



# Methane Emissions from Process Equipment at Natural Gas Production Sites in the United States: Pneumatic Controllers

David T. Allen,<sup>\*,†</sup> Adam P. Pacsi,<sup>†</sup> David W. Sullivan,<sup>†</sup> Daniel Zavala-Araiza,<sup>†,∇</sup> Matthew Harrison,<sup>‡</sup> Kindal Keen,<sup>‡</sup> Matthew P. Fraser,<sup>§</sup> A. Daniel Hill,<sup>||</sup> Robert F. Sawyer,<sup>⊥</sup> and John H. Seinfeld<sup>#</sup>

<sup>†</sup>Center for Energy and Environmental Resources, University of Texas at Austin, 10100 Burnet Road, Building 133, M.S. R7100, Austin, Texas 78758, United States

<sup>‡</sup>URS Corporation, 9400 Amberglen Boulevard, Austin, Texas 78729, United States

<sup>§</sup>School of Sustainable Engineering and the Built Environment, Arizona State University PO Box 875306, Tempe, Arizona 85287, United States

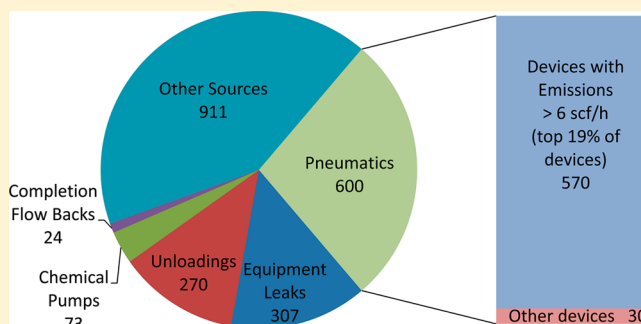
<sup>||</sup>Department of Petroleum Engineering, Texas A&M University, 3116 TAMU, College Station, Texas 77843-3116, United States

<sup>⊥</sup>Department of Mechanical Engineering, Mail Code 1740, University of California, Berkeley, California 94720-1740, United States

<sup>#</sup>Department of Chemical Engineering, California Institute of Technology, M/C 210-41, Pasadena, California 91125, United States

## Supporting Information

**ABSTRACT:** Emissions from 377 gas actuated (pneumatic) controllers were measured at natural gas production sites and a small number of oil production sites, throughout the United States. A small subset of the devices (19%), with whole gas emission rates in excess of 6 standard cubic feet per hour (scf/h), accounted for 95% of emissions. More than half of the controllers recorded emissions of 0.001 scf/h or less during 15 min of measurement. Pneumatic controllers in level control applications on separators and in compressor applications had higher emission rates than controllers in other types of applications. Regional differences in emissions were observed, with the lowest emissions measured in the Rocky Mountains and the highest emissions in the Gulf Coast. Average methane emissions per controller reported in this work are 17% higher than the average emissions per controller in the 2012 EPA greenhouse gas national emission inventory (2012 GHG NEI, released in 2014); the average of 2.7 controllers per well observed in this work is higher than the 1.0 controllers per well reported in the 2012 GHG NEI.



## INTRODUCTION

Natural gas production in the United States is increasing; the U.S. Energy Information Administration projects that by 2040, total natural gas production in the United States will increase by 40%.<sup>1</sup> With increased production, natural gas is displacing other fuels,<sup>2</sup> and this fuel switching has implications for greenhouse gas emissions.

Natural gas may have a lower greenhouse gas footprint than other, more carbon intensive, fossil fuels (coal and petroleum), since the carbon dioxide emissions associated with natural gas combustion are less than those associated with the combustion of coal and petroleum. For example, for identical heat releases on combustion, natural gas generates less than half of the carbon dioxide emissions of a typical coal.<sup>3</sup> The greenhouse gas benefits of natural gas relative to other fossil fuels may be eroded, however, by natural gas leaks in the supply chain. Methane, the principal component of natural gas, is a potent, but short-lived greenhouse gas. Because one kg of methane emissions is equivalent to between 28 and 120 kg of CO<sub>2</sub> emissions, depending on the time scale over which impacts are

assessed (100-year to immediate time horizons),<sup>4,5</sup> methane emissions in the natural gas supply chain amounting to more than a few percent of natural gas use can change the greenhouse gas footprint of natural gas, relative to other fossil fuels.<sup>5–9</sup> Thus, to characterize the greenhouse gas footprint of natural gas, it is important to determine the magnitude of methane emissions in the natural gas supply chain.<sup>10</sup>

Methane emissions in the natural gas supply chain have been estimated using two basic approaches, commonly referred to as top-down and bottom-up approaches. Top-down approaches for estimating methane emissions from the natural gas supply chain involve measuring ambient concentrations of methane near emission sources. These concentrations can be measured using fixed ground monitors,<sup>11,12</sup> mobile and vehicle mounted ground monitors,<sup>13,14</sup> aircraft based instruments<sup>15–17</sup> or

Received: August 18, 2014

Revised: November 11, 2014

Accepted: November 21, 2014

Published: December 9, 2014



**Table 1. Sample Population, Categorized by Controller Application and Region (AP= Appalachian; GC = Gulf Coast; MC = Mid-Continent; RM = Rocky Mountain)**

region	separator	number of controllers sampled, categorized by application							total
		process heater	compressor	wellhead	plunger lift	dehydration system	flare	sales	
AP	14	13	0	24	1	0	0	0	52
GC	73	0	13	11	7	17	1	1	123
MC	48	11	7	0	11	0	0	0	77
RM	51	21	0	32	11	8	2	0	125
total	186	45	20	67	30	25	3	1	377

satellite instruments.<sup>18</sup> Brandt et al.<sup>19</sup> and Miller et al.<sup>20</sup> have summarized recent top-down estimates of methane emissions and conclude that top-down emission estimates are generally higher than current bottom-up inventories of methane emissions, and some of this difference may be due to methane emissions from the natural gas supply chain. However, these analyses do not reveal which of the many potential sources of methane emissions along the natural gas supply chain might be incorrectly estimated.

Complementing top-down measurements, bottom-up measurements of methane emissions are made directly at the emission sources. In this approach, emission measurements are made at a representative sample of sources; the measurements from the sample population are then extrapolated to larger regional or national populations. The advantage of “bottom-up” approaches is that they can gather much more detail about the emission sources, and therefore can identify which source categories, among many, are responsible for emissions. For example, Allen, et al.<sup>21</sup> concluded that emissions from well completion flowbacks are overestimated, while emissions from pneumatic controllers may be underestimated, in current inventories of emissions. Both top-down and bottom-up approaches can contribute to an improved understanding of methane emissions from the natural gas supply chain. The work reported here uses bottom-up measurements to improve understanding of emissions from pneumatic controllers on natural gas production sites.

Pneumatic controllers use gas pressure to control the operation of mechanical devices, such as valves. The valves, in turn, control process conditions such as levels, temperatures, and pressures. When a pneumatic controller identifies the need to change liquid level, pressure, temperature or flow, it will open or close a control valve in order to return to a desired set point. The opening and closing of the valve can occur either through discrete (on/off) changes, or through changes that are proportional in magnitude to the deviation from the set point (throttling). Controllers can deliver this type of service (on/off and throttling) through either continuously venting or intermittent venting of gas. Thus, controllers can be grouped into four categories, depending on the type of service (on/off or throttling) and the type of venting (continuous or intermittent). In estimating emissions, the U.S. EPA uses the categories of low continuous bleed (<6 scf/h of gas vented), high continuous bleed (>6 standard cubic feet per hour (scf/h) of gas vented) and intermittent controllers.<sup>22</sup> Finally, controllers can also be categorized based on equipment manufacturer, model number, and the type of application (e.g., separator level control) in which they are used. In this work, the primary categorization of controllers will be as either continuous vent or intermittent vent based on the pattern observed during measurement; data on applications, service

types, and EPA categorization for the controllers sampled in this work are provided in Supporting Information (SI).

The U.S. EPA<sup>22</sup> reports 477 606 pneumatic controllers are in use at natural gas production sites in the United States. These controllers are estimated to emit 334 Gg/yr of methane (17.4 billion cubic feet (bcf) methane), for an average of 0.7 Mg device<sup>-1</sup> yr<sup>-1</sup> or 4.2 scf/h methane device<sup>-1</sup>. These estimated emissions from pneumatic controllers have been based on relatively limited measurements;<sup>23</sup> recent field measurements have suggested that these emissions may be understated.

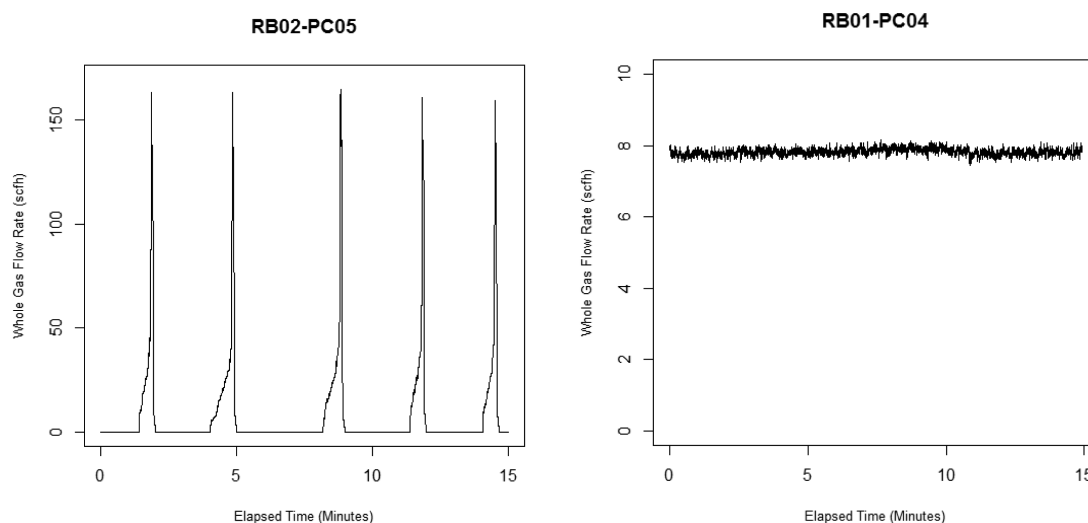
Allen et al.<sup>21</sup> made measurements of emissions from 305 pneumatic controllers on well sites in the United States where the wells had been hydraulically fractured. Average emissions were 10.5 scf/h of methane, approximately double the average emission rate per device in the current EPA national emission inventory. Measurements of emissions from 581 pneumatic controllers, made in British Columbia and Alberta, averaged 9.2 scf/h of whole gas,<sup>24</sup> an emission rate similar to that reported by Allen et al.<sup>21</sup> In both of these studies, emissions from controllers exhibited wide ranges. In both sets of measurements, a small subset of controllers accounted for most of the emissions.

While the measurements at hydraulically fractured gas wells in the United States<sup>21</sup> and the measurements in British Columbia and Alberta<sup>24</sup> recorded emissions higher than the average emissions per device in the EPA national emission inventory, the sampled populations for these two sets of measurements were not necessarily broadly representative of U.S. national populations of pneumatic controllers. The measurements reported by Allen et al.<sup>21</sup> were made exclusively in shale gas production regions, and at sites where the wells had been hydraulically fractured. Many of the sites were recently completed wells, which initially tend to have higher liquid production rates, and therefore may have more frequent actuation of certain types of pneumatic devices than the average for the entire population of gas wells in the United States, leading to potentially higher emissions. The Canadian measurements<sup>24</sup> were made exclusively in one production region and on devices with manufacturer specified emission rates in excess of 4.2 scf/h.

The goals of the work presented here were (i) to measure emissions from pneumatic controllers at a wider population of wells, geographically distributed across the United States, including conventional gas wells, shale gas wells and a limited number of oil wells, and (ii) to characterize the features of the controllers with high emissions, which previous work<sup>21,24</sup> has found to be the major contributor to emissions.

## ■ MATERIALS AND METHODS

**Sampled Population.** A total of 377 pneumatic controllers were sampled at 65 sites (some with multiple wells) throughout the United States (an average of 5.8 pneumatic controllers per



**Figure 1.** Representative time series for supply gas measurements for intermittent vent (left) and continuous vent (right) controllers; the intermittent vent controller (RB02-PC05) had a total of five actuations during the sampling period and an average emission rate, over the 15 min period of 7.9 scf/h; the continuous vent controller (RB01-PC04) had nearly constant emissions of 8.0 scf/h.

site, 2.7 controllers per well). Measurements were made primarily at natural gas production sites (351 of 377 controllers), and at a limited number of oil sites (26 controllers). Because the definitions of oil and gas wells vary, largely depending on gas to oil production ratios, the data will be treated as a single set. Sampling sites were selected from well sites owned by companies participating in the study using a process designed to yield a random sampling of participant sites (see SI, Section S1). For each well site that was visited, all controllers on the site were sampled using supply gas meters, unless operating conditions, safety issues or other factors prevented sampling. A total of 333 controllers had measurements made using the supply gas meters; 97 controllers could not be measured using the supply gas meter; of the 97 that were not sampled with the supply gas meter, 44 were sampled using exhaust gas measurements, leading to a total of 377 controllers in the sampled population. The applications that the controllers were used in (e.g., separator level control, compressor pressure control) are shown in Table 1. Details of the regions, device types, associated well types, operating methods and other characteristics of each of the 377 controllers sampled in this work are provided in SI, Section S4.

**Emission Measurement Methods.** Emissions from pneumatic controllers can be determined either by measuring the supply of gas entering the controller or by measuring the gas discharged from the controller. Both approaches were used in this work, and since there is no accumulation of gas in the controller, both measurement approaches should lead to equivalent measurements, if there are no leaks in the equipment downstream of the controller.

Measurements of the gas entering the controller were made by one of three Fox flow meters (model #FT2A); flow meters were inserted into the supply gas line for the controller. This supply gas measurement was the primary measurement method used in this work, and was used to measure emission rates on 333 of the 377 controllers in the sample population (the remainder were sampled by measuring gas emitted by the controller using a HiFlow Sampler described later in this section, see SI for comparisons between the supply gas and exhaust gas measurements). The flow meters reported flows at a sampling frequency of 10 Hz. Two of the Fox model #FT2A

instruments (labeled A and C in this study) had a range of operation of 0–300 scf/h, with a precision of  $\pm 1\%$  of flow, and the third Fox model #FT2A (labeled B in this study) had a range of operation of 0–1200 scf/h, with a precision of  $\pm 1\%$  of flow. The Fox model #FT2A instruments A and C were used whenever possible because of their greater absolute precision, however, if any instantaneous reading on the A or C Fox model #FT2A was greater than 300 scf/h, the measurement on the pneumatic device was repeated with the B meter to ensure that high leak rates were measured accurately. This happened only once during the measurement campaign, and for this single controller, the flow exceeded 300 scf/h only during a few seconds when the flow from an actuation was peaking (average whole gas flow rate over 15 min of sampling was 3.06 scf/h, a value lower than the average emissions per controller in the sampled population). A repeat test with the B instrument did not detect any actuations.

For each controller measurement using the supply gas flow meter, a site operator depressurized and disconnected the controller supply gas line; the flow meter was inserted and the system was reconnected, repressurized, and allowed to stabilize for several minutes before measurements began. Once the system had stabilized, measurements were made for approximately 15 min. Longer sampling times may have allowed a more complete measurement of emission rates from devices with relatively fewer controller actuations, but would have limited the number of controllers that could be sampled. Figure 1 shows representative 15 min emission time series for pneumatic controllers measured using the supply gas measurement.

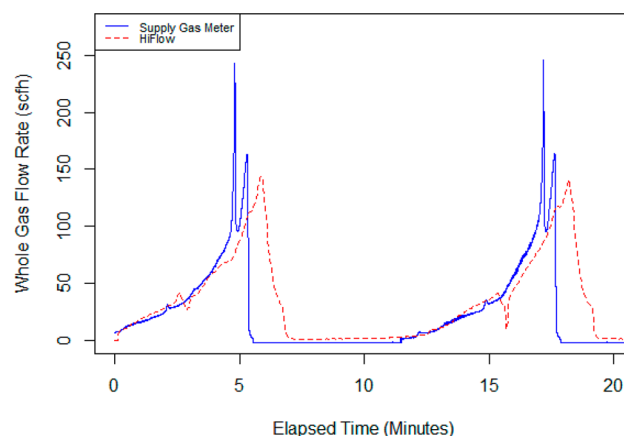
All three Fox flow meters were calibrated by the instrument manufacturer and in laboratory testing, using methane. The instruments measure flow based on a thermal conductivity measurement. In this work, since gas composition information was available for each site where measurements were made, site specific correction factors were employed to estimate methane and whole gas emission rates. The method is described in SI Section S2. Results in this work are reported as both methane and whole gas emission rates, based on site specific gas composition data.



For some pneumatic controllers, it was not possible or safe to disrupt the supply gas to insert the supply gas flow meter, so exhaust gas measurements were used as the primary measurement on that subset of devices. Exhaust gas flow rate was measured using a Hi Flow instrument similar to that described by Allen et al.<sup>21</sup> Briefly, the Hi Flow Sampler is a portable, intrinsically safe, battery-powered instrument that has been used for several decades in measuring emissions of methane in the natural gas supply chain.<sup>25–27</sup> An emission source is enclosed, using attachments that come with the instrument; leak rate is measured by drawing air from the enclosure, through the sampler, at a high flow rate (up to 8–10 cfm) to capture all the gas emitted by the component, along with a certain amount of entrained surrounding air. By accurately measuring the flow rate of the sampled stream and the background corrected natural gas concentration within the sampled stream, the gas leak rate is calculated. Methane is measured, at concentrations less than approximately 5%, by a catalytic oxidizer unit coupled with a thermal conductivity detector. At methane concentrations greater than approximately 5%, concentrations are measured directly using a thermal conductivity detector. The instrument was calibrated using pure methane and a mixture containing 2.5% methane. The instrument reading based on the methane calibration was corrected for gas composition using site specific gas composition data and laboratory data, as described in SI Section S2. The commercial Hi Flow instrument is designed primarily to measure methane leaks that have a relatively steady flow and flow rates are not normally automatically recorded at high frequency. For this work, the instrument software was modified by the manufacturer to output data every 2–3 s. A 0.3–0.5 Hz reporting frequency was selected based on residence times expected in the leak enclosures at the maximum flow rate of the Hi Flow device (at a 10 cubic feet per minute sample flow, gas in a 1 ft<sup>3</sup> sample enclosure has a residence time of 6 s). As with the in-line supply gas measurement, Hi Flow data were collected for approximately 15 min for each controller. A time series from the Hi Flow device, along with a parallel measurement made using a supply gas meter, is shown in Figure 2. The Hi Flow device, because it entrains ambient air in a long sample loop, dampens some of the peak rate. Therefore, the Hi Flow is not able to resolve high frequency actuations as well as the in-line supply measurement. For 24 controller measurements, both supply gas and Hi Flow measurements were made to compare the two measurement methods (for these controllers, the supply gas flow was treated as the primary measurement). The detailed results are provided in SI Section S3. To summarize, 11 of the 24 simultaneous measurements had emissions of less than 0.005 scfh (46%), as recorded by the in-line supply gas meter (the primary measurement device). For five devices which had an average emission rate greater than 6 scfh (measured by the supply gas meter) the supply gas meter to Hi Flow measurement ratio was between 0.7 and 1.1.

## RESULTS AND DISCUSSION

Methane emissions from 377 controllers were measured in this work and details of each of the individual measurements are available in SI Section S4. A relatively small subset of devices accounts for a majority of the emissions. At the high end of the emission rate distribution, 20 percent of devices accounted for 96% of whole gas and methane emissions. The 19% of devices that had emissions in excess of 6 standard cubic feet whole gas



**Figure 2.** Comparison of supply gas meter (blue line) and Hi Flow measurements (red line) for device LB07-PC04, which was a water level control on a separator. The average emission rate measured by the supply gas meter was 27.0 scf/h as compared to 33.9 scf/h measured by the Hi Flow. Note that the time lag, longer period of emission detection, and the reduced maximum flow rate associated with the Hi Flow measurement is expected because of the dilution that occurs with ambient air in the exhaust enclosure and the flow through the instrument.

per hour (scf/h) accounted for 95% of all whole gas and methane emissions. At the low emission rate end of the distribution, more than half (51%) of the controllers had an emissions rate less than 0.001 scf/h over the 15 min sampling period; 62% had an emissions rate less than 0.01 scf/h over the 15 min sampling period.

The average emission rate for the 377 devices is 5.5 scf/h of whole gas (4.9 scf/h of methane), however, this average emission may be influenced by the estimated emission rates for devices that had no emissions over the 15 min sampling period. If the devices with no emissions detected over 15 min are assigned the lowest emission rate detected (0.001–0.01 scf/h), there is no change in the average emission rate. However, using this minimum detection limit approach may underestimate potential emissions for devices that had little to no detectable emissions over 15 min. Some of these devices may have relatively infrequent actuations that were not sampled. In principle, any device actuating less than four times per hour may not have been detected over a 15 min sampling period. To estimate the emissions from devices with no emissions detected over a 15 min sampling period, the average emission per actuation was calculated for controllers in each application. The average emissions per actuation were multiplied by an estimated frequency of actuation. For example, for separator level controllers, the average volume per actuation was estimated by averaging observed volumes per actuation for separator level controllers; the average frequency of actuation for devices, for which no actuations were observed, was estimated by extrapolating observed actuation frequency data for controllers in separator level control service. A variety of assumptions can be made in extrapolating actuation frequencies. Details of a variety of approaches are available in SI Section S5. Using a variety of approaches, the estimated average emissions associated with devices with no emissions recorded over a 15 min sampling increases the population average emissions by 2–11%. Because this increase is relatively small, for clarity, all of the data reported in this work are based on

**Table 2. Whole Gas Emissions from Controllers (scf/h), Categorized by Region and Application<sup>a</sup>**

region	all devices	average whole gas emission rates from controllers (scf/h), categorized by the application								
		separator	process heater	compressor	wellhead	plunger lift	dehydration system	flare	sales	avg. w/o compressors
AP	1.7	0.3	1.3		2.8	0.0				1.7
GC	11.9	16.3		10.6	0.0	7.3	4.3	0.0	0.0	12.0
MC	5.8	4.9	0.0	20.2		6.5				4.4
RM	0.8	1.5	0.2		0.4	0.1	0.0	0.0		0.8
average	5.5	8.1	0.5	14.0	1.2	4.1	3.0	0.0	0.0	5.0

<sup>a</sup>Numbers of devices sampled in each category are reported in Table 1.

actual measurements, not including additions to the emissions for devices with low (0.001–0.01 scf/h) observed emissions.

To estimate an uncertainty bound on the overall average, a bootstrapping process was used.<sup>28</sup> In the bootstrapping procedure, the original data set of 377 devices was recreated by making 377 random device selections, with replacement, from the data set. A total of 1000 of these resampled data sets were created and the mean value of the emissions for each resampled data set was determined. The 95% confidence interval for the average whole gas emission estimate of 5.5 scf/h is 4.0–7.2 scf/h, where the bounds represent the 2.5% and 97.5% percentiles of the means in the 1000 resampled data sets. Similarly, the 95% confidence interval for the average methane emissions estimate of 4.9 scf/h is 3.6–6.5 scf/h.

The measurements showed significant variations among regions, the controller application, and whether the device was continuous vent or intermittent vent. Table 2 summarizes the distribution of emission rates among controllers in various applications, and shows the regional distribution of controller emissions. Measurements made on pneumatics in service on compressors had average emission rates of 14.0 scf/h (12.4 scf/h methane), compared to an average whole gas emission rate of 5.5 scf/h (4.9 scf/h methane) for all devices. Devices in use for level control on separators averaged 8.1 scf whole gas/h (7.1 scf methane/h). Overall, 76% of devices measured with whole gas emission rates greater than 6 scf/h were in service on compressors or as level controllers on separators. Emission rates for continuous vent controllers (57 devices, average emissions of 24.1 scf/h whole gas, 21.8 scf/h methane) were higher than for intermittent vent devices (2.2 scf/h whole gas, 1.9 scf/h methane).

In addition to varying by application and controller type (continuous vent or intermittent vent), emissions also varied by region. Emissions were highest in the Gulf Coast and Mid-Continent regions and were lowest in the Rocky Mountain and Appalachian regions (see SI, Section S4, for geographical boundaries of regions). Controllers on compressors, with high average emissions, were only observed on sampling sites in the Gulf Coast and Mid-Continent regions, so some of the regional differences can be attributed to the presence of compressors. As shown in Table 2, however, if average emissions by region are recalculated without including controllers associated with compressors, the Gulf Coast and Mid-Continent regions still had average emissions greater than those observed in the Rocky Mountain and Appalachian regions.

Another factor that may account for regional differences in emission rates is frequency of actuation. For example, controllers on separators in the Gulf Coast could actuate more frequently due to higher liquid production rates, which could explain higher emission rates in that region. However, the frequencies of actuation for the devices in Gulf Coast were similar to those in most other regions, indicating a larger

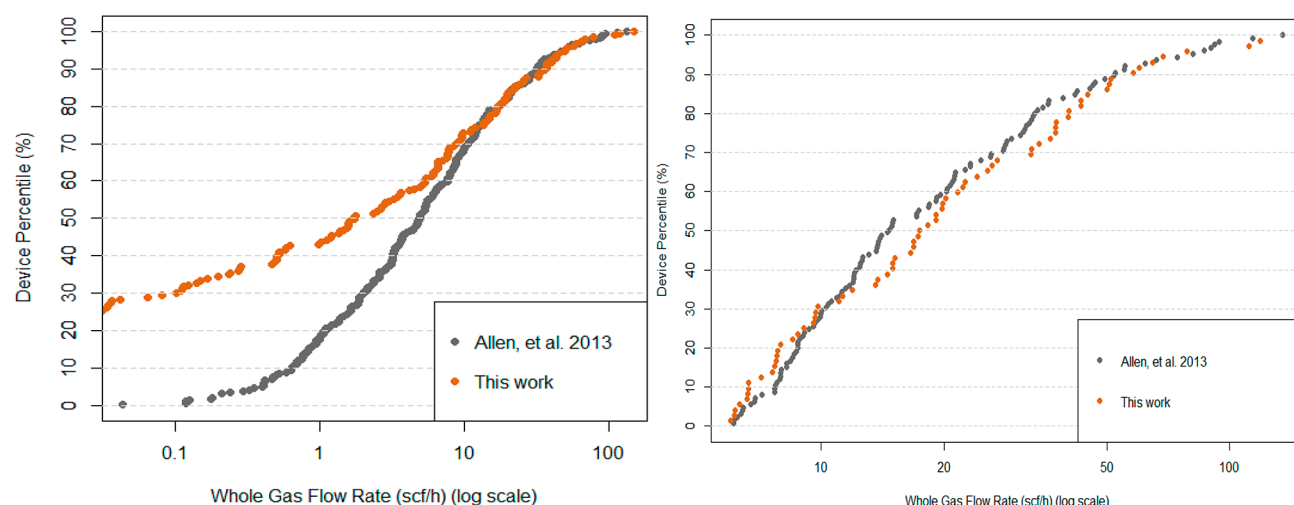
emission per actuation for the devices in the Gulf Coast, rather than more frequent actuation. In contrast, the Appalachians showed a considerably higher frequency of actuations and a smaller emission rate, indicating a smaller emission per actuation for those devices (Table 3). Thus, regional differences

**Table 3. Frequency of Actuations and Emissions from Intermittent Vent Controllers Where Actuations Were Observed, Categorized by Region**

region	count of devices	frequency of actuations (#/min)	avg. emission rate (scf/h)
AP	8	2.42	4.85
GC	30	0.37	20.5
MC	17	0.93	5.05
RM	25	0.43	1.72
total	80	average: 0.73	average: 9.76

in pneumatic controller emission rates cannot be completely explained by frequency of actuation of controllers, or by controllers associated with compressors and separator level control (Table 2); much of the difference may be due to differences in controller type (continuous vent vs intermittent vent) among regions. Continuous vent devices, with average whole gas emissions of 24.1 scf/h, were 21% of the controllers in the Gulf Coast and Mid-Continent regions, but only 9% in the Appalachian and Rocky Mountain regions.

This data set of emissions from pneumatic devices has elements that are similar to and different from the previous data sets reported for the United States,<sup>21</sup> and for British Columbia and Alberta.<sup>24</sup> The primary similarity is that all three data sets indicate that a small population of devices dominates total emissions. In this work, 19% of devices with emissions greater than 6 scf/h of whole gas account for 95% of the whole gas and methane emissions. In the previous measurements reported by Allen et al.,<sup>21</sup> 20% of devices account for 80% of the whole gas emissions and 41% of devices with emissions greater than 6 scf/h of whole gas, account for 90% of the whole gas emissions (88% of the methane emissions). In the measurements for British Columbia and Alberta<sup>24</sup> (referred to here as the British Columbia data), which were restricted to pneumatic devices with manufacturer reported bleed rates greater than 4.2 scf/h, 44% of devices with emissions greater than 6 scf/h accounted for 91% of emissions. Both the British Columbia data and the measurements reported in this work had large numbers of devices for which no emissions were detected during the sampling period. For the British Columbia data (again, focused on devices with manufacturer reported bleed rates in excess of 4.2 cfh), 31% of measurements had no detectable emissions over a 30 min sampling period; in this work 62% of devices had emissions less than 0.01 scf/h over the 15 min sampling period.



**Figure 3.** Distributions of emissions for subsets of controllers (38% of devices measured in this work) venting greater than 0.01 scf/h (left) and subsets of controllers venting greater than 6 scf/h of whole gas (right) as reported in this work (the 19% of devices that account for 95% of emissions) and Allen et al.<sup>21</sup>

The overall average emission rates reported in this work are lower than the previous data sets reported by Allen et al.<sup>21</sup> for the United States, and for British Columbia and Alberta.<sup>24</sup> For the British Columbia data this can be attributed to the sampling design for that data set, which selected devices with manufacturer reported bleed rates in excess of 4.2 scf/h. These controller types tend to be found in particular applications. When the emissions from the British Columbia data set are compared to the emissions reported in this work, for devices in similar applications, the results are in reasonable agreement. For example, for the separator controllers that were the most frequent application observed in this work, the British Columbia data report an average emissions rate of approximately 7.8 scf/h (level control) while the average for this work was 8.1 scf/h (separator application).

The lower average emission rates reported in this work, compared to those reported by Allen et al.<sup>21</sup> is primarily due to the number of controllers with no emissions detected over the sampling period. Figure 3 compares emission rates for controllers sampled in this work, with emissions rates reported by Allen et al.<sup>21</sup> The results show reasonable agreement between the two studies for controllers with emissions above 6 scf/h. These controllers accounted for 95% of the emissions in this work and 90% of the emissions in the sample reported by Allen et al. (2013).

The primary reason for the differences in the average emission rates reported in this work and in Allen et al.<sup>21</sup> is the higher percentage of low emission devices (<0.01 scf/h) observed in this work. This could be due to multiple factors. In this work, all controllers on-site were sampled, regardless of whether they would be reported through emission inventories. For example, emergency shut-down (ESD) controllers represented 12% of the sampled population in this work. These controllers do not have planned actuations, so they would not have been sampled in the work of Allen et al.,<sup>21</sup> and they may or may not be included in controller counts in national emission inventories. In addition, in the work of Allen et al.<sup>21</sup> about 40% of the inventoried controllers on sites were sampled; while these were intended to be selected randomly from inventoried controllers, there may have been an

unintentional bias toward devices that were observed, with an infrared camera, to have emissions.

**Implications for National Emission Estimates.** As shown in Table 4, if regional average emission rates determined

**Table 4. National Emission Estimates, Based on Regional Device Counts for Pneumatic Controllers and Regional Average Emissions Measured in This Work**

region	count of devices	avg. emission rate whole gas(scfh)	avg. emission rate methane (scfh)	regional emissions (Gg/yr)
AP	77 261	1.70	1.65	21.5
GC	53 436	11.80	10.61	95.4
MC <sup>a</sup>	222 684	5.80	4.87	182.5
RM <sup>b</sup>	124 225	0.75	0.67	14.0
total	477 606			313.4

<sup>a</sup>MC totals include equipment counts for Mid-Continent and Southwest regions reported in the 2012 EPA GHG NEI. <sup>b</sup>RM totals include equipment counts for Rocky Mountain and West Coast regions reported in the 2012 EPA GHG NEI.

in this work are multiplied by regional controller counts reported in the 2012 EPA national greenhouse gas emission inventory (2012 GHG NEI, released in 2014), the national methane emission estimate for pneumatic controllers in natural gas service is 313 Gg/yr (within 10% of the 2012 GHG NEI estimate of 334 Gg). If the national average of the emission rates measured in this work (5.5 scf/h of whole gas, or 4.9 scf/h of methane) is multiplied by the total national equipment count in the 2012 GHG NEI (477 606 controllers) the national methane emission estimate is 394 Gg/yr, 17% higher than the 2012 GHG NEI estimate of 334 Gg. Adding an additional 2–11% to the estimated emission totals to account for potential emissions from controllers that had less than 0.01 scf/h of emissions over 15 min, would only slightly change these comparisons with the 2012 GHG NEI. This estimate may represent a lower bound on national emissions, however, since the average emissions per controller observed in this work includes some low- or nonemitting devices, such as ESD controllers, that may not be included in the count of national controllers. If the average emissions per controller from this



work were recalculated with ESD controllers excluded, the average emissions would increase by approximately 15% (see SI, Section S7).

The inclusion or exclusion of ESD controllers in national pneumatic controller counts is just one part of the uncertainty associated with the total count of controllers. The average number of controllers per well observed in this work (2.7 controllers per well) was higher than the average number of controllers per well (1.0 controllers per well) reported in the 2012 GHG NEI, potentially indicating an under-count of controllers in the GHG NEI. Some of the difference between the controllers per well observed in this work and the average pneumatic controllers per well in the GHG NEI is due to wells that use mechanical or other nonpneumatic controllers; as an example of how an alternative controller count could influence national emission estimates, if 75% of wells in the United States have an average of 2.7 pneumatic controllers per well (the remainder having nonpneumatic controllers), and if 75% of the controllers on well sites are inventoried as having emissions, the total count of pneumatic controllers would double the level in the current inventory, roughly doubling emissions to 600 Gg (see SI). It was beyond the scope of this work to develop new national pneumatic controller counts, but the data reported here indicate that this is a topic that merits attention.

**Characteristics of High Emitting Devices.** Because average emissions are strongly influenced by the highest emitting devices, the characteristics of the 40 controllers with highest emissions rates were examined in detail by experts in pneumatic device operation. These characterizations included the service type, region of use, device type, the numbers of actuations and other temporal features of the emission time series. Based on these analyses, many of the devices in the high emitting group were behaving in a manner inconsistent with the manufacturer's design. For example, some devices not designed to bleed continuously had continuous emissions. This could be the result of a defect in the system, such as a crack or hole in the end-device's (control valve's) diaphragm actuator, or a defect in the controller itself, such as fouling or wear. No additional troubleshooting analysis was performed on these high emitters, so the actual root causes are not known with certainty. The results, however, do indicate that some of the high emissions were caused by repairable issues. Details are provided in the SI (Section S8).

## ■ ASSOCIATED CONTENT

### ■ Supporting Information

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

## ■ AUTHOR INFORMATION

### Corresponding Author

\*Phone: 512-475-7842; e-mail: [allen@che.utexas.edu](mailto:allen@che.utexas.edu).

### Present Address

<sup>†</sup>Environmental Defense Fund, 301 Congress Avenue, Suite 1300 Austin, Texas 78701

### Notes

The authors declare the following competing financial interest(s): Lead author David Allen serves as chair of the Environmental Protection Agency's Science Advisory Board, and in this role is a paid Special Governmental Employee. He is also a journal editor for the American Chemical Society and has served as a consultant for multiple companies, including

Eastern Research Group, ExxonMobil, and Research Triangle Institute. He has worked on other research projects funded by a variety of governmental, nonprofit and private sector sources including the National Science Foundation, the Environmental Protection Agency, the Texas Commission on Environmental Quality, the American Petroleum Institute and an air monitoring and surveillance project that was ordered by the U.S. District Court for the Southern District of Texas. Adam Pacsi and Daniel Zavala-Araiza, who were graduate students at the University of Texas at the time the work in this paper was done, have accepted positions at Chevron Energy Technology Company and Environmental Defense Fund, respectively. John Seinfeld served as a consultant for Shell in 2012. A. Daniel Hill owns ExxonMobil, BP, and ConocoPhillips stock, serves on the Advisory Board for Sanchez Oil and Gas, for which he is compensated, and has been a consultant for Schlumberger and numerous oil and gas operating companies..

## ■ ACKNOWLEDGMENTS

We thank the sponsors of this work for financial support, technical advice and access to sites for sampling. The sponsors were Environmental Defense Fund (EDF), Anadarko Petroleum Corporation, BG Group plc, Chevron, ConocoPhillips, Encana Oil & Gas (USA) Inc., Pioneer Natural Resources Company, SWEPI LP (Shell), Southwestern Energy, Statoil, and XTO Energy, a subsidiary of ExxonMobil. Funding for EDF's methane research series, including the University of Texas study, is provided for by Fiona and Stan Druckenmiller, Heising-Simons Foundation, Bill and Susan Oberndorf, Betsy and Sam Reeves, Robertson Foundation, Alfred P. Sloan Foundation, TomKat Charitable Trust, and the Walton Family Foundation. We thank the experts who reviewed the data on high emitting devices.

## ■ REFERENCES

- (1) US Energy Information Administration. *Annual Energy Outlook*; U.S. Department of Energy: Washington D.C, 2014.
- (2) Energy Information Administration. *Annual Electric Generation Data*, January 2013. U.S. Department of Energy, Washington, DC, 2013; [www.eia.doe.gov/cneaf/electricity/page/eia860.html](http://www.eia.doe.gov/cneaf/electricity/page/eia860.html).
- (3) Allen, D. T. Atmospheric emissions and air quality impacts from natural gas production and use. *Annu. Rev. Chem. Biomol. Eng.* **2014**, *5*, 55–75.
- (4) Intergovernmental Panel on Climate Change. *Fifth Assessment Report*; Intergovernmental Panel on Climate Change: Geneva, 2014.
- (5) Alvarez, R. A.; Pacala, S. W.; Winebrake, J. J.; Chameides, W. L.; Hamburg, S. P. Greater focus needed on methane leakage from natural gas infrastructure. *Proc. Natl. Acad. Sci. U.S.A.* **2012**, *109*, 6435–6440.
- (6) Hayhoe, K.; Kheshgi, H. S.; Jain, A. K.; Wuebbles, D. J. Substitution of natural gas for coal: Climatic effects of utility sector emissions. *Clim. Change* **2002**, *54*, 107–139.
- (7) Lelieveld, J.; Lechtenböhmer, S.; Assonov, S. S.; Brenninkmeijer, C. A. M.; Dienst, C.; Fischelick, M.; Hanke, T. Low methane leakage from gas pipelines. *Nature* **2005**, *434*, 841–842.
- (8) Wigley, T. M. L. Coal to gas: The influence of methane leakage. *Clim. Change* **2011**, *108*, 601–608.
- (9) MacKay, D. J. C.; Stone, T. *Potential Greenhouse Gas Emissions Associated with Shale Gas Extraction and Use*; Department of Energy and Climate Change: London, September 9, 2013.
- (10) Laurenzi, I. J.; Jersey, G. R. Life cycle greenhouse gas emissions and freshwater consumption of Marcellus Shale Gas. *Environ. Sci. Technol.* **2013**, *47*, 4896–4903.
- (11) Zavala-Araiza, D.; Sullivan, D. W.; Allen, D. T. Atmospheric hydrocarbon emissions and concentrations in the Barnett Shale natural gas production region. *Environ. Sci. Technol.* **2014**, *48*, 5314–5321.

- (12) Katzenstein, A. S.; Doezeema, L. A.; Simpson, I. J.; Blake, D. R.; Rowland, F. S. Extensive regional atmospheric hydrocarbon pollution in the southwestern United States. *Proc. Natl. Acad. Sci. U.S.A.* **2003**, *100*, 11975–11979.
- (13) Herndon, S. C.; Floerchinger, C.; Roscioli, J. R.; Yacovitch, T. I.; Franklin, J. F.; Shorter, J. H.; Kolb, C. E.; Subramanian, R.; Robinson, A. L.; Molina, L. T.; Allen, D. Measuring methane emissions from industrial and waste processing sites using the dual tracer flux ratio method. In *Annual Meeting the American Geophysical Union*, San Francisco, December 2013.
- (14) Herndon, S. C.; Jayne, J. T.; Zahniser, M. S.; Worsnop, D. R.; Knighton, B.; Alwine, 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.* **2005**, *130*, 327–339.
- (15) Petron, G.; Frost, G.; Miller, B. R.; Hirsch, A. I.; Montzka, S. A.; Karion, A.; Trainer, M.; Sweeney, C.; Andrews, A. E.; Miller, L.; Kofler, J.; Bar-Ilan, A.; Dlugokencky, E. J.; Patrick, L.; Moore, C. T.; Ryerson, T. B.; Siso, C.; Kolodzey, W.; Lang, P. M.; Conway, T.; Novelli, P.; Masarie, K.; Hall, B.; Guenther, D.; Kitzis, D.; Miller, J.; Welsh, D.; Wolfe, D.; Neff, W.; Tans, P. Hydrocarbon emissions characterization in the Colorado Front Range: A pilot study. *J. Geophys. Res.* **2012**, *117* (D4), D04304.
- (16) Peischl, J.; Ryerson, T. B.; Brioude, J.; Aikin, K. C.; Andrews, A. E.; Atlas, E.; Blake, D.; Daube, B. C.; de Gouw, J. A.; Dlugokencky, E.; Frost, G. J.; Gentner, D. R.; Gilman, J. B.; Goldstein, A. H.; Harley, R. A.; Holloway, J. S.; Kofler, J.; Kuster, W. C.; Lang, P. M.; Novelli, P. C.; Santoni, G. W.; Trainer, M.; Wofsy, S. C.; Parrish, D. D. Quantifying sources of methane using light Alkanes in the Los Angeles Basin, California. *J. Geophys. Res. Atmos.* **2013**, *118*, 4974–4990.
- (17) Karion, A.; Sweeney, C.; Pétron, G.; Frost, G.; Hardesty, R. M.; Kofler, J.; Miller, B. R.; Newberger, T.; Wolter, S.; Banta, R.; Brewer, A.; Dlugokencky, E.; Lang, P.; Montzka, S. A.; Schnell, R.; Tans, P.; Trainer, M.; Zamora, R.; Conley, S. Methane emissions estimate from airborne measurements over a western United States natural gas field. *Geophys. Res. Lett.* **2013**, *40*, 1–5 DOI: 10.1002/grl.50811.
- (18) Wecht, K. J.; Jacob, D. J.; Sulpizio, M. P.; Santoni, G. W.; Wofsy, S. C.; Parker, R.; Bosch, H.; Worden, J. Spatially resolving methane emissions in California: Constraints from the CalNex aircraft campaign and from present (GOSAT, TES) and future (TROPOMI, geostationary) satellite observations. *APCD* **2014**, *14*, 4119–4148.
- (19) Brandt, A. R.; Heath, G. A.; Kort, E. A.; O'Sullivan, F.; Pétron, G.; Jordaan, S. M.; Tans, P.; Wilcox, J.; Gopstein, A. M.; Arent, D.; Wofsy, S.; Brown, N. J.; Bradley, R.; Stucky, G. D.; Eardley, D.; Harriss, R. Methane leaks from north american natural gas systems. *Science* **2014**, *343*, 733–735.
- (20) Miller, S. M.; Wofsy, S. C.; Michalak, A. M.; Kort, E. A.; Andrews, A. E.; Biraud, S. C.; Dlugokencky, E. J.; Eluszkiewicz, J.; Fischer, M. L.; Janssens-Maenhout, G.; Miller, B. R.; Miller, J. B.; Montzka, T.; Nehrkorn, T.; Sweeney, C. Anthropogenic emissions of methane in the United States. *Proc. Natl. Acad. Sci. U.S.A.* **2013**, *110*, 20018–20022.
- (21) Allen, D. T.; Torres, V. M.; Thomas, J.; Sullivan, D.; Harrison, M.; Hendler, A.; Herndon, S. C.; Kolb, C. E.; Fraser, M. P.; Hill, A. D.; Lamb, B. K.; Miskimins, J.; Sawyer, R. F.; Seinfeld, J. H. Measurements of methane emissions at natural gas production sites in the United States. *Proc. Natl. Acad. Sci. U.S.A.* **2013**, *110*, 17768–17773.
- (22) U.S. Environmental Protection Agency (EPA). *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2012*, April 2014.
- (23) U.S. Environmental Protection Agency (EPA). *Oil and Natural Gas Sector Pneumatic Devices*, Report for Review Panel; Office of Air Quality Planning and Standards, April 2014.
- (24) Prasino Group. *Final Report for determining bleed rates for pneumatic devices in British Columbia*. Report to British Columbia Ministry of Environment, December 2013.
- (25) Harrison, M. R.; Shires, T. M.; Wessels, J. K.; Cowgill, R. M. *Methane Emissions from the Natural Gas Industry*, Vols 1–15, Final Report, GRI-94/0257 and EPA-600/R-96-080; Gas Research Institute and US Environmental Protection Agency, June 1996.
- (26) 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.
- (27) 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* **1997**, *35*, 1365–1390.
- (28) Efron, B.; Tibshirani, R. *An Introduction to the Bootstrap*; CRC Press: Chapman & Hall, 1993.