneous production rate at the time of the measurement or as lifetime well production volume. In this work, the lifetime well production will be reported as an estimated ultimate recovery (EUR), over 30 years, for each well. The EUR is estimated for each well using the age of the well at the time of the measurement, the instantaneous production at the time of the measurement, and an assumed production decay rate over 30 years of production, as described in the Supporting Information (see the Supporting Information, Section S3). Sensitivity analyses, using instantaneous production rates, shorter well life, and alternative production decay assumptions, are also reported (see the Supporting Information, Sections S8−S10).

Results in this work are reported as a ratio of emissions to total production. For emissions from wells in routine production, the numerator in the ratio is total estimated emissions, over 30 years, based on a measured instantaneous emission rate and an assumption that emissions (except for well completions) remain constant over 30 years. The denominator is the EUR from the well, over 30 years. For completion events, which are assumed to only occur once over the life of a well, the numerator is the emissions from the single event. The denominator is the estimated ultimate recovery from the well, over 30 years.

RESULTS AND DISCUSSION

Emissions from Wells in Routine Production. Table 1 reports emissions from pneumatic controllers, pneumatic pumps, and leaks from equipment sampled by Allen et al.[12,13] Allocated emissions for individual sites are shown in the Supporting Information, Sections S4−S7. To arrive at total emissions for each site, emissions from leaks, pneumatic controllers and pneumatic pumps are summed. All leaks at sites were sampled, but not all controllers or pumps. For controllers and pumps, the average emission rate for each site was multiplied by the total number of devices reported for that site. Averages over multiple sites are taken on a production weighted basis, as described in the Methods section. Emissions assigned to each product are summed over all sites and divided by the production rate of that product, summed over all sites.

If the instantaneous emission rate is normalized by instantaneous production rate at the time of the measurement, emissions from pneumatics and leaks from wells in routine production are 0.0003 scf of methane per scf of produced natural gas, compared to the 0.0011−0.0012 scf of methane per scf of produced natural gas reported in Table 1, when emissions over 30 years and an EUR based on a well lifetime of 30 years is assumed (see the Supporting Information, Section S9). The difference is due to the relatively young age of the wells in the data set (average age of wells, weighted by gas production is 1.3 years), and the assumption that emissions stay constant over the life of the well. Measurements of emissions from pneumatic controllers, pneumatic pumps, and equipment leaks performed by Allen et al.[12,13] showed no proportional relationship between emissions and production, indicating that emissions from these categories can be considered as relatively constant throughout the life of the well. Therefore, for the purposes of this analysis, emissions during the life of a well are assumed to be constant. If the assumed well life is reduced to 15 years, emissions are reduced by half but production is only reduced by, on average, 22% (see the Supporting Information, Section S8). This leads to an estimated natural gas emission rate of 0.0007 scf of methane per scf of produced natural gas. Section S10 of the Supporting Information provides additional EUR calculations, under different assumptions about the rate of decline in production. Under such scenarios, estimated natural gas emissions range from 0.0006 to 0.0013 scf of methane per scf of produced natural gas, which are similar in magnitude to the results for the base case production decay assumption. Large basin-specific historical production datasets would allow refining the EUR calculations presented in this work. Because the goal of this work is to characterize the direction and magnitude of normalized emission changes under various allocation schemes, the remainder of this work will use the base case production decay assumption.

Emissions from Completion Flowback Events. Allen et al.[12,13] also made direct measurements from 27 well completion events. Production data were available for 19 of these 27 wells; 12 report both gas and oil production, and 7 report production of gas only. Table 2 shows the average ratio of methane emissions from the 19 completions, divided by the total estimated ultimate recovery of each component, over 30 years, from the 19 wells. This average represents the sum of emissions divided by the sum of production across the 19 measured events. (Ratios and allocated emissions for each separate event are shown in the Supporting Information, Section S11.)

Total Measured Emissions. Table 3 shows the sum of the average completion, pneumatic device and leak measurements. The contribution of completion events is small compared to the emissions from wells in routine production. For emissions allocated to natural gas, the three allocation methods show similar results. The mass based allocation is used in the rest of

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Table 4. Description of Emission Sources Not Directly Measured by Allen et al.[12] That Are Considered in the EPA GHG National Inventory, Showing How the Emissions Are Allocated to the Products Considered in This Work[a]

EPA GHG inventory activity	net emissions (Gg methane/yr)	allocate emissions to: NG + NGL + oil[b]	NG + NGL[c]	oil only
refractures	143	x		
gas wells without HF	13	x		
gas wells with HF	15	x		
separators	57	x		
meters/piping	54		x	
heaters	18	x		
dehydrators	16		x	
workovers without HF	0.3	x		
liquids unloading (without plunger lifts)	149	x		
liquids unloading (with plunger lifts)	108	x		
Kimray pumps	185		x	
condensate tanks without controls	94			x
condensate tanks with controls	52			x
gas engines	227		x	
dehydrators vents	41		x	
small reciprocating compressors	49		x	
large reciprocating compressors			x	
large reciprocating stations			x	
pipeline leaks	90		x	
well drilling	0.4	x		
vessel blowdowns	0.4		x	
pipeline blowdowns	2		x	
compressor blowdowns	2		x	
compressor starts	3		x	
pressure relief valves	0.4		x	
mishaps	1	x		
total emissions allocated to NG		429 Gg	630 Gg	

[a]NG = natural gas, NGL = natural gas liquids. The column showing net emissions refers to total emissions from each category before the allocation to the corresponding products. Last row shows total emissions after allocation. (Rationale for the disaggregation of each category is provided in Table S11-1 (Supporting Information).) [b]For categories that allocate emissions to all three final products (NG + NGL + oil), 85.1% of the emissions from each category are allocated to natural gas (percentage based on the previously described energy basis allocation). [c]For categories that allocate emissions to natural gas and natural gas liquids only (NG + NGL), 94.1% (percentage based on the previously described energy basis allocation) of the emissions from each category are allocated to natural gas.

the analyses presented in this work. The economic value allocation presents challenges due to the variability and potential fluctuation of the prices of each product.

Emissions from pneumatic controllers, pneumatic pumps, leaks from equipment and well completion flowback events (categories that were directly measured by Allen et al.[12,13]) represent 0.11%−0.12% (vol) of the total natural gas production when emissions are allocated among natural gas, natural gas liquids and oil. For those same categories, the EPA national greenhouse gas inventory (data for calendar year 2012, released in 2014) reports a total of 792 Gg of methane/yr or 0.17% (a difference of approximately 50%), on a volume basis of total dry natural gas production (24.1 trillion cubic feet of natural gas produced in the US in 2012[11]) when allocation to coproducts is not considered.

Table 4 lists additional sources of methane emissions from natural gas production that are quantified in the EPA national greenhouse gas inventory, but were not measured or were measured in very small sample sizes by Allen et al.[12,13] Table 4 disaggregates these emissions, based on whether the equipment handles natural gas (NG) and natural gas liquids (NGL) alone (NG + NGL), oil alone, or all products from the well (NG + NGL + oil). The rationale for the disaggregation of each source into the three products is shown in Table S12-1 of the Supporting Information. For categories that allocate emissions to all three final products (NG + NGL + oil), 85.1% of the emissions from each category will be allocated to natural gas. This 85.1% is based on the ratio of the average mass content in natural gas to the mass in all well products, over all wells on which measurements were made. For source categories that allocate emissions to natural gas and natural gas liquids only (NG + NGL), 94.1% of the emissions are allocated to natural gas. This value is based on the ratio of the average mass content in natural gas to the combined mass NG and NGL products, over all wells on which measurements were made. Combined, these mass based allocations for natural gas production sources not directly estimated by Allen et al.,[12,13] allocate an estimated 1060 Gg of methane emissions to natural gas product, which is 0.24% (volume) of the 22.9 trillion cubic feet of dry natural gas produced in the US in 2011. Because these national estimates capture a population of wells of all ages, it is assumed that the percentage of natural gas emitted (0.24%) would be the same for current emissions from all wells of all ages divided by current production of all wells of all ages or well lifetime emissions divided by EUR.

If these additional emissions are added to the allocated emissions from pneumatics, equipment leaks, and completion flowbacks described in this work (0.12% of natural gas

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Table 5. Methane Emissions Allocated to Each Salable Product (Natural Gas, Natural Gas Liquids, and Oil), Total Estimated Ultimate Recovery (EUR) for Each Product and Ratio of Emissions to Production, Based on a Mass Allocation[a]

	Appalachians (164 wells)	Gulf Coast (146 wells)	Mid Continent (76 wells)	Rocky Mountains (78 wells)
total methane emissions (Mg)	15,801	28,143	15,975	4272
methane emissions allocated to natural gas (Mg)	15,736	21,288	13,994	2175
methane emissions allocated to natural gas liquids (Mg)	64	1928	1201	1394
methane emissions allocated to oil (Mg)	2	4927	780	703
EUR natural gas (Mg)[b]	23,770,765	20,371,250	2,729,781	363,904
EUR natural gas liquids (Mg)[b]	38,492	2,505,534	138,572	265,674
EUR oil (Mg)[b]	1037	5,014,564	114,540	210,754
emitted methane (scf)/dry natural gas (scf) (%)	0.07	0.10	0.51	0.60
emitted methane (Mg)/produced natural gas liquids (Mg) (%)	0.16	0.08	0.86	0.51
emitted methane (Mg)/produced oil (Mg) (%)	0.19	0.10	0.68	0.33

[a]The values represent the ratio of the sum of emissions divided by the sum of production across all sites where each specific product is produced, considering a 30 year well lifetime and for each region. For each region, the number of sampled wells (within the sampled sites) is shown. [b]Cumulative EUR per wells sampled.

production), the total methane emissions would represent 0.36% (vol) of total natural gas production, a value that is lower than the 0.52 vol %,which is the ratio of the 2300 Gg of methane emissions reported by Allen et al.,[12] not accounting for coproduct allocation, divided by the 2011 US total dry natural gas production (22.9 trillion cubic feet[15]). This is a difference of over 40%.

Regional Analysis. Allen et al.[12,13] reported measured emissions from natural gas production sites from four different regions in the United States: Appalachian (45 sites), Gulf Coast (58 sites), Mid Continent (23 sites), and Rocky Mountains (18 sites). Methane emissions (sum of emissions from pneumatics and equipment leaks) are also allocated on a regional basis (mass based allocation), with the intention of highlighting regional differences in emission rates as well as in total production, and the effects that the proposed coallocation scheme would have on each region's emissions. Table 5 shows allocated methane emissions for each region.

The Appalachians region shows the biggest difference between gas and oil production; where combined production for natural gas and natural gas liquids is 4 orders of magnitude larger than the oil production (for the measured unconventional gas wells). In the case of the Gulf Coast region and the Mid Continent region, combined NG and NGL production is just 1 order of magnitude larger than oil production (from gas wells), whereas for the Rocky Mountains, gas and oil production are of the same order of magnitude. The national combined production of natural gas (47 236 000 Mg) and natural gas liquids (2 948 000 Mg) for all of measured wells is 1 order of magnitude larger than the oil production (5 341 000 Mg), with a higher production of oil relative to natural gas liquids. Consequently, a majority of the emissions (85%) are allocated to natural gas while 10% are allocated to oil, and 5% to natural gas liquids.

For the Appalachian region, over 99% of the emissions are allocated to natural gas, with almost all the remaining emissions being allocated to natural gas liquids. In the case of the Gulf Coast region, 70% of the emissions are allocated to natural gas; 18% is allocated to oil and 12% to natural gas liquids. For the Mid Continent, 92% of the emissions are allocated to natural gas, with remaining emissions being allocated in similar amounts to natural gas liquids and oil. The Rocky Mountains represent the region where the smallest fraction of emissions is allocated to natural gas (43%), with 32% of the emissions allocated to natural gas liquids, and 25% to oil. This is driven by the similar masses produced for the three products.

The ratios of methane emissions allocated to natural gas production divided by its production are 5 to 6 times higher for the Mid Continent (0.0051) and Rocky Mountains (0.0060) than for the other two regions (Appalachian, 0.0007; Gulf Coast, 0.0010).

Wells in the Appalachian and Gulf Coast regions have a similar EUR (Cumulative EUR per wells sampled) for salable natural gas; 144 900 Mg per well and 139 500 Mg per well, respectively. These values are considerably higher than for the Mid Continent, where per well EUR for natural gas is 35 900 Mg. For the Rocky Mountain region EUR for natural gas is 4700 Mg per well. The higher ratios of well life emissions divided by EUR observed in the Mid Continent and Rocky Mountains are driven by a similar order of magnitude of emissions per well across all regions but a significantly lower production per well.

Comparison to Other Databases. The National Energy Technology Laboratory (NETL) prepared a life cycle assessment (LCA) for natural gas,[5] both for conventional and unconventional sources. Under the life cycle stage called "Raw Material Acquisition", the source categories include emissions from well construction, well completions, liquid unloading, workovers, other point source emissions (gas released from wellhead and gathering equipment), and other fugitive emissions. Data for the LCA comes primarily from EPA emission factors and inventories, which have been subject to uncertainty due to the lack of direct measurements. Estimates are based on the EPA GHG inventory released in 2011, which were higher than the EPA estimates released in 2014.[19]

NETL reports 0.366 kg of methane emitted per million BTU of natural gas extracted or 0.0196 scf methane/scf of extracted natural gas from the raw material acquisition stage from shale gas wells (or 0.0244 scf methane/scf of extracted natural gas if the dry natural gas production is considered instead of the total natural gas production; NETL uses a natural gas energy content of 1027 BTU per cubic feet of natural gas, the same value used in the present work (Supporting Information, Table S2-1)). This emission rate is almost 1 order of magnitude higher than the value presented in this work (0.0036 scf of methane emissions over the life of a well allocated to natural gas/scf of produced natural gas over a well lifetime) and higher than the value reported by Allen et al. (0.0052 scf current emissions of

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methane/scf of dry natural gas production) for unallocated emissions.

Argonne National Laboratory prepared a LCA comparing shale gas to conventional natural gas.[20] The report is based on EPA estimates and emission factors, and acknowledges the uncertainty in EPA's estimates and modifications and revisions. Estimates are based on the EPA GHG inventory released in 2011 which were higher than EPA estimates released in 2013.[16] Based on EPA's revisions to the inventory, as well as recently released reports of measurements of methane emissions, such as Allen et al.,[12] Argonne updated their inventories.[17]

Argonne's GREET (Greenhouse gases, Regulated Emissions, and Energy use in Transportation) model is used for the analysis, developing a specific pathway for shale gas. The production stage includes emissions from well completions, workovers, liquid unloading, and well equipment (field separation equipment, gathering compressors, normal operations, condensate collection, compressors venting and upsets). The current inventory estimates a total of 120.7 g of methane per million BTU of natural gas, or a total of 0.0064 scf of methane/scf of natural gas produced for the production stage (or 0.0080 scf methane/scf of produced natural gas if the dry natural gas production is considered instead of the total natural gas production), which represents roughly 50% of the emissions estimated with the previous version of the GREET model, and which is a roughly a factor of 3 higher than the value reported in this work.

CONCLUSIONS

Mass, energy, and economic value basis allocation methods have been used to assign methane emissions from natural gas production from shale formations to the three main products of production activity: salable natural gas, oil, and natural gas liquids. Results for the three allocation methods are similar; however, the mass and energy allocation methods are preferred due to potential price fluctuations and variability that would affect the economic value approach.

On a national scale, approximately 85% of the emissions from the well site are assigned to natural gas (mass allocation), but regional variability is observed, thus, this work demonstrates the need to track natural gas sources by both formation type and production region. Methane emissions allocated to salable natural gas reported in this work are roughly 2 to 7 times lower than those reported in commonly used LCA data sets.

Care must be taken when allocating emissions across the natural gas supply chain. The results from the present work and the comparison to other studies indicate that allocation methods can have significant impact on interpretation of sources of emissions.

ASSOCIATED CONTENT

Supporting Information

Additional information as described in the text. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

Daniel Zavala-Araiza, who was a graduate student at the University of Texas at the time the work in this paper was done, has accepted a position at Environmental Defense Fund. David Allen has served as chair of the Environmental Protection Agency's Science Advisory Board (through January 2015), and in this role was a paid Special Governmental Employee. He is also a journal editor for the American Chemical Society and has served as a consultant for multiple companies, including Eastern Research Group, ExxonMobil, and Research Triangle Institute. He has worked on other research projects funded by a variety of governmental, nonprofit and private sector sources including the National Science Foundation, the Environmental Protection Agency, the Texas Commission on Environmental Quality, the American Petroleum Institute and an air monitoring and surveillance project that was ordered by the U.S. District Court for the Southern District of Texas. Gilbert Jersey is retired from ExxonMobil Research and Engineering.

The authors declare the following competing financial interest(s): One of the authors (D.T.A.) has research support from a consortium of the Environmental Defense Fund and 11 natural gas producers to measure methane emissions from natural gas production activities. One of the authors (D.Z.A.) performed analyses for this work as part of his graduate studies while at the University of Texas at Austin. After completing this research, D.Z.A. accepted a position with Environmental Defense Fund. One of the authors (G.R.J.) was formerly an ExxonMobil employee.

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