RESEARCH ARTICLE

University of Texas study underestimates national methane emissions at natural gas production sites due to instrument sensor failure

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Abstract

The University of Texas reported on a campaign to measure methane (CH_4) emissions from United States natural gas (NG) production sites as part of an improved national inventory. Unfortunately, their study appears to have systematically underestimated emissions. They used the Bacharach Hi-Flow® Sampler (BHFS) which in previous studies has been shown to exhibit sensor failures leading to underreporting of NG emissions. The data reported by the University of Texas study suggest their measurements exhibit this sensor failure, as shown by the paucity of high-emitting observations when the wellhead gas composition was less than 91% CH_4, where sensor failures are most likely; during follow-up testing, the BHFS used in that study indeed exhibited sensor failure consistent with under-reporting of these high emitters. Tracer ratio measurements made by the University of Texas at a subset of sites with low CH_4 content further indicate that the BHFS measurements at these sites were too low by factors of three to five. Over 98% of the CH_4 inventory calculated from their own data and 41% of their compiled national inventory may be affected by this measurement failure. Their data also indicate that this sensor failure could occur at NG compositions as high as 97% CH_4, possibly affecting other BHFS measurement programs throughout the entire NG supply chain, including at transmission sites where the BHFS is used to report greenhouse gas emissions to the United States Environmental Protection Agency Greenhouse Gas Reporting Program (USEPA GHGRP, U.S. 40 CFR Part 98, Subpart W). The presence of such an obvious problem in this high profile, landmark study highlights the need for increased quality assurance in all greenhouse gas measurement programs.

Introduction

The climatic benefits of switching from coal to natural gas (NG) depend on the magnitude of fugitive emissions of methane (CH_4) from NG production, processing, transmission, and distribution [12, 13, 27]. This is of particular concern as the United States increasingly exploits NG from shale formations: a sudden increase in CH_4 emissions due to increased NG production could trigger climate "tipping points" due to the high short-term global warming potential of CH_4 (86× carbon dioxide on a 20-year time scale) [19]. The United States Environmental Protection Agency (USEPA) estimates CH_4 emissions from the NG supply chain by scaling up individual ground-level measurements, mostly collected by reporting from industry [26]. However, some recent studies have questioned whether these "bottom-up" inventories are too low, since airborne measurements indicate that CH_4 emissions from NG production regions are higher than the inventories indicate [5, 14, 17, 20, 21].

In order to help determine the climate consequences of expanded NG production and use, and to address the apparent discrepancy in top-down and bottom-up measurements, the University of Texas (UT) at Austin and the

Environmental Defense Fund launched a large campaign to measure CH_4 emissions at NG production sites in the United States [1]. This study used both existing EPA GHG inventory data and new measurements to compile a new national inventory of CH_4 emissions from production sites. Forty-one percent of this new inventory was based on measurements made by [1], which included measurements of emissions from well completion flowbacks as well as measurements of emissions from chemical injection pumps, pneumatic devices, equipment leaks, and tanks at 150 NG production sites around the United States already in routine operation (measurements from tanks were not used for inventory purposes). However, the measurements of emissions at well production sites already in operation (which comprised 98% of the new inventory developed by [1]) were made using the Bacharach Hi-Flow Sampler (BHFS; Bacharach, Inc., New Kensington, PA) and recent work has shown that the BHFS can underreport individual emissions measurements by two orders of magnitude [10]. This anomaly occurs due to sensor transition failure that can prevent the sampler from properly measuring NG emission rates greater than ~0.4 standard cubic feet per minute (scfm; 1 scfm = 1.70 m^3 h^{-1} or 19.2 g min^{-1} for pure CH_4 at 60°F [15.6°C] and 1 atm; these are the standard temperature and pressure used by the U.S. NG industry). Although this failure is not well understood, it does not seem to occur when measuring pure CH_4 streams, but has been observed in four different samplers when measuring NG streams with CH_4 contents ranging from 66% to 95%. The sampler's firmware version and elapsed time since last calibration may also influence the occurrence of this problem [10, 18].

This paper presents an analysis of the UT [1] emissions measurements that were made with the BHFS, and shows that high emitters (>0.4 scfm [0.7 m^3 h^{-1}]) were reported very rarely at sites with a low CH_4 content in the wellhead gas (<91%), consistent with sensor transition failure. It also details testing of the exact BHFS instrument used in that study and shows the occurrence of this sensor failure at an NG production site with a wellhead composition of 91% CH_4 (the highest CH_4 concentration site available during testing). Finally, the downwind tracer ratio measurements made by [1] at a subset of their test sites are reexamined and indicate that the BHFS measurements made at sites with low wellhead CH_4 concentrations were too low by factors of three to five.

Evidence of BHFS Sensor Transition Failure in the UT Dataset

The Allen et al. [1] UT dataset is unique due to the large number of BHFS measurements made across a wide geographic range, the variety of emissions sources (equipment leaks, pneumatic devices, chemical injection pumps, and tanks) and the wide range of NG compositions (67.4–98.4% CH_4) that were sampled. As such, the UT study provides an important opportunity to evaluate the occurrence of sensor transition failure in the BHFS as well as the impact of this issue on emission rates and emissions factors based on measurements in other segments of the NG supply chain.

The BHFS uses a high flow rate of air and a loose enclosure to completely capture the NG-emitting from a source, with the emission rate calculated from the total flow rate of air and the resulting sample NG concentration, after the background NG concentration is subtracted. The sampler uses a catalytic oxidation sensor to measure sample concentrations from 0% to 5% NG in air, but must transition to a thermal conductivity sensor in order to accurately measure sample concentrations higher than 5%. It is the failure of the sampler to transition to the higher range that has been previously observed by Howard et al. [10] and which can prevent the sampler from correctly measuring emission rates larger than 0.3–0.5 scfm (0.5–0.9 m^3 h^{-1}) (corresponding to sampler flow rates of 6–10 scfm [10–17 m^3 h^{-1}]). Figure 1 summarizes data
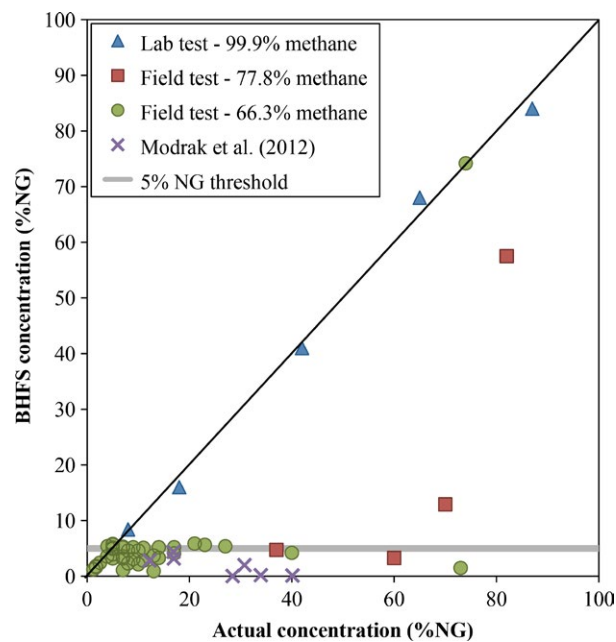

Figure 1. Occurrence of sensor transition failure in BHFS instruments with natural gas of varying CH_4 content from field and lab testing and from emission measurement studies (data from [10, 18]). NG concentrations in the BHFS sampling system measured by the BHFS internal sensor are compared to independent measurements of the sample NG concentrations. The 5% NG sample concentration threshold is the approximate concentration above which sensors should transition from catalytic oxidation to thermal conductivity. BHFS, Bacharach Hi-Flow Sampler; NG, natural gas.

showing the occurrence of sensor transition failure in several BHFS instruments during both field and laboratory testing as well as an example of the failure that occurred during an emission measurement study [10, 18].

Figure 2 presents the BHFS emission measurements from [1] as a function of percent CH_4 in wellhead gas at each site. Figure 2 also shows a line corresponding to emission rates of 0.3–0.5 scfm (0.5–0.9 m^3 h^{-1}), which represents the range of emission rates that would require transition from the catalytic oxidation sensor to the thermal conductivity sensor at sample flows ranging from 6 to 10 scfm (10–17 m^3 h^{-1}).

As seen in Figure 2, there are very few measurements in the thermal conductivity sensor range (above ~0.4 scfm [0.7 m^3 h^{-1}]) at sites where the wellhead gas composition of CH_4 is less than 91%, and this is true across all source categories. Raw data for sample flow and concentration from the BHFS were not provided in [1] supplemental information, so for this analysis, an average BHFS sample flow rate of 8 scfm (14 m^3 h^{-1}) has been assumed, which is the lower of the two sampling flows specified by the Bacharach operating manual [4]. At this sample flow rate, an emission source of 0.4 scfm (0.7 m^3 h^{-1}) corresponds with a sample concentration of 5% NG in air, above

which point the sampler would need to transition to the thermal conductivity sensor to allow for accurate measurements. For sites with CH_4 concentrations less than 91%, only four out of 259 measurements (1.5%) exceeded 0.4 scfm (0.7 m^3 h^{-1}), while for sites with CH_4 concentrations greater than 91%, 68 out of 510 measurements (13.3%) exceeded 0.4 scfm (0.7 m^3 h^{-1}). Consequently, there were almost nine times fewer measurements in the thermal conductivity range at sites with wellhead gas compositions of <91% CH_4 (Fig. 2). If the sample flow rate were 6 scfm (10 m^3 h^{-1}) (due to a flow restriction or reduced battery power), the threshold for transition to the thermal conductivity range would be 0.3 scfm (0.5 m^3 h^{-1}); this would still mean that there were almost seven times fewer measurements in the thermal conductivity range at sites with wellhead gas compositions of <91% CH_4 than at sites with >91% CH_4. Although it is well known that a small percentage of NG emission sources account for most of the total emissions from any given population [9, 15, 25], it is unlikely that almost all the significant emitters at NG production sites would occur only at sites with well head gas compositions >91% CH_4. It is also unlikely that the emission rates of all of the source categories surveyed by [1], which had diverse emission mechanisms such as equipment leaks, pneumatic controllers, chemical injection pumps, and tanks, would all have a ceiling of ~0.4 scfm (0.7 m^3 h^{-1}) at sites with lower wellhead gas CH_4 concentrations. Consequently, the low occurrence of high emitters at sites with lower wellhead gas CH_4 concentrations in [1] indicates that sensor transition failure occurred at sites with CH_4 content <91% and is consistent with the BHFS sensor failure found by Howard et al. [10].

Alternative Theories for the Emission Rate Pattern

Other possible causes of the emission rate pattern in the UT BHFS measurements were considered, including: regional operating differences at production sites; lighter gas densities resulting in higher emission rates; and improved detection of emissions by auditory, visual, and olfactory (AVO, e.g., [24]) methods at sites with heavier hydrocarbon concentrations.

Regional operating differences

Allen et al. [1] point out that air pollution regulations in Colorado which required installation of low bleed pneumatic devices in ozone nonattainment areas after 2009 might have led to lower emission rates in the Rocky Mountain region, which also had the lowest average concentration of CH_4 in the wellhead gas. However, if the
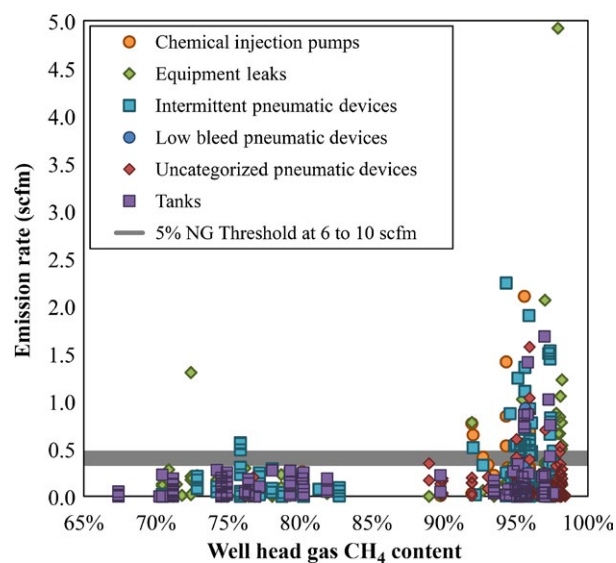

Figure 2. Emission rates of various sources measured by BHFS at NG production sites versus CH_4 concentration of the wellhead gas (data from [1]). The solid line indicates the maximum emission rate that could be measured by the catalytic oxidation sensor only (i.e., in the case of sensor transition failure). For sites with a NG composition greater than 91% CH_4, 13.3% of the measurements are in the TCD sensor range, assuming a sampler flow rate of 8 cubic feet per minute. For sites with less than 91% CH_4, only 1.5% of the measurements are in the TCD range. BHFS, Bacharach Hi-Flow Sampler; NG, natural gas; TCD, thermal conductivity detector.

Rocky Mountain region is removed from the analysis, the occurrence of emitters >0.4 scfm (0.7 m^3 h^{-1}) at sites with wellhead gas <91% CH_4 was still only four out of 129 measurements (3.1%), while for sites with CH_4 concentrations greater than 91%, there remain 68 out of 510 measurements (13.3%) that exceeded 0.4 scfm (0.7 m^3 h^{-1}) (there were no Rocky Mountain sites with CH_4 >91%). Consequently, even if the Rocky Mountain region is removed from consideration, the occurrence of emitters >0.4 scfm (0.7 m^3 h^{-1}) was almost four times less at sites with less than 91% CH_4 than at sites with greater than 91% CH_4, so air quality regulations in Colorado do not appear to be the cause of the emission rate trend shown in Figure 2.

Beyond air pollution regulations, other unknown regional operating practices unrelated to CH_4 concentration might coincidentally cause the apparent relationship of site CH_4 concentrations with the occurrence of high emitters. However, as shown in Figure 3, the increase in leaks >0.4 scfm (0.7 m^3 h^{-1}) directly correlates with the increase in the average regional CH_4 concentration. Because there are four regions and two variables (site CH_4 concentration and the percent of leaks >0.4 scfm [0.7 m^3 h^{-1}]), the likelihood that regional operating characteristics would coincidentally cause the increase in occurrence of leaks >0.4 scfm (0.7 m^3 h^{-1}) to mirror the increasing regional site CH_4 concentration is only one in 24 (four factorial), or ~4%.

Other known operating characteristics of the regions, such as average site pressure and average site age, are not related to the occurrence of equipment leaks >0.4 scfm (0.7 m^3 h^{-1}): average site pressures show no correlation, and average site age is negatively correlated with the occurrence of equipment leaks >0.4 scfm (0.7 m^3 h^{-1}).

Another argument against regional differences comes from the air quality study conducted by the City of Fort Worth ([6]; or the Ft. Worth study). Ft. Worth is part of the Mid-Continent region defined by [1], where the occurrence of equipment leaks only (as opposed to all BHFS measurement categories) >0.4 scfm (0.7 m^3 h^{-1}) observed by [1] was 2.0% of the total equipment leaks in that region. However, equipment leaks >0.4 scfm (0.7 m^3 h^{-1}) were 9.9% of the equipment leaks measured in the Ft. Worth study. This was determined using the Ft. Worth study categories of valves and connectors; their remaining category of "other", which included pneumatic control devices, had an even higher occurrence of sources >0.4 scfm (0.7 m^3 h^{-1}) of 27.0%. Previous work [10] has shown that although sensor transition failure likely occurred in the Ft. Worth study, these incidents were limited compared to those in [1]. Consequently, the much lower occurrence of leaks >0.4 scfm (0.7 m^3 h^{-1}) in the Mid-Continent region in [1] compared to the Ft. Worth study indicates that sensor transition failure was responsible for the low occurrence of emitters <0.4 scfm (0.7 m^3 h^{-1}) as opposed to regional differences.

Gas density

Wellhead gas with a lower CH_4 and a greater heavier hydrocarbon content will be denser than gas with higher CH_4 content. Since gas flow through an opening is inversely related to the square root of the gas density, streams with lower CH_4 content would have a lower flow rate if all other conditions were the same. However, this would cause at most a 20% decrease for the lowest CH_4/highest heavier hydrocarbon streams compared to the highest CH_4/lowest heavier hydrocarbon streams observed in the UT study. This would also result in a gradual increase in emissions as CH_4 content increased, as opposed to the dramatic increase in emissions observed over a very narrow range of CH_4 concentrations (Fig. 2).

AVO detection

AVO methods might improve for gas streams with a greater proportion of heavier hydrocarbons, since those streams would have greater odor and might leave more visible residue near a leak. However, Figure 4 presents the occurrence of emitters >0.4 scfm (0.7 m^3 h^{-1}) as a function of site CH_4 concentrations in the Appalachia
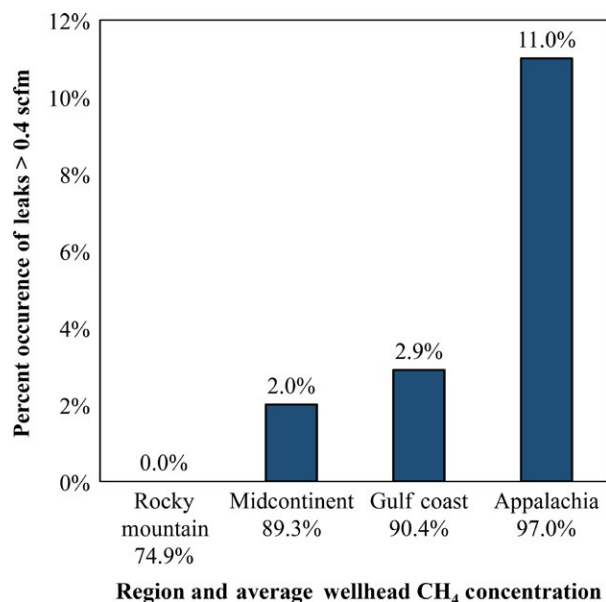
Figure 3. Occurrence of equipment leaks >0.4 scfm in each region of the [1] equipment leak data set. The odds of the occurrence of leaks >0.4 scfm being positively correlated with site CH_4 concentration are one in 24, which makes it unlikely this trend is due to regional operating effects.
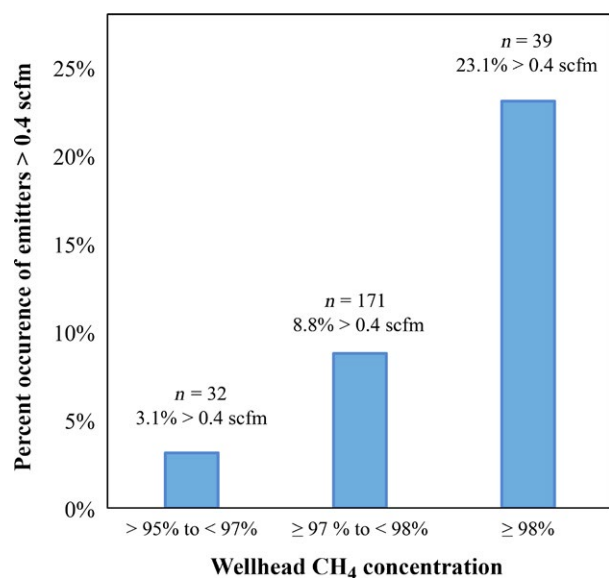
Figure 4. Occurrence of emitters >0.4 scfm as a function of site wellhead gas composition in [1] for the Appalachia region. An emission rate of greater than 0.4 scfm would require the transition from catalytic oxidation sensor to the thermal conductivity sensor for an average sample flow rate of 8 scfm. The dramatic increase in emitters >0.4 scfm over a narrow concentration range argues against the possibility that auditory, visual, and olfactory leak detection is the cause of the emission rate pattern seen in the [1] data set.

region alone. This region had the highest average CH_4 concentration in wellhead gas of any of the regions sampled in [1]. As seen in Figure 4, even over a very narrow range of site CH_4 concentrations (from 95% to >98% CH_4), there is a dramatic increase in emitters >0.4 scfm (0.7 m^3 h^{-1}) with increasing CH_4 concentration. It is unlikely that AVO methods would become so much more efficient over such a narrow range of high CH_4 concentrations where the gas streams are likely odorless and would leave little residue. This dramatic increase in high emitters at sites with high CH_4 concentrations within the Appalachia region alone also argues against the previously discussed regional operating differences hypothesis in general, since this trend is within a single region. Additionally, although the Rocky Mountain region surveyed by UT [1] had the lowest average site CH_4 concentration (74.9%) and heaviest hydrocarbon content, it actually had the highest number of equipment leaks (of any size) per well of all the regions, and there were 25% more leaks per well in that region than in the Appalachia region, which had the highest average site CH_4 concentration (97.0%) and therefore the lowest heavier hydrocarbon content. If AVO methods were more effective due to the presence of heavier hydrocarbons, it seems unlikely the region with the heaviest hydrocarbon concentrations would have the highest rate of overall leak occurrences.

Field Testing of the UT BHFS

Because the trend in the [1] data was consistent with sensor transition failure in the BHFS and no other explanation seemed plausible, I partnered with UT to test the sampler used by [1]. During that field program, the UT sampler had a version of firmware earlier than version 3.03, and older firmware versions have been shown to exhibit sensor transition failure [10]. However, the possible effect of the sampler's firmware version on the sensor failure was not known before this testing of the UT sampler, and at the time of my testing its firmware had been upgraded to a custom version (3.04).

As previously explained, the BHFS uses a catalytic oxidation sensor to measure sample stream concentrations from 0% to ~5% NG, and a thermal conductivity sensor for concentrations from ~5% to 100% NG. The catalytic oxidation sensor is typically calibrated with 2.5% CH_4 in air and the thermal conductivity sensor is calibrated with 100% CH_4 [4]. The manufacturer recommends sensor calibration every 30 days, a process which adjusts the response of the instrument. The calibration may also be checked ("bump-tested") periodically by the user, which does not adjust the instrument response. It is important to note that the description of the BHFS sensor operation in the supplemental information of [1] is incorrect, as they state that:

[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 0.05–100% 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.

However, the BHFS manual [4] clearly states that the catalytic oxidation sensor is used to measure concentrations from 0% to 5% CH_4 and the thermal conductivity sensor from 5% to 100% CH_4. This is a critical distinction because understanding that the BHFS uses a different sensor for each range and that it must transition from the catalytic oxidation sensor to the thermal conductivity sensor in order to conduct accurate measurements is critical to understanding the problem of sensor transition failure.

I initially conducted field testing of the UT sampler in conjunction with the UT team at a NG production site with a wellhead gas CH_4 concentration of 90.8%. NG composition analysis (via gas chromatograph-flame ionization detector) of wellhead gas at this site was conducted by the host company just prior to the sampler testing. The tests were conducted by metering known flow rates of NG into the BHFS inlets through a rotameter (King Instrument Company, Garden Grove, CA; 0–10 scfm air

scale). The sample concentration indicated by the internal BHFS sensor was recorded and compared to an external gas concentration monitor used to measure the actual NG concentration at the sampler exhaust (Bascom-Turner Gas Sentry CGA 201, Norwood, MA). The Gas Sentry unit was calibrated with 2.5% and 100% CH_4 prior to the testing; exhaust concentrations measured using this unit agreed with concentrations calculated using the sampler flow rate and amount of NG metered into the inlet to within an average of ±6%.

This field testing was conducted in March of 2014 and is described by [10]; the UT sampler is identified therein as BHFS No. 3. At the time of this testing, the UT BHFS had firmware Version 3.04 (September 2013); this sampler had been calibrated 2 weeks prior to the field test and had been used for emission measurements at production sites since that time. The response of the sensors was checked ("bump-tested") by the UT field team but not calibrated prior to the start of testing. This was apparently consistent with the UT field program methodology: the sampler had been used for measurements with only sensor bump tests, but without the actual calibration unless the sensors failed the bump tests (as was acceptable according to the manufacturer's guidelines) during their ongoing field measurement program and was provided to me for these measurements "ready for testing".

Although the UT sampler's internal sensors initially measured the sample concentration correctly, after ~20 min of testing the sampler's sensors failed to transition from the catalytic oxidation scale (<5% NG) to the thermal conductivity scale (>5% NG), resulting in sample concentration measurements that were 11–57 times lower than the actual sample concentration (Fig. 5). Because sample concentration is directly used to calculate emission measurements made by the sampler, this would result in emission measurements that are too low. After this sensor transition failure occurred, the UT BHFS was calibrated (not simply "bump-tested") and thereafter did not exhibit any further sensor transition failures even during a second day of testing at sites with wellhead CH_4 concentrations as low as 77%. Two other BHFS that were not part of the UT program were also tested using the same procedure; these instruments had the most updated firmware commercially available (Version 3.03) and were put through an actual calibration sequence by the instrument distributor's representative prior to any testing. Neither of these instruments exhibited sensor transition failure at any of the sites. These results combined with the sensor transition failure previously observed in instruments with earlier versions of firmware suggest that the combination of updated firmware and frequent actual calibrations might reduce sensor failure, although this has not been proved conclusively [10, 11].
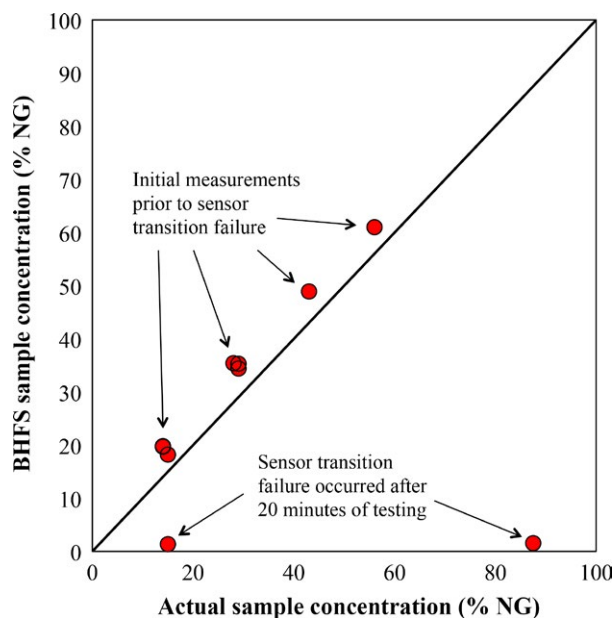

Figure 5. Performance of the BHFS used during the [1] study with NG composed of 90.8% CH_4; instrument firmware had been upgraded to version 3.04 after that study but before this testing; calibration was 2 weeks old. Sensor transition failure set in after ~20 min of testing; this failure was eliminated once the BHFS was put through a calibration sequence (as opposed to just a response test). BHFS, Bacharach Hi-Flow Sampler; NG, natural gas.

The UT recently published a follow-up study of pneumatic device emissions [2]. As part of this work, Allen et al. [2] conducted laboratory testing of the UT BHFS by making controlled releases of both 100% CH_4 and a test gas of 70.5% CH_4 mixed with heavier hydrocarbons into the UT BHFS and did not report any sensor transition failures during these tests, but during this laboratory testing the sampler (with the updated firmware version 3.04) was calibrated (not 'bump-tested") immediately prior to any testing. Consequently, the absence of sensor failure during their laboratory testing is consistent with the results observed during the March 2014 field tests, where calibrating the instrument eliminated the sensor failure.

Allen et al. [3] have suggested that the protocol during their field campaign was to check the calibration of the UT BHFS anytime it was turned on and that not following this protocol led to the sensor transition failure observed during this testing. However, in this instance, the sensor failure occurred both prior to and after the instrument was restarted. Additionally, the UT team observing the testing process did not suggest a calibration check when the instrument was turned back on for further testing. It was only after the sensor failure was observed that they checked and calibrated the instrument, so it

does not appear that their protocol was to check the instrument calibration anytime it was turned on.

In summary, because the firmware for the UT sampler was updated prior to this testing (and therefore not the same as the version used during the UT field campaign [1]), and updated firmware may be a factor in reducing sensor failure, it is not expected that these test results are representative of how frequently sensor transition failure might have occurred during the UT study [1]. However, these results do clearly demonstrate that sensor transition failure could occur while using the UT BHFS.

Comparison With Other Pneumatic Device Studies

Two other recent studies have measured emission rates from pneumatic devices by installing meters into the supply gas lines of the devices, as opposed to measuring emissions using the BHFS as was done by Allen et al. [1]. Prasino [22] used the meter installation technique to study emissions from pneumatic controllers in British Columbia, and the UT follow-up study [2] installed meters to measure emission rates from pneumatics in the four regions surveyed in the previous UT study [1].

Unfortunately, it is not possible to compare the pneumatic device emission factors from [1] to those from either the Prasino study, or from [2], because even though [1] sought to randomly sample pneumatic devices, the result was clearly an emitter data set (measurements focused on pneumatic devices that were emitting), while the Prasino data set was made with a random selection of devices and [2] made comprehensive measurements of all devices that could be measured safely at each site. This difference can be demonstrated by comparing the percentage of emitting intermittent pneumatic devices occurring in [1] to that in [2]. In [1], 95.3% (123 out of 129 intermittent devices) were greater than zero, with the smallest nonzero emitter equal to 0.12 scfh (0.0034 m^3 h^{-1}). In [2], only 57.5% (184 out of 320 intermittent devices) were greater than zero. This percentage of nonzero measurements drops further if the lowest nonzero emitter (0.12 scfh; 0.0034 m^3 h^{-1}) observed by [1] is used as a threshold, in which case only 21.3% (68 out of 320) would be considered emitters. Since this threshold of 0.12 scfh (0.0034 m^3 h^{-1}) is 25 times lower than the typical minimum range of the Fox FT2A meters by [2], the reported emitters below this threshold are most likely instrument noise caused by the meter's thermal elements inducing convection currents [7].

Consequently, although the intent of [1] was to survey randomly selected devices, their approach actually resulted in a data set comprised almost exclusively of emitting devices; this possibility is acknowledged by [2]. Therefore,

average emissions and emission factors for pneumatic devices calculated from [1] cannot be compared to those calculated from data collected by random or comprehensive sampling, such as presented in [22] or [2], because the emitter data set removes almost all the zero emitters and would result in much higher average emissions.

However, both [1] and [2] provide the CH_4 composition of the wellhead gas at the sites surveyed. This allows a comparison of emission rate patterns as a function of CH_4 concentration between devices measured by the BHFS [1] and by installed meters [2]. If the scarcity of high emitters measured by BHFS at sites with lower CH_4 concentrations in the initial UT study [1] was not an artifact caused by sensor transition failure, then the same concentration pattern should be present whether measured by the BHFS or by installed meters.

For this analysis, I removed the Rocky Mountain region to eliminate any bias from current or impending regulations that might have affected emission rates. Additionally, I focused on emissions from intermittent pneumatics because that provides the most complete data set from the two studies. Finally, as noted previously, the pneumatic device measurements from [1] apparently focused on emitting devices, whereas the devices surveyed in [2] were sampled as comprehensively as possible so the occurrences of high emitters in each study cannot be directly compared. Consequently, it is the ratio of the occurrences of high emitters at low CH_4 sites compared to high CH_4 sites within each study that must be compared.

As seen in Table 1, when measured by [1] via BHFS, the occurrence of emitters >0.4 scfm (0.7 m^3 h^{-1}) (on a percentage basis) at sites with wellhead gas compositions <91% CH_4 is almost a factor of five less than at sites with CH_4 >91%, consistent with BHFS sensor failure. Conversely, when measured via installed meters [2], the occurrence of emitters >0.4 scfm (0.7 m^3 h^{-1}) at sites with wellhead gas compositions <91% CH_4 is almost a factor of three higher than at sites with >91% CH_4, indicating a complete reversal in this trend. This stark difference between BHFS measurements and installed meter measurements corroborates that the scarcity of high emitters at sites with lower wellhead gas CH_4 content present in [1] was an artifact due to sensor failure in the BHFS.

## Focused Analysis of the UT Study Equipment Leaks

In order to better understand the threshold of wellhead gas CH_4 concentrations at which sensor transition failure might occur, I conducted further analysis focused only on the equipment leak measurements in [1]. Equipment leaks were targeted because they are expected to be short term, steady state measurements, whereas emissions

Table 1. Occurrence of intermittent pneumatic device high emitters as a function of wellhead gas composition, measured by Bacharach Hi-Flow Sampler (BHFS) and installed meters (Rocky Mountain region excluded).

	No. of devices measured	No. of devices with emissions >0.4 scfm	% of devices with emissions >0.4 scfm
Allen et al. [1] (Measured by BHFS sampler)			
Wellhead gas composition >91% CH_4	85	28	32.9
Wellhead gas composition <91% CH_4	44	3	6.8
Ratio of frequency of high emitters at sites with wellhead gas compositions <91% CH_4 to sites with wellhead gas compositions >91% CH_4			0.21
Allen et al. [2] (Measured by installed meters)			
Wellhead gas composition >91% CH_4	106	3	2.8
Wellhead gas composition <91% CH_4	97	8	8.2
Ratio of frequency of high emitters at sites with wellhead gas compositions <91% CH_4 to sites with wellhead gas compositions >91% CH_4			2.9

reported from pneumatic devices and chemical injection pumps are likely to be an average of several measurements, and emissions from tanks may have an NG composition different from the reported wellhead composition.

Figure 6 presents the occurrence of equipment leaks in [1] that are >0.4 scfm (0.7 m³ h⁻¹) as a function of site CH_4 concentrations. At sites with gas compositions of >97% CH_4, 11.7% of the leaks were >0.4 scfm (0.7 m³ h⁻¹). At sites with wellhead compositions between 90% and 97% CH_4, only 2.7% of the leaks were >0.4 scfm
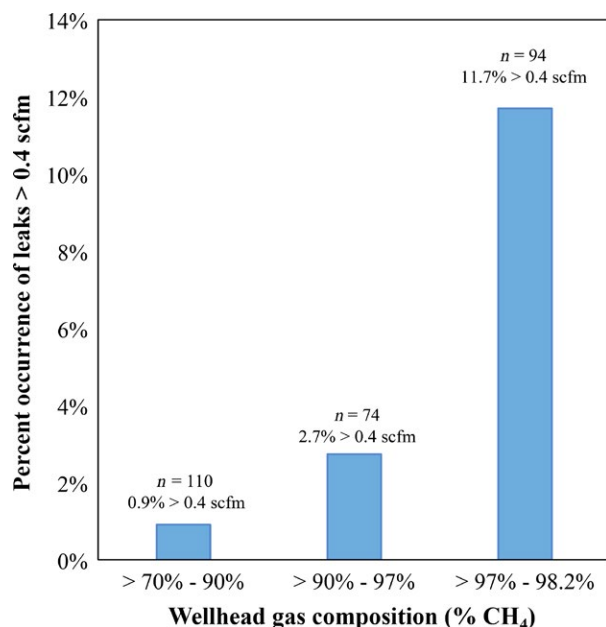
Figure 6. Occurrence of equipment leaks >0.4 scfm as a function of site well head gas CH_4 content in the [1] study. Leaks >0.4 scfm would require the transition from catalytic oxidation sensor to the thermal conductivity sensor for an average sample flow rate of 8 scfm. The large increase in the occurrence of leaks >0.4 scfm at sites with CH_4 content >97% indicates sensor transition failure below that threshold.

(0.7 m³ h⁻¹), and this occurrence dropped to less than 1% at sites with wellhead gas compositions of <90% CH_4, indicating that the sampler's ability to measure leaks >0.4 scfm (0.7 m³ h⁻¹) declined dramatically with decreasing concentrations of CH_4 in the wellhead gas (Fig. 6). This analysis indicates the BHFS may underreport emitters >0.4 scfm (0.7 m³ h⁻¹) even when making measurements of NG streams with CH_4 content up to 97%, and provides a valuable refinement of the possible CH_4 concentration threshold where sensor failure may occur, since the highest CH_4 wellhead content available for direct field testing of the BHFS was only 91.8%.

Comparison of the UT Study Downwind Tracer Ratio Measurements to On-Site Measurements

Allen et al. [1] also made emission measurements using a downwind tracer ratio method at 19 sites for comparison to their on-site measurements. Their emissions from on-site measurements were calculated by using direct measurements of equipment leaks and pneumatic devices that were made by the UT team combined with estimates of emissions from any sources at the well pad that were not measured. These unmeasured sources included all tanks and compressors (compressors were a small source in comparison to all other sources) as well as any pneumatics that was not directly measured during the site survey. For CH_4 emissions from tanks and compressors, the authors used "standard emissions estimation methods" [1]. For pneumatic devices that were not surveyed, they applied their own emission factors based on the measurements of pneumatic devices collected during the UT study.

The tracer ratio measurements were made by releasing a tracer gas at a known rate to simulate the emissions from the site being measured. Simultaneous downwind measurements were then made of the concentrations of both the tracer gas and CH_4, and then the emission rate

of CH_4 was calculated after correcting for background CH_4 and tracer concentrations. The tracer ratio method allows for the calculation of CH_4 emissions from the entire production site by accounting for the dilution of CH_4 as it is transported into the atmosphere from the source to the receptor.

In summarizing their tracer ratio measurements, [1] state: "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." However, upon further examination, I found that the downwind tracer measurements do in fact indicate the occurrence of sensor transition failure in their BHFS measurements.

Table 2 summarizes the characteristics of the sites surveyed by [1] using both the BHFS and the tracer ratio method. As described above, the on-site total is a combination of the measurements made by BHFS and estimates for any sources not actually measured by the UT team. I calculated the ratio of actual BHFS measurements to

the total reported on-site emissions (estimated and measured) using the supplemental information provided by [1]. Actual measured emissions ranged from 1% to 79% of the total reported on-site emissions and the on-site total emissions range from 13% to 3500% of the downwind tracer ratio measurements (Table 2).

Table 3 compares the tracer ratio measurements to the on-site emissions, categorized by CH_4 content in the wellhead gas and by the fraction of actual BHFS measurements that comprise the on-site emissions. As shown in Table 3, when comparing all sites without separating them into these categories, the total of the tracer ratio measurements does agree closely to the on-site emissions, as [1] concluded. However, four of the sites had wellhead gas compositions of ≥97% CH_4, at which the BHFS would be expected to make accurate measurements. The remaining 15 sites had wellhead gas compositions of <82% CH_4, at which sensor transition failure might occur and the BHFS would underreport emissions measurements.

Once the sites are categorized by these wellhead gas compositions, a deficit between the on-site emissions and the tracer ratio measurements appears in sites with lower CH_4 concentrations, and this deficit becomes more

Table 2. Sites surveyed by Allen et al. [1] using both Bacharach Hi-Flow Sampler (BHFS) and downwind tracer methods.

Tracer site name[1]	BHFS site name[1]	Wellhead gas CH_4 concentration (%)	On-site total[2] (BHFS measurements and estimates) (scfm CH_4)	BHFS measurements/on-site total[3]	Leaks measured by BHFS/on-site total[3]	Tracer ratio emission rate (scfm CH_4)	On-site total/ tracer ratio emission rate
MC-1	MC-1	70.9	1.89	0.12	0.12	2.32	0.815
MC-2	MC-14	78.1	0.99	0.34	0.01	2.00	0.495
MC-3	MC-20	77.2	1.63	0.45	0.18	2.95	0.552
MC-4	MC-5	74.2	2.31	0.19	0.14	3.36	0.687
MC-5	MC-16	79.3	1.85	0.56	0.18	4.16	0.445
RM-1	RM-7	81.9	0.22	0.11	0.09	0.584	0.368
RM-2	RM-8	74.5	4.43	0.02	0.02	1.70	2.60
RM-3	RM-1	76.4	0.13	0.67	0.69	0.442	0.303
RM-4	RM-3	74.9	0.11	0.21	0.00	0.839	0.137
RM-5	RM-2	74.5	0.09	0.35	0.33	0.240	0.392
RM-6	RM-5	74.5	0.74	0.41	0.42	0.421	1.75
RM-7	RM-14	74.5	0.27	0.26	0.26	0.368	0.736
RM-8	RM-19	76.2	0.29	0.82	0.79	1.08	0.266
RM-9	RM-12	74.5	0.38	0.05	0.05	0.864	0.436
RM-10	RM-4	76.2	2.86	0.01	0.00	0.080	35.7
AP-2	AP-23	97.6	1.28	0.68	0.35	0.270	4.74
AP-3	AP-43	97.0	4.75	0.62	0.59	4.12	1.15
AP-4	AP-37	97.0	1.36	0.44	0.42	0.709	1.92
AP-5	AP-18	97.0	0.39	0.74	0.69	0.288	1.37

[1]MC, Midcontinent; RM, Rocky Mountain; AP, Appalachia. Different site numbers were used to identify the same sites in the [1] supplemental information depending on whether BHFS or tracer ratio measurements were under discussion.
[2]On-site totals were calculated by [1] by combining measurements made by the BHFS with estimates of any sources not measured; these estimates were made using mathematical models for tanks as well as emission factors for compressors and any pneumatic controllers not directly measured.
[3]Calculated by this author from [1] supplemental information.

pronounced as the amount of the on-site emissions actually measured by the BHFS becomes a larger fraction of the total on-site emissions (measured and estimated). As seen in Table 3, for the high CH_4 sites where the sampler should function properly, the on-site measurements and estimates exceed the tracer measurements, but approach a ratio of one (complete agreement) as the amount of actual measurements increases. For the two sites with wellhead gas compositions ≥97% where the measured equipment leaks (which should produce steady emissions as compared to pneumatic devices which might be intermittent) averaged 64% of the total on-site measurements and estimates, the on-site total still exceeds the tracer measurements but are within 17% (Table 3). However, for the sites with wellhead gas CH_4 concentrations <82%, there is a clear trend of increasing deficit of the on-site emissions compared to the tracer ratio measurements as the actual BHFS measurements become a larger part of the on-site total. For instance, for the nine sites with at least 20% of on-site emissions from BHFS measurements (for an average of 45% of the total on-site emissions measured by the BHFS), the on-site emissions are only 49% of the tracer measurements (Table 3). For the two sites that had greater than 67% of on-site emissions data actually measured by the BHFS (for an average of 75% of on-site emissions data measured by the BHFS), the on-site emissions are only 28% of the tracer measurements (Table 3).

Comparing the on-site data to the downwind tracer measurements provides two valuable insights. First, there were six sites in the Rocky Mountain region for which at least 20% of the on-site emissions were measured by the BHFS (for an average of 45% actual BHFS measurements) (Table 2). For these six sites, the on-site emissions average 48% of the tracer data. For the two sites in this region with at least 67% of on-site emissions from actual BHFS measurements (and with BHFS measurements averaging 75% of the total on-site data), the on-site emissions were only 28% of the tracer measurements (Table 2). This provides clear evidence that the sampler actually did fail in the Rocky Mountain region, as opposed to any possible regional differences (discussed previously) that might have created an emission pattern of no high emitters at sites with lower CH_4 concentrations in the wellhead gas.

Additionally, the tracer measurements provide a method to estimate the magnitude of errors introduced in the data collected by [1] due to BHFS sensor transition failure. For all of the sites with wellhead gas compositions ≥97% CH_4 (where the sampler should operate correctly), the emission rates determined by on-site measurements exceeded those determined by the downwind tracer ratio measurements. Assuming that the tracer method accurately measured the total emissions from the sites surveyed (e.g., [8, 15, 16]), I concluded that the methods used in [1] overestimated the on-site sources that were not directly measured. Therefore, I calculated the error in BHFS measurements at sites with low CH_4 wellhead gas composition by assuming the tracer ratio measurements are correct. I have also assumed for this analysis that the estimates of any on-site sources made by [1] are also correct, even though the tracer data indicate they may be too high, because this is conservative in the sense that correcting for this overestimate would increase the BHFS error calculated below. Given these assumptions, subtracting the on-site estimated emissions from the tracer ratio emissions gives the expected measurement total that should have been reported from the BHFS measurements. Comparing this expected measurement total to the actual

Table 3. Comparison of on-site measurements to tracer ratio measurements made by Allen et al. [1] categorized by wellhead gas CH_4 concentration.

Site category (number of sites in parentheses)	Average percentage of on-site emissions reported by BHFS	Total on-site emissions (reported by BHFS and estimated) (scfm CH_4)	Total emissions measured by tracer (scfm CH_4)	Ratio of on-site emissions to emissions measured by tracer
All sites (19)	37	26.0	26.8	0.97
Sites where BHFS measurements are expected to be accurate (wellhead gas composition ≥97% CH_4)				
All sites (4)	62	7.78	5.39	1.44
Sites with >50% BHFS measurements (3)	68	6.42	4.68	1.37
Sites with >50% equipment leaks (2)	64 (equipment leaks/on-site total)	5.14	4.41	1.17
Sites where BHFS measurements are expected to underreport high emitters (wellhead gas composition <82% CH_4)				
All sites (15)	28	18.2	21.4	0.85
Sites with ≥5% BHFS measurements (13)	35	10.9	19.6	0.56
Sites with ≥20% BHFS measurements (9)	45	6.10	12.5	0.49
Sites with >50% BHFS measurements (3)	69	2.27	5.68	0.40
Sites with >67% BHFS measurements (2)	75	0.42	1.52	0.28

BHFS, Bacharach Hi-Flow Sampler.

measurement total reported by the BHFS provides an estimate of the error in BHFS measurements made by Allen et al. [1].

Table 4 presents the results of this analysis, and shows that for the 13 sites with wellhead gas compositions <82% CH_4 and with at least 5% actual BHFS measurements (with an average of 35% of emission sources measured by BHFS; bottom half of Table 3), the actual measurement total of the BHFS is less than one-third of the expected total, and this appears consistent as sites with greater fractions of actual BHFS measurements are examined. For these sites, the emission rates for equipment leaks and pneumatics devices presented by [1] are approximately equal, so it is not possible to assign a larger error to one category or another. Additionally, the errors introduced by the sensor failure would be expected to vary from site to site depending on how many emitters were present with emission rates exceeding the sensor transition threshold ceiling. Nevertheless, for these 13 sites, the BHFS underreported emissions for equipment leaks and pneumatic devices on average by more than a factor of 3 (Table 4).

Although the magnitude of error due to BHFS sensor failure is not known for all the sites in [1], the tracer ratio measurements make clear that the BHFS measurements for sites with lower CH_4 content in the wellhead gas could be at least a factor of three too low. More precise estimates of errors in [1] are not possible because of the nature of the sensor failure. Unlike a simple calibration error, for which it might be possible to correct, when sensor transition failure occurs, it is not possible to know for any particular measurement if the failure has occurred, and if it has, what the resulting error was, since the reported emission rates could range from 20% to two orders of magnitude too low.

Implications

Sensor transition failure is clearly apparent in the BHFS measurements made in the UT study by Allen et al. [1], as evidenced by the rare occurrence of high emitters at sites with lower CH_4 (<91%) content in the wellhead gas. The occurrence of this sensor transition failure was corroborated by field tests of the UT BHFS during which it exhibited this sensor failure, as well as by tracer ratio measurements made by [1] at a subset of sites with lower wellhead gas CH_4 concentrations. At this subset of sites, the tracer ratio measurements indicate that the BHFS measurements were too low by at least a factor of three. Because BHFS measurements were the basis of 98% of the inventory developed by [1] using their own measurements (and 41% of their total compiled inventory), the inventory clearly underestimates CH_4 emissions from production sites. However, the extent of this error is difficult to estimate because the underreporting of emission rates due to BHFS sensor transition failure at any given site would vary depending on sampler performance and on how many high emitters were present at that site. Estimating this error is further complicated by the fact that the data set collected for pneumatic devices by [1] was an emitter data set; this might offset the effect of underreported high emitters in their pneumatic device emission factors. Finally, although real differences may exist in regional emission rates, the UT data set [1] should not be used to characterize them because the occurrence of sensor failure clearly varied between regions due to variations in wellhead CH_4 compositions, which may mask any actual regional differences that existed.

Although the performance of the BHFS may vary between instruments or with sensor age or calibration vintage, this analysis of the [1] data set shows that measurements made using a BHFS for NG streams with CH_4 content

Table 4. Estimation of underreporting in Allen et al. [1] BHFS measurements of CH_4 emission rates at sites with low CH_4 well head gas composition (<82%), using downwind tracer measurements (from Table 3).

Minimum percentage of on-site emissions reported by BHFS	Average percentage of on-site emissions reported by BHFS	No. of sites	Total emissions measured by tracer (scfm CH_4)	On-site emissions estimated by UT (excludes BHFS measurements) (scfm CH_4)	Expected BHFS measurement total (tracer – on-site estimates) (scfm CH_4)	Emissions reported by BHFS (scfm CH_4)	Ratio of reported BHFS to expected BHFS
≥5	35	13	19.63	7.09	12.54	3.81	0.30
≥20	45	9	12.50	3.34	9.16	2.76	0.30
>50	69	3	5.68	0.71	4.97	1.56	0.31
>67	75	2	1.52	0.11	1.42	0.31	0.22

BHFS, Bacharach Hi-Flow Sampler; UT, University of Texas.

up to 97% could lead to severe underreporting of NG leaks. That this failure can occur at such high CH_4 concentrations, which are close to the higher end of those found in transmission and distribution systems, indicates that past measurements in all segments of the NG supply chain could have been affected by this problem. Because the BHFS sensor transition failure phenomenon is not fully understood, it is not known how much this error may have affected past measurements of CH_4 emission rates. Two factors preclude this: first, the performance of any individual BHFS may vary, and second, once sensor transition failure occurs, there is no way to determine the magnitude of the measurement error in the absence of an independent flux or concentration measurement.

If BHFS sensor transition failure has occurred during industry monitoring at transmission, storage, and processing compressor stations where the BHFS is approved for leak measurements mandated by the USEPA Subpart W Greenhouse Gas Reporting Program (GHGRP) [23], then these errors could be larger than those observed at production sites. Leaks at transmission, storage, and processing compressor stations commonly exceed 0.4 scfm (0.7 m^3 h^{-1}) (the approximate threshold for BHFS sensor transition failure) and in some cases may range from 10 to over 100 scfm. Because the largest 10% of leaks typically account for 60–85% of the total leak rate at a given facility [9, 25], sensor transition failure in the BHFS could bias CH_4 emission inventories compiled by the USEPA GHGRP substantially low since the most significant leaks could be underreported. Additionally, leak measurements using the BHFS may be used to guide repair decisions at NG facilities, and underreporting of leaks could compromise safety if large leaks remain unrepaired as a result.

Finally, it is important to note that the BHFS sensor failure in the UT study [1] went undetected in spite of the clear artifact that it created in the emission rate trend as a function of wellhead gas CH_4 content and even though the authors' own secondary measurements made by the downwind tracer ratio technique confirmed the BHFS sensor failure. That such an obvious problem could escape notice in this high profile, landmark study highlights the need for increased vigilance in all aspects of quality assurance for all CH_4 emission rate measurement programs.

Acknowledgments

Conflict of Interest

The author is the developer of high flow sampling technology (US Patent RE37, 403) and holds a license to use it for any purpose; however, he does not sell high flow samplers nor was he involved in the development of the Bacharach Hi-Flow Sampler.

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