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SoCalGas Attachments to Comments, 15-IEPR-04

SoCalGas Attachment 1 of 2 to comments filed for June 1, 2015 Workshop.

Additional submitted attachment is included below.

Corrections

PERSPECTIVE

Correction for "Theory of mass-independent fractionation of isotopes, phase space accessibility, and a role of isotopic symmetry," by Rudolph A. Marcus, which appeared in issue 44, October 29, 2013, of *Proc Natl Acad Sci USA* (110:17703–17707; first published June 28, 2013; 10.1073/pnas.1213080110).

The author notes that, on page 1, middle column, lines 11–16 "Fewer accidental resonances mean less energy sharing and so less statistical behavior with a consequence that they are in equilibrium with the population of accessible states of O_3^* at low pressures, as discussed later." should instead appear as "Fewer accidental resonances mean less energy sharing and so less statistical behavior with a consequence of a shorter lifetime of O_3^* at low pressures, as discussed later."

On page 2, middle column, first full paragraph, lines 11–14 "This major difference in the pressure effect indicates a difference in the role of the collisions in these two distant phenomena." should instead appear as "This major difference in the pressure effect indicates a difference in the role of the collisions in these two distinct phenomena."

On page 3, middle column, first paragraph, lines 4–7 "The overall deviation from statistical theory for the recombination rate constant was (N. Ghaderi) perhaps a factor of 2." should instead appear as "The overall deviation from statistical theory for the recombination rate constant was (N. Ghaderi) less than a factor of 2."

On page 3, middle column, first full paragraph, lines 22–26 "Any chaos in the form of higher-order resonances within a volume element h^N would be coarse gained and so presumably contribute to quantum chaos." should instead appear as "Any chaos in the form of higher-order resonances within a volume element h^N would be coarse gained and so presumably not contribute to quantum chaos."

Both the online article and the print article have been corrected.

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Correction for "Measurements of methane emissions at natural gas production sites in the United States," by David T. Allen, Vincent M. Torres, James Thomas, David W. Sullivan, Matthew Harrison, Al Hendler, Scott C. Herndon, Charles E. Kolb, Matthew P. Fraser, A. Daniel Hill, Brian K. Lamb, Jennifer Miskimins, Robert F. Sawyer, and John H. Seinfeld, which appeared in issue 44, October 29, 2013, of *Proc Natl Acad Sci USA* (110:17768–17773; first published September 16, 2013; 10.1073/pnas.1304880110).

The authors note that upon publication their conflict of interest statement was not complete. The updated disclosure statement is as follows, "Jennifer Miskimins holds a joint appointment with Barree & Associates and the Colorado School of Mines. She has also served as an advisor to Nexen in 2012. David T. Allen served as a consultant for the Eastern Research Group and ExxonMobil in 2012, and is the current chair of the Science Advisory Board for the EPA. John H. Seinfeld has served as a consultant for Shell in 2012. David T. Allen, Matthew Harrison, Charles E. Kolb, and Robert F. Sawyer variously serve as members of scientific advisory panels for projects supported by Environmental Defense Fund and companies involved in the natural gas supply chain. These projects are led at Colorado State University (on natural gas gathering and processing), Washington State University (on local distribution of natural gas), and the University of West Virginia (on CNG fueling and use in heavy duty vehicles)."

Both the online article and print article have been corrected.

Correction for "mitoBK$_{Ca}$ is encoded by the *Kcnma1* gene, and a splicing sequence defines its mitochondrial location," by Harpreet Singh, Rong Lu, Jean C. Bopassa, Andrea L. Meredith, Enrico Stefani, and Ligia Toro, which appeared in issue 26, June 25, 2013, of *Proc Natl Acad Sci USA* (110:10836–10841; first published June 10, 2013; 10.1073/pnas.1302028110).

PNAS notes that a conflict of interest statement was omitted during publication. PNAS declares that "The editor, Ramon Latorre, is a recent coauthor with the authors of this publication, having published a paper with them in 2012."

Additionally, the authors note:

"Although Figs. 1 and S1 display the same sequence template, the analyses of LC/MS/MS data were performed against the respective databases, rat for Fig. 1, and mouse for Fig. S1. Sequence alignment of rat (NCBI:Q62976.3; UniProtKB: Q62976-1 V.3, which differs by 3 amino acids near the N terminus with that of Figs. 1 and S1) and mouse (NCBI: NP_001240298.1) isoforms show 98.9% amino acid identity with differences circumscribed to the extreme N and C termini. Peptides identified by LC/MS/MS have the exact sequence in rat and mouse as shown in Figs.1 and S1."

"In published Fig. 7, panels *E* and *F* show slices of the same heart in each condition; to better display the infarcted vs. healthy portions, these images were scaled to approximately the same size. We noticed that some data points in panel *G* were slightly moved during figure preparation. The revised Fig. 7 now shows heart slices at their original magnification (*E* and *F*) and the correct panel *G*. The corrected figure and its legend appear below."


Fig. 7. BK$_{Ca}$ protects the heart from ischemic injury. (*A*) Ischemia/reperfusion protocol. (*B* and *C*) Function traces of hearts preconditioned with vehicle (DMSO, control) or with NS1619 (10 μM) in WT and *Kcnma1*$^{−/−}$ mice. (*D*) NS1619 significantly improved mean RPP in WT but not in *Kcnma1*$^{−/−}$ mice. (*E* and *G*) WT hearts preconditioned with NS1619 exhibited less infarct size (white) compared with the control. (*F* and *G*) In *Kcnma1*$^{−/−}$, infarct size was not reduced with NS1619. (*H*–*J*) Mitochondrial Ca^{2+} uptake. NS1619 preconditioning increased the amount of Ca^{2+} needed to induce a large Ca^{2+} release in WT but not in *Kcnma1*$^{−/−}$ samples. Black arrows, addition of mitochondria. Blue arrows, 40 nmol Ca^{2+} pulses. Arrowheads, massive release of Ca^{2+}. *$P < 0.05$ vs. control (Ctrl); CRC, Ca^{2+} retention capacity.

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Measurements of methane emissions at natural gas production sites in the United States

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Engineering estimates of methane emissions from natural gas production have led to varied projections of national emissions. This work reports direct measurements of methane emissions at 190 onshore natural gas sites in the United States (150 production sites, 27 well completion flowbacks, 9 well unloadings, and 4 workovers). For well completion flowbacks, which clear fractured wells of liquid to allow gas production, methane emissions ranged from 0.01 Mg to 17 Mg (mean = 1.7 Mg; 95% confidence bounds of 0.67–3.3 Mg), compared with an average of 81 Mg per event in the 2011 EPA national emission inventory from April 2013. Emission factors for pneumatic pumps and controllers as well as equipment leaks were both comparable to and higher than estimates in the national inventory. Overall, if emission factors from this work for completion flowbacks, equipment leaks, and pneumatic pumps and controllers are assumed to be representative of national populations and are used to estimate national emissions, total annual emissions from these source categories are calculated to be 957 Gg of methane (with sampling and measurement uncertainties estimated at ±200 Gg). The estimate for comparable source categories in the EPA national inventory is ~1,200 Gg. Additional measurements of unloadings and workovers are needed to produce national emission estimates for these source categories. The 957 Gg in emissions for completion flowbacks, pneumatics, and equipment leaks, coupled with EPA national inventory estimates for other categories, leads to an estimated 2,300 Gg of methane emissions from natural gas production (0.42% of gross gas production).

greenhouse gas emissions | hydraulic fracturing

Methane is the primary component of natural gas and is also a greenhouse gas (GHG). In the US national inventories of GHG emissions for 2011, released by the Environmental Protection Agency (EPA) in April 2013 (1), 2,545 Gg of CH_4 emissions have been attributed to natural gas production activities. These published estimates of CH_4 emissions from the US natural gas industry are primarily based on engineering estimates along with average emission factors developed in the early 1990s (2, 3). During the past two decades, however, natural gas production processes have changed significantly, so the emission factors from the 1990s may not reflect current practices. This work presents direct measurements of methane emissions from multiple sources at onshore natural gas production sites incorporating operational practices that have been adopted or become more prevalent since the 1990s.

Horizontal drilling and hydraulic fracturing are among the practices that have become more widely used over the past two decades. During hydraulic fracturing, materials that typically consist of water, sand and, additives, are injected at high pressure into low-permeability formations. The injection of the hydraulic fracturing fluids creates channels for flow in the formations (often shale formations), allowing methane and other hydrocarbon gases

and liquids in the formation to migrate to the production well. The well and formation is partially cleared of liquids in a process referred to as a completion flowback, after which the well is placed into production. Production of natural gas from shale formations (shale gas) accounts for 30% of US natural gas production, and this percentage is projected to grow to more than 50% by 2040 (4).

Multiple analyses of the environmental implications of gas production using hydraulic fracturing have been performed, including assessments of water contamination (5–8), criteria air pollutant and air toxics releases (9–11), and greenhouse gas emissions (11–18). Greenhouse gas emission analyses have generally been based on either engineering estimates of emissions or measurements made 100 m to a kilometer downwind of the well site. This work reports direct on-site measurements of methane emissions from natural gas production in shale gas production regions.

Significance

This work reports direct measurements of methane emissions at 190 onshore natural gas sites in the United States. The measurements indicate that well completion emissions are lower than previously estimated; the data also show emissions from pneumatic controllers and equipment leaks are higher than Environmental Protection Agency (EPA) national emission projections. Estimates of total emissions are similar to the most recent EPA national inventory of methane emissions from natural gas production. These measurements will help inform policymakers, researchers, and industry, providing information about some of the sources of methane emissions from the production of natural gas, and will better inform and advance national and international scientific and policy discussions with respect to natural gas development and use.

Methane emissions were measured directly at 190 natural gas production sites in the Gulf Coast, Midcontinent, Rocky Mountain, and Appalachian production regions of the United States. The sites included 150 production sites with 489 wells, all of which were hydraulically fractured. In addition to the 150 production sites, 27 well completion flowbacks, 9 well unloadings, and 4 well workovers were sampled; the sites were operated by nine different companies. The types of sources that were targeted for measurement account for approximately two-thirds of methane emissions from all onshore and offshore natural gas production, as estimated in the 2011 national greenhouse gas emission inventory (1). A summary of the scope of the study, along with a rationale for the inclusion or exclusion of sources for direct measurement efforts, is provided in *SI Appendix*. Sampling was conducted from May 2012 through December 2012 at sites throughout the United States (see *SI Appendix* for a map and for the number of sampling sites in each region). All nine companies that participated in the study provided sites for sampling, and at least three companies provided sites in each of the regions (*SI Appendix*).

The data presented in this report represent hundreds of measurements of methane emissions from several types of onshore natural gas production activities; however, the sites sampled still represent a small fraction of the total number of sites nationwide (Table 1). This dataset is designed to be representative of the participating companies' activities and practices, but not necessarily all activities and practices. Multiple methods were used to minimize the potential for bias in the sample set, as described in *SI Appendix*.

Results

Emission measurements were performed for 27 well completion flowbacks, 9 liquids unloadings, 4 well workovers, and 150 production sites with 489 hydraulically fractured wells (Table 1 and *SI Appendix*). Data are summarized here for the well completion flowbacks, liquids unloading, and production site emissions. *SI Appendix* provides additional details. The data on well workovers, collected for workovers without hydraulic fracturing, are not presented because the data set was small and emission estimates for workovers without fracturing represent less than 0.1% of national emission estimates.

Well Completion Flowbacks. After a well is drilled, the well is "completed." Completion is the process of making a well ready for continuous production. Specifically, after drilling and fracturing, before natural gas production can begin, the well must be cleaned of sand and liquid of various types that had been injected into the well. The recovery of these liquids is referred to as a flowback, and gas, including methane, can be dissolved or entrained in the flowback liquids. Some of the methane in the liquids can be sent to sales or emission control devices, but some can be emitted.

Measurements were made of methane emissions during 27 completion flowback events. Emissions data for each of the 27 events is provided in *SI Appendix*. Five of the flowbacks were in the Appalachian region, seven in the Gulf Coast region, five in the Midcontinent region, and 10 in the Rocky Mountain region. The durations of the completions ranged from 5 to 339 h (2 wk). Measured methane emissions over an entire completion flowback event ranged from less than 0.01 Mg to more than 17 Mg, with an average value of 1.7 Mg and a 95% confidence interval of 0.67–3.3 Mg. Measurement and sampling uncertainty are included in the confidence interval; uncertainties due to a limited sample size dominate the overall uncertainty estimate. Methods for determining the confidence intervals are described in *SI Appendix*.

The completions with the lowest emissions were those in which the flowback from the well was sent immediately, at the start of the completion, to a separator, and all of the gases from the separator were sent to sales. The only emissions from these completions were from methane dissolved in liquids (mostly water) sent from the separator to a vented tank. The completion flowback with the highest total emissions, 17 Mg, was the longest in duration (339 h) and had initial flowback into a vented tank with very high methane concentrations. Some of the other relatively high emission completion flowbacks (~3 Mg to 6 Mg of methane) involved large amounts of flared gas (up to 130 Mg of methane to the flare, which was assumed to combust the methane at 98% efficiency, *SI Appendix*). Another completion with emissions of 4 Mg of methane was one in which all gases, for the entire event, were vented to the atmosphere. This type of venting for the entire duration of the completion was observed in 9 of the 27 completions. However, the nine completions of this type showed a wide range of emissions (4 Mg of methane for one completion and 0.5 Mg of methane for another completion of this type for an adjacent well).

These data provide extensive measurements on methane emissions from well completions that can be used in national emission estimates. Current national inventories of methane emissions have been assembled, based on simple engineering models of the completion process. In the most recent EPA national greenhouse gas emission inventory (2011 inventory, released April 2013) (1), 8,077 well completions with hydraulic fracturing are estimated to result in 654 Gg per year of emissions, for an average of 81 Mg of methane per completion flowback (compared with 1.7 Mg per flowback for the events reported here). To understand the reasons for the much lower emissions per event reported in this work, it is useful to define a potential emission for each flowback. The potential of a flowback to emit is defined here, and in the EPA national inventory (1), as the methane that would be emitted if all of the methane leaving the wellhead during the flowback were vented to the atmosphere. Potential emissions for the wells in this work ranged from 0.2 Mg to more than 1 Gg methane, with an average of 124 Mg. The average from the EPA national inventory is slightly higher at 151 Mg. Net emissions are calculated, in the EPA national inventory, by reducing potential emissions by estimates of methane captured or controlled

Table 1. Comparison of sample set size to emission source populations

Source	No. of events/locations sampled	Total no. of events/locations
Well completions	27	8,077*
Gas well unloading	9	35,828[†]
Well workovers	4	1782 (11,663)[‡]
Wells	489	446,745[§]

*Completions, with hydraulic fracturing reported in the 2011 National GHG Emission Inventory (1).
[†]Wells without plunger lift that have unloading events (the type of event sampled in this work) reported in the 2011 National GHG Emission Inventory (1).
[‡]Workover events with (and without) hydraulic fracturing reported in the 2011 National GHG Emission Inventory (1).
[§]Gas wells with and without hydraulic fracturing reported in the 2011 National GHG Emission Inventory (1); 513,000 on-shore natural gas wells are reported by the Energy Information Administration (20); see *SI Appendix*.

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because of regulatory or voluntary emission reductions. In the current national inventory, emission reductions are roughly one-half of potential emissions (*SI Appendix*). In this work, net or measured emissions for the total of all 27 completions are 98% less than potential emissions. This large difference between the net emissions measured in this work and the net emissions estimated in the national inventory is due to several factors. First, consistent with emerging regulatory requirements (21) and improved operating practices, 67% of the wells sent methane to sales or control devices. Second, for those wells with methane capture or control, 99% of the potential emissions were captured or controlled. Finally, the wells with uncontrolled releases had much lower than average potential to emit. Of the nine wells in this work that had uncontrolled venting of methane, the average potential to emit was 0.83 Mg, which is 0.55% of the average potential to emit in the national inventory. The relative importance of these factors is discussed in *SI Appendix*.

Unloadings. Gas wells often produce liquid hydrocarbons and water along with natural gas. In most new wells, the velocity of natural gas up the production tubing of the well is sufficient to lift any produced water out of the well with the gas. As gas production declines, the velocity may no longer be sufficient to lift the liquids, which begin to accumulate in the wellbore and eventually restrict gas flow from the producing formation. Liquids accumulation therefore needs to be removed to allow the well to continue to produce gas at optimal rates.

There are multiple methods of unloading a gas well, some of which do not result in emissions. In this work, sampling was performed for unloadings in which an operator manually bypasses the well's separator. Unlike automated plunger lift methods, these manual unloading events could be scheduled, allowing the study team adequate time to install measurement equipment. As the flow to the separator, which typically operates at pressures of multiple atmospheres, is bypassed, flow is diverted to an atmospheric pressure tank. This diversion allows the well to flow to a lower pressure destination (the atmospheric pressure tank, rather than the pressurized separator). This lower pressure end point allows more gas to flow, increasing velocity in the production tubing and lifting the liquids out of the well. Gas is discharged from the tanks through the tank vent, unless the tanks have an emissions control system such as a combustor.

The nine unloading events reported in this work were varied in their characteristics. Methane emissions ranged from less than 0.02 Mg to 3.7 Mg. Some unloadings lasted 2 h (or more) and had relatively uninterrupted flow. Other unloadings were as short as 10–15 min with uninterrupted flow, and still others had intermittent flow for short periods and periods of no flow for much of the unloading period. Some of the wells sampled only unloaded once over the current life of the well, whereas others were unloaded monthly. The average emission per unloading event was 1.1 Mg of methane (95% confidence limits of 0.32–2.0 Mg). If the emissions per event for each well are multiplied by the event frequency (events per year) reported by the well operators, the average emission per well per year was 5.8 Mg (an average of 5.9 events per unloaded well per year). The sampled population reflected a wide range of emission rates, with a population of high emitting wells and a population of low emitting wells. When emissions are averaged per event, emissions from four of the nine events contribute more than 95% of the total emissions. *SI Appendix* provides more information about individual unloading events.

Because the characteristics of the unloading events sampled in this work are highly variable, and because the number of events sampled is small, extrapolating the results to larger populations should be done with caution. One source of data on larger populations of wells with unloadings, to which the population sampled in this work can be compared, is a survey reported by the American Petroleum Institute and America's Natural Gas Alliance (API/ANGA) (22). In this survey, more than 20 companies provided data and well characteristics for 40,000–60,000 wells (with the number in the sample depending on the type of emission event). These API/ANGA data were used by the EPA to arrive at 2011 national inventory emission estimates for 35,828 wells without plunger lift and 22,866 with plunger lift, which vent for unloading. Unloading emissions for the wells in the API/ANGA survey were estimated based on well characteristics such as well bore volume, well pressure, venting time, and gas production rate (3). For the unloading events without plunger lift, 100 of the 2,901 wells (3%) in the survey account for 50% of the estimated emissions. Ninety percent of the estimated emissions in the API/ANGA survey are due to one-half of the wells. Because a small population of wells (3%) accounts for one-half of the emissions, if this relatively small population of high emitting wells is not adequately sampled, it is not possible to accurately estimate national emissions. The wells sampled in this work unloaded relatively infrequently. In contrast, some wells in the API/ANGA survey, including some of the highest emitting wells, unload with a daily or weekly frequency. An average frequency of unloading for the wells in the API/ANGA survey is 32.57 events per year, compared with an average observed in this work of 5.9.

Because a small number of unloading events accounts for a large fraction of emissions in the API/ANGA survey (22), and because some of these wells had frequencies of unloading higher than any of the events observed in this work, the sample set of nine events reported in this work is not sufficient for accurately estimating emissions from unloading at a national scale. Nevertheless, the data reported here provide valuable insights for the design of future sampling campaigns.

One important result from the measurements reported here is that current EPA estimation methods overpredict measured emissions. If the emission estimation method (3) used in the API/ANGA survey is applied to the events sampled in this work, estimates are 5 times higher than measured emissions. Estimates of the emissions for the nine events are 5.2 Mg per event versus measured emissions of 1.1 Mg per event. Emissions were overestimated for every event. The percentage by which emissions are overestimated increases as emissions per event decrease (*SI Appendix*). Possible causes of the overestimate include the assumptions in the estimation method that the entire well bore volume is released in an unloading and that the gas flow during an unloading is continuous.

Overall, the implication of all of these issues is a large uncertainty bound in the national emissions from gas well unloading. If the per well annual emissions from this work are used, a national emission estimate based on counts of wells that undergo unloading is in reasonable agreement with emissions in the EPA national inventory (1). In contrast, another estimate of unloading emissions, based on the per event emissions observed in this work and an estimate of national unloading events (22), would lead to a national estimate five times the estimate based on well counts. This estimate is not supported by the available data, given that the national event count is dominated by high frequency unloading events and the wells observed here unloaded far less frequently with much higher emission estimates per event. A lower estimate of unloading emissions could be suggested based on national well counts, emission estimates, and the finding that emission estimation methods, used in many EPA inventory estimates, overestimate observations made in this work by a factor of 5. All of these methods, however, assume a single scalar value represents a wide range of unloadings; the data presented in this work and in the API/ANGA survey (22) suggest that refined emission estimation methods, taking into account well and unloading characteristics, will be required. Additional measurements of unloading emissions are needed, both to resolve the

Table 2. National emission estimates for the natural gas production sector, based on this work and the 2011 national inventory

Category	2011 EPA GHG inventory net emissions,* Gg of methane/yr	Emission estimates from this report,[†] Gg of methane/yr	Comments
Sources with emissions measurements from this work used to generate national emission estimates			
Completion flowbacks from wells with hydraulic fracturing	654*	18[‡] (5–27)[§]	Decrease in national emission estimate
Chemical pumps	34*	68 (35–100)[§]	Increase in national emission estimate
Pneumatic controllers	355*	580[‡] (518–826)[§]	Increase in national emission estimate; if national emission factors derived from this work are used, this estimate becomes 790 Gg (*SI Appendix*)
Equipment leaks	172–211*,[¶]	291[‡] (186–396)[§]	Increase in national emission estimate; this comparison is based on equivalent categories of equipment, not all equipment leaks[¶] (*SI Appendix*)
Subtotal, national emissions, estimated based on this work	1215–1254[†#]	957 ± 200 [#]	Decrease of ~250 Gg for national emission estimate
Sources with limited measurements; national emissions not estimated			
Unloadings (nonplunger lift)	149* (EPA inventory)		Highly diverse events; small data set collected in this work; preliminary national emission estimates have a broad range of values (25–206 Gg; see text)
Workovers (without hydraulic fracturing)	0.3* (EPA inventory)		Measurements in this work included only one recompletion and three swabbing events (see text)
Other sources, not measured in this work			
Unloadings (plunger lift)	108* (EPA inventory)		No measurements made in this work
Workovers (with hydraulic fracturing)	143* (EPA inventory)		No measurements made in this work; equipment configurations are similar to completion flowbacks for wells with hydraulic fracturing; if emissions per event are comparable to completion flowbacks, current inventories may overestimate emissions
Other sources, not measured in this work	891–930*,[¶] (EPA inventory)		Includes potential emissions of sources not measured less prorated regulatory and voluntary emission reductions*
Total methane, Gg	2,545	2,300	Decrease of ~250 Gg for estimate
Methane emissions,*,* % [percent of gross gas production]	0.47% [0.59%]	0.42% [0.53%]	Brackets: gross gas emitted/gross gas produced (assuming produced gas is 78.8% methane)

*Emissions from EPA national inventory are based on reported potential emissions less reductions; when reductions are reported for combined source categories, identical percentage reductions of potential emissions are assumed to apply across source categories (*SI Appendix*, section S5).

[†]Emission factors used to estimate national inventories are designed to be representative of the participating companies' activities and practices, but not necessarily all activities and practices.

[‡]National emissions based on a regionally weighted average (*SI Appendix*, section S5).

[§]Ranges are based on 95% confidence bounds of emission factors; activity factors are identical to those used in EPA inventory. Uncertainties in activity factors (e.g., device counts) are not included. Uncertainties associated with whether regional or national averaging is performed are included in the uncertainty estimate (*SI Appendix*, section S5.4).

[¶]Sampling in this work included compressors on well sites, but not all gathering compressors. Well site and gathering compressors are combined in the national inventory. Range reported for national inventory for equipment leaks and "other" sources reflect uncertainty in attributing compressor emissions from national inventory to a specific source category.

[#]Uncertainty bound assumes uncertainties for completion flowbacks, pneumatic pumps and controllers and leaks are independent, and consequently, the combined uncertainty is the square root of the sum of the squares of the individual uncertainties.

**US total gross gas production (oil and coal bed, gas, and shale, onshore and offshore): 547,000 Gg.

differences between estimates and measurements and to better characterize the population of wells with unloading emissions.

Finally, it is also clear from the data that properly accounting for unloading emissions will be important in reconciling emission inventories with regional ambient measurements. Average methane emission rates for a single unloading ranged from roughly 100 g/min to in excess of 30,000 g/min. These rates are much larger than emission rates for production sites (typically tens of grams of methane per minute per well) or from completions (typically a few hundred grams per event per minute). At these emission rates, a single unloading event could, during the short period that it is occurring, result in emissions that are the equivalent of just a few wells in routine production to the equivalent of up to several thousand wells in routine production. Therefore, reconciliation between instantaneous ambient measurements and emission inventories will need to carefully represent the emissions from unloadings.

Well Sites in Routine Production. A well site contains one or more wellheads and may contain separators, pneumatic controllers, water tanks, hydrocarbon tanks (oil or condensate), and possibly other devices such as dehydrators, compressors, and flares. In this work, measurements were made from pneumatic controllers and pumps, because these devices release methane as part of their routine operation, and from equipment leaks detected by using an infrared camera (*SI Appendix*) at well sites.

Emissions for equipment on well sites, in routine production, that were targeted for measurements had much narrower uncertainty bounds than well completion flowbacks or well unloadings. Emissions from pneumatic chemical injection pumps measured in this work averaged 3.7 ± 1.6 g of methane per minute per pump, 9% lower than the EPA emission factor (*SI Appendix*, section S2). Intermittent and low bleed pneumatic devices measured in this work averaged 5.9 ± 2.4 and 1.7 ± 1.0 g

Table 3. Measurement methods used in the study

Source	Direct measurement methods	Mobile downwind sampling
Well completions	Measurements from flowback tanks made by using enclosures and temporary stacks with measurements of flow rate and composition	Downwind tracer ratio methods: Metered release of C_2H_2 and N_2O on site and downwind measurements of methane to C_2H_2 and methane to N_2O concentration ratios
Gas well unloading	Temporary stack with measurements of flow rate and composition	
Well workovers	Measurements from flowback tanks made by using enclosures and temporary stacks with measurements of flow rate and composition	
Production sites	Infrared (FLIR) camera surveys of sites and flow rate measurements using a HiFlow device	Metered release of C_2H_2 and N_2O on site and downwind measurements of methane to C_2H_2 and methane to N_2O concentration ratios

of natural gas per device per minute, 29% and 270% higher than EPA emission factors, respectively (*SI Appendix*, section S2). No high bleed pneumatic devices were identified at the sampling sites, and the average emission rate for the population of pneumatic controllers sampled in this work was 3.36 ± 0.65 g of methane per min (3.8 ± 0.69 g of natural gas per min). Equipment leaks measured in this work averaged 1.23 ± 0.44 g of methane per minute per well, which can be compared with an EPA estimate of potential emissions (no regulatory or voluntary emission reductions) of 1.37–1.67, derived from EPA's inventory for similar equipment types (wellheads, separators, heaters, meters/piping, and dehydrator fugitives), with the range reflecting whether small compressors are added to the comparison (*SI Appendix*, section S5). Comparing to net emissions is challenging because EPA does not assign emission reductions to specific equipment categories. Additional information is provided in *SI Appendix*.

There was significant geographical variability in the emissions rates from pneumatic pumps and controllers, but these regional differences were not as pronounced for equipment leaks. Emissions per pump from the Gulf Coast are statistically significantly different and roughly an order of magnitude higher than from pumps in the Midcontinent. Emissions per controller from the Gulf Coast are highest and are statistically significantly different from controller emissions in the Rocky Mountain and Appalachian regions. Emissions per controller in the Rocky Mountain region are lowest and an order of magnitude less than the national average (*SI Appendix*).

Implications for National Emission Estimates. If the average emissions reported in this work for well completion flowbacks, pneumatic devices, and equipment leaks are assumed to be representative of national populations and are applied to national counts of completions, pneumatic devices, and wells in EPA's national inventory, emissions from these source categories would be calculated as 957 Gg (with sampling and measurement uncertainties estimated at ±200 Gg), compared with 1,211–1,250 Gg methane per year in the 2011 EPA national inventory (1) for the same source categories. A large emissions decrease associated with completion flowbacks is partially offset by emission increases from pneumatic controllers and equipment leaks. Reasons for these differences are described in *SI Appendix*.

The estimated uncertainty in the national emission estimates based on this work is ~20% (200 Gg). The sources of uncertainty include measurement uncertainty, uncertainty introduced by the selection of sites, and uncertainty due to choices in performing regional or national averaging of equipment counts and emission factors. These components of the quantified uncertainty are described in *SI Appendix*. The uncertainty estimate does not include factors such as uncertainty in national counts of wells or equipment and the issue of whether the companies that provided sampling sites are representative of the national population.

The 957 ± 200 Gg in emissions for completion flowbacks, pneumatics, and equipment leaks, coupled with national inventory estimates for other categories, leads to an estimated 2,300 Gg of methane emissions from natural gas production (0.42% of gross gas production). A summary is provided in Table 2, and details of the calculations are available in *SI Appendix*.

Total emissions estimated based on measurements in this work (2,300 Gg) are comparable with the most recent EPA national GHG inventory (2,545 Gg in the 2011 inventory, released in April 2013) (1). Table 2 also compares emissions in specific source categories, estimated based on the measurements made in this work, to EPA estimates of the same categories in the national inventory (1). For some emission categories, such as completion flowbacks and pneumatic controllers, conclusions can be drawn from the comparisons. Specifically, measured emissions from completion flowbacks are roughly 600 Gg lower than the completion flowback emissions in the current inventory; measured emissions from pneumatic controllers are 150–500 Gg higher than in the current inventory. For other emission categories, such as equipment leaks and pneumatic pumps, however, drawing conclusions is more difficult. For these source categories, the national inventory reports potential emissions for each category, but aggregates emission reductions, creating uncertainty in the net emissions in these categories (see *SI Appendix*, section S5.5 for more details).

It should also be noted that the national inventory has changed in recent years based on evolving regulations (21) and understanding of emission sources. In this work, comparisons are made to the most recent release of the inventory (2011 final version, released in April 2013) and back casts to previous years by using consistent calculation methodologies. Emissions were estimated as 2,545 Gg in 2011, compared with 2,948 Gg in 2009 and 2,724 Gg in 2010. The work presented here suggests practices such as combusting or capturing emissions from completion flowbacks, as required by New Source Performance Standards subpart OOOO and the revised National Emission Standards for Hazardous Air Pollutants subpart HH (21), are resulting in reduced methane emissions. Other source categories require more data to produce national emission estimates, and adjustments in the inventory may emerge as more emission measurements are performed. Emission estimates may be adjusted downward if workovers with hydraulic fracturing are found to have emissions per event that are similar to completion flowbacks and may be adjusted either upward or downward as more emissions data are collected for liquids unloading or pneumatic devices.

Finally, an emissions intensity of 0.42% is reported in Table 2. The intensity expresses a methane emission per unit of gross gas production. This intensity should be interpreted with caution, because it includes only production operations and implicitly attributes all methane emissions from natural gas wells to natural gas production, although natural gas wells produce substantial amounts of natural gas liquids and oil. The intensity is reported here because it facilitates comparisons with other analyses that have appeared in the literature (23).

Methods

Multiple independent and complementary techniques were used to measure methane emissions. The primary procedures involved direct measurements of CH_4 emissions at their source. A variety of different procedures were used for direct source measurements, depending on the type of source being sampled and the type of natural gas production equipment being used. Table 3 summarizes the direct source methods used in the study; detailed descriptions of the methods are provided in *SI Appendix*.

In addition to direct source measurements, tracer ratio measurements, designed to estimate the total methane emissions from a site, were made at 20% of the well completion flowbacks and 13% of the production sites. The tracer release method was developed in the 1990s to quantify methane emissions from a wide range of natural gas system components (24, 25). Sites for tracer releases were selected for their steady, moderate winds and downwind access. Measurements for sites without downwind access could not be made. Table 3 also summarizes these measurement methods, which are described in detail in *SI Appendix*. In brief, tracer compounds were released at a known rate on-site; downwind measurements of methane (minus

background) and the tracer (minus background) were assumed to be equal to the ratio of emission rates, allowing methane emissions to be estimated. These measurements were performed for a subset of the sampling locations that had relatively open terrain and steady winds, producing well-defined emission plumes downwind of the sites. The tracer studies allowed for an independent measurement of emissions that were also measured by using direct source methods. For completion flowbacks, emission estimates based on the downwind measurements were generally within a factor of 2 of the direct source measurements, supporting the conclusion that emissions from completion flowbacks are roughly 97% below the most recent national estimates and that emissions from completion flowbacks without methane control or recovery equipment, observed in this work, are well below the average potential emissions in current national inventories (1). For the production sites, emissions estimated based on the downwind measurements were also comparable to total on-site measurements; however, because the total on-site emissions were determined by using a combination of measurements and estimation methods, it is difficult to use downwind measurements to confirm the direct source measurements. Tracer study results are summarized in *SI Appendix*.

1. US Environmental Protection Agency (EPA) (2013) *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990-2011* (Environ Protect Agency, Washington, DC). EPA 430-R-13-001.
2. Harrison MR, Shires, TM, Wessels JK, Cowgill RM (1996) *Methane Emissions from the Natural Gas Industry, Volumes 1–15,* Final Report. (Gas Res Inst and Environ Protec Agency, Washington, DC) GRI-94/0257 and EPA-600/R-96-080, Appendix B-1.
3. Code of Federal Regulations, Title 40 Section 98.233, Calculating GHG Emissions, 75 FR 74488, November 30, 2010, as amended at 76 FR 80575, December 23, 2011.
4. Energy Information Administration (2013) Annual energy outlook, early release. Available at www.eia.gov/forecasts/aeo/er/index.cfm. Accessed January 31, 2013.
5. U.S. Environmental Protection Agency (2011) Plan to study the potential impacts of hydraulic fracturing on drinking water resources (Environ Protect Agency, Washington, DC). Available at www2.epa.gov/hfstudy/plan-study-potential-impacts-hydraulic-fracturing-drinking-water-resources-epa600r-11122. Accessed August 27, 2013.
6. Osborn SG, Vengosh A, Warner NR, Jackson RB (2011) Methane contamination of drinking water accompanying gas-well drilling and hydraulic fracturing. *Proc Natl Acad Sci USA* 108(20):8172–8176.
7. Vidic RD, Brantley SL, Vandenbossche JM, Yoxtheimer D, Abad JD (2013) Impact of shale gas development on regional water quality. *Science* 340(6134):1235009.
8. Rahm B, Riha S (2012) Toward strategic management of shale gas development: Regional, collective impacts on water resources. *Environ Sci Policy* 17:12–23.
9. McKenzie LM, Witter RZ, Newman LS, Adgate JL (2012) Human health risk assessment of air emissions from development of unconventional natural gas resources. *Sci Total Environ* 424:79–87.
10. Litovitz A, Curtright A, Abramzon S, Burger N, Samaras C (2013) Estimation of regional air quality damages from Marcellus Shale natural gas extraction in Pennsylvania. *Environ Res Lett* 8:014017.
11. Pacsi AP, Alhajeri NS, Zavala-Araiza D, Webster MD, Allen DT (2013) Regional air quality impacts of increased natural gas production and use in Texas. *Environ Sci Technol* 47(7):3521–3527.
12. Hayhoe K, Kheshgi HS, Jain AK, Wuebbles DJ (2002) Substitution of natural gas for coal: Climatic effects of utility sector emissions. *Clim Change* 54:107–139.
13. Lelieveld J, et al. (2005) Greenhouse gases: Low methane leakage from gas pipelines. *Nature* 434(7035):841–842.
14. Wigley TML (2011) Coal to gas: The influence of methane leakage. *Clim Change* 108:601–608.
15. Howarth RW, Santoro R, Ingraffea A (2011) Methane and the greenhouse-gas footprint of natural gas from shale formations. *Clim Change* 106:679–690.
16. Kargbo DM, Wilhelm RG, Campbell DJ (2010) Natural gas plays in the Marcellus Shale: Challenges and potential opportunities. *Environ Sci Technol* 44(15):5679–5684.
17. Kerr RA (2010) Energy. Natural gas from shale bursts onto the scene. *Science* 328(5986):1624–1626.
18. Laurenzi IJ, Jersey GR (2013) Life cycle greenhouse gas emissions and freshwater consumption of Marcellus shale gas. *Environ Sci Technol* 47(9):4896–4903.
19. International Energy Agency (2012) Golden rules for a golden age of gas. Available at www.worldenergyoutlook.org/media/weowebsite/2012/goldenrules/WEO2012_GoldenRulesReport.pdf. Accessed August 15, 2012.
20. Energy Information Administration (2012) Summary statistics for natural gas in the United States, 2007-2011. Available at www.eia.gov/naturalgas/data.cfm#production. Accessed January 31, 2013.
21. Code of Federal Regulations 40, Part 60 Subpart OOOO and NESHAP 40, CFR 63 Subpart HH, 77 FR 49490, August 16, 2012.
22. American Petroleum Institute and America's Natural Gas Alliance (API/ANGA) (2012) Characterizing pivotal sources of methane emissions from natural gas production summary and analyses of API and ANGA survey responses, final report, updated September 2012. Available at www.api.org/news-and-media/news/newsitems/2012/oct-2012/~/media/Files/News/2012/12-October/API-ANGA-Survey-Report.pdf. Accessed February 28, 2013.
23. Alvarez RA, Pacala SW, Winebrake JJ, Chameides WL, Hamburg SP (2012) Greater focus needed on methane leakage from natural gas infrastructure. *Proc Natl Acad Sci USA* 109(17):6435–6440.
24. Lamb BK, et al. (1995) Development of atmospheric tracer methods to measure methane emissions from natural gas facilities and urban areas. *Environ Sci Technol* 29(6):1468–1479.
25. Shorter JH, et al. (1997) Collection of leakage statistics in the natural gas system by tracer methods. *Environ Sci Technol* 31:2012–2019.

ENVIRONMENTAL SCIENCES

Measurements of Methane Emissions at Natural Gas Production Sites in the United States

Supporting Information

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Table of Contents

This Supporting Information uses industry standard units of standard cubic feet (scf).
One scf of methane contains 19.2 g methane.

S1   Direct Source Measurements: Well Completion Flowbacks

S1.1   Methods

Methane emissions were measured directly, at the point of release. Data for 27 well completion events are reported.   Section S4 describes measurements of methane concentrations that were made downwind of 6 of the completion events; these downwind measurements were used to confirm that all of the major emission sources were being measured.

The sources of well-site methane emissions during a completion depend on the equipment used in the completion. In this work, the surface configurations will be classified into five categories, each with different types of surface configurations. Figure S1-1 shows a simplified flow diagram for one type of surface equipment configuration used during completion flowback (labeled as Configuration 1 in this work). There are several stages in the flowback process that utilize the equipment shown in Figure S1-1. In the first stage (Step 1 in Figure S1-1), reservoir gases mixed with water, sand and fracturing liquid flow from the high pressure well head, through a choke, to either an open top tank or an enclosed tank with open vents. In either case, the tank gases are vented to the atmosphere. Figure S1-2 shows examples of open-top tanks, used in Step 1. To measure emissions from open-top tanks, a temporary plastic cover was placed over the open-top tank, secured by clamping to the edge of the tank. A hand-held infrared camera, designed with filters and banded wavelengths to visualize hydrocarbon plumes, was used to check for leakage around the seal. The gases were vented through a plenum that had exit stacks of two diameters. The smaller diameter stack was used during periods of low flow and the larger stack was used during periods of high flow. Switching between the stacks was done with pneumatic controllers operated remotely. Gas velocity in the stack was measured using a pitot tube in the center of the stack. Total volumetric flow was calculated by multiplying the stack cross-sectional area by 80% of the gas velocity at the stack centerline. The factor of 0.8 was used to convert the centerline velocity in the stack to an estimated average velocity in the stack.[1] Gas samples for composition analysis were drawn from the temporary stack, through tubing to a sampling port 10-20 meters from the tank. Gas samples were drawn into evacuated tedlar bags for subsequent analysis using gas chromatography. If an enclosed (vented) tank was used, then no plastic cover was used and a temporary stack was placed over the tank hatch. Gas velocities and compositions were measured using the same methods as used for the open top tanks.

Figure S1-1. Flowback surface equipment configuration including an open top tank and oil and water flowback tanks, venting to atmosphere; in this configuration, emissions occur from the open top tank, the water and hydrocarbon flowback tank hatches, and the flare
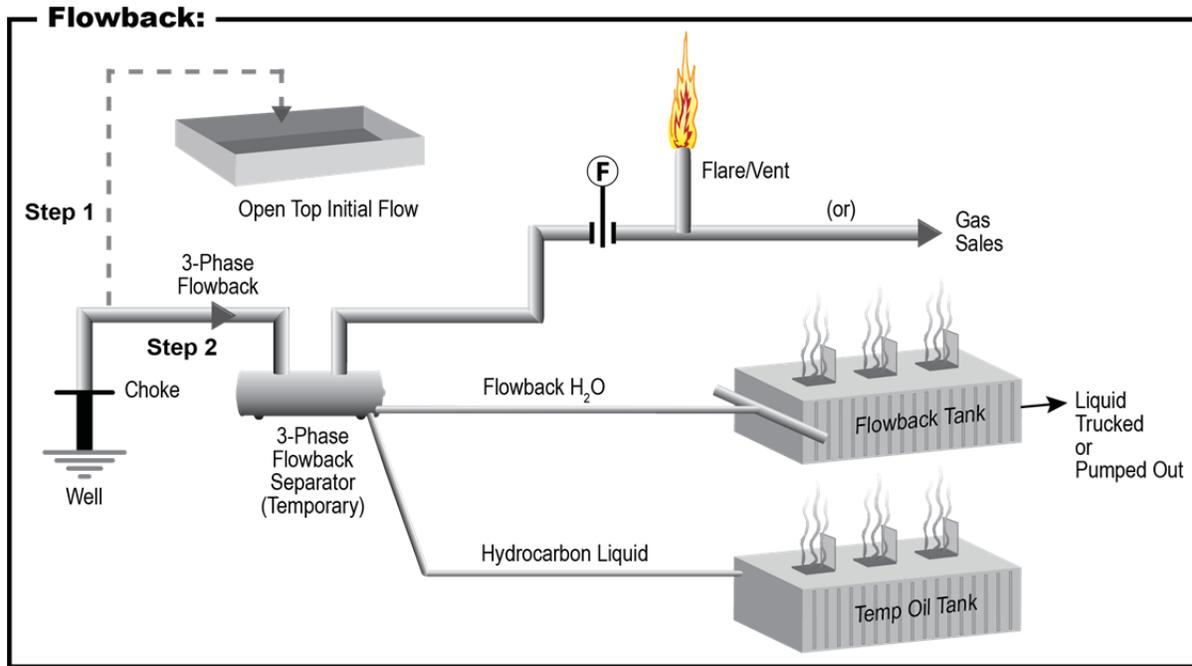
Note: Sand Filter may be installed upstream of Separator. Also, there may be 2-stage (HP, LP) Separation.

Figure S1-2. Open top tank used in Step 1 of flowback using the equipment configuration shown in Figure S1-1. Upper Left: line leading from well to tank; upper right: temporary plastic cover installed and clamped to edge of tank, with exhaust stacks on ground adjacent to tank; lower: Conceptual diagram of sampling system.
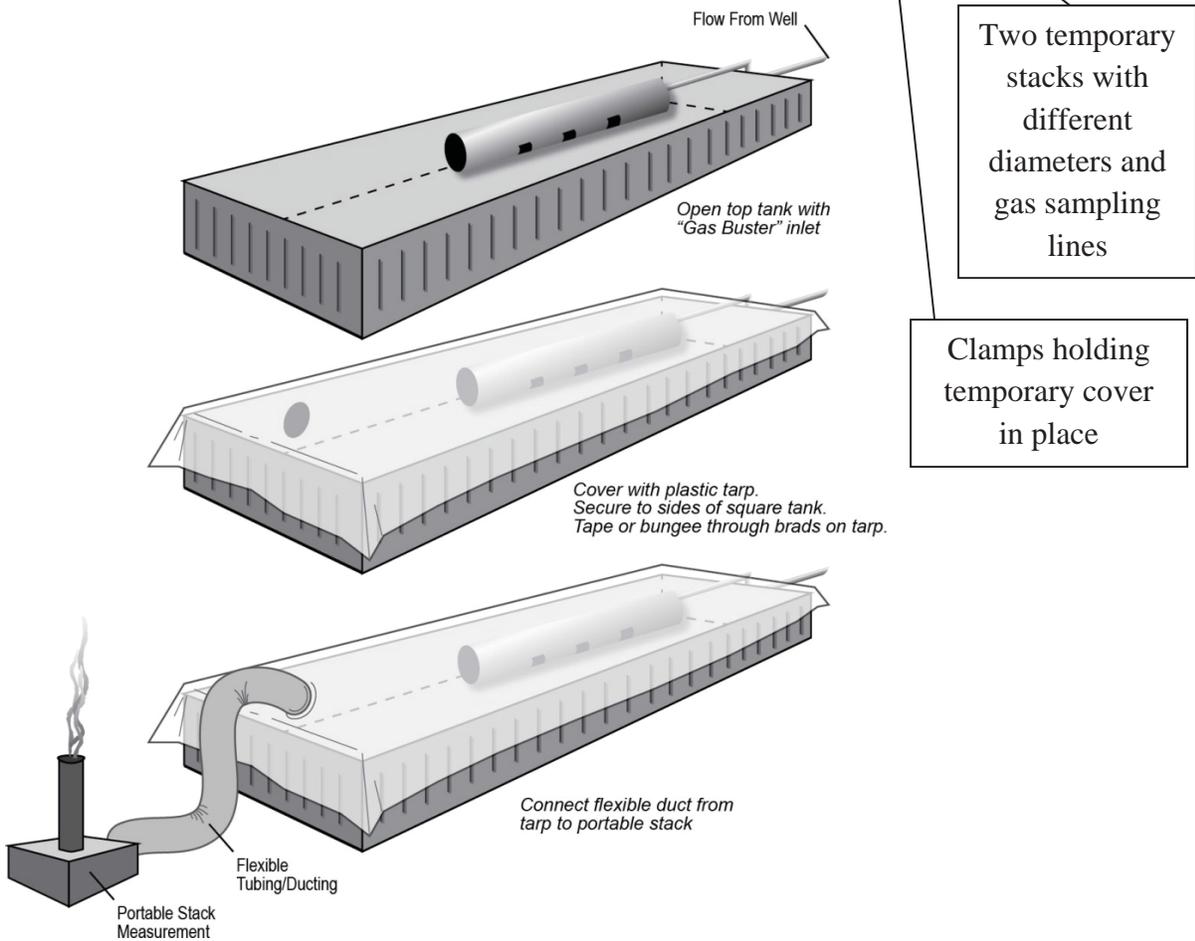
The initial step of the completion flowback to the open-top or vented tank lasted until sufficient volumes and concentrations of natural gas were present, allowing the completion to proceed to the next step. This initial period ranged from an hour to multiple days. In some completions, Step 2 of the completion consisted of flow to a separator (sometimes with a sand trap between well and separator). Separator pressures ranged, over the completion events sampled in this work, from less than 100 to more than 1000 psi. Gas and liquid streams (sometimes separate water and hydrocarbon liquid streams) flow from the separator. The water and hydrocarbon streams were fed to water and hydrocarbon flowback tanks, shown in Figure S1-3. The flowback tanks were generally enclosed, with hatches allowing venting to the atmosphere. As shown in Figure S1-3, temporary stacks, similar to those used in Step 1, recorded the volumes of gas exiting the flowback tanks. Tubing was used to draw gas samples to a remote sampling port, where again the samples were drawn into evacuated tedlar bags for subsequent gas analysis. The gas stream from the separator was routed, through a flow meter, to a flare, or sometimes to sales. If the gas was sent to a flare, the flow rate and gas composition analysis, reported by the operator of the site, were used to determine the flow of flared methane. A combustion efficiency of 98% was assumed, based on standard EPA emission factors[2,3].

The period of flowback to the separator and enclosed flowback tank lasted from a few hours to more than a week, depending on the characteristics of the well. After this phase of the completion, gas was routed to sales lines and the well entered production.

Figure S1-3. Oil and water flowback tanks. Upper and middle: Hatches in the tanks allowed gases to vent to the atmosphere; temporary stacks were installed on the hatches to measure gas flow. Samples for gas composition analyses were drawn from the stack, through tubing, to a remote sampling port.

Lower: Conceptual diagram of sampling system.

Open

Oil Flow From Separator

Water Flow From Separator

Stack on open top tank only operational during Step 1

Oil

Oil

To Trucks

H₂O

H₂O

H₂O

H₂O

To Trucks

To Trucks

Cover Over Closed Hatch

Open Hatch

Temporary Stack Measurement Device

Vapor communication between tanks connected by a liquid line was minimal since the fluid in the active tank was above the truck loading line and not at the tank bottom
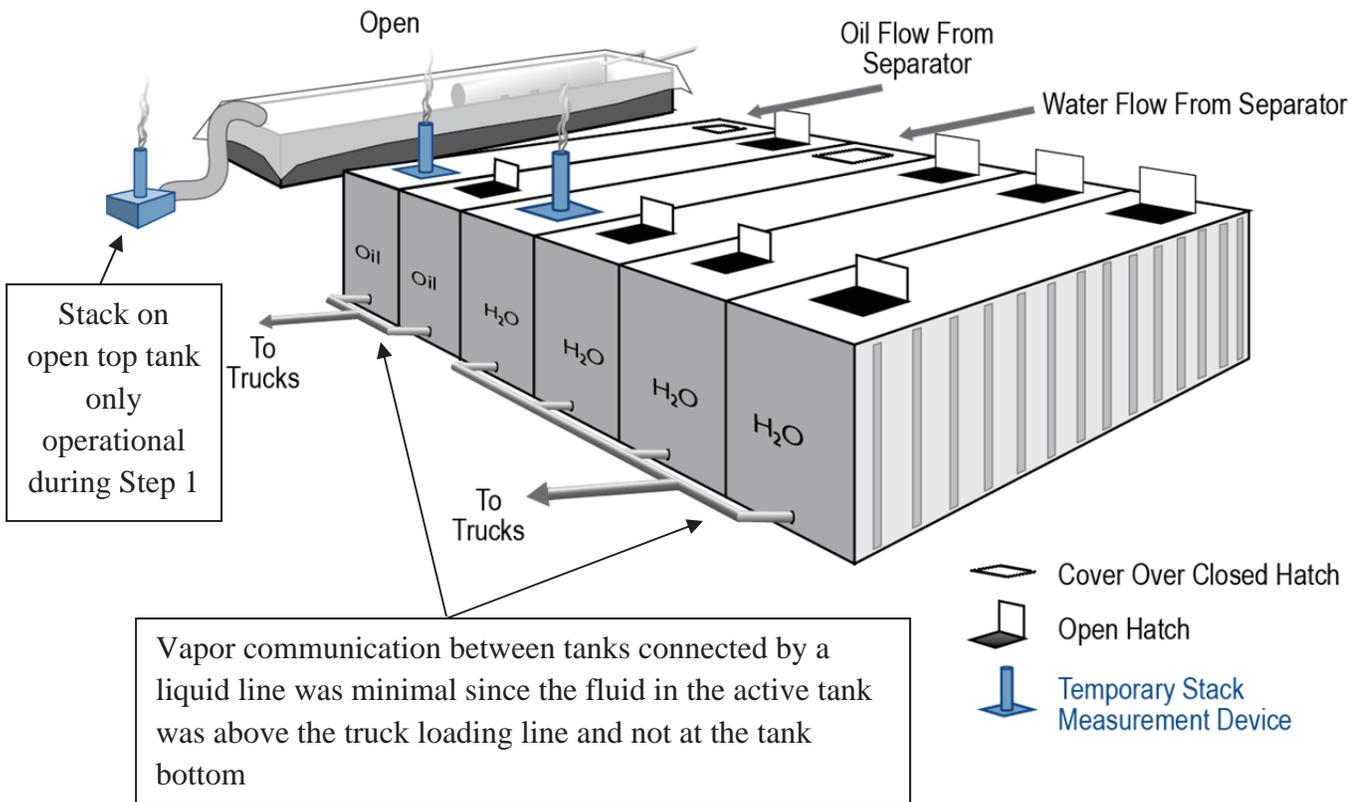
The completion flowback configuration shown in Figure S1-1 was one of multiple surface equipment configurations encountered by the Study Team over the course of the study. The flowback configurations, and the frequency with which they were observed, are summarized in Table S1-1. Not all of the surface configurations in each of the 5 categories were exactly identical. For example, in some configurations, gas from a separator was routed to a flare; in other cases the gas was routed to sales and the flare, and in still other cases the gas from the separator was routed exclusively to sales. The categorizations shown in Table S1-1 are distinguished by the type of surface equipment used, rather than the fate of the streams from particular pieces of surface equipment. Thus, Table S1-1 is a summary, rather than a complete inventory of surface configurations.

Table S1-1. Surface equipment configurations for completions

Configuration Number	Description of surface equipment and completion process	Frequency of configuration in completions sampled in this work (%)
1	Initial flow from the well to an **open or vented tank**, with gases vented to the atmosphere; after this initial phase flow is routed to a **separator** or multiple (high and low pressure) separators. Water and hydrocarbon liquids are sent to **water and oil flowback tanks that vent** to the atmosphere; gas from the separator is metered and sent to a flare or sales. (See Figure S1-1)	9 (33%)
2	Initial flow from the well to an **open or vented tank**, with gases vented to the atmosphere; after this initial phase flow is routed to a **separator** or multiple (high and low pressure) separators. Water is sent from the separator to a vented flowback tank. The vented gases may be released or metered and sent to a flare. **Hydrocarbon liquids are sent from the separator to a sealed flowback tank, and the vented gases are sent to a combustor.**	4 (15%)
3	**Flow directly from the well to a separator** or multiple separators, with no initial flowback to an open tank; gases from the separator either to sales or flare; **liquids from the separator to a flowback tank**	5 (18%)
4	**Flow from the well to an open or vented tank**, with gases vented to the atmosphere, **for the entire duration of the completion**	9 (33%)
5	**Other***	0 (0%)

*The other category is included to facilitate comparisons with national data on equipment configurations used in completion flowbacks

These multiple equipment configurations reflect the wide range of production characteristics of wells and can be expected to lead to different emissions. However, there are common

elements in the completions which are similar across multiple configurations. These elements include:

1. Flow of a mixture of sand, water, gas and fracturing liquid from the well to an open tank, where the gas is vented.
2. Flow of pressurized hydrocarbon liquid, with dissolved methane, from a separator to a tank where gas flashes from the liquid and is either vented or sent to a combustion device
3. Flow of pressurized water, with dissolved methane, from a separator to a tank where gas flashes from the liquid and is either vented or sent to a combustion device
4. Flow of gas, including methane, from a separator to a sales line or to a flare which is designed to destroy 98+% of the combustible gases

In addition, during some of the completions there were other small venting events. In completions that used sand filter vessels, the sand filter was occasionally blown down to a vented or open top tank to discharge the collected sand. These small emission events were not possible to directly measure. In cases where it was anticipated that emissions from these sources could be significant, estimates of these quantities were added to the completion emissions.

The focus in the completion flowback emissions reported here is on actual emissions, however, in order to understand the differences in emissions between the different surface equipment categories, it will be necessary to distinguish between potential and actual emissions. The concept of potential emissions, as opposed to actual emissions, is used by the US EPA in its national emission inventory.[4] In this work, the potential emissions from a completion flowback will include the emissions that would occur if all of the methane flowing from the well during the completion flowback was emitted to the atmosphere. Configurations 1, 2 and 3 all involve some level of emission control, so actual emissions will be lower than potential emissions. In contrast, for Configuration 4, a configuration that will not be permissible under recent EPA New Source Performance Standards (NSPS) (Subpart OOOO regulations), there are no emission controls, so potential emissions and actual emissions are equal.

Section S1.2 reports total methane emission data for each completion sampled in this work, and methane emissions for each of the elements that was in place for the sampled completions.

S1.2   Results and Discussion

A total of 27 completion flowback events were sampled. Completion flowback events were defined as beginning with the initiation of the flow of liquids and gases from the well and ending at the point at which the completion contractor's report stated that it ended. Often this end point was when gases were routed to sales or to a centralized gas processing facility, however, the end point was not uniformly defined. For example, some completion flowbacks were routed from the well to a temporary separator, and the operator defined the end of the completion as the point at which flow was routed to a permanent, rather than temporary separator, even though the gases from the temporary separator went to sales. In other cases, the end of completion flowback was the point at which flow ended to temporary flowback equipment. In all cases for this study, the end of the completion flowback was at the termination time stated in the completion contractor's report.

Of the 27 completions sampled in this work, five were in the Appalachian region, seven in the Gulf Coast region, five in the Mid-Continent region, and ten in the Rocky Mountain region. Summaries of the methane emission estimates are provided in Tables S1-2 through S1-5.

Methane emissions over an entire completion flowback event, summed over all emission sources for each event (e.g., tank vents, uncombusted methane from flares), ranged from a few thousand scf to more than 800,000 scf, with an average value of 90,000 scf.   The durations of the completions ranged from 5 to 339 hours (2 weeks). The completions with the lowest emissions were those where the flowback from the well was sent immediately, at the start of the completion, to a separator, and all of the gases from the separator were sent to sales. The only emissions were from methane dissolved in liquids (mostly water) sent from the separator to a vented flowback tank. The completion with the highest total emissions, 880,000 scf, was the longest completion (339 hours) and also was a completion in which the initial flowback from the well went directly into a vented tank, and where that initial flow was very high in methane. Some of the other relatively high emission events (~200,000 to 300,000 scf methane) were completions with large amounts of flared gas (up to 7 million scf of methane sent to the flare). Another completion with emissions in excess of 200,000 scf of methane was one in which all gases, for the entire event, were vented to the atmosphere. This type of venting for the entire duration of the completion was observed in 9 completions. However, the 9 completions of this type showed a wide range of emissions (200,000 scf methane for one completion (Midcontinent Completion 1) and 27,000 scf methane for another completion of this type for an adjacent well completed during the same time period (Midcontinent Completion 2 – see Table S1-4)).

Many of the completions sampled in this study either sent gases directly to sales and/or used a flare on-site to combust gases vented from separators. In some cases where a flare was present, the assumed volume of uncombusted methane from the flare dominated the total methane emissions from the completion event (Gulf Coast Completions 1-4– see Table S1-3). For flowbacks using flares, it was assumed that 98% of the methane fed to the flare was

combusted and 2% of the methane fed to the flare remains un-combusted and escaped into the atmosphere[2,3]. Figure S1-4 shows an example of the methane flow to the flare at a completion, which had the surface equipment configuration shown in Figure S1-1. In this completion (Gulf Coast Completion 1), a total of 5,000,000 scf of methane (6.4 million scf of total gas) was fed to the flare during the multi-day completion. Flow to the flare begins, after hour 4, when the transition is made from flow to the open top tank (Step 1) to flow to the separator. Flow to the flare ends when the completion ends and gases are routed to sales. If the 5,000,000 scf of methane (6,400,000 scf of gas) fed to the flare (counted as a potential emission in this completion) is combusted at 98% efficiency, methane emissions from the flare will be 100,000 scf. In this completion, all other methane emissions during the completion event totaled 5,000 scf methane. The assumed methane emissions from the flare (estimated at 100,000 scf) dominate total methane emissions during this completion event.

Figure S1-4. Flow of gas from well completion separators to a flare (Gulf Coast Completion 1)
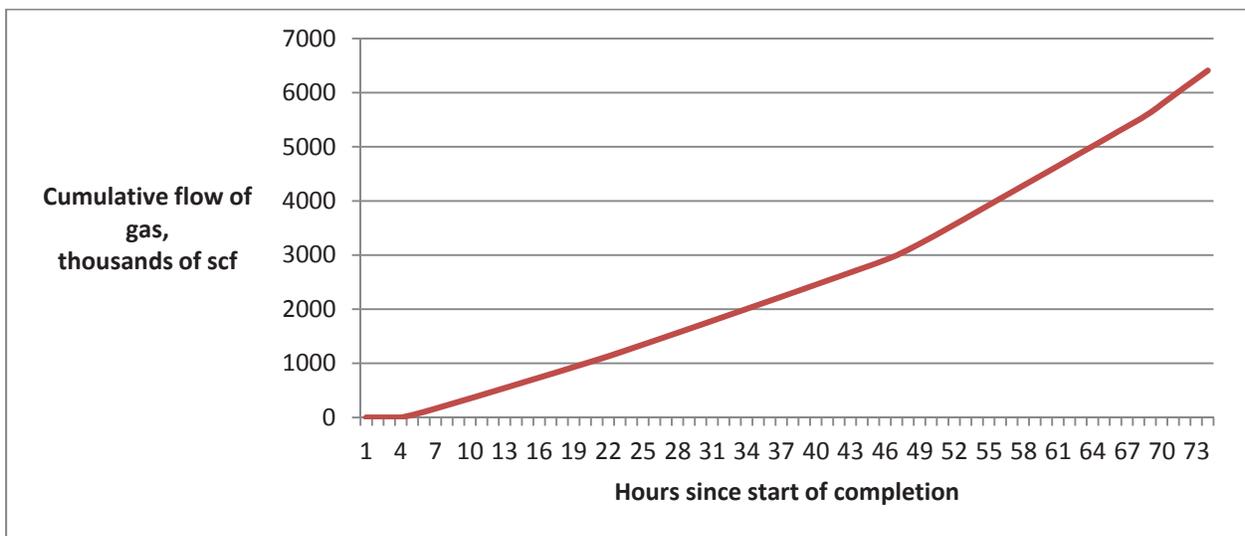
Another source of methane emissions in many completions was methane that flowed from a separator, dissolved in hydrocarbon phase or aqueous phase liquids, which subsequently flashed in an oil or water flowback tank. The flow from the separator to the flowback tank is not constant. The flow varies as the separator periodically builds hydrocarbon liquid level to a set point, then discharges the liquid to the flowback tank. This results in the type of periodic flow shown in Figures S1-5 and S1-6.

Figure S1-5. Methane venting through temporary stack in an oil flowback tank in Gulf Coast Completion 1. Two hours of data are shown. Approximately 40 separator discharge events occurred during this period (20 per hour).

minutes
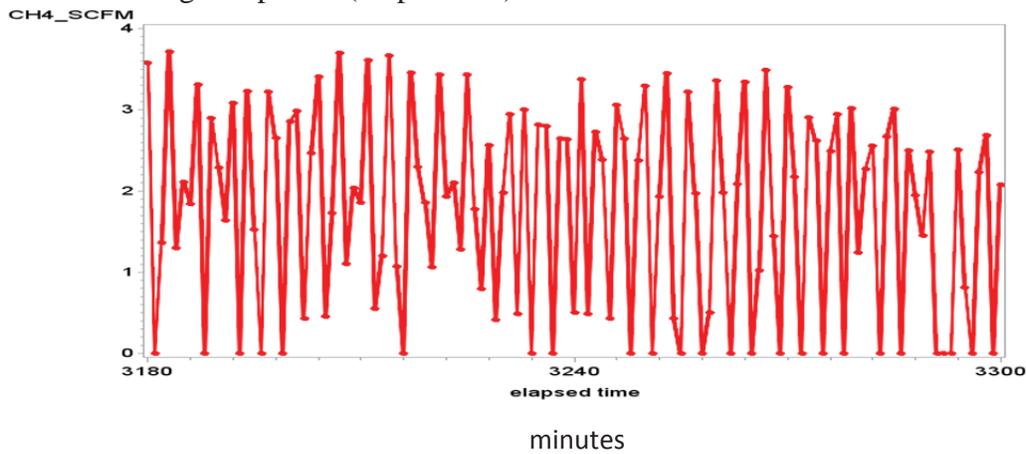
Figure S1-6. Methane venting through temporary stack in a water flowback tank for Gulf Coast Completion 1. Six hours of data are shown; 24 discharge events occurred during this period (4 per hour).

minutes
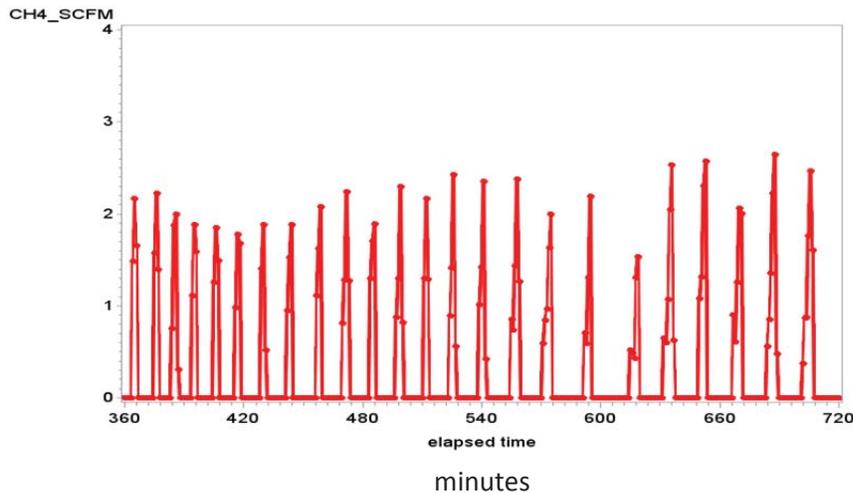
The percentage of methane in the gases vented from flowback tanks in separator discharge events such as those shown in Figures S1-5 and S1-6 varied over the course of the flowback. There are a number of factors that can cause the concentration of methane in the vent gas to vary. For example, methane concentration in the stack of the flowback tank will vary based on the oil and water level in the flowback tank, since the methane flashing from the separator discharge is diluted by the existing air in the vapor space of the flowback tank and dilution changes as vapor space changes. These liquid levels change, depending on the schedule

for emptying tanks of their liquids. In addition, oil and water composition can vary over the course of a flowback, changing the methane solubility. Because of these and other factors, detailed temporal analysis of the methane emissions from the flowbacks was not performed; instead, time integrated analyses were done.

Volumetric flow of vent gas was recorded each minute. For each one-minute record of volumetric flow, a percentage of methane was determined using linear interpolation between the most recent composition measurement before and the most recent composition measurement after the flow measurement. Compositions were measured approximately hourly during initial phases of completion flowbacks; as completions extended into multiple days and flows became steady, composition measurements were made every 4-8 hours. To assess the magnitude of the uncertainty associated with using linearly interpolated methane concentrations, two sensitivity analyses were performed. In one sensitivity analysis, the methane concentration for each minute of flow data was assumed to be the lower of the most recent composition measurement before and the most recent composition measurement after the flow measurement. In a second sensitivity analysis, the methane concentration for each minute of flow data was assumed to be the higher of the most recent composition measurement before and the most recent composition measurement after the flow measurement. For the estimate of the lower bound on emissions, it was assumed that the methane percentage in the gas at the start of the completion was equal to half of the detection limit (0.18%, equal to half of the smallest concentration recorded in the chromatographic analyses (0.36%) during the entire study) and it was assumed that the final gas composition persisted from the time of the measurement until the end of the completion. For the estimate on the higher bound on concentration, the methane concentration at the start of the completion was assumed to be equal to the initial concentration measurement and it was assumed that the final gas composition persisted from the time of the measurement until the end of the completion. These two sensitivity analyses provide a quantification of the uncertainty associated with using discrete, rather than continuous methane analyses. Methane concentrations are not expected to change rapidly based on physical arguments. The size of the vapor space in a half full flowback tank is more than 1000 scf, so each separator discharge event only displaces a few percent of available vapor space.

The uncertainty ranges reported in Tables S1-2 to S1-5 are a combination of the uncertainty bounds based on using intermittent, rather than continuous composition analyses, and an estimated 10% uncertainty bound for the flow through the temporary stacks.[5] In arriving at an overall uncertainty estimate, it is assumed that the uncertainties in composition measurements and flow are independent. Not included in the uncertainty estimates for the measurements are uncertainties in combustion efficiencies in flares and combustors (assumed to be 98%[2]) and uncertainties in the flow measurements of gas flows to sales or flares. The total quantified measurement uncertainties are approximately 20% of the total emission estimates.

Table S1-2. Methane emissions (scf) from Appalachian well completions: results from 5 sampling events (Dark shading indicates that data were not used in determining average emission factors[a])

Emission Source	1 Company AP-A	2 Company AP-B	3 Company AP-B	4 Company AP-C	5[a] Company AP-C
(duration of completion flowback event, hr)	Configuration 1* (62.5 hr)	Configuration 3*** (37.8 hr)	Configuration 3*** (12.5 hr)	Configuration 1** (339.2 hr)	Configuration 1** (228 hr)
Flowback to open top tank; gases vented	12,700 ± 10,000 scf	6,700 ± 800 scf	Not applicable	1,105,000 ± 320,000 scf	240,000 ± 122,000 scf
<u>Atmospheric</u> Vent from Tank handling liquid HC stream from Completion Separator	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
<u>Controlled</u> (combusted) Vent from Tank handling liquid HC stream from Completion Separator	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
<u>Atmospheric</u> Vent from Tank handling liquid water stream from Completion Separator	Included in the flowback to open tank	Included in the flowback to open tank	63,500 ± 6,000 scf	Included in the flowback to open tank	Included in the flowback to open tank
<u>Controlled</u> (combusted) Vent from Tank handling liquid water stream from Completion Separator	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
Gas from overhead of completion separator, sent to flare (assumed 2.0% of methane is uncombusted in flare)	16,000 scf	1,000 scf	44,000 scf[b]	Not applicable	Not applicable
Total (based on temporary stack cross sectional area x centerline velocity)	29,000 scf	7,700 scf	108,000	1,105,000 scf	240,000 scf
Total (based on temporary stack cross sectional area x centerline velocity x 0.8)	26,000 ± 8,000 scf	6,400 ± 700 scf	95,000± 5,000 scf	880,000 ± 300,000 scf	190,000 ± 100,000 scf

[a]Because of partial data loss, there is significant uncertainty, difficult to quantify, in the results from this completion; the data from this completion were not used in calculating averages or in regional and national extrapolations

[b]Includes 4,000 scf from flare and 40,000 scf from venting of separator;

*Configuration 1 (from Table S1-1): Initial flowback went to an open-top tank. After the initial period, the flow was sent to a separator. Gas from the separator was sent to a flare. Liquids from the separator were sent to flowback tanks that were vented

** Configuration 1 (from Table S1-1): Initial flowback went to an open-top tank. After the initial period, the flow was sent to a separator. Gas from the separator was sent to sales. Liquids from the separator were sent to flowback tanks that were vented

***Configuration 3 (from Table S1-1): Flowback to a separator; gas from the separator to sales; liquid from the separator to a vented flowback tank

Table S1-3. Methane emissions (scf) from Gulf Coast well completions: results from 7 sampling events

Emission Source (duration of completion flowback event, hr)	1 Company GC-A Configuration 1* (74.9 hr)	2 Company GC-A Configuration 1* (74.9 hr)	3 Company GC-B Configuration 2** (28.0 hr)	4 Company GC-B Configuration 2** (27.9 hr)	5 Company GC-C Configuration 4**** (13.8 hr)
Flowback to open top tank; gases vented	1300 ± 180 scf	500 ± 400 scf	40,000 ± 30,000 scf	13,000 ± 10,000 scf	21,600 ± 12,000 scf
Atmospheric Vent from Tank handling liquid HC stream from Completion Separator	3700 ± 550 scf	4800 ± 900 scf	Not applicable	Not applicable	Not applicable
Controlled (combusted) Vent from Tank handling liquid HC stream from Completion Separator	Not applicable	Not applicable	14,000 scf	20,000 scf	Not applicable
Atmospheric Vent from Tank handling liquid water stream from Completion Separator	600 ± 120 scf	200 ± 100 scf	60,000 scf	60,000 scf	Not applicable
Controlled (combusted) Vent from Tank handling liquid water stream from Completion Separator	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
Gas from overhead of completion separator, sent to flare (assumed 2.0% of methane is uncombusted in flare)	100,000 scf	85,000 scf	150,000 scf	90,000 scf	Not applicable
Total (based on temporary stack cross sectional area x centerline velocity)	106,000 scf	91,000 scf	264,000 scf	180,000 scf	21,600 scf
Total (based on temporary stack cross sectional area x centerline velocity x 0.8)	105,000 ± 600 scf	90,000 ± 800 scf	260,000 ± 30,000 scf	180,000 ± 8,000 scf	17,300 ± 10,000 scf

*Configuration 1(from Table S1-1): Initial flowback went to an open-top tank. After the initial period, flow was sent to a high pressure separator. Gas from the high pressure separator was sent to a flare; water from the high pressure separator was sent to a vented flowback tank. Hydrocarbon liquids from the high pressure separator were sent to a low pressure separator. Gas from the low pressure separator was sent to a flare; hydrocarbon liquids from the low pressure separator were sent to a vented flowback tank
**Configuration 2 (from Table S1-1): Initial flowback went to an open-top tank. After the initial period, the flow was sent to a separator. Gas from the separator was sent to a flare or to sales. Hydrocarbon liquids from the separator were sent to a flowback tanks that was vented to a combustion device.
****Configuration 4 (from Table S1-1): Flowback went to a vented tank.

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Table S1-3 (continued). Methane emissions (scf) from Gulf Coast well completions: results from 7 sampling events (Dark shading indicates that data were not used in determining average emission factors[a])

Emission Source	6[a] Company GC-A Configuration 2** (164 hr)	7[a] Company GC-A Configuration 2** (108 hr)
(duration of completion flowback event, hr)		
Flowback to open top tank; gases vented	1,000 scf	1,000 scf
Atmospheric Vent from Tank handling liquid HC stream from Completion Separator	Not applicable	Not applicable
Controlled (combusted) Vent from Tank handling liquid HC stream from Completion Separator	Not applicable	Not applicable
Atmospheric Vent from Tank handling liquid water stream from Completion Separator	3,000	3,000
Controlled (combusted) Vent from Tank handling liquid water stream from Completion Separator	Not applicable	Not applicable
Gas from overhead of completion separator, sent to flare (assumed 2.0% of methane is uncombusted in flare)	243,000 scf	86,000 scf
Total (based on temporary stack cross sectional area x centerline velocity)	247,000 scf	90,000 scf
Total (based on temporary stack cross sectional area x centerline velocity x 0.8)	247,000 scf	90,000 scf

[a]Because of partial data loss, there is significant uncertainty, difficult to quantify, in the results from this completion; the data from this completion were not used in calculating averages or in regional and national extrapolations

**Configuration 2 (from Table S1-1): Initial flowback went to an open-top tank. After the initial period, the flow was sent to a separator. Gas from the separator was sent to a flare. Hydrocarbon liquids from the separator were sent to a flowback tanks that was vented to a combustion device.

Table S1-4. Methane emissions (scf) from Mid-Continent well completions: results from 5 sampling events

Emission Source	1 Company MC-A	2 Company MC-A	3 Company MC-B	4 Company MC-B	5 Company MC-B
(duration of completion flowback event, hr)	Configuration 4**** (144.7 hr)	Configuration 4**** (147.2 hr)	Configuration 3*** (138.0 hr)	Configuration 3*** (138.0 hr)	Configuration 3*** (138.0 hr)
Flowback to open top tank; gases vented	250,000 ± 32,000 scf	34,000 ± 5,000 scf	Not applicable	Not applicable	Not applicable
Atmospheric Vent from Tank handling liquid HC stream from Completion Separator	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
Controlled (combusted) Vent from Tank handling liquid HC stream from Completion Separator	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
Atmospheric Vent from Tank handling liquid water stream from Completion Separator	Not applicable	Not applicable	3,400 scf	3,000 scf	3,400 scf
Controlled (combusted) Vent from Tank handling liquid water stream from Completion Separator	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
Gas from overhead of completion separator, sent to flare (assumed 2.0% of methane is uncombusted in flare)	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
Total (based on temporary stack cross sectional area x centerline velocity)	250,000 scf	34,000 scf	3,400 scf	3,000 scf	2,600 scf
Total (based on temporary stack cross sectional area x centerline velocity x 0.8)	200,000 ± 30,000 scf	27,000 ± 4,000 scf	2,700 scf	2,400 scf	2,100 scf

***Configuration 3 (from Table S1-1): Flowback to a separator; gas from the separator to sales; liquid from the separator to a vented flowback tank
****Configuration 4 (from Table S1-1): Flowback went to a vented tank.

Table S1-5. Methane emissions (scf) from Rocky Mountain well completions: results from 10 sampling events

Emission Source	1 Company RM-A	2 Company RM-A	3 Company RM-B	4 Company RM-B	5 Company RM-B
(duration of completion flowback event, hr)	Configuration 4**** (30.2 hr)	Configuration 4**** (30.1 hr)	Configuration 4**** (44.5 hr)	Configuration 4**** (34.3 hr)	Configuration 4**** (68.4 hr)
Flowback to open top tank; gases vented	30,000 ± 10,000 scf	16,400 ± 3,000 scf	13,000 ± 7,000 scf	37,000 ± 10,000 scf	49,000 ± 30,000 scf
Atmospheric Vent from Tank handling liquid HC stream from Completion Separator	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
Controlled (combusted) Vent from Tank handling liquid HC stream from Completion Separator	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
Atmospheric Vent from Tank handling liquid water stream from Completion Separator	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
Controlled (combusted) Vent from Tank handling liquid water stream from Completion Separator	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
Gas from overhead of completion separator, sent to flare (assumed 2.0% of methane is uncombusted in flare)	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
Total (based on temporary stack cross sectional area x centerline velocity)	30,000 scf	16,400 scf	13,000 scf	37,000 scf	49,000 scf
Total (based on temporary stack cross sectional area x centerline velocity x 0.8)	24,000 ± 8,000 scf	13,000 ± 2,000 scf	10,400 ± 6,000 scf	30,000 ± 8,000 scf	39,000 ± 30,000 scf

****Configuration 4 (from Table S1-1): Flowback went to a vented tank.

Table S1-5 (continued). Rocky Mountain methane emissions (scf) from well completions: results from 10 sampling events

Emission Source	6 Company RM-B	7 Company RM-C	8 Company RM-C	9 Company RM-C	10 Company RM-C
(duration of completion flowback event, hr)	Configuration 4**** (23.7 hr)	Configuration 1* (4.8 hr)	Configuration 1* (15.1 hr)	Configuration 1* (20.5 hr)	Configuration 1* (34.1 hr)
Flowback to open top tank; gases vented	42,000 ± 4,000 scf	40 scf	6,000 ± 2,000 scf	50,000 ± 5,000 scf	39,000 ± 11,000 scf
<u>Atmospheric</u> Vent from Tank handling liquid HC stream from Completion Separator	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
<u>Controlled</u> (combusted) Vent from Tank handling liquid HC stream from Completion Separator	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
<u>Atmospheric</u> Vent from Tank handling liquid water stream from Completion Separator	Not applicable	Included in the flowback to open tank	Included in the flowback to open tank	Included in the flowback to open tank	Included in the flowback to open tank
<u>Controlled</u> (combusted) Vent from Tank handling liquid water stream from Completion Separator	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable
Gas from overhead of completion separator, sent to flare (assumed 2.0% of methane is uncombusted in flare)	Not applicable	440 scf	9,000 scf	4,300 scf	6,500 scf
Total (based on temporary stack cross sectional area x centerline velocity)	42,000 scf	500 scf	15,000 scf	54,000 scf	45,500 scf
Total (based on temporary stack cross sectional area x centerline velocity x 0.8)	34,000 ± 3,000 scf	500 scf	12,000 ± 2,000 scf	44,000 ± 4,000 scf	37,700 ± 9,000 scf

*Configuration 1 (from Table S1-1): Initial flowback went to an open-top tank. After the initial period, the flow was sent to a separator. Gas from the separator was sent to a flare or to sales. Water and hydrocarbon liquids from the separator were sent to flowback tanks that were vented
****Configuration 4 (from Table S1-1): Flowback went to a vented tank.

Tables S1-2 to S1-5 provide data on 27 completion flowback events. Of these, 24 will be used to establish emission averages. The three completion flowbacks that were not considered in establishing averages (AP-5, GC-6 and GC-7) all had initial flowbacks into open top tanks, with gases vented to the atmosphere. In these completion flowbacks, the study team was unable to collect complete emission data for the initial flow to the open tank. Existing methods for estimating emissions during these initial flows do not provide reliable estimates, therefore, these completion flowbacks are not included in averages. Completion flowbacks MC-3, MC-4 and MC-5 also had some missing data, but in this case the completion flowbacks were included in the averaging. These completions involved no initial flow to an open top tank. Flowback went directly to a temporary separator; gas from the separator went to sales, and liquids from the separator went to a vented flowback tank (Configuration 3). The study team made several days of measurements, but the arrival of a hurricane necessitated removing the temporary stacks. The flowbacks continued throughout the hurricane. The study team used the completion reports to extrapolate data that had already been collected on the vent from the flowback tank. Because the study team was able to develop an extrapolation based on emission behavior that had already been directly measured for several days, the data were included.

Additional data for each of the 27 completions are provided in Table S1-6. Table S1-6 includes potential emissions for each of the completions, and compares net to potential emissions. The concept of potential, as opposed to net emissions is used by the US EPA in its national emission inventory.[4] In this work, the potential emissions from a completion flowback include the emissions that would occur if all of the methane flowing from the well during the completion flowback was emitted to the atmosphere. Configurations 1, 2 and 3 all involve some level of emission control, so measured emissions will be lower than potential emissions. In contrast, for Configuration 4, there are no emission controls so potential emissions and measured emissions are equal. The average fraction of emissions controlled was 98.6%, where:

Fraction of emissions controlled = 1- (Σ measured emissions / Σ potential emissions)
with the summation taken over 24 of the 27 emission events

Table S1-6. Potential and actual methane emissions for completion flowbacks

Completion flowback	Configuration (see Table S1-1)	Potential emissions (scf methane)	Measured emissions (scf methane)	Measured/ potential	Initial production (10^6 scf/day)[a]
AP-1	1	788,000	26,000	0.03	<0.01
AP-2	3	57,000	6,400	0.11	6.0
AP-3	3	390,000	95,000	0.24	6.8
AP-4	1	54,000,000	880,000	0.02	2.2
AP-5	1	48,000,000	190,000	0.004	7.5
GC-1	1	5,000,000	105,000	0.02	3.0
GC-2	1	4,250,000	90,000	0.02	2.7
GC-3	2	21,500,000	260,000	0.01	1.3
GC-4	2	13,000,000	180,000	0.01	0.8
GC-5	4	17,300	17,300	1	5.4
GC-6	2	12,200,000	247,000	0.02	6.1
GC-7	2	4,320,000	90,000	0.02	1.5
MC-1	4	200,000	200,000	1	1.3
MC-2	4	27,000	27,000	1	1.3
MC-3	3	20,500,000	2,700	0.0001	3.2
MC-4	3	17,500,000	2,400	0.0001	3.9
MC-5	3	18,700,000	2,100	0.0001	3.8
RM-1	4	24,000	24,000	1	Not avail.
RM-2	4	13,000	13,000	1	Not avail.
RM-3	4	10,400	10,400	1	0.3
RM-4	4	30,000	30,000	1	0.3
RM-5	4	39,000	39,000	1	0.4
RM-6	4	34,000	34,000	1	0.1
RM-7	1	22,000	500	0.02	3.2
RM-8	1	440,000	12,000	0.03	3.2
RM-9	1	254,000	44,000	0.17	1.7
RM-10	1	358,000	37,700	0.11	3.2
Average		6,500,000**	90,000**	0.014*	

[a]Average daily gas production for first 30 days after completion

*1- Σ measured emissions/ Σ potential emissions, with the summation taken over 24 of the 27 emission events, excluding AP-5, GC-6 and GC-7

**Average taken over 24 of the 27 emission events, excluding AP-5, GC-6 and GC-7

The data reported in Tables S1-2 to S1-6 are the first extensive measurements reported to date on methane emissions from well completion flowbacks. However, national inventories of methane emissions have been performed .[4] In the most recent EPA national greenhouse gas emission inventory,[4] a total of 8077 well completions with hydraulic fracturing are estimated to result in 63.6 billion scf of methane emissions for an average of 7.87 million scf of potential methane emissions per event. EPA then reduced their potential emission estimates due to assumed reductions from regulatory and voluntary controls. In the national inventory, EPA combines reductions associated with well completion flowbacks and workovers with hydraulic fracturing. In order to allow a comparison between the emissions reported in this work and an average emission per completion flowback in the national inventory, the same percentage reduction to potential emissions was applied to workovers and completion flowbacks. Specifically, since potential emissions for completion flowbacks (63.6 billion scf) and workovers with hydraulic fracturing (13.8 billion scf) totaled 77.4 billion scf, and since total reductions were 36 billion scf, the percentage reduction applied to potential emissions for both completion flowbacks and workovers with hydraulic fracturing was 46.4%. This leads to an estimate of 34 billion scf of net methane emissions for completion flowbacks and 7.4 billion scf of net emissions for workovers with hydraulic fracturing. The average net completion flowback emissions, per event is 4.2 million scf of methane. In this work, the average emission per completion flowback is 0.09 million scf per event, a reduction of 98% relative to the 4.2 million scf average for actual emissions in the EPA national inventory.

This large difference between the net emissions measured in this work and the net emissions estimated in the national inventory is due to several factors. First, the average potential emissions for completion flowbacks, measured in this work, are 20% lower than estimated by EPA. Second, 67% of the wells sent methane to sales or control devices. Third, for those wells with methane capture or control, 99% of the potential emissions were captured or controlled. Combined, these three factors account for approximately 80% of the reduction in emissions relative to the EPA inventory. Finally, the wells with uncontrolled releases had much lower than average potential to emit. Of the 9 wells in this work that had uncontrolled venting of methane, the average potential to emit was 43,000 scf (0.83 Mg), which is 0.55% of the average potential to emit in the national inventory. This accounts for the remainder of the emissions difference.

S1.3   Uncertainty Estimates

Confidence limits for the completion flowback emissions were estimated using two complementary approaches. As noted earlier in this section, uncertainties associated with composition and flow measurements were estimated and combined into an overall measurement uncertainty. For the completion flowbacks, this resulted in uncertainty bounds that were in the range of 20% of emissions. A complementary bootstrapping method[6] was employed to develop

an estimate of the combined sampling and measurement uncertainties. In the bootstrapping procedure, the original data set of 24 flowbacks was recreated by making 24 random event selections, with replacement, from the data set. A total of 1000 of these re-sampled data sets were created and the mean value of the emissions for each re-sampled data set was determined. The 95% confidence interval for the emission estimate of 90,000 scf is 35,000-173,000 scf, where the bounds represent the 2.5% and 97.5% percentiles of the means in the 1000 re-sampled datasets. The combined measurement and sampling uncertainty estimate from the bootstrapping procedure leads to a much larger uncertainty range than would be estimated from the uncertainty associated with the measurement alone. Therefore the overall uncertainty in the completion flowback emission estimate is reported as the uncertainty determined from the bootstrapping method.

S2   Direct Source Measurements: Wells in Routine Production

S2.1 Methods

Source types

Emission sources on production sites include pneumatically powered equipment, such as pumps and controllers, leaks from piping and equipment and flashing of methane from storage tanks. In addition, some sites may have equipment such as compressors that may have methane in their exhaust. The focus in this work was on measuring emissions from pneumatic pumps and controllers and measuring leaks from equipment, pipes, flanges and fittings. These sources were chosen for measurement because they are currently estimated to contribute over 20 bcf of the EPA national inventory from natural gas production.[4] Figure S2-1 shows a representative well site configuration with potential emission sources identified.

Figure S2-1. Gas Well Production Site
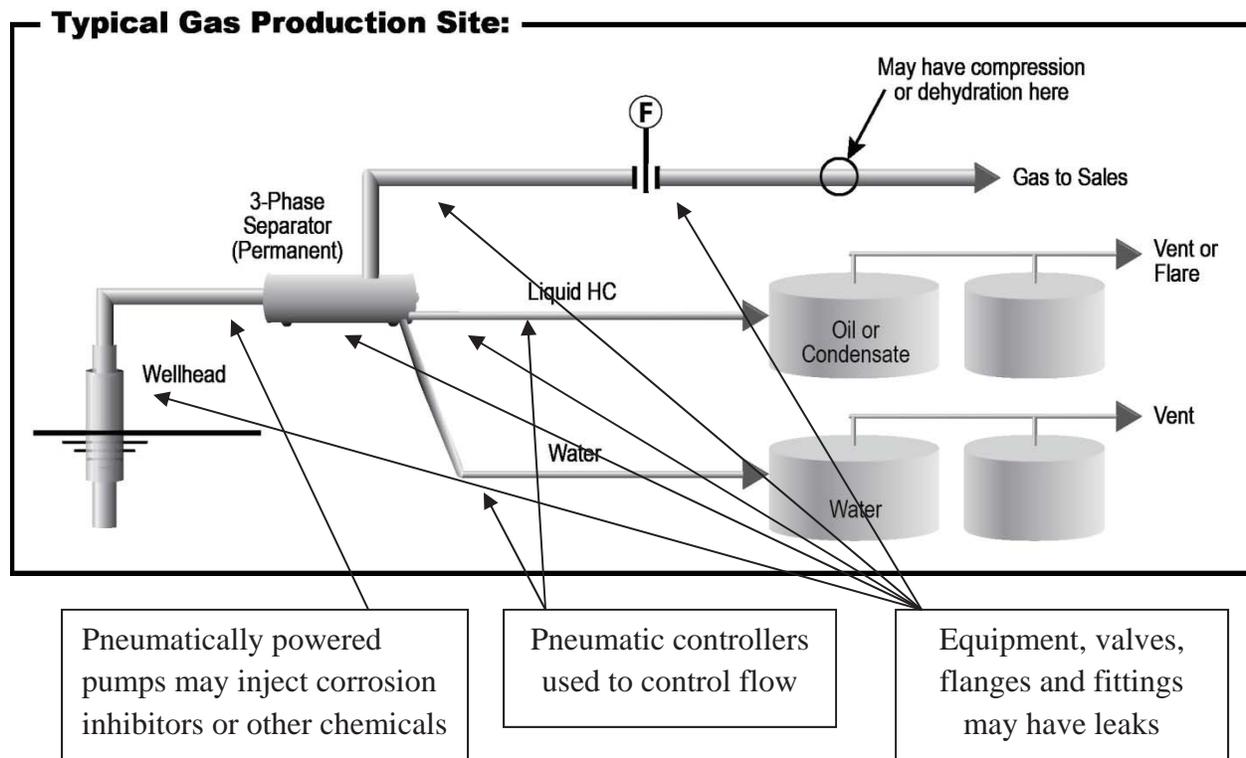
The equipment present on individual well sites can be highly variable. Sites could contain one or multiple wells. Some sites isolate wells from separators and their controllers and in these cases, a site may have no wells.   At sites with multiple wells, the wells might each have their own separator and tank system, or separator and tank systems servicing multiple wells might be in place. Additional equipment such as dehydrators and compressors were present on

some sites but not others. Some sites had solar powered devices (e.g., chemical injection pumps) or combustion control devices that reduced or eliminated emissions even if the equipment associated with production was the same.

This heterogeneity in the configuration of well sites has been documented in other studies. For example, in a study by the City of Fort Worth,[7] which reports on emissions from 375 well sites in the Barnett Shale production region (sites were randomly selected from the well sites that were within the City of Fort Worth), 30% of the sites had one well, 63% had between 2 and 6 wells, and one site had 13 wells. Similarly, while 78% of the sites had between 1 and 4 tanks, 16% had more than 4 tanks, and one site had 20 tanks. The potential sources of fugitive emissions, such as valves and flanges, varied by an order of magnitude or more between sites. Ten percent of the sites had less than 62 valves, but 10% had more than 446 valves. Ten percent of the sites had 390 or less connectors (such as flanges), but 10% had more than 3571.

Because of the heterogeneity of individual well sites, this study will not focus on average emissions per site. Instead, the data analysis reported here will be on individual equipment types and emissions per well. Specifically, emissions for chemical injection pumps and pneumatic controllers will be reported per device. The equipment leak measurements included leaks from wellhead equipment, piping, flanges, fittings, valves, separators, dehydrators, and non-exhaust emissions from compressors. Since the equipment count is expected to scale with the number of wells, emissions from equipment leaks are reported per well. Emissions for tanks were not examined because access to the multiple potential leak sites on tanks would have required a lift at each site, severely limiting the number of sites that could have been visited.   Measurements from exhaust gases (e.g., from compressor exhaust) were also not included.

For the pneumatic pumps and pneumatic controllers, emissions are reported as regional and national averages per device. Equipment leak emissions at a site are divided by the number of wells at a site to arrive at emissions per well. Emissions per well at each site were averaged on both a regional and national basis. These per device and per well emission factors are used in the extrapolation of the data reported here to regional and national estimates, as described in Section S5.

Table S2-1 summarizes the measurements made for each source type.

Table S2-1. Summary of equipment and sites sampled

Equipment type	Numbers of devices sampled in each production region				Total
	Appalachian	Gulf Coast	Midcontinent	Rocky Mtn.	
Chemical Injection Pump	0	21	41	0	62
Pneumatic Devices	133	106	51	15	305
Equipment leaks*	100	69	50	59	278
Number of distinct sites	47	58	26	19	150
Number of wells	168	157	85	79	489

*Includes leaks from wellhead equipment, piping, flanges, fittings, valves and separators; does not include flashing from tanks or engine exhaust gases

Measurement methods

       The initial step in the measurements was to scan the site using an infrared camera[8] to identify potential leak sources. Scanning with an infrared camera is an approved alternative work practice (40CFR60.18) used in identifying leaking equipment. In the alternative work practice, the threshold for detecting a leak, consistent with the practices used by the study team, is 30 g/hr (0.026 scf/m).   The threshold for detection of a leak with an infrared camera can depend, however, on operator interpretation of visual images and site specific parameters such as the background in the image of the potentially leaking component.

       Once the site was scanned with the infrared camera, all identified leaks were measured with a Hi-Flow Sampler.[9] The Hi-Flow Sampler is a portable, intrinsically safe, battery-powered instrument designed to determine the rate of gas leakage around various pipe fittings, valve packings, and compressor seals found in natural gas production, transmission, storage, and processing facilities. The Hi-Flow instrument has been used for several decades in measuring emissions of methane in natural gas production.[5,10,11] The instrument is packaged inside a backpack, thus leaving the operator's hands free for climbing ladders or otherwise accessing locations. The instrument comes with attachments for enclosing leaking devices and is controlled by a handheld unit consisting of an LCD and a 4-key control pad, which is attached to the main unit via a 6 foot coiled cord.

       A component's leak rate is measured by sampling at a high flow rate so as to capture all the gas leaking from the component along with a certain amount of surrounding air. By accurately measuring the flow rate of the sampling stream and the natural gas concentration within that stream, the gas leak rate can be calculated (see Equation below). The instrument

automatically compensates for the different specific gravity values of air and natural gas, thus assuring accurate flow rate calculations.

$$Leak = Flow * (Gas\ sample - Gas\ background) * 10^{-2}$$

Where: Leak = Rate of gas leakage from source (cfm)
              Flow = Sample flow rate (cfm)
              Gas sample = Concentration of gas from leak source (volume %)
              Gas Background = Background gas concentration (volume %)

The gas sample is drawn into the main unit through a flexible 1.5 inch I.D. hose. Various attachments connected to the end of the sampling hose provide the means of capturing all the gas that is leaking from the component under test.

The main unit consists of an intrinsically safe, high-flow blower that pulls air, at up to 10 scf/m, from around the component being tested through a flexible hose and into a gas manifold located inside the unit. The sample is first passed through a restrictor where the measured pressure differential is used to calculate the sample's actual flow rate. Next, a portion of the sample is drawn from the manifold and directed to a combustibles sensor that measures the sample's methane concentration in the range of 0.05 to 100% gas by volume. The combustibles sensor consists of a catalytic oxidizer, designed to convert all sampled hydrocarbons to CO_2 and water. A thermal conductivity sensor is then used to determine CO_2 concentration. A second identical combustibles sensor channel measures the background methane level within the vicinity of the leaking component.

The instrument was calibrated using samples consisting of pure methane in ambient air. However, when natural gas emissions are measured, the instrument will encounter additional hydrocarbons (typically ethane, propane, butane and higher alkanes). To account for the effect of these species on the measurements, gas composition data were collected for each natural gas production site that was visited. Typically this gas analysis was provided by the site owner. Based on the gas composition, provided for each site in the study data set, the percentage of carbon accounted for by methane, in the sample stream, was determined. This percentage, multiplied by the total gas flow rate reported by the instrument, was the methane flow.

The final element in the sampling system is a blower that exhausts the gas sample back into the atmosphere away from the sampling area. The measured flow rate and the measured methane levels (both leak and background levels) are used to calculate the leak rate of the component being tested, with all measured and calculated values being displayed on the handheld control unit.

Once the equipment leak emissions, detected by the infrared camera were quantified, emissions from pneumatic chemical injection pumps and pneumatic controllers were measured with the Hi-Flow Sampler. All operating pneumatic Chemical Injection Pumps were sampled.

Some sites had solar powered electrical pumps, which did not emit methane in normal operation as pneumatic pumps do. Other sites had pneumatic pumps installed but not in operation. Still other sites did not have pumps. Both solar powered and non-operating pneumatic pumps were only sampled if leaks were detected using the infrared camera.

Because many of the devices sampled had intermittent flows (e.g., pneumatic pumps and controllers), a variety of methane concentrations were encountered by the Hi-Flow measurement system as the operation cycle for a pump or controller was sampled. Because of this intermittency in flow, determining the detection limit for the measurement system is not simple. It can be quantified based on the smallest non-zero emission rate measured. In this work, the smallest non-zero emission rate measured by the Hi-Flow system was 0.00048 scf/m and therefore the detection limit will be assumed to be less than or equal to that value.

Measurements were made on a total of 305 pneumatic controllers, representing an estimated 41% of the controllers, randomly sampled from the controllers associated with the wells that were sampled. This approach of random sampling was adopted after the first sites had already been visited. For the first sites, only pneumatic controllers that were observed to be actively emitting methane were sampled. Statistical analysis of the data collected using the two approaches showed no systematic difference so the data for the controllers were treated as one dataset.

Data analysis methods and uncertainty reporting

Average methane emission rates, by equipment type, will be the primary method of data reporting in this section. The uncertainty in these average emission estimates is dominated by the uncertainty in the representativeness in the sample set. There are hundreds of thousands of natural gas production wells in the United States, and the number of sites sampled in this work, while large in comparison to other emission data sets, is small relative to the total number of sites available. Therefore, the uncertainties reported in this section will characterize the expected uncertainty in the emission means, using a method referred to as bootstrapping.[6]

In the bootstrapping procedure, a data set was re-sampled at random (with replacement). For example, for Chemical Injection Pumps, the original data set of 62 pumps was recreated by making 62 random pump selections, with replacement, from the data set. A total of 1000 of these re-sampled data sets were created and the mean value of the emissions for each re-sampled data set was determined. The bounds reported here represent the 2.5% and 97.5% percentiles of the means in the 1000 re-sampled datasets. This bootstrapping procedure was used to establish uncertainty estimates for chemical injection pumps, pneumatic controllers and equipment leaks.

S2.2   Results and Discussion

Pneumatic Chemical Injection Pumps

Pneumatic Chemical Injection Pumps use the pressure from on-site natural gas to drive pumps that inject anti-corrosion and other liquids into the produced gas stream. Table S2-2 reports emission rates, by region, and a national average, for Chemical Injection Pumps.

Not all wells had active Chemical Injection Pumps. For example, no operating Chemical Injection Pumps were encountered at active production sites in the Appalachian or Rocky Mountain regions. When Chemical Injection Pumps were present, some were solar powered (no routine methane emissions), and some wells had pneumatic injection pumps that had been installed but were not in operation (e.g., because the liquids, such as anti-corrosion additives, were not required by the well at that point in the well life).

Table S2-2 reports both "whole gas" emission rates, and methane emission rates. The methane emissions rate is based on the Hi-Flow Sampler measurement.   Whole gas (natural gas) emissions are reported here since emission factors are expressed in US EPA emission factors as whole gas emissions per device.

Table S2-2. Emissions from Chemical Injection Pumps

	Emissions per Pneumatic Chemical Injection Pump*				
	Appalachian	Gulf Coast	Midcontinent	Rocky Mtn.	Total
Number sampled		21	41		62
Emissions rate (scf methane/min/device)**		0.476 ± 0.200	0.047 ± 0.013		0.192 ± 0.085
Emissions rate (scf whole gas/min/device, based on site specific gas composition)**		0.506 ± 0.209	0.050 ± 0.014		0.204 ± 0.089

*Solar powered pumps, and pneumatic pumps that were present but not in operation are not included in the total

**Uncertainty characterizes the variability in the mean of the data set (as described in Section S2.1), rather than an instrumental uncertainty in a single measurement

The average values of emissions per pump for Chemical Injection Pumps reported here are similar to the emission factor suggested by EPA[3] for use in estimating methane emissions (13.3 scf whole gas per pump per hour vs. 12.2 (9% lower) reported here). As described in Section S5, however, if estimated emission reductions are applied to potential emissions, the net EPA estimate will be less per pump than the values reported here.

There is significant geographical variability in the emissions rate from Chemical Injection Pumps between production regions. Emissions per pump from the Gulf Coast are statistically different (higher) than emissions from pumps in the Midcontinent region. The difference in average values is roughly an order of magnitude.

A number of hypotheses were examined to attempt to explain the differences in emissions. Volume of liquid pumped was not a good predictor of emissions. Well head and separator pressure were considered since the pumps must overcome these pressures to drive liquid flow. These variables also were not good predictors of emissions. Company specific practices were also considered. While roughly 90% of the samples came from two companies, one from each region (see Section S6), a total of 6 companies provided data, 3 in the Gulf Coast and 3 in the Midcontinent, and for all of these companies the same regional differences (Gulf Coast emissions > Midcontinent) were observed. Mean values of emissions, by company, were similar in each of the regions.   Other possibilities, that have not yet been investigated, but that may be pursued in follow-up work, include pump design or local regulatory requirements.

Pneumatic Controllers

Pneumatic Controllers use the pressure from on-site natural gas to drive devices that actuate valves controlling flow from units such as separators to units such as tanks. Table S2-3 reports emission rates, by region and a national average, for Pneumatic Controllers.

Table S2-3. Emissions from Pneumatic Controllers

	Emissions per Pneumatic Controller*				
	Appalachian	Gulf Coast	Midcontinent	Rocky Mtn.	Total
Number sampled	133	106	51	15	305
Emissions rate (scf methane/min/device)**	0.126 ± 0.043	0.268 ± 0.068	0.157 ± 0.083	0.015 ± 0.016	0.175 ± 0.034
Emissions rate (scf whole gas/min/device, based on site specific gas composition)**	0.130 ± 0.044	0.289 ± 0.071	0.172 ± 0.086	0.021 ± 0.022	0.187 ± 0.036

*Intermittent and low bleed controllers are included in the total; no high bleed controllers were reported by companies providing controller type information
**Uncertainty characterizes the variability in the mean of the data set (as described in Section S2.1), rather than an instrumental uncertainty in a single measurement

The average values of emissions per device for Pneumatic Controllers reported here are comparable to the values suggested by EPA[3] for use in estimating methane emissions (1.39, 37.3 and 13.5 scf whole gas per device per hour for low bleed, high bleed and intermittent bleed controllers vs. 11.2 reported here for a mix of intermittent and low bleed controllers). No high bleed controllers were reported by the companies that provided controller type information. At a total of 55 sites, site operators reported only intermittent controllers and at 24 sites, site operators reported only low bleed controllers. These sites, where potential mis-identification of controller type is less likely to be a confounding factor, can be used to establish separate emission factors for intermittent and low-bleed devices. These emission factors are 0.290±0.120 scf natural gas per device per minute (17.4 scf/h, 5.9±2.4 g scf/m assuming a natural gas density of 20.3 g/scf, as measured in this work) for intermittent controllers and 0.085±0.049 scf/m (5.1 scf/h, 1.7±1.0 g scf/m assuming a natural gas density of 20.3 g/scf, as measured in this work) for low bleed controllers. For intermittent and low bleed controllers, the measured emission factors are 29% and 270% higher than the EPA emission factors (expressed in units of scf whole gas per hour), respectively.

There is significant geographical variability in the emissions rate from pneumatic controllers between production regions. Emissions per controller from the Gulf Coast are highest and are statistically different than emissions from controllers in Rocky Mountain and Appalachian regions. The Rocky Mountains have the lowest emissions. The difference in average values is more than a factor of ten between Rocky Mountain and Gulf Coast regions.

Some of the regional differences in emissions may be explained by differences in practices for utilizing low bleed and intermittent controllers. For example, new controllers installed after February 1, 2009 in regions in Colorado that do not meet ozone standards, where most of the Rocky Mountain controllers were sampled, are required to be low bleed (or equivalent) where technically feasible (Colorado Air Regulation XVIII.C.1; XVIII.C.2; technical feasibility criterion under review as this is being written). However, observed differences in emission rates between intermittent and low bleed devices (roughly a factor of 3) are not sufficient to explain all of the regional differences. A number of additional hypotheses were examined to attempt to explain the differences in emissions. For datasets consisting entirely of intermittent or entirely of low-bleed devices, the volume of oil produced was not a good predictor of emissions. Well head and separator pressure were also not good predictors of emissions. The definition of low-bleed controllers may be issue, however. All low bleed devices are required to have emissions below 6 scf/hr (0.1 scf/m), but there is not currently a clear definition of which specific controller designs should be classified as low bleed and reporting practices among companies can vary. Other possibilities for explaining the low-bleed emission rates observed in this work, that have not yet been investigated, but that may be pursued in follow-up work, include operating practices for the use of the controllers.

Emissions from equipment leaks

Emissions from leaks in piping, valves, separators, wellheads, and connectors located on site are reported in Table S2-4. The data are reported as emissions normalized by the number of wells on each site. Out of the 150 sites visited, 146 had wells on the sites. The remaining 4 sites, all in the Gulf Coast region, had separators and other equipment on site, but no wells. Some companies operating in the Gulf Coast region isolate wells from separators and aggregate separators for multiple wells on a single site. Because these sites did not include all of the equipment associated with natural gas production, and because the wells associated with the separators were not sampled, these four sites were excluded in the data averaging. The equipment at the four sites with no wells was estimated to be associated with 11 off-site wells, making a well count of 478 for 146 sites. The average emissions per well for these four sites (assuming one well per separator located at the site) were all less than the average per well emissions reported for the Gulf Coast.

Emissions are reported per well because the variability in the number of wells and the type of equipment located on well sites makes averaging emissions per site a less useful way to represent equipment leak data than average emissions from leaks per well (leaks at a site divided by the number of wells at the site). Further, the number and type of equipment that could be potential leak sources generally scales with the number of wells.

Table S2-4. Emissions from equipment leaks

	Emissions per Well*				
	Appalachian	Gulf Coast	Midcontinent	Rocky Mtn.	Total
Number of sites with wells visited (number of sites with leaks detected)	47 (30)	54 (31)	26 (19)	19 (17)	146 (97)
Emissions rate (scf methane/min/well)**	0.098 ± 0.059 scf/m/well	0.052 ± 0.030 scf/m/well	0.046 ± 0.024 scf/m/well	0.035 ± 0.026 scf/m/well	0.064 ± 0.023 scf/m/well
Emissions rate (scf whole gas/min/well, based on site specific gas composition)**	0.100 ± 0.060 scf/m/well	0.058 ± 0.033 scf/m/well	0.055 ± 0.034 scf/m/well	0.047 ± 0.034 scf/m/well	0.070 ± 0.024 scf/m/well

*All leaks detected with the FLIR camera, not including pneumatic pumps and controllers are included in the total

**Uncertainty characterizes the variability in the mean of the data set (using a bootstrapping method as described in Section 2.3), rather than an instrumental uncertainty in a single measurement

The average values of equipment leak emissions per well reported here are similar to the average values of potential emissions per well for gas wells, separators, heaters, piping and dehydrator leaks (0.072 scf methane/min/well), calculated by dividing the potential emissions in these categories in the EPA national inventory by the number of wells.[4] Two issues confound this comparison, however. First, measurements made in this work included non-exhaust emissions from compressors that were located on well sites. These compressors can perform a variety of functions, including lift and compression for delivery into sales lines. The national inventory groups fugitive emissions from all of these types of compressors into a category for gathering compressors (3.5 billion scf/year; 0.015 scf/m per well). It would be appropriate to include some of these emissions in the comparisons to the measurements made in this work, but not all of the emissions, since this work did not collect data on all gathering compressors for the wells that were sampled. A second factor confounding comparisons with the national inventory is that the EPA calculates net emissions in the national inventory by subtracting reductions from potential emissions. The equipment leak reductions are reported as an aggregate reduction that also includes reductions associated with blowdowns, pressure relief valves, some coal-bed methane categories and other source categories (see Section S5). If these reductions are assumed to be the same percentage of potential emissions for these categories, the emissions in the national inventory (not including compressors) are 9 billion scf (172 Gg, 0.04 scf/m per well). These estimated net emissions from equipment leaks are roughly half to two-thirds (depending on how compressors are included) of the emissions measured in this work.

S2.3   Uncertainty Estimates

Confidence limits for the emissions were estimated using two complementary approaches. Uncertainties associated with composition and flow measurements were estimated as approximately 10% of emissions. A complementary bootstrapping method[6] was employed to develop an estimate of the combined sampling and measurement uncertainties. In the bootstrapping procedure, the original data set of was recreated by making random event selections, with replacement, from the data set. A total of 1000 of these re-sampled data sets were created and the mean value of the emissions for each re-sampled data set was determined. The 95% confidence interval for the emission estimate represents the 2.5% and 97.5% percentiles of the means in the 1000 re-sampled datasets. The combined measurement and sampling uncertainty estimate from the bootstrapping procedure leads to a much larger uncertainty range than would be estimated from the uncertainty associated with the measurement alone. Therefore the overall uncertainty in the emission estimate is reported as the uncertainty determined from the bootstrapping method.

S3   Direct Source Measurements: Gas Well Liquids Unloading

S3.1   Methods

The method used to measure emissions from manual liquid unloading of a gas well, by well blowdown, is similar to the method used to measure emissions from flowback tanks, described in Section S1. Flow is directed through a portable stack installed on top of the tank vent on the blowdown site tanks. Figure S3-1 shows a temporary stack in use. Grounded metal or metal lined tubing was used to prevent static discharge. Flow rate through the temporary stack was measured continuously, near the centerline of the temporary stack, using a pitot tube.

Figure S3-1. Temporary Stack on Blowdown Site Tank Hatches

Where there were multiple tanks manifolded together, either all of the blowdown was routed to a single tank with a temporary stack, or temporary stacks were placed on all of the tanks that were vented.   Total volumetric flow was calculated by multiplying the cross-sectional area of each stack by 80% of the gas velocity at the stack centerline. The factor of 0.8 was used to convert the centerline velocity in the stack to an estimated average velocity in the stack, accounting for the change in velocity profile from friction near the stack walls.[1]

Since the gas vented is the produced natural gas, the methane fraction of the vented gas will be assumed to be equal to the methane fraction in the normally produced gas. This was presumed to be a more accurate indicator of total emissions than measurements of the gas composition made through the temporary stack. The gas exiting through the temporary stack during the blowdown period is a combination of the blowdown gas and the gas initially in the tank (typically much lower in methane than the site's produced gas). At the end of the

blowdown, the tank will contain more methane, from the blowdown, than was in the tank at the start of the blowdown. This methane, which is associated with the blowdown event, will eventually be released as part of normal tank operations. Multiplying vented gas volume by production gas methane fraction captures these emissions that occur because of the blowdown but that are not released during the period when the tank is actively venting through the thief hatch.

Uncertainty in these measurement methods is estimated at 10% of the measured emissions and this estimate is dominated by the assumed uncertainty in the flow (10%). Variability in the gas composition from the well is expected to be much less than 10%. As described later in this section, these measurement uncertainties are small compared to the combined sampling and measurement uncertainty.

S3.2   Results and Discussion

Emissions were measured for a total of 9 gas well liquid unloading events for non-plunger lift wells. Measurements were made in the Appalachian, Gulf Coast, and Rocky Mountain production regions. No data were taken in the Midcontinent region because there were no unloadings at the visited fields during the measurement campaign. Data are presented in Tables S3-1 and S3-2. Unloading events 1a-1c were performed on three different wells at a single well site and unloading events 2a-c were also performed at three different wells at a single well site, in a different production region than Events 1a-c.

The unloadings were heterogeneous in their characteristics. Methane emissions ranged from less than 1,000 scf to 191,000 scf. Some unloadings lasted two hours (or more) and had relatively uninterrupted flow (Events 1a-b). Other unloadings were as short as 10-15 minutes (e.g, Events 2b and 3) with uninterrupted flow and still others had intermittent flow for short periods and periods of no flow for much of the unloading period (e.g., Events 2a, 2c).

The data from the unloading events can be averaged in multiple ways. One method for averaging the emissions is to consider emissions per event. Total emissions for the nine events are summed and divided by the number of events (9 events). This leads to an average of 57,000 scf of methane per event and a median value of 5,000 to 11,000 scf. Bootstrapping methods (see Section S1) established 95% confidence bounds of 17,000-105,000 scf. The emissions from four of the 9 events contribute over 95% of the total emissions, so if this sample is representative, there is a population of high emitting events and a population of low emitting events.

A second method for analyzing the data recognizes that average emissions are often used to establish an annual emission estimate for unloading for individual wells. An annual emission estimate will multiply the emissions per event by a frequency (events per year) of the events. These calculations are reported in Table S3-2. For the nine wells for which data were available, this average was 300,000 scf per well per year (95% confidence limit of 100,000-620,000 scf). This per well average of unloading emissions is comparable to the 215,000 scf average emissions

per well per year for unloading without plunger lifts in EPA's national inventory (7,734 million scf for unloadings without plunger lifts for 35,828 wells with this type of unloading[4]) and an estimate of 240,000 scf methane based on a survey conducted by the American Petroleum Institute and America's Natural Gas Alliance.[12]   Again, however, the data are skewed with three very low emitting wells in the Rocky Mountain region, and much higher emissions per well in the Gulf Coast and Appalacian regions.

Table S3-1. Emissions and well data for measurements of manual well unloading

Event (Region)	Volume vented, scf Raw and (corrected)	Methane in produced gas (vol %)	Methane emitted per event (scf)[b]	Duration of blowdown[c] (hr)	Volume of well bore (ft³)	Well shut-in pressure (psia)	Normal production rate for well (scf/hr)	Events per year for well[g]
1a (GC)	248,500[a] (199,000)[b]	96%	191,000	2.77[d]	10,906	300	374,000	7
1b (GC)	208,100[a] (166,000)[b]	96%	159,000	1.904[d]	10,906	300	374,000	1
1c (GC)	85,800[a] (68,600)[b]	96%	65,900	0.63[d]	10,906	300	374,000	1
2a (RM)	1,810[a] (1,450)[b]	92.9%	1,350	0.75[e]	1,875	527	295,000	2
2b (RM)	1,770[a] (1,420)[b]	92.9%	1,320	0.2[d]	1,876	642	169,000	4
2c (RM)	1,270[a] (1,020)[b]	92.9%	950	1.25[e]	1,900	1116	304,000	2
3 (AP)	14,550[a] (11,600)[b]	97.4%	11,300	0.25[d]	1,404	890	208,000	12
4 (GC)	5670[a] (4540)[b]	84.4%	3,800	1.1[f]	1,977	1500	25,000	12
5 (GC)	121,200[a] (97,000)[b]	81.4%	79,000	1.25[d]	1,977	1450	16,700	12
Avg.	76,500 (61,200)	93%	57,000	1.0	4,900	780	240,000	5.9

[a]based on temporary stack cross sectional area * centerline velocity
[b]based on temporary stack cross sectional area * centerline velocity * 0.8
[c]measured based on the time of first appearance of gas flow in temporary stack to end of gas flow in temporary stack
[d]Once gas flow began, flow was continuous until the end of the unloading
[e]An initial burst of flow for ~5 minutes, flowed by a period of no flow, followed by a burst of flow for ~5-15 minutes
[f]Flow for 1 hour 5 minutes with 4 bursts of flow of up to 15 minutes, periods of no flow of up to 35 minutes.
[g]Reported by companies that provided the wells for sampling

Table S3-2. Emissions estimates per well per year for manual well unloading

Event	Region[a]	Methane emitted per event (scf)[b]	Events per year for well	Emissions per year for well (scf)
1a	GC	191,000	7	1,337,000
1b	GC	159,000	1	159,000
1c	GC	65,900	1	65,900
2a	RM	1,350	2	2,700
2b	RM	1,320	4	5,280
2c	RM	950	2	1,900
3	AP	11,300	12	136,000
4	GC	3,800	12	45,600
5	GC	79,000	12	948,000
Avg.		57,000	5.9	300,000

[a]GC: Gulf Coast; RM: Rocky Mountain; NE: Northeast
[b]based on temporary stack cross sectional area * centerline velocity * 0.8

Since the number of events sampled is very small relative to the total number of wells and unloading events (35,828 wells with unloading events without plunger lifts in the 2013 EPA national inventory), the characteristics of the wells sampled in this work should be compared to wider populations. One source of data is a survey reported by the American Petroleum Institute and America's Natural Gas Alliance.[12] In this survey, over 20 companies provided unloading data on 40,000-60,000 wells (with the number in the sample depending on the type of emission event). Based on these survey data, API/ANGA estimate national totals of 28,863 wells without plunger lift that vent for unloading and 36,806 wells with plunger lift that vent for unloading. For the non-plunger lift wells, API/ANGA report an average of 32.57 events per well per year, higher than the average of 5.9 in this work. The average duration is 1.90 hours, which is roughly double the average time of 1.0 hr for the unloadings sampled in this work. The average release for wells without plunger lift (based on data in Appendix C of API/ANGA[12]) is 304,000 scf of gas or 240,000 scf methane per well per year, assuming that gas is 78.8% methane. This is consistent with the data reported in this work (300,000 scf methane per well per year), however, while the per well annual emission rates for the 9 wells sampled in this work are consistent with the per well annual emissions in the API/ANGA data, there are significant differences between the two populations. One major difference is the frequency of unloading. The wells in the API/ANGA survey have an average of 32.57 unloadings per year, while in this work the average is 5.9. This means that the average per event, accounting for the different frequency of unloading of individual wells, is 9300 scf gas (7350 scf methane) in the API/ANGA survey and 57,000 scf methane in the observations reported here. The API/ANGA dataset contains more wells that unload with high frequency, but lower emissions per event, than the data reported here.

Another difference between the API/ANGA survey reports and the data reported here is that the API/ANGA dataset relies on estimated, rather than measured emissions. The emissions were estimated using the method suggested for unloading events in EPA's Greenhouse Gas Reporting Program (GHGRP).[3] Methodology 2 for unloading without plunger lifts in the GHGRP[3] assumes that the volume in the entire length of the pressurized well is vented to the atmosphere. This is assumed to occur during the first hour of the blowdown, if the blowdown lasts more than one hour, and any gas flow beyond 1 hour is assumed to occur at the normal well gas flow rate production rate. If the blowdown lasts for less than one hour, the emissions are assumed to be equal to the volume in the pressurized well. The equation (W-8) provided by EPA is:

$$E_{s,n} = \sum_{p=1}^{W}\left[V_p \times\left(\left(0.37\times10^{-3}\right)\times CD_p^2 \times WD_p \times SP_p\right) + \sum_{q=1}^{V_p}\left(SFR_p \times\left(HR_{p,q} - 1.0\right)\times Z_{p,q}\right)\right]$$

Where:

$E_{s,n}$ = Annual natural gas emissions at standard conditions, in cubic feet/year; this work assumes one event and reports the results per event

W = Total number of wells with well venting for liquids unloading for each sub-basin = 1 in this work.

0.37×10^{-3} = {3.14 (π)/4}/{14.7*144} (psia converted to pounds per square feet).
CD_p = Casing internal diameter for each well, p, in inches.
WD_p = Well depth from either the top of the well or the lowest packer to the bottom of the well, for each well, p, in feet.
SP_p = For each well, p, shut-in pressure or surface pressure for wells with tubing production or casing pressure for each well with no packers in pounds per square inch absolute (psia); or casing-to-tubing pressure ratio of one well with no packer from the same sub-basin multiplied by the tubing pressure of each well, p, in the sub-basin, in pounds per square inch absolute (psia); in this work the product of 0.37×10^{-3}* CD_p* WD_p* SP_p is obtained by multiplying the well volume (in ft^3, from Table S3-1), by the shut-in pressure (in psia, from Table S3-1) and dividing by 14.7

V_p = Number of unloading events per year per well, p; assumed equal to 1 in this work
.
SFR_p = Average flow-line rate of gas for well, p, at standard conditions in cubic feet per hour; for this work these data are reported in Table S3-1.

$HR_{p,q}$ = Hours that each well, p, was left open to the atmosphere during each unloading event, q; for this work these data are reported in Table S3-1.

1.0 = Hours for average well to blowdown casing volume at shut-in pressure.

$Z_{p,q}$ = If $HR_{p,q}$ is less than 1.0 then $Z_{p,q}$ is equal to 0. If $HR_{p,q}$ is greater than or equal to 1.0 then $Z_{p,q}$ is equal to 1.

Data for all of the input variables for EPA equation W-8 (above) were collected from each study participant on the wells where direct measurements were made and are reported in Tables S3-2 and S3-3. Table S3-3 reports the results of applying this estimation method to the 9 well unloadings (without plunger lift) sampled in this work.

Table S3-3. Comparison of measured and estimated gas volumes emitted during well blowdown

Event number	Measured Volume vented (scf)	Total Emission Estimate per event based on Equation W-8 (scf)	Total Emission Estimate per well per year based on Equation W-8 and events/yr (Table 4-2) (scf)	Emissions based on well bore volume from Equation W-8 (scf)	Emissions, after hour 1, based on production rate (scf)
1a	248,500[a] (199,000)[b]	884,600	6,192,600	222,600	662,000
1b	208,100[a] (166,000)[b]	559,200	559,200	222,600	336,600
1c	85,800[a] (68,600)[b]	222,600	222,600	222,600	0
2a	1,810[a] (1,450)[b]	67,200	134,400	67,200	0
2b	1,770[a] (1,420)[b]	81,900	327,600	81,900	0
2c	1,270[a] (1,020)[b]	144,200	288,400	144,200	0
3	14,550[a] (11,600)[b]	85,000	1,020,000	85,000	0
4	5670[a] (4540)[b]	204,200	2,450,200	201,700	2,500
5	121,200[a] (97,000)[b]	199,200	2,390,000	195,000	4,200
Avg.	76,500[a] (61,200)[b]	270,000	1,500,000	159,000	111,000

[a]based on temporary stack cross sectional area * centerline velocity
[b]based on temporary stack cross sectional area * centerline velocity * 0.8

In general, a simplified model assuming that the entire volume of the pressurized well is emitted during an unloading appears to work in some cases (e.g., Events 1a and 1b), but not in others (e.g., Events 2a-c). Further, the detailed temporal patterns of gas flow observed in this

work do not support the concept of a transition in the mechanism of flow after a one hour time period.

Overall, the average emission estimate, employing EPA emission estimation methods, for the 9 unloadings reported here (270,000 scf methane), is roughly five times the measured average per event of 57,000 scf. If the estimated emissions are calculated by well (multiplying the emissions per event by the events per year for the well), the average is 1,500,000 scf methane, six times the average in the API/ANGA survey.

All of these averaging methods assume a single scalar value represents a wide range of unloadings; the data presented in this work and in the API/ANGA survey suggest that refined emission estimation methods, taking into account well and unloading characteristics, will be required. Additional measurements of unloading emissions are needed, both to resolve the differences between estimates and measurements, and to better characterize the population of wells with unloading emissions.

Finally, it is also clear from the data that properly accounting for unloading emissions will be important in reconciling emission inventories with regional ambient measurements. Average methane emission rates for a single unloading ranged from roughly a hundred grams per minute (5 scf/m) to in excess of 30,000 grams per minute (1500 scf/m), with a mean value of approximately 10,000 g/min (500 scf/m). Values for specific unloadings can be calculated from the data in Table S3-1. The unloading emission rates are much larger than emission rates for production sites (typically approximately 1 scf/m per well) or from completions (typically tens of scf/m per event). At these emission rates, a single unloading event could, during the very short period that it is occurring, result in emissions that are the equivalent of just a few wells in routine production to the equivalent of up to several thousand wells in routine production. This indicates that reconciliation between instantaneous ambient measurements and emission inventories will need to very carefully represent the emissions from unloadings.

S3.3   Uncertainty Estimates

Confidence limits for the unloading emissions were estimated using two complementary approaches. As noted earlier in this section, uncertainties associated with composition and flow measurements were estimated as approximately 10% of emissions. A complementary bootstrapping method[6] was employed to develop an estimate of the combined sampling and measurement uncertainties. In the bootstrapping procedure, the original data set of 9 unloadings was recreated by making 9 random event selections, with replacement, from the data set. A total of 1000 of these re-sampled data sets were created and the mean value of the emissions for each re-sampled data set was determined. The 95% confidence interval for the emission estimate of 57,000 scf is 17,000-100,000 scf, where the bounds represent the 2.5% and 97.5% percentiles of the means in the 1000 re-sampled datasets. The combined measurement and sampling uncertainty estimate from the bootstrapping procedure leads to a much larger uncertainty range than would be estimated from the uncertainty associated with the measurement alone. Therefore

the overall uncertainty in the unloading emission estimate is reported as the uncertainty determined from the bootstrapping method.

S4   Downwind Tracer Ratio Measurements at Natural Gas Production Sites

S4.1   Measurement Description and Objective

The overall goal of the sampling downwind of natural gas production sites was to perform instantaneous and time integrated measurements of the total methane emissions from natural gas production sites. The resulting emissions measurements represent a site aggregated emission estimate and complement on-site measurements of emissions from multiple emission sources. The objective of these downwind measurements is to determine whether the direct source measurements are capturing all significant sources of emissions, and to assess the magnitude of emissions of methane that were not directly measured using the methods employed in this study, such as emissions that are part of the exhaust gas of devices such as flares.

The measurements employed tracer release methodologies to quantify the total methane emission rate coming from a site. Tracer species were emitted at a controlled rate, on site, at locations as close as possible to methane releases. The tracer species and methane were measured at downwind locations (100 m to more than 1 km). Upwind concentrations were measured, as required, if downwind mobile sampling indicated that concentrations did not return to baseline values outside of detected plumes. If it is assumed that the tracer disperses in a manner equivalent to the methane, the ratio of the far field concentrations of the tracer gas and the sample gas will be the same as the ratios of their emission rates. Thus, the unknown methane emission rate is obtained from the well-known tracer release rate and the ratio of the methane concentration to the tracer concentration detected sufficiently far downwind, as shown in Equation S4-1.

Methane emission rate = Tracer emission rate * (downwind – upwind concentration of methane) / (downwind – upwind concentration of tracer) (Eqn. S4-1)

Prior work has demonstrated that the "tracer flux ratio" quantification approach can accurately quantify the total emissions from emissions from industrial sites[13] and landfills.[14,15] Prior research has also shown the tracer flux ratio method to be useful at quantifying the sum of small emission rates stemming from a large area where individual measurements would be challenging.[16-19]   The primary assumption underlying the use of the tracer flux ratio approach is the assumption of equivalent dispersion of the tracer and methane. Therefore, in addition to reporting methane emissions estimated using this technique, a series of experiments were conducted to assess the accuracy of the equivalent dispersion assumptions.

Application of this method depends on winds that will form a well-developed plume, detectable at ground level, downwind of the site. The method also requires accessibility of downwind locations for sampling. For this work, since measurements were done using a mobile van, a downwind road network was required. Because of these constraints, downwind sampling was only performed at a subset of the sites. The exact fraction of sites that are amenable to this

type of sampling will depend on meteorological conditions, which may vary by season, as well as topographic conditions.

S4.2 Methods

S4.2.1 Mobile Laboratory

The mobile laboratory is a 25' long truck equipped with commercial and research grade instrumentation designed to operate while in motion. The majority of the instruments on board are in-situ sampling instruments used to characterize the composition of the sampled air. Other instruments include wind speed and direction, vehicle position and orientation using the global positioning system (GPS), atmospheric pressure and temperature. Examples of the application of this type of mobile laboratory to atmospheric measurements are described by Kolb et al. [20] and Herndon et al.[21,22].

Inlet description

Through an inlet system at the front of the vehicle, ~ 15 standard liters per minute (slpm) of ambient sample are continuously drawn from a common inlet to various instruments that subsample in parallel and series (depending on the needs of the specific instrument). Generally, each of the instruments has a response time of less than 1 s. The lag time from sample passage into the inlet and measurement by the instrument varied for each instrument. For compounds of interest in this work, the lag time was only ~ 1 s because of an 18 fold pressure drop induced just after the sample line had been brought into the truck. The additional measurements that were not associated with the primary or auxiliary tracer or methane had longer lag times, 5-9 seconds, but retained the rapid time response in the instrument itself. Reported time series data from the instruments with a longer lag time were consequently time shifted to a common inferred inlet time without averaging. The timescale of the plume encounters in the atmosphere were 15 seconds to a minute or longer. Since the time spent in the instrument was typically less than 1 second, the nominal timeshift to common inlet time does not introduce bias when interpreting the concomitant increases in various species.

The inlet system also had an "overblow" line attached where gas from the truck interior could be added within 6 cm of the inlet tip (or the atmosphere). The line was used to trigger periodic zero gas, used to time the respective inlet lag as well as define instrument zero. The flow rate of the zero of calibration gas was set to induce less than a 0.2% pressure change at the first instrument in the manifold and still "overflow" the inlet. Thus, the instrumentation was operated in the same mode between calibration and sample. The overblow line was also drawn with a minor flow (90 sccm) at its interior terminus in order to prevent the contents of the line from slowly diffusing or turbulently burping into the sample.

Instrument overview

The mobile lab heading and position were determined using a Hemisphere V110 GPS Compass operated at 1 Hz. Wind speed and direction were measured several ways. Fixed portable rotary vane anemometers were deployed at the release site. On the mobile lab, the apparent wind was measured using a Vaisala WM30 rotary vane anemometer and an AirMar LB150 sonic anemometer. The LB150 device also employs an internal GPS to correct for the motion of the vehicle to report a true wind. Atmospheric pressure was logged using a calibrated 1000 Torr MKS pressure transducer and temperature was measured. A Vacuubrand MD4 diaphragm pump was used to draw sample through the CO_2 Licor instrument and to other diagnostic equipment. It was also used to induce small draws on the lines used for periodic zero and calibration. A Varian Triscroll 600 was used to draw 9 slpm through the tunable infrared differential absorption spectrometers in series.

The primary analytical composition measurements in this work were performed using tunable infrared laser differential absorption spectroscopy (TILDAS). The primary external tracer in this work was nitrous oxide (N_2O) and was measured using absorption lines at 2199.737 cm^{-1}. The auxiliary external tracer in this work was acetylene (C_2H_2) and was measured using an absorption line at 1342.349 cm^{-1}. Methane (CH_4) was measured using the minor (^{13}C) and major carbon isotope absorption lines at 1294.196 and 1294.379 cm^{-1}. In the same continuous wave quantum cascade laser modulation cycle as N_2O, an absorption line of carbon monoxide (CO) was also included. A summary of the analytical instrumentation is provided in Table S4-1.

Table S4-1. Measurements deployed during the tracer release campaign

Measurement	Rate	Instrument
Methane (CH_4)	1 s	Quantum Cascade Laser Sys. (1294 cm^{-1})
Carbon Monoxide (CO)	1 s	Quantum Cascade Laser System (2230 cm^{-1})
Acetylene/Ethyne (C_2H_2)	1 s	Quantum Cascade Laser System (1342 cm^{-1})
Nitrous Oxide (N_2O)	1 s	Quantum Cascade Laser System (2200 cm^{-1})
Carbon Dioxide (CO_2)	1 s	Licor or QCL

S4.2.2 Tracer Release Stand Description

The flow of tracer gas is regulated and monitored using an MKS 247 4-Channel Controller (SN 01000079) coupled with MKS 1179 A Mass Flow Controllers (SN's 001347521, 001773651, 020003843). The MKS 1179 A Mass Flow Controller operates on the principle that by monitoring the temperature flux of a gas and using a known specific heat constant for that gas one can monitor the mass flow rate. The MKS 247 unit acts as a processor for the MFCs; its data output is logged on to the release stand computer. The mass flow controllers are calibrated and checked throughout each campaign to ensure accuracy. Once the release gas has been regulated by the mass flow controller it passes through a series of emergency shut off valves then to release.

The tracer release base unit was deployed with two Porta-Met Meteorological stations to collect information about the release environment. The Porta-Met is a relatively compact, tripod mounted unit that is able to measure wind speed, wind direction, temperature, pressure, GPS location, solar intensity, and relative humidity. Units were placed at positions appropriate to the site scale wind transport. Wireless RS 232 radios fed data to the on-site tracer release stand. After periodic archiving of the data, the tracer release stand distilled and bundled the results for a radio transmission to the Mobile Laboratory. The transmission was done using a pair of Freewave RS 232 wireless transmitters.

Two tracers were employed in this work. The primary use of the dual tracers in this work was to assess the accuracy of the fundamental assumption in the tracer flux ratio method, that the tracer compound and the species of interest for quantification (in this work, methane) undergo equivalent atmospheric dispersion prior to downwind sampling. The observed downwind concentration ratio of the two tracers can be compared to the known ratios of mass emission rates to provide a characterization of measurement uncertainty.

S4.2.3 Data analysis

Typical data generated in tracer flux measurements are shown in Figure S4-1. An aerial image of a natural gas production site is shown; winds out of the southeast are shown as blue lines on the image. The tracer release points for N_2O and acetylene are shown. The N_2O release point was located adjacent to an on-site compressor. The acetylene release point was located adjacent to a tank battery. The mobile van made measurements of methane, acetylene and N_2O along a road north of the production sites. The path of the van is shown as a series of dots, with the size and color of the dots indicating the magnitude of the methane concentration. Measured concentration distributions are shown for N_2O, acetylene and methane.
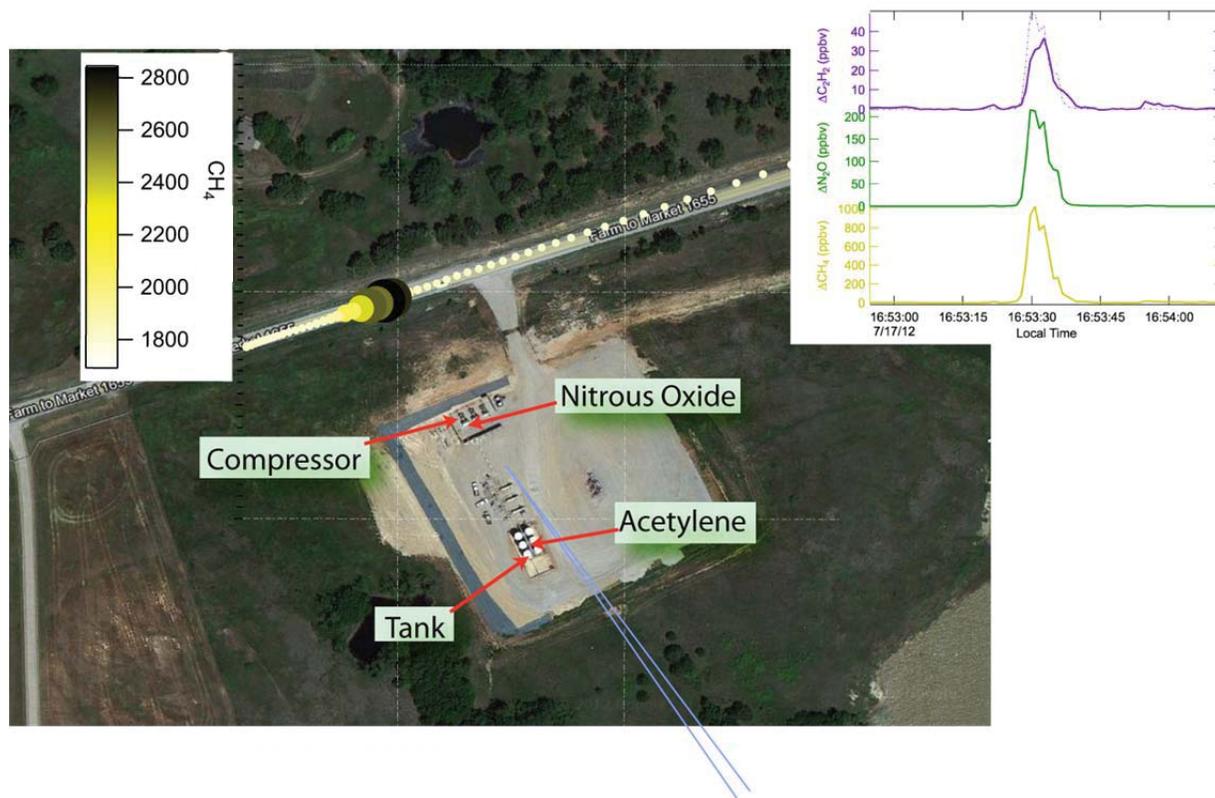
Figure S4-1. Summary of data collected during downwind sampling at a natural gas production site. Shown are a satellite image of the site, wind direction at multiple points in time (from the southeast, shown in blue as two lines, indicating variations in wind direction), downwind methane measurements along the measurement van path (shown as dots), and concentration distributions of tracers and methane along the transect path (upper right).

The analysis of the concentration distributions is summarized in Figure S4-2 and S4-3. A first observation from the concentration distributions is that the acetylene and N_2O concentration profiles are different, based on locations of the tracer release sites. Also shown in Figure S4-2 is a hypothetical distribution of acetylene that would be anticipated, if the releases were co-located. The differences in the acetylene and N_2O concentration distributions can, in principle, be used to estimate separately emissions from the compressor and tank battery. In this work, however, only total site measurements are reported and the dual tracers were used to estimate plume capture (as described below).
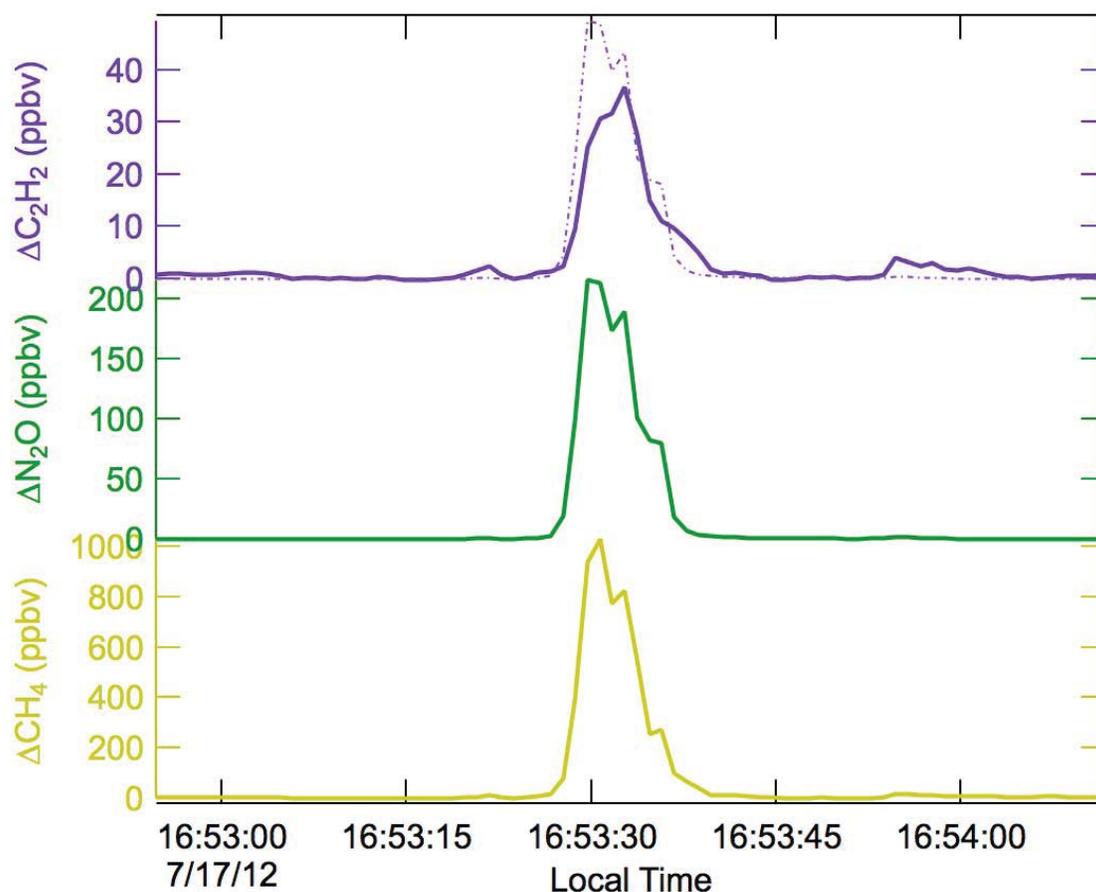
Figure S4-2. Acetylene, N$_2$O and methane concentration profiles (background corrected) along the path traversed by the measurement van (horizontal axis is local measurement time). Also shown is a hypothetical distribution of acetylene (dashed blue line) that would be anticipated, if the releases were co-located.

The total methane release rate for the site is based on the N$_2$O tracer.

Methane emission rate = N$_2$O tracer emission rate * (downwind – upwind concentration of methane) / (downwind – upwind concentration of N$_2$O tracer) (Eqn. S4-2)

Figure S4-3 shows multiple instances of the instantaneous methane concentration and the N$_2$O tracer concentration at the same time. The average ratio of the instantaneous concentrations (4.4:1, derived from the slope of the line) and the known N$_2$O release rate gives an estimated methane release rate of 1.26 g CH$_4$ per second.
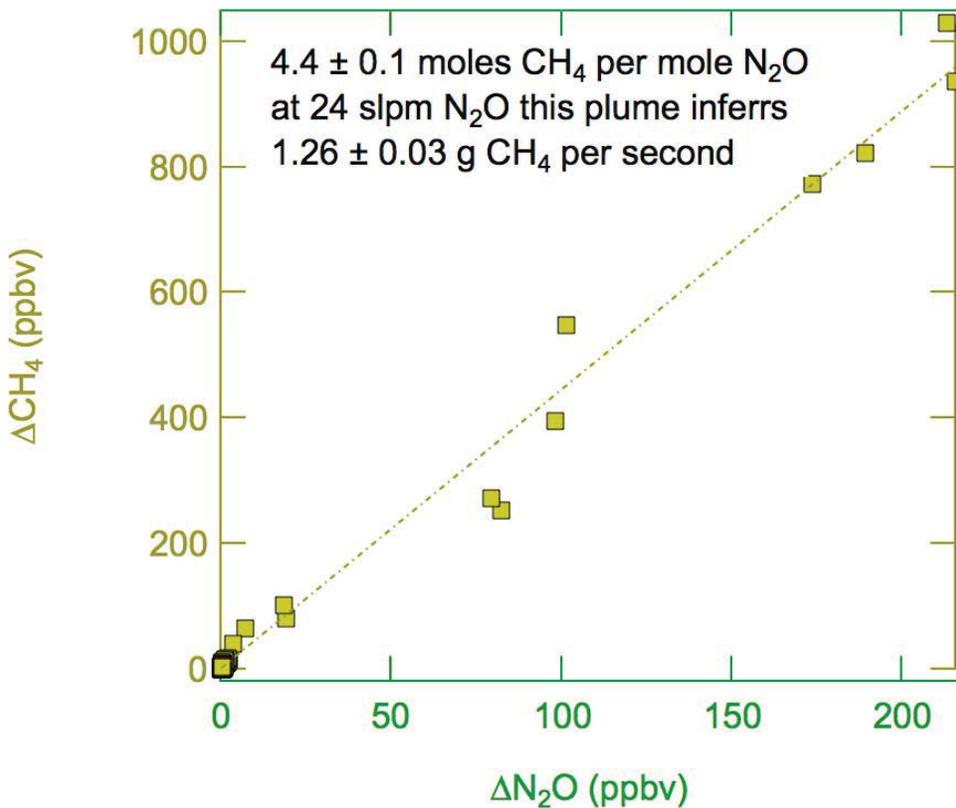
Figure S4-3. Comparison of the instantaneous methane concentration (measurement – background) associated with the N₂O tracer shown in Figure S4-1, and the N₂O tracer concentration (measurement – background) at the same time. The slope of the line shown in the Figure equals the ratio of the emission rate of methane to emission rate of tracer.

The degree of plume capture is assessed by comparing the concentration ratios of acetylene to N₂O, observed second by second, in the plume, to the known ratio of the emission rates of the two tracers. Figure S4-4 shows this comparison for a typical production site where the tracer releases were co-located. For this site, the concentration for the two tracers and methane were very highly correlated in second by second observations in the plume. The average of the methane to N₂O concentration ratios, observed second by second in the plume, indicated by the slope of the line in Figure S4-4b, was consistent throughout the plume, and was used in Equation S4-2, along with the known N₂O tracer emission rate, to calculate methane emissions. The average of the acetylene to N₂O concentration ratios, observed second by second in the plume, indicated by the slope of the line in Figure S4-4c, was compared to the known ratio of emission rates for the two tracers. This calculation tested the assumption of equivalent dispersion of the two plumes. The 0.8% error reported in Figure S4-4c indicates the difference between the ratio of the observed tracer concentrations in the plume (determined from the slope of the line shown in Figure S4-4c) and the ratios of the tracer release rates for this site, divided by the ratio of the tracer releases.

When the observed ratios of tracer concentrations in the plumes at all sites are compared to the known ratios of tracer releases at all sites, the distribution is normally distributed with a standard deviation of 15%. The ratio shows a positive bias of 4%. Overall, this leads to an empirical uncertainty estimate of 20% for the dual tracer measurements performed in this work. This uncertainty would be expected to be sensitive to the meteorological conditions, topography and downwind access, and so is likely not generalizable beyond the work reported here.
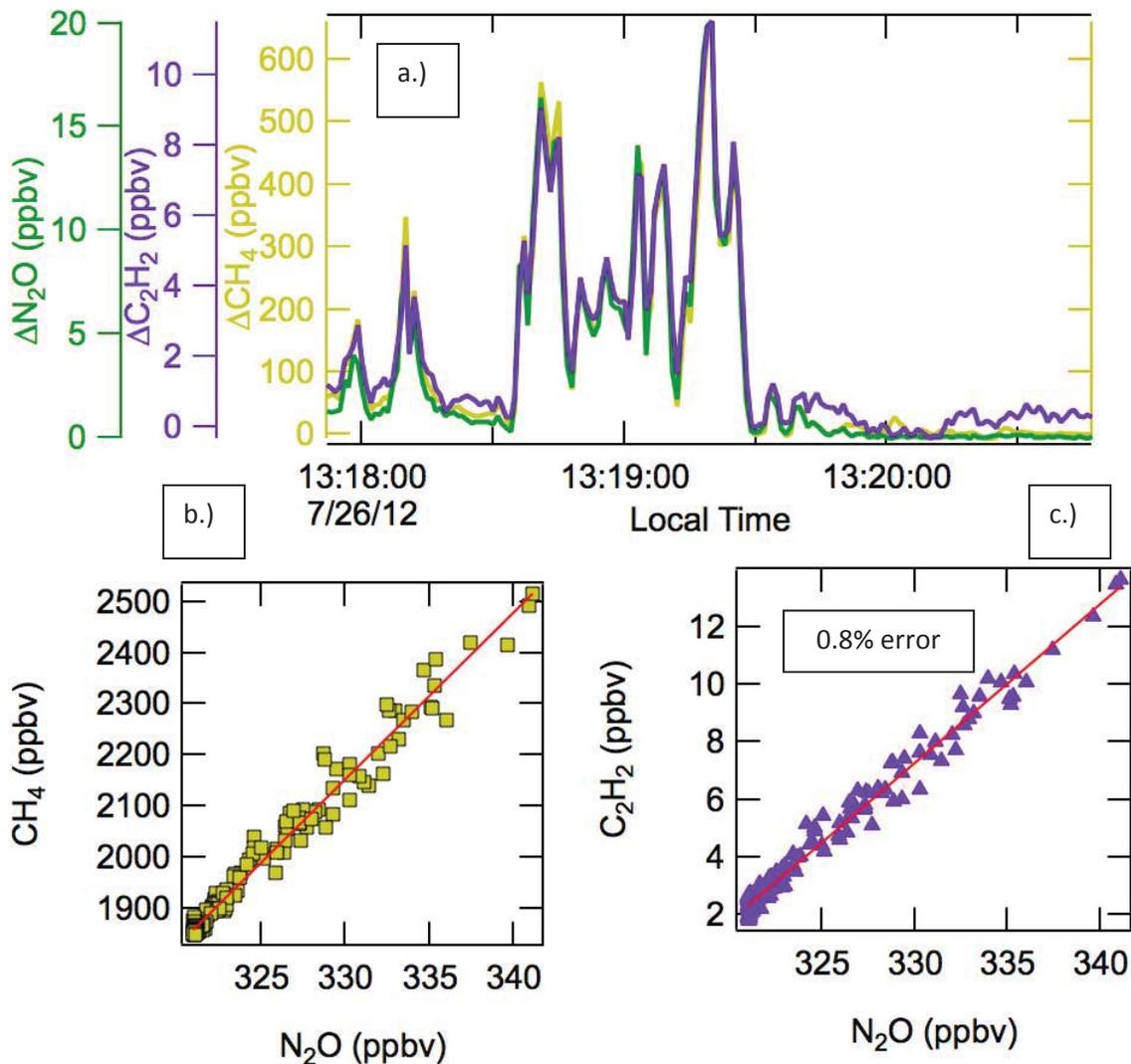
Figure S4-4 a.) Methane, acetylene and N_2O plumes observed downwind of a production site; tracers were co-located; b.) the average ratio of methane to N_2O in the plume, determined using second by second observations of methane and N_2O is indicated by the slope of the line; this ratio was used in Equation S4-2, with the known release rate for N_2O, to estimate methane emissions; c.) the average ratio of acetylene to N_2O in the plume determined using second by second observations of acetylene and N_2O in the plume is indicated by the slope of the line; the 0.8% error indicates the difference between the ratios of the observed concentrations in the plume and the ratios of the tracer release rates for this site.

S4.3 Results and Discussion

The results from the downwind sampling are presented in two sections: (i) emissions fluxes from well completion activities, and (ii) emission fluxes from natural gas production sites.

Emission fluxes from well completion flowback activities

Measurements were made downwind of 6 well completion flowbacks. Five of the six flowbacks were of the type reported as Configuration 4 in Table S1-1 – flowbacks in which the entire flow was sent to a vented tank. There are several reasons for a focus on these types of completions. First, these types of completions have the greatest uncertainty in their emissions. Other types of completions, involving separators, have well completion reports that typically include gas volumes, oil volumes and water volumes released by the separators. Reasonably accurate engineering methods can be used to estimate the emissions arising from these separator flows when they reach the tanks. In contrast, engineering estimates of emissions associated with flow directly from a well head have a high degree of uncertainty and the direct source measurements made in this work provide data that cannot be routinely estimated. An additional reason for focusing on this category of completions is that the completion involves a single emission point. This reduces the uncertainty in the emission estimates based on the tracer release method.

Comparisons of the direct source measurements and the emission estimates based on the dual tracer method, for the 5 flowbacks that utilized Configuration 4, are provided in Table S4-2 and Table S4-3. Table S4-2 reports comparisons for Rocky Mountain completions 3, 4 and 5, which occurred sequentially. Table S4-3 reports comparisons for Midcontinent completions 1 and 2, which occurred concurrently. Since the downwind van is only able to sample when meteorological conditions set up a plume measurable downwind, the results are presented in time windows when the downwind measurements were made.   In general, downwind measurements are reported for periods when the van was consistently (but not necessarily continuously) in the plume. During these periods, the ratio of methane to tracer concentration in the plume was plotted, and the average of the slope of was determined. The slope is multiplied by the known release rate for the tracer to yield an emission rate for methane. No attempt was made to estimate transport times since typically the measurement interval was long compared to the transport times.

Tables S4-2 and S4-3 compare dual tracer emission estimates to direct source measurements in two ways. In one calculation method, non-zero direct source emission estimates were calculated by summing all emissions for the measurement period, then dividing by the time period during the van measurements when emissions were non-zero. In addition, Tables S4-2 and S4-3 report the average total direct source emissions over the measurement period divided by the total measurement time. The downwind measurement is expected to be between these two values, since plume capture was not complete for the sampling period. In addition, a low bias in the downwind measurements, when compared to direct source, non-zero

emissions might be expected, since plume spreading downwind would cause the van to record shorter zero emission periods than the direct source measurements.

Table S4-2. Direct source methane emission measurements for Rocky Mountain Completions 3, 4 and 5 and methane emission estimates based on downwind measurements.

Completion	Measurement van start and stop times, expressed as time from start of completion		Tracer emission estimate (scf/m)	Direct source emission measurement (scf/m)		Direct source to trace emission estimate ratio (1.0=match)
	Start	End		Averaged over Entire period	Averaged over non-zero emissions	
RM-3	33 hr 42 min	34 hr 35 min	9.8±1.5	1.4	12.6	0.14-1.3
RM-3	34 hr 44 min	45 hr 26 min*	19.7±3	4.6	24	0.23-1.2
RM-4	27 hr 18 min	29 hr 25 min	37.1±5.6	30.4	32.5	0.82-0.87
RM-4	30 hr 37 min	33 hr 59 min	7.4±1.1	10.2	20.0	1.3-2.7
RM-5	40 hr 18 min	44 hr 44 min	4.4±0.7	19.3	42.6	4.3-9.6

*time period extends a few minutes past the end of the completion

Table S4-3. Direct source methane emission measurements for combined Midcontinent Completions 1 and 2 (emissions occurred concurrently on the same site) and methane emission estimates based on downwind measurements.

Completion	Measurement van start and stop times, expressed as time from start of completion (both started at same time)		Tracer emission estimate (scf/m)	Direct source emission measurement (scf/m)		Direct source to trace emission estimate ratio
	Start	End		Averaged over Entire period	Averaged over non-zero emissions	
MC-1 + MC-2	3 hr 58 min	7 hr 34 min	10.7±1.6	2.4	10.9	0.22-1.0
MC-1 + MC-2	90 hr 51 min	93 hr 31 min	12.1±1.8	13.6	28.8	1.1-2.3

The sixth completion flowback for which downwind measurement were made (AP-3 in Table S1-2) was selected because of the presence of a flare. Flow from the well was routed to a separator; liquids from the separator went to a vented tank, where methane would flash. This flow was measured and is reported in Table S4-4 in the column labeled vented flowback tank emissions. Gas from the separator initially went to sales, then was vented for a period reported as 75 minutes in the completion report; after the venting period ended, the gas was sent from the

separator to a flare where the gas was combusted. Flow rates to the flare were reported hourly in the completion report, so hourly emission estimates were made for the flare, based on methane flow to the flare and an assumed combustion efficiency of 98%. Each of the three flows (flowback tank vent, separator vent and flare emissions) are reported in Table S4-4. The temporal resolution of the direct source measurements is set by the separator vent and flare flow information in the completion report, which is in hourly blocks. The 75 minute vent from the separator began 30 minutes into the 9-10AM block on the day of the flowback and ended at 10:45 in the 10-11AM block. Flaring began at 10:45. Table S4-4 compares emissions estimated based on the dual tracer downwind measurement method to the sum of the direct source measurements.

Table S4-4. Direct source methane emission measurements for Appalachian Completion Flowback 3 (emissions from multiple sources) and methane emission estimates based on downwind measurements.

Time	Direct emission measurements (scf)				Dual tracer measurement (scf per min)	Direct/Dual tracer
	Vented flowback tank	Separator vent	Flare	Total Direct measurements (scf per min)		
8:30-9:00	717	0	0	24	128	0.19
9:00-9:30	2800	0	0	92	290	0.32
9:30-10:00	2800	15,600	0	612	421	1.45
10:00-10:30	4400	10,200	0	558	390	1.07
10:30-11:00	5700	7,400	150*	390	366	1.07
Total emitted				50,300 scf	47,900 scf	1.05

* Flow rates to the flare were reported hourly; emission estimates were made for the flare emissions, based on methane flow to the flare and an assumed combustion efficiency of 98%

Agreement between direct source emission measurements and emission estimates based on downwind tracer measurements is generally within a factor of 2. Because of the challenges associated with comparing intermittent direct source measurements with plume measurements taken up to a kilometer or more downwind, more precise comparisons are not justified using the methods employed in this work.   Nevertheless, the comparison of direct source measurements of completions, with an independent downwind measurement, provides strong support for the conclusion that methane emissions from completion flowbacks are roughly 97% below the most recent national estimates and that emissions from completion flowbacks without methane control or recovery equipment, observed in this work, are well below the average potential emissions in current national inventories[4]. Even if emissions based on the direct source measurements were

doubled, projected national emissions from completion flowbacks would still be 95% less than current estimates.[4]

One exception to the general agreement, within a factor of two, between the downwind measurements and the direct source measurements is Rocky Mountain Completion 5. For this completion, the downwind dual tracer measurements indicate a much lower emission rate than the direct source measurements, but this is likely due to the temporal resolution of the composition measurements in the direct source method. In this completion, during this time period, methane concentrations were dropping rapidly. The direct source emission estimate is based on gas composition measurements at the beginning and end of the sampling period and the direct source emissions are based on a linear extrapolation of the concentration. The van records high emission rates initially, then a rapid drop to lower emissions for most of the measurement period. If the direct source measurements are based on the gas composition at the end of the period, rather than a linear interpolation, the ratio of the direct source measurements to the tracer measurements would be similar to other ratios in Table S4-2.

It should also be noted that the downwind dual tracer measurements for completion flowback AP-3 provide some evidence of flare combustion efficiencies. At approximately 10:45, methane that had been vented from the separator was routed to the flare. The downwind emission estimate dropped in less than a few minutes from 325 scf/m to 9.1 scf/m. If it is assumed that the emissions from the flowback tank vent that had mixed downwind with the flare emissions were typical of the flowback tank emissions during the 30 minute period from 10:30-11:00 (5700 scf/30 minutes) the total emissions measured downwind are less than the emissions directly measured from the flowback tank vent, suggesting the flare emissions are near zero (100% combustion efficiency). Even if it is assumed that the downwind measurements are only detecting methane emissions from the flare, combustion efficiencies would be estimated as at least 97.2% ((1-(9.1/325))*100). These are preliminary observations, and given the uncertainties in the direct source and dual tracer measurements, do not support the use of a new emission factor for flares. Nevertheless, the data are supportive of the assumption of 98% combustion efficiency used throughout this work.

Emission fluxes from natural gas production sites

Measurements were made downwind of a total of 20 production sites. Results are summarized in Table S4-5. Table S4-5 lists the number of wells per site, the emissions from various pieces of equipment on each site, the total emissions from the site based on direct source measurements and emission estimation methods, and the total site emissions based on downwind measurements.

Note that emissions from the exhaust of compressors and emissions from tanks are estimated using standard emission estimation methods, rather than measurements. The emissions from compressors are small, relative to total emissions, however, the emissions from tanks are relatively large for many sites. These tank emissions are due to methane dissolved in hydrocarbon liquid and water at separator conditions, which subsequently flashes when the liquids are sent to atmospheric pressure tanks. The amount of methane dissolved in the liquids is calculated based on a solubility estimate, which depends on the composition of the gas, the API gravity of the oil, and the separator pressure. In some regions (e.g., the Rocky Mountains), gases vented from hydrocarbon liquid (condensate) tanks are not vented directly to the atmosphere, but instead are sent to a combustor. The combustor was assumed to have a 98% combustion efficiency.

Table S4-5. Comparison of direct source and downwind emission estimates for production sites.

Site (# of wells on site)	Emissions from on site measurements or estimation (scf/m)					Tracer emission estimate (scf/m)	Direct source to tracer emission estimate
	Pneumatic Controls and pumps[a]	Fugitives[a]	Compressors[b]	Tanks[b]	Total		
MC-1 (2)	1.41	0.23	2.23E-04	0.25	1.89	2.32	0.815
MC-2 (1)	0.97	0.01	5.91E-05	0.01	0.99	2.00	0.495
MC-3 (3)	1.09	0.29	5.25E-04	0.25	1.63	2.95	0.552
MC-4 (2)	0.61	0.33	7.60E-06	1.38	2.31	3.36	0.687
MC-5 (2)	1.41	0.34	2.31E-04	0.11	1.85	4.16	0.445
RM-1 (8)	0.19	0.02	0	0.01	0.22	0.584	0.368
RM-2 (8)	0.05	0.10	0	4.28	4.43	1.70	2.60
RM-3 (1)	0.05	0.09	0	0.00	0.13	0.442	0.303
RM-4 (7)	0.11	0.00	0	0.00	0.11	0.839	0.137
RM-5 (2)	0.06	0.03	0	0.01	0.09	0.240	0.392
RM-6 (6)	0.42	0.31	0	0.01	0.74	0.421	1.75
RM-7 (1)	0.19	0.07	0	0.02	0.27	0.368	0.736
RM-8 (1)	0.04	0.23	0	0.02	0.29	1.08	0.266
RM-9 (4)	0.00	0.02	0	0.36	0.38	0.864	0.436
RM-10 (6)	0.03	0.01	0	2.82	2.86	0.080	35.7
AP-1 (6)	0.13	0.01	0	0.29	0.43	*	*
AP-2 (6)	0.61	0.45	0	0.22	1.28	0.270	4.74
AP-3 (6)	0.14	2.82	0	1.80	4.75	4.12	1.15
AP-4 (5)	0.03	0.57	0	0.76	1.36	0.709	1.92
AP-5 (6)	0.02	0.27	0	0.10	0.39	0.288	1.37

[a]Based on direct source measurements or averages of direct source measurements
[b]Based on emission estimation methods
*No plume captured downwind

If site RM-10 is excluded, the average of the ratio of direct source measurements to tracer measurements is 1.1, suggesting reasonable agreement between the dual tracer emission estimates and the direct source measurements and emission estimates (for those sources not measured). It is important to note, however, that for some sites, the total on site measurements are dominated by tanks and these are estimated, rather than calculated emissions. Most of the Rocky Mountain sites had combustors on the vents to the hydrocarbon tanks, resulting in much lower tanks emissions, but the extent of agreement between on-site emission measurements and estimates and emissions estimated based on downwind measurements is still dependent on calculations rather than measurements, in this case the control efficiency assumed for the combustor. For example, for RM-10, the site owner reports that tank vents are not sent to a combustor. If a combustor were in place, or if the separator did not send liquid to the tank while

the measurements were being made, the ratio of on-site to tracer based measurements would be similar to the other Rocky Mountain sites.

Another factor that is likely small, but difficult to account for, is that the on-site direct source measurements altered the methane emitted from the sites, since the HiFlow analyzer combusted methane as it was making measurements. This bias is believed to be small because the Hi-Flow instrument only analyzed one source at a time and because the instrument was not operated continuously for the entire downwind sampling period.

Overall, the downwind measurements do not suggest any reason to expect that there are large systemic biases or errors in the direct source measurements.

## S5	Nationally Scaled Emissions Estimates

### S5.1	Methods

The method used to scale up an emission measurement from a limited set of samples to a larger regional or national total is to multiply the average result of the emission measurement times the number of times that emission occurs in the larger scale. Often the emission measurement is thought of as an "emission factor" or EF, and is applied to a particular source in some discreet increment (such as an emission per event, an emission per device, and emission per component, or emission per location). The term used to scale up the emissions is called the activity factor (AF), and is the count or population of the source or event at the scale of interest, such as regionally or nationally. This can be shown as:

$$EF_i * AF_i = ER_i \hspace{4cm} \text{(Equation S5-1)}$$

Where:

EF_i = Emission Factor for region i

AF_i = Activity Factor for region i

ER_I = resulting Emission Rate total for region i

This can be done at any scale. It may be done regionally, if that is the primary differentiator, then the regions can be summed to produce a national total. Alternately, it may be done nationally in a single calculation.

S5.2   Activity data

Activity factors used in this work are drawn from EPA's 2011 national emission inventory[4] and are shown in Table S5-1.

Table S5-1. National Activity Factors from EPA Inventory

Year 2011 AF's	National Count for 2011
Count of new gas well completion events	8077 with hydraulic fracturing
Count of gas well unloading events performed	Not reported as events, EPA does report 35,828 non-plunger wells that unload
Count of applicable gas well workovers	11,663 workovers without hydraulic fracturing 13,445 total workovers
Count of gas powered pneumatic controllers	447,379
Count of injection pumps	35,013
Count of gas wells (not including associated gas wells)	446,745[a]

[a]Estimate from U.S. Department of Energy, Energy Information Administration is 513,000 on-shore gas wells[23]

No uncertainty bounds are reported for activity factors in the EPA national inventory, and therefore no uncertainty bounds are reported in Table S5-1. It is beyond the scope of this work to perform a detailed analysis of the uncertainty in the activity factor counts reported in Table S5-1, however, a variety of calculation scenarios can provide a rough characterization of the uncertainties associated with applying these counts to estimating national emissions. Specifically, for well completions and equipment leaks, regional activity counts can be multiplied by regionally averaged emission factors or national activity counts can be multiplied by nationally averaged emission factors. For pneumatic devices, emissions averaged by device type (e.g., low bleed and intermittent pneumatic controllers) can be multiplied by activity counts by device type, or total equipment counts can be multiplied emission factors averaged over all device types. Regional or national averaging can also be performed. These calculation scenarios are reported in Section S5.3.

S5.3 Emission Factors and National Emission Estimates

Table S5-2 provides central estimates of national emission rates, by source category, based on the activity factors reported in Table S5-1.

Table S5-2. Central estimates of National Emission Rates by Source Categories

Source Category	Emission Factor (based on measure-ments in this work)	EF Units	Activity Factor	AF Units	AF Source	National Emissions Billion scf/yr CH$_4$ (Gg)
New Gas Well Completion Flowbacks	115,000[a]	scf CH$_4$/ event	8077	events/yr	EPA 2011 Inventory	0.93 bcf/yr (18 Gg)
Gas Well Liquids Unloadings (non-plunger)	48,000-300,000[b]	scf CH$_4$/ well/yr	35,828	Wells that unload	EPA 2011 Inventory	1.3-10.7 bcf/yr (25- 205 Gg)
Pneumatic Devices	67,400[c]	scfy CH$_4$ /device	447,379	devices	EPA 2011 Inventory	30.2 bcf/yr[d] (580 Gg)
Chemical Injection Pumps	101,000	scfy CH$_4$ /device	35,013	devices	EPA 2011 Inventory	3.5 bcf/yr (68 Gg)
Equipment Leaks	33,900	scfy CH$_4$ /well	446,745	gas wells	EPA 2011 Inventory	15.1[e] bcf/yr (291 Gg)

[a]Regional averages for the completion flowbacks were calculated (252,000 Appalachia; 130,000 Gulf Coast; 47,000 Midcontinent, 24,000 Rocky Mountain) and multiplied by the fraction of completion events per region (U.S. EPA[4], 0.19 Appalachia; 0.36 Gulf Coast; 0.20 Midcontinent, 0.17 Rocky Mountain; 0.08 West Coast and Southwest) to arrive at a regionally weighted emission; for West Coast and Southwest regions, the national average emission factor (90,000) was used.

[b]An emission factor based on the limited measurements reported in this work is 300,000 scf per well per year; an emission factor based on the emissions reported by API/ANGA[12] and multiplied by a ratio of measured to estimated emissions found in this work is 48,000 scf per well per year

[c]U.S. EPA[4] reports 447,379 pneumatic devices but does not report an activity factor by device type; the emission factor used here is based on an average value of the samples taken in this work.

[d]Using regional emission factors of 0.126 (Appalachia), 0.268 (Gulf Coast), 0.157 (Midcontinent) and 0.015 (Rocky Mountain) scf methane/min/device, multiplied by the number of devices per region (74,136 Appalachia; 53,049 Gulf Coast; 140,041 Midcontinent; 122,878 Rocky Mountain; 57,275 West Coast and Southwest) results in a regionally weighted emission factor of 67,400 scf methane/device/yr and a national estimate of 30.2 billion scf/yr; for West Coast and Southwest regions, the national average emission factor (92,000) was used.

[e]Regional averages for the equipment leaks per well were calculated (0.098 Appalachia; 0.058 Gulf Coast; 0.046 Midcontinent, 0.035 Rocky Mountain) and multiplied by the fraction of wells per region (U.S. EPA[4], 0.34 Appalachia; 0.17 Gulf Coast; 0.20 Midcontinent, 0.19 Rocky Mountain; 0.10 West Coast and Southwest) to arrive at a regionally weighted emission; for West Coast and Southwest regions, the national average emission factor was used.

Alternative scenarios for estimating national emissions were considered for each source category.

Completion flowbacks Estimates of national emissions for completion flowbacks can be based on either regional average activity counts, as reported in Table S5-2, or on national activity counts and a national average emission factor. The national averaging leads to an emission estimate that is 23% lower than the regional estimates (0.73 bcf; 90,000 scf/event, 8077 events). Regionally averaged emission estimates were used as the central estimate in this work because there were differences in the emission factors observed by region and because the frequency of U.S. well completion activity (highest number of completions in the Gulf Coast) did not match the distribution well completions sampled in establishing emission factors (highest number of events sampled in the Rocky Mountains).

Pneumatic devices Estimates of national emissions for pneumatic controllers can be based on either regional average activity counts, as reported in Table S5-2, or on national activity counts and a national average emission factor. The national averaging leads to an emission estimate that is 36% higher than the regional estimates (41.1 bcf; 92,000 scf/device, 447,379 devices). Regionally averaged emission estimates were used as the central estimate in this work because there were differences in the emission factors observed by region. Pneumatic controllers could also be aggregated by device type, with national emissions estimated by multiplying the numbers of devices by the emission factor per device. This approach was not used in this work because the EPA national inventory[4] does not report device counts by type, and because when device counts by type are reported, some high bleed devices are typically part of the count; no high bleed devices were reported by the companies that provided controller type information in this work.

Pneumatic pumps   Estimates of national emissions for pneumatic pumps can be based on either regional average activity counts, or on national activity counts and a national average emission factor, as reported in Table S5-2. Nationally averaged emission estimated were used as the central estimate in this work, even though there were differences in the emission factors observed by region, because of the limited number of regions in which pneumatic pumps were sampled in this work. An estimate of the uncertainty introduced by using a national averaging will be assumed to be comparable to the difference between regional and national averaging for pneumatic controllers (36%).

Equipment leaks Estimates of national emissions for equipment leaks can be based on either regional average activity counts, as reported in Table S5-2, or on national activity counts and a national average emission factor. The national averaging leads to an emission estimate that

is identical (to two significant figures) to the regional estimates (15.1 bcf; 33,900 scf/well, 446,745 wells).

Measurements made on liquids unloading and workovers, collected as part of this work, were not used to develop national emission estimates.

Workovers For workovers, four events, all done without hydraulic fracturing, were sampled. Since the data set was very small and since workovers without hydraulic fracturing represent less than 0.1% of total emissions in the EPA national inventory, these data were not used to estimate emissions for workovers (without hydraulic fracturing) at a national scale. Workovers with hydraulic fracturing are a more significant source of estimated emissions, accounting for approximately 10% of emissions in the EPA national inventory.[4] The emissions associated with flowback from workovers with hydraulic fracturing may be expected to have emissions comparable to completion flowbacks, and in the EPA national inventory, completion flowbacks and workovers with hydraulic fracturing are sometimes grouped together.[4] Therefore, an emission factor for workovers with hydraulic fracturing comparable to the emission factor for completion flowbacks may be appropriate. If the emission factor for workovers with hydraulic fracturing were assumed to be the same as the emissions factor for completion flowbacks that is based on the measurements presented in this work, total workover emissions would be reduced by 97%, compared to the current estimate in the national emission inventory. This assumption is not made in this work since no direct measurements of workovers with hydraulic fracturing were made.

Liquid unloadings Only well unloadings without plunger lifts were measured in this work. If the per well annual emissions from unloadings without plunger lifts, determined in this work (300,000 scf/well), are multiplied by the national counts of wells with unloadings without plunger lift, the national emission estimate is in reasonable agreement with the EPA inventory[4] and the API/ANGA estimate.[12] In contrast, another estimate of unloading emissions, based on the per event emissions observed in this work and a count of unloading events from the API/ANGA survey,[12] would lead to a national estimate 5 times the estimate based on unloading emissions per well. A lower estimate of unloading emissions could be suggested based on national event counts, emission estimates, and the finding that emission estimation methods over-estimate observations made in this work by a factor of 5. All of these methods, however, assume a single scalar value represents a wide range of unloadings; the data presented in this work and in the API/ANGA survey suggest that refined emission estimation methods, taking into account well and unloading characteristics, will be required. Additional measurements of unloading emissions are needed, both to resolve the differences

between estimates and measurements, and to better characterize the population of wells with unloading emissions.

S5.4 Central estimates and overall assessment of uncertainties in national emission estimates

Uncertainties in national emission estimates calculated in this section are a combination of the uncertainties associated with emission factors, activity factors and the methods used to combine activity and emission factors. For each source category for which national emissions were estimated, combined uncertainty ranges were estimated.

Well completions: The 95% confidence bound on national average emission factor measurement was 35,000-173,000 scf/event (±66% around the midpoint). Assuming that the uncertainty due to activity data can be roughly characterized as the range of values derived from national and regional estimates (±12% around the midpoint) and that the emission factor measurements are independent of the activity data, leads to a net uncertainty of ±67% around the midpoint. The overall range for the estimate is 0.28-1.4 bcf with a regionally weighted central estimate of 0.93 bcf.

Pneumatic controllers The 95% confidence bound on national average emission factor measurement was ±19% around the midpoint. Assuming that the uncertainty due to activity data can be roughly characterized as the range of values derived from national and regional estimates (±14% around the midpoint) and that the emission factor measurements are independent of the activity data, leads to a net uncertainty of ±23% around the midpoint. The overall range for the estimate is 27-43 bcf with a regionally weighted central estimate of 30.2 bcf.

Pneumatic pumps The 95% confidence bound on national average emission factor measurement was ±44% around the midpoint. Assuming that the uncertainty due to activity data can be roughly characterized as the same as for pneumatic controllers (±14% around the midpoint) and that the emission factor measurements are independent of the activity data, leads to a net uncertainty of ±46% around the midpoint. The overall range for the estimate is 1.8-5.2 bcf with a nationally weighted central estimate of 3.5 bcf.

Equipment leaks The 95% confidence bound on national average emission factor measurement was ±36% around the midpoint. Assuming that the uncertainty due to activity data can be roughly characterized as the range of values derived from national and regional estimates (±1% around the midpoint) and that the emission factor measurements are independent of the activity

data, leads to a net uncertainty of ±36% around the midpoint. The overall range for the estimate is 9.7-20.7 bcf with a regionally weighted central estimate of 15.1bcf.

S5.5 Comparisons with National Emission Inventory

The national emission estimates, based on the measurements made in this work, can be compared to emissions reported in the EPA's national emission inventory.[4] In assembling the national emission inventory, EPA first estimates potential emissions for source categories, then reduces the potential emissions by estimated voluntary reductions and reductions required by regulations. All regulatory reductions and some voluntary reductions are assigned to specific sources, however, some voluntary reductions are aggregated by source category. For example, the EPA estimates a total of 36 billion scf (691 Gg) of emission reductions for well completion flowbacks and workovers with hydraulic fracturing, combined. In cases such as this, allocation of combined reductions was assumed to be proportional to the potential emissions. Specifically, since the potential emissions for completion flowbacks and workovers with hydraulic fracturing were 63.6 and 13.8 billion scf, respectively (1221 and 266 Gg), the reductions were 46% of the combined potential emissions (77.4 billion scf). This percentage reduction was applied uniformly for all of the aggregated source categories. In situations where a source had both regulatory and voluntary reductions (e.g., dehydrator vents and condensate tanks), potential emissions were first reduced by reductions required by regulations, then the voluntary reductions were apportioned based on the remaining emissions. Table S5-3 reports potential emissions and the reductions applied for each source category. This Table uses units of Gg methane, the units in which the national inventory is reported, to facilitate comparison with the national inventory.

Table S5-3 National emission inventory emission estimates by source category (potential emissions, reductions and net emissions in Gg methane per year); source categories labeled in gray had aggregated voluntary emission reductions (see text)

EPA GHG Inventory Activity	Potential Emissions (Gg)[a]	Emission Reductions (Gg)[a]	Net Emissions (Gg)
Completions with Hydraulic Fracturing	1221[b]	567[b]	654
Refractures (Workovers with hydraulic fracturing)	266[b]	124[b]	143
Pneumatic Device Vents	1,134	779	355
Chemical Injection Pumps	64	30[c]	34
Equipment leaks: Gas Wells without HF	24	11[c]	
Equipment leaks: Gas Wells with HF	28	13[c]	
Equipment leaks: Separators	107	50[c]	172[d]
Equipment leaks: Meters/Piping	102	48[c]	
Equipment leaks: Heaters	33	15[c]	
Equipment leaks: Dehydrators	31	15[c]	
Workovers without HF	0.6	0.3[c]	0.3
Liquids Unloading (without plunger lifts)	149	0	149
Liquids Unloading (with plunger lifts)	108	0	108
Kimray Pumps	365	180	
Condensate Tanks without Controls	261	167[c]	
Condensate Tanks with Controls	52	0	
Gas Engines	276	49	
Dehydrators Vents	114	73[c]	
Small Reciprocating Compressors	68		
Large Reciprocating Compressors	15	35	
Large Reciprocating Stations	1		
Pipeline Leaks	170	80[c]	
Completions without Hydraulic Fracturing	0	0	
Well Drilling	0.8	0.4[c]	930[e]
Vessel Blowdowns	0.7	0.3[c]	
Pipeline Blowdowns	3	1[c]	
Compressor Blowdowns	3	1[c]	
Compressor Starts	6	3[c]	
Pressure Relief Valves	0.7	0.3[c]	
Mishaps	2	1[c]	
Emissions from Coalbed Methane and Offshore Production			
Powder River Coal Bed Methane Produced Water	46	21[c]	
Black Warrior Coal Bed Methane Produced Water	13	6[c]	
Offshore Platforms	266	125[c]	
Deepwater Platforms	23	11[c]	
TOTAL	**4,949**	**2,405**	**2,545**

[a]Potential emissions data are from U.S. GHG Inventory 13 Annex 3, Table A-134. Emission reductions data are from Tables A-132 and A-133.[4]

[b]EPA reports aggregated emission reductions for completions and workovers with hydraulic fracturing. This work divides these emission reductions between completions and workovers proportional to their potential emissions.

[c]EPA reports 551 Gg of aggregated emission reductions for the 23 activities shaded in the table. This work divides these emission reductions among the activities proportional to their potential emissions.

[d]Table 2 in the main text reports 172-211 Gg; the 211 Gg includes an additional 39 Gg due to small compressors (68 Gg of potential emissions minus 29 Gg of reductions assigned to small compressors out of 35 Gg of reductions assigned to all compressors

[e]Table 2 in the main text reports 891-930 Gg; the 891 Gg does not include 39 Gg due to small compressors (68 Gg of potential emissions minus 29 Gg of reductions assigned to small compressors out of 35 Gg of reductions assigned to all compressors

S6   Site Selection and Representativeness

Methane emissions were measured directly at 190 natural gas production sites in the Gulf Coast, Midcontinent, Rocky Mountain and Appalachian production regions of the United States. The sites included 150 production sites associated with 489 wells (146 sites had wells directly on the site that were examined in this work; 4 sites were fed by 11 off-site wells, the equipment on these 4 sites was examined but not the off-site wells; see p. S-32). In addition to the 150 production sites, 27 well completion flowbacks, 9 well unloadings, and 4 well workovers were sampled; the sites were operated by 9 different companies. The types of sources that were targeted for measurement account for approximately two-thirds of methane emissions from all onshore and offshore natural gas production, as estimated in the 2011 national greenhouse gas emission inventory.[4] A summary of the scope of the study, along with a rationale for the inclusion or exclusion of sources for direct measurement efforts, is provided in Table S6-1. Sampling was conducted at sites throughout the United States (see Figure S6-1). Table S6-2 lists the number of sampling sites in each region. Of the nine companies that provided sites for sampling, at least three companies provided sites in each of the regions.

While the data presented in this report represents one of the most extensive datasets available on methane emissions from current natural gas production activities, the sites sampled still represent a small fraction of the total number of sites nationwide.   Representative sampling was believed to be achieved by:

- Selecting a large number of participant companies
- Selecting a range of geographic areas to sample
- Setting minimum number of sampling targets in each area

Table S6-1. Sources of CH$_4$ Emissions from Natural Gas Well Sites[*, †, §]
(measurements were made for sources shaded light gray; dark gray shading indicates limited measurements)

	Activity or Source	Included in Current Work?	Rationale for Inclusion/Exclusion
Pre-production	Construction of Well site	N	No emissions from the natural gas formation occur during this process. Surface construction equipment combustion emissions were not part of the measurement target.
	Well Drilling	N	While emissions from the gas formation are possible during drilling fluid circulation, they are considered to be small and not included (except for combustion) by the EPA in the GHG Reporting Program
	Hydraulic Fracturing	N	Liquids are going into the formation during this process, and at extremely high pressures. No gas releases are expected. Surface trucking and pumping equipment combustion emissions were not part of the measurement target.
	Well Completion (Flowback)	Y	34 bcf in the 2011 EPA national emission inventory of methane emissions from natural gas production
	Well Testing	N	Considered to be a minor source, and rarely involving venting.
Routine Production	Pneumatic Devices	Y	18.5 bcf in the 2011 EPA national emission inventory of methane emissions from natural gas production
	Pneumatic Pumps	Y	1.8 bcf in the 2011 EPA national emission inventory of methane emissions from natural gas production
	Condensate Tanks	Partial	Emissions from these tanks, while a potentially large source, are considered well defined and known, with working models and equations of state. Therefore these were not a primary study target, but were measured in some opportunistic cases.
	Produced Water Tanks	Partial	
	Dehydrator Vents	N	These complex sources were considered relatively well defined and modeled.
	Surface Equipment (Leaks)	Y	~ 10 bcf in the 2011 EPA national emission inventory of methane emissions from for equipment sampled in this work
	Surface Equipment (Leaks from Compressors)	Partial	The scope of the study excluded central facilities (mimicking the GHG Reporting Program), compressor facilities were not targeted. Some on well sites were measured.
	Amine Units (Gas sweetening)	N	These complex sources were considered a small national contributor and also well defined and modeled.
Maintenance	Well liquids unloading	Y	13.4 bcf in the 2011 EPA national emission inventory of methane emissions from natural gas production
	Well workovers	Y	7.5 bcf in the 2011 EPA national emission inventory of methane emissions from natural gas production
	Blowdowns	N	This intermittent source of equipment depressuring for maintenance is manually calculated and not a measurable event. It was not a target of this study.

[*] Study scope is limited to CH$_4$ emissions from well sites and any off-site equipment up to the point of comingling of multiple well streams (in which case the centralized separation facility is included). Emissions from gas processing plants, transmission pipelines, gas storage, and local distribution systems are beyond the scope of the study.

[†] Other sources excluded from direct measurements include all combustion sources and other miscellaneous sources: flaring, compressor engine exhaust, drill rig engines, trucks, well abandonment, upsets and others.
Upwind/Downwind measurements inform an assessment of total emissions from excluded sources.

[§] Covered sources account for 85 bcf of 133 bcf CH$_4$ (65%) from the natural gas production stage (onshore and offshore).[4]

Figure S6-1. Methane emissions were measured at well-sites in the Appalachian, Midcontinent, Gulf Coast, and Rocky Mountains Regions. Regions shaded in blue indicate American Association of Petroleum Geologists (AAPG)[24] basins where sampling was done.
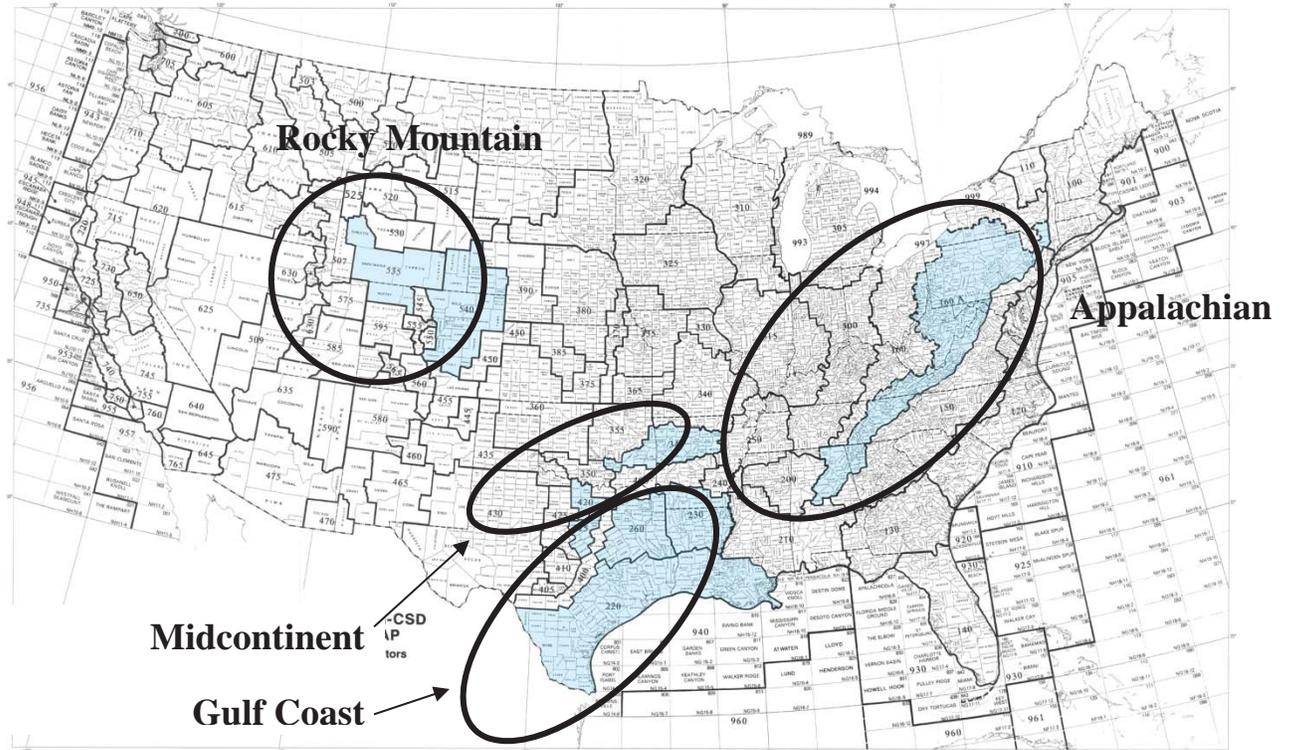
Table S6-2. Distribution of sampling locations, by region

Region	Production Sites	Unloadings	Workovers	Completion Flowbacks
Gulf Coast	58 (157 wells)	5	0	7
Midcontinent	26 (85 wells)	0	1	5
Rocky Mountain	19 (79 wells)	3	3	10
Appalachian	47 (168 wells)	1	0	5
Total	150 (489 wells)	9	4	27

The nine companies that participated in this study included mid-size and large companies. While there are thousands of oil and gas companies in the U.S., and small companies were not part of the participants, the participants do represent a sizable sample of overall U.S. production and well count, as shown in Tables S6-3 and S6-4. Participants account for almost 12% of all U.S. gas wells, account for 16% of gross gas production, and almost half of the new well completions. Representativeness cannot be completely assured, however, since companies volunteered, and were not randomly selected.

Table S6-3. Participant Portion of National Gas Wells

Region (from Fig 2-3)	Participant Company Total Gas Wells, 2011	DOE EIA Gas Wells, Onshore, L48, 2011	Percentage of Gas Wells operated by our participants
Appalachian	8,739	202,788	4.30%
Midcontinent	18,117	125,295	14.5%
Gulf Coast	9,941	81,247	12.2%
Rocky Mountain	23,805	103,643	23.0%
Total	60,602	513,000	11.8%

Table S6-4. Participant Portion of National Gross Gas Production

Region (from Fig 2-3)	Participant Co. Total Gas Production (million scf), 2011	DOE EIA Gas Production, Gross (million scf) 2011*	Percentage of Production operated by our participants
Appalachian	282,798	2,357,792	12.0%
Midcontinent	893,010	7,125,555	12.5%
Gulf Coast	1,271,450	7,498,590	17.0%
Rocky Mountain	1,379,408	6,262,666	22.0%
Total	3,826,667	23,264,162 (15,577,188) (gas well prod + shale gas prod)	16.4%

*Excludes U.S. offshore and Alaska

Regional sampling was done to account for differences in practices that may occur due to field or state differences, or practices that may vary even within a company across the U.S. The Study Team sampled in four geographical regions (Appalachian, Gulf Coast, Midcontinent and Rocky Mountain), and with multiple companies in each region. All nine of the sponsoring companies provided sampling sites during the Study. The details of the distribution of samples in the data set, for production sites, by company, region and equipment type is provided in Table S6-5.

Table S6-5 – Distribution of samples in study dataset: Counts, per company, of Chemical Injection Pumps, Pneumatic Devices (Controllers), fugitive leaks, number of geographically distinct sites, and number of wells, and production sites and wells. Each of the 9 companies providing sites and is assigned a randomly chosen letter identifier. Data are reported for the aggregated study data set and by region.

	Host Company									
Number count	D	F	H	L	N	Q	R	S	W	total
Chemical Injection Pump	0	1	0	1	1	0	17	39	3	62
Pneumatic Devices	12	33	32	15	33	32	60	26	62	305
Other Fugitives	3	27	16	32	9	45	88	19	39	278
Number of distinct sites	6	11	10	14	16	13	45	12	23	150
Number of wells	10	27	33	88	54	49	106	49	73	489

	Appalachian Region Host Company									
Number count	D	F	H	L	N	Q	R	S	W	total
Chemical Injection Pump										
Pneumatic Devices	12		32		28	32			29	133
Other Fugitives	3		16		4	45			32	100
Number of distinct sites	6		10		10	13			8	47
Number of wells	10		33		36	49			40	168

	Rocky Mountain Host Company									
Number count	D	F	H	L	N	Q	R	S	W	total
Chemical Injection Pump										
Pneumatic Devices				4			11			15
Other Fugitives				10			49			59
Number of distinct sites				4			15			19
Number of wells				25			54			79

Number count		Gulf Coast Host Company								
	D	F	H	L	N	Q	R	S	W	total
Chemical Injection Pump				1			17		3	21
Pneumatic Devices		13		11			49		33	106
Other Fugitives		1		22			39		7	69
Number of distinct sites		3		10			30		15	58
Number of wells		9		63			52		33	157

Number count		Midcontinent Host Company								
	D	F	H	L	N	Q	R	S	W	total
Chemical Injection Pump		1			1			39		41
Pneumatic Devices		20			5			26		51
Other Fugitives		26			5			19		50
Number of distinct sites		8			6			12		26
Number of wells		18			18			49		85

For completions, seven of the nine companies provided sampling sites. For the remaining two companies, attempts were made to schedule completion sampling, however, it was not possible to identify completions that the study team could sample, due to scheduling of the completions and the study team's other sampling commitments. In each of the Appalachian, Gulf Coast and Rocky Mountain regions, three different companies provided completion events. These companies are identified as AP-A to AP-C, GC-A to GC-C and RM-A to RM-C in this report. In the Midcontinent region, two different companies provided completion events. These companies are identified as MC-A and MC-B in this report. Note that company A in one region is not necessarily the same as company A in another region.

For liquid unloadings, 5 events were sampled in the Gulf Coast, 3 in the Rocky Mountains and 1 in the Appalachian region. These events were provided by 4 different companies.

The selection of specific sites was randomized to the extent possible. For completions, the study team provided time windows when the measurement team would be available in certain regions and host companies identified completions that would begin as soon as possible after the study team arrived. In most cases this scheduling completely determined which sites would be sampled. To illustrate this, consider that the total number of well completions, nationwide in 2011, for all the participating companies combined, averaged roughly 10 per day. That meant that in any given production region, on any particular day, just one or two new completions, for all of the companies combined, was likely to be starting.

The time commitment associated with sampling completions was extensive. Completions lasted up to two weeks; sampling equipment set up and tear down by the study team required a day

before and a day after the completion. Unloading, workover and production site sampling was much shorter in duration, typically a few hours to a half day. Consequently, sites selected for unloading, workover and production site sampling were selected based on proximity to completion sampling. Typically, a list of candidate sites was provided by the host company. If the list was too long to be entirely sampled in the allotted time, the study team selected sites based on an ability to sample as many sites as possible in the time available.

One exception to this pattern was for Gulf Coast sites, where the study team, based in Austin, Texas, could make day trips to production sites. For these sites, the study team randomly selected from hundreds of potential sites provided by host companies. A second exception was for unloadings. These events were difficult to schedule since they were often done, by site operators, immediately as needed. This often did not allow the study team to travel to the site and set up equipment prior to the unloading occurring. Therefore, special efforts were made to identify and sample unloadings that could be scheduled. Unloadings numbered 1a-1c and 2a-2c were selected in this manner.

The downwind sampling described in S4 was performed only at a subset of sites. This subset was selected based on the expectation of stable, moderate winds at the sites and the availability of a dense road network that would allow the van access to downwind sampling points. Based on these criteria, the downwind sampling team was tasked to mirror the sampling efforts of the direct source measurement team to the extent possible given road networks downwind of sampling sites and the prevailing meteorology on the day sampling was done. Sampling was done in the Appalachian, Midcontinent and the Rocky Mountain regions.

A final potential bias is changes to the U.S. gas production system during the measurement effort. The system of wells and oil and gas operations continued to grow during the test period, but the technology was considered fairly constant during the test period. The study participants were aware that the EPA's new Volatile Organic Compound (VOC) controls, in the forms of new regulations for National Emission Standards for Hazardous Air Pollutants (NESHAP) and New Source Performance Standards (NSPS Part OOOO) that went into effect in 2012 and would potentially affect new sources in 2012. The regulations require controls on certain VOC sources that also result in control of methane. Since most of these controls will affect tank vents and dehydrators, which were not sampled by this study, the effect was considered minimal. The regulations do require minimization of high-bleed rate pneumatics on new facilities, and this may have had an effect on the study pneumatic measurements.

References

(1) Bird, R.B.; Stewart, W.E.; Lightfoot, E.N.; "Transport Phenomena", Wiley, New York, 1960 pg 175

(2) U.S. Environmental Protection Agency (EPA) AP-42 Emission Factors, Section 13.5, Industrial Flares, Dated 9/91, available at http://www.epa.gov/ttnchie1/ap42/ch13/final/c13s05.pdf , accessed February, 2013.

(3) Code of Federal Regulations (CFR), Title 40 Section 98.233, Calculating GHG Emissions, July 1, 2012.

(4) U.S. Environmental Protection Agency (EPA). *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990-2011.* EPA 430-R-13-001, April 2013. http://www.epa.gov/climatechange/ghgemissions/usinventoryreport.html and (accessed April, 2013)

(5) Harrison, M.R.; Shires, T.M.; Wessels, J.K.; Cowgill, R. M; Methane Emissions from the Natural Gas Industry, Volumes 1 – 15, Final Report, GRI-94/0257 and EPA-600/R-96-080, Gas Research Institute and US Environmental Protection Agency, June 1996.

(6) Efron B.; Tibshirani, R. An Introduction to the Bootstrap, Chapman & Hall, CRC Press, (1993).

(7) Eastern Research Group; Sage Environmental, City of Fort Worth Natural Gas Air Quality Study, Final Report, July 13, 2011, available at http://fortworthtexas.gov/uploadedFiles/Gas_Wells/AirQualityStudy_final.pdf , accessed January 2013.

(8) FLIR, Forward Looking Infrared Cameras. http://www.flir.com/thermography/americas/us/content/?id=18358 (accessed January, 2013).

(9) Bacharach, HiFlow Sampler, http://www.bacharach-inc.com/hi-flow-sampler.htm (accessed January, 2013)

(10) Harrison, M.R.; Shires, T.M.; Wessels, J.K.; Cowgill, R.M.; Methane Emissions from the Natural Gas Industry, EPA/600/SR-96/080 June 1997.

(11) Kirchgessner, D.A.; Lott, R.A. Cowgill, R.M.; Harrison, M.R.; Shires, T.M. Estimate of Methane Emissions from the U.S. Natural Gas Industry, Chemosphere, 35, 1365-1390, 1997.

(12) American Petroleum Institute and America's Natural Gas Alliance (API/ANGA) Characterizing Pivotal Sources of Methane Emissions from Natural Gas Production,: Summary and Analyses of API and ANGA Survey Responses, Final Report, updated September, 2012, available at: http://www.api.org/news-and-media/news/newsitems/2012/oct-

2012/~/media/Files/News/2012/12-October/API-ANGA-Survey-Report.pdf (accessed, February 2013)

(13) Lamb, B.; Shorter, J.H.; McManus, J.B.; Kolb, C.E.; Mosher, B.W.; Harriss, R.C.; Allwine, E.J.; Blaha, D.; Siverson, R.; Howard, T.; Lott, R.A.; Siverson, R.; Westberg, H.; Zimmerman, P.; Development of Atmospheric Tracer Methods to Measure Methane Emissions from Natural Gas Facilities and Urban Areas, Environ. Sci. Technol., 29, 1468-1479, 1995.

(14) Czepiel, P. M.; Mosher, B.; Shorter, J. H., McManus, J. B.; Kolb, C. E.; Allwine, E.; Lamb, B. K.; Harriss, R.C.; Landfill Methane Emissions Measured by Static Enclosures and Atmospheric Tracer Methods, J. Geophys. Res. 101, 16,711-16,719, 1996.

(15) Czepiel, P. M.; Shorter, J. H.; Mosher, B.; Allwine, E.; McManus, J. B.; Harriss, R. C.; Kolb, C. E.; Lamb, B. K.; The Influence of Atmospheric Pressure on Landfill Methane Emissions, Waste Manage. 23, 593-598, 2003.

(16) Shorter, J. H.; McManus, J.B.; Kolb, C.E.; Allwine, E.J.; Siverson, R.; Lamb, B.K.; Mosher, B.W.; Harriss, R.C.; Howard, T.; Lott, R. A.; Collection of Leakage Statistics in the Natural Gas System by Tracer Methods, Environ. Sci. Technol., 31, 2012-2019, 1997.

(17) Mosher, B.W.; Czepiel, P. M.; Shorter, J. H.; Allwine, E.; Harriss, R.C.; Kolb, C.E.; Lamb, B.; Mitigation of Methane Emissions at Landfill Sites in New England, USA, Energy Conversion Manage. 37, 1.093-1098, 1996.

(18) Mosher, B.W.; Czepiel, P.M.; Harriss, R.C.; Shorter, J.H.; Kolb, C.E.; McManus, J.B.; Allwine, E.; Lamb. B. K.; Methane Emissions at Nine Landfill Sites in the Northeastern United States, Environ. Sci. Technol. 33, 2088-2094, 1999.

(19) Blaha, D.; Mosher, B.; Harriss, R.C.; Kolb, C.E.; McManus, J.B.; Shorter, J.H.; Lamb, B.; Mapping Urban Sources of Atmospheric Methane, Geo. Info. Systems, 4, 34-38, 1994.

(20) Kolb, C.E.; Herndon, S.C.; McManus, J.B.; Shorter, J.H.; Zahniser, M.S.; Nelson, D.D.; Jayne, J.T.; Canagaratna, M.R.; Worsnop, D.R.; Mobile Laboratory with Rapid Response Instruments for Real-Time Measurements of Urban and Regional Trace Gas and Particulate Distributions and Emission Source Characteristics, Environ. Sci. Technol., 38, 5694–5703, 2004.

(21) Herndon, S. C.; Jayne, J. T.; Zahniser, M. S.; Worsnop, D. R.; Knighton, B.; Allwine, E.; Lamb, B. K.; Zavala, M.; Nelson, D. D.; McManus, J. B.; Shorter, J. H.; Canagaratna, M. R.; Onasch, T. B.; Kolb, C. E.; Characterization of urban pollutant emission fluxes and ambient concentration distributions using a mobile laboratory with rapid response instrumentation, Faraday Discuss., 130, 327–339, 2005a.

(22) Herndon, S. C.; Shorter, J.H; Zahniser, M.S.; Wormhoudt, J.; Nelson, D.D.; Demerjian, K.L; Kolb, C.E.; Real-time Measurements of SO_2, H_2CO and CH_4 Emissions from in-use

Curbside Passenger Buses in New York City using a Chase Vehicle, Environ. Sci. Technol. 39, 7984-7990, 2005b.

(23) Energy Information Administration, Summary Statistics for Natural Gas in the United States, 2007-2011, available at: http://www.eia.gov/naturalgas/data.cfm#production (accessed January, 2013).

(24) American Association of Petroleum Geologists, Contiguous United States: AAPG-CSD Geologic Provinces Codes Map, 1991