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Air Impacts of Increased Natural Gas Acquisition, Processing, and Use: A Critical Review

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ABSTRACT: During the past decade, technological advancements in the United States and Canada have led to rapid and intensive development of many unconventional natural gas plays (e.g., shale gas, tight sand gas, coal-bed methane), raising concerns about environmental impacts. Here, we summarize the current understanding of local and regional air quality impacts of natural gas extraction, production, and use. Air emissions from the natural gas life cycle include greenhouse gases, ozone precursors (volatile organic compounds and nitrogen oxides), air toxics, and particulates. National and state regulators primarily use generic emission inventories to assess the climate, air quality, and health impacts of natural gas systems. These inventories rely on limited, incomplete, and sometimes outdated emission factors and activity data, based on few measurements. We discuss case studies for specific air impacts grouped by natural gas life cycle segment, summarize the potential benefits of using natural gas over other fossil fuels, and examine national and state emission regulations pertaining to natural gas systems. Finally, we highlight specific gaps in scientific knowledge and suggest that substantial additional measurements of air emissions from the natural gas life cycle are essential to understanding the impacts and benefits of this resource.

INTRODUCTION

Natural gas currently accounts for 26% of primary energy consumption in the U.S., compared to 20% for coal and 36% for petroleum and other liquids.[1] Although the percentage of U.S. energy obtained from natural gas is expected to rise modestly to 28% during the next 30 years, the production of natural gas is expected to increase to the point where the U.S. will be a net exporter of natural gas by 2020.[1] A decrease in conventional on-shore gas production since the 1980s has been the impetus in the U.S. for developing unconventional natural gas plays (areas targeted for exploration and production) that have low permeability—such as sandstones (tight-sand gas), shales (shale gas), and coal (coal-bed methane).[1] Between 2000 and 2011, the share of U.S. natural gas production from unconventional formations increased from 31% to 67% and is expected to reach 80% by 2040.[1] In particular, annual shale gas production is expected to double from 7.9 trillion cubic feet (Tcf) in 2011 to 16.7 Tcf by 2040.[1]

Between 2000 and 2011, the number of producing gas wells in the U.S. increased by 50%,[2] reaching 514 637. This surge in exploration and production from unconventional sources has been accompanied by public concerns about various environmental issues—including air quality, water quantity and quality, and human health impacts.[3−9] Moreover, with this fast-moving industry, scientists have been struggling to obtain adequate funding and data access for research studies, and regulators have been grappling with the development of new rules and policies along with limited resources for enforcement during the surge in drilling.[7,10,11] Decision and rule making at the state and national levels in the U.S. have been informed in part by limited, out of date, and sometimes incomplete emission inventories[11] and self-reported industry data. Further confounding the ability to adequately assess the industry's environmental impacts are a number of other factors including (1) a lack of independent field measurements to evaluate assumptions, quantify risks, and assess actual impacts; (2) contradictory scientific results; and (3) polarizing political and sociological dichotomies (i.e., jobs vs environmental stewardship).

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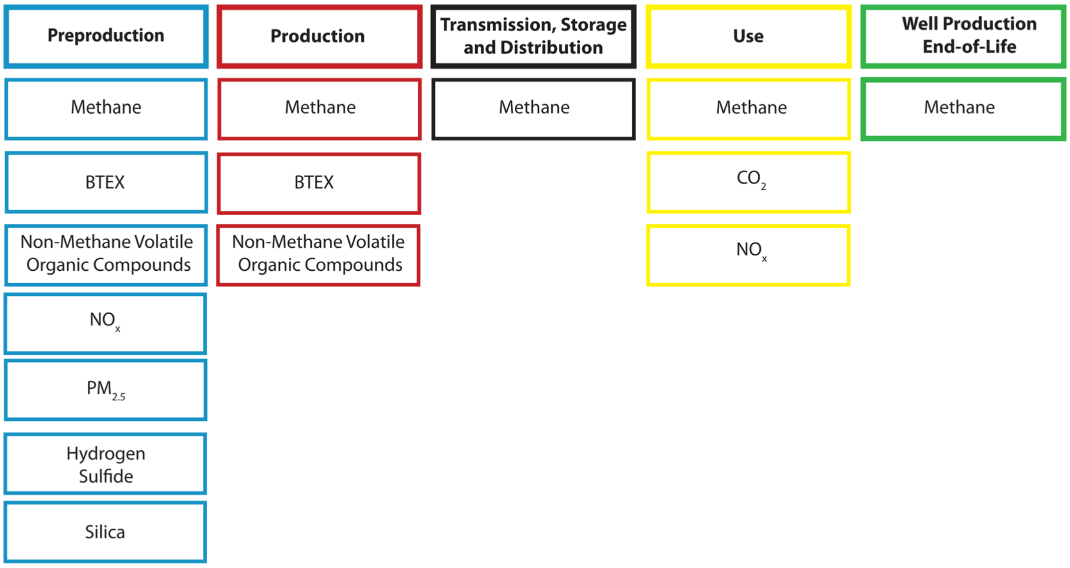

Figure 1. Potential species emitted to the atmosphere during specific stages of the natural gas life cycle.

To lay the foundation for a clear, concise discussion of the issues, we begin by defining a consistent vocabulary. Unconventional oil and natural gas development in general is often referred to as "fracking".[12,13] Instead, we separate the process of drilling, often undertaken 1−2 km horizontally and kilometers underground, from the more scientifically accurate term "hydraulic fracturing", which describes the process of fracturing low permeability rocks using water mixed with sand and proprietary chemicals pumped into the borehole under high pressure.[7,12,14] Hydraulic fracturing originated in the 1940s, but the pressures and volumes used today are much higher than in the past. The process of hydraulic fracturing typically lasts only a few days to a few weeks.[15,16] Both unconventional and conventional natural gas wells typically produce commercially for a few decades.[17] Therefore, a true evaluation of the air quality impacts of natural gas production and use must expand to all areas of the natural gas life cycle.

Throughout this critical review, we will refer to five stages of the natural gas life cycle using the terminology of Branosky et al.:[18] (1) preproduction; (2) natural gas production; (3) natural gas transmission, storage, and distribution; (4) natural gas end-use; and (5) well production end-of-life (Figure 1). In terms of the life cycle, unconventional natural gas differs from conventional natural gas in three main ways. First, extraction of unconventional natural gas often requires directional or horizontal drilling. Second, well-completion (hydraulic fracturing) procedures for unconventional natural gas are much more extensive than for conventional wells. Third, unconventional natural gas wells typically have a sharper production decline curve and a less well constrained total volume of natural gas recovered per well (based on both economical and practical constraints).[19,20] Once out of the ground, however, unconventional natural gas is subject to the same fate (e.g., processing, transport, end-use) as conventional natural gas, and the atmospheric impacts are indistinguishable between the two forms.

Much of the earlier scientific work on unconventional natural gas has focused on evaluating the potential climate impacts and benefits of developing unconventional natural gas reservoirs and switching from coal or oil burning to using natural gas.[21] These studies typically focus on climate forcing impacts and their conclusions range from small benefits (<6% greenhouse gas reduction) for the switch to unconventional from conventional natural gas, to potentially large benefits (>30% greenhouse gas reduction) for the switch to natural gas over coal[22] for power generation. The air-quality benefits of switching from coal to natural gas are extensive for pollutants such as mercury and sulfur dioxide (SO_2). These benefits may be less so for nitrogen oxides (NO_x), important ozone precursors for which life cycle emissions appear to be similar for natural gas and coal[23,24] unless natural gas combined-cycle (use of two heat engines) technology is used to generate electricity.[25]

When possible, we will distinguish between conventional and unconventional natural gas in this review, which is organized into five sections. In the first section, we present a review of studies on methane (CH_4) leakage from the entire natural gas life cycle. The second section includes a synthesis of available studies on the non-methane air quality impacts of natural gas, which include emissions of the hazardous air pollutants benzene; toluene; ethylbenzene; and xylenes (BTEX); other non-methane volatile organic compounds (NMVOCs) and NO_x, both precursors of surface ozone; and particulate matter. We summarize the current understanding of the benefits and impacts of switching from coal or oil to gas in the third section. In the final two sections, we discuss current air emission regulations at the state and national levels and identify key areas for future research on the air quality impacts of unconventional natural gas.

■ ESTIMATES OF LIFE CYCLE METHANE LEAKAGE FROM NATURAL GAS

As the primary chemical constituent of natural gas (70−90% by volume for raw natural gas from the well and >90% by volume for pipeline quality natural gas),[26,27] CH_4 can alter global atmospheric chemistry and is a powerful greenhouse gas.[28] Combined, natural gas systems are the highest emitters of CH_4 of any anthropogenic sector in the U.S.[30] and may be partially responsible for a renewed increase in global CH_4 levels since 2006.[28,31] CH_4 is an important atmospheric constituent in that it has been shown to influence background ozone concentrations at the Earth's surface,[32] although it reacts very slowly in the lower atmosphere (8–9 year global average lifetime). The Fifth Assessment of the Intergovernmental Panel on Climate Change (IPCC) estimates that CH_4 has a global warming potential 28–34 times that of CO_2 over a 100-year time frame and 84–86 times greater on a 20-year time frame.[33] Surface level CH_4 in the global atmosphere is about 1.8 ppm, making it the second largest contributor (after CO_2) to the total direct radiative forcing due to long-lived greenhouse gases.[34]

Raw natural gas produced from wells distributed across a basin is gathered via a network of pipelines and compressor stations. It then is processed at centralized plants to remove contaminants, such as water and acids, and to separate CH_4 from natural gas liquids and condensate or oil. Processed natural gas that enters the pipeline distribution network for consumers is comprised primarily of CH_4 and ethane (C_2H_6), with the addition of an odorant, mercaptan, to help customers detect leaks in their homes or neighborhoods. C_2H_6 is left in the natural gas stream, at typically ~5%, to maintain the minimum energy content of the gas. Its lifetime in the atmosphere is much shorter than that of CH_4, typically only a few months.

Each year since 1998,[35] the U.S. Environmental Protection Agency (US EPA) has released an updated national inventory (NI) of greenhouse gas (GHG) sources and sinks and submitted it to the United Nations Framework Convention on Climate Change. National estimates for CH_4 emissions from natural gas systems are modeled and calculated annually from 1990 to two years prior to the release year based on 80 different emission factors (emissions per unit process or component) determined from direct measurements made at ~200 sites in the early 1990s.[11,30,36] Additional emissions or activity data for the estimates are supplied by states and the industry.[37,38]

Uncertainties in this inventory approach are illustrated by a series of methodological changes that US EPA implemented during the past four years to estimate CH_4 emissions from natural gas systems[30,39] (Figure 2). Based on the US EPA approach, leakage estimates for natural gas across the entire life cycle ranged from as high as 2.8% of domestic natural gas production (2011 and 2012 GHG NI releases) to as low as 1.65% in the 2013 US EPA GHG NI release (6.9 million metric tons lost out of 418 million metric tons CH_4 produced[29]). This

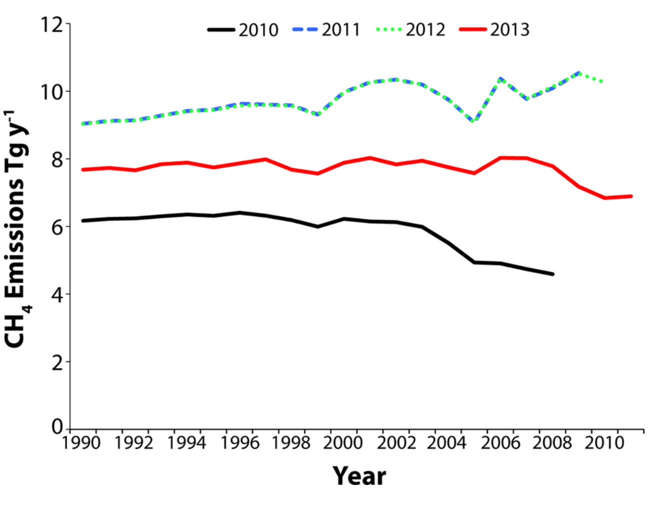

Figure 2. U.S. natural gas systems methane emission estimates from 1990 to present based on the 2010, 2011, 2012, and 2013 releases of the US EPA GHG NI.

range in values is important because an analysis by Alvarez et al.[40] concluded that CH_4 leakage of 3.2% or less would provide immediate net climate benefits for electricity production from natural gas compared to coal.

Two recent scientific studies have found that U.S. total CH_4 emissions are underestimated in current inventories.[41,42] Miller et al.[41] published a top-down estimate of CH_4 emissions in the U.S. based on long-term aircraft and tower observations conducted by U.S. government laboratories (National Oceanic and Atmospheric Administration and Department of Energy) in 2007 and 2008. The authors concluded that the US EPA inventory underestimated CH_4 anthropogenic emissions by ~50%. Brandt et al.[42] reached a similar conclusion of ~50% underestimation by US EPA based on a meta-analysis of published results. Based in part on the distribution of emissions excess observed especially in the southern U.S. and on the content of propane in the air, both studies suggest that some of the missing emissions in the inventory could be explained by larger emissions from oil and gas production and processing.

A few regional atmospheric studies in the U.S. have shown elevated levels of methane and other hydrocarbons in oil and gas producing regions.[43−45] Karion et al.[45] estimated that 8.9% ± 2.8% of the methane produced in the Uintah Basin gas field of Utah was lost to the atmosphere based on airborne measurements on one day in 2012. This is more than twice the average loss rate estimated by Pétron et al.[44] (average, 4%; range, 2.3–7.7%) for an oil and gas field in northeastern Colorado in 2008, based on a mix of methane and propane tower and ground-based measurements and inventory data.

Recent emission factors derived by Allen et al.[15] for three natural gas production source categories (gas well completion flowbacks, production site equipment leaks, and venting of pneumatic pumps and controllers) suggest that average CH_4 emissions for well completions using reduced-emissions flowback procedures are less than estimated in the US EPA inventory. The study, however, found higher emissions on average from pneumatic devices and pumps and production site leaks than assumed in the US EPA GHG NI. The direct emission measurements conducted by Allen et al.[15] at 190 onshore production sites—in partnership with operators—in four different U.S. regions were averaged and extrapolated to the national level for comparison with the US EPA GHG NI. At the national level, they estimated that 0.42% of natural gas gross production leaked to the atmosphere, which is lower than in the 2013 US EPA GHG NI estimate for 2011 (0.49%).

Transmission, storage, and distribution of natural gas includes hundreds of thousands of kilometers of pipeline, > 1400 compressor stations, and approximately 3.5 Tcf (~equivalent to two months of national consumption) of underground storage throughout the U.S.[46] According to the 2013 US EPA inventory, transmission is the stage of the natural gas life cycle with the highest emission of CH_4. Emissions during transmission, storage, and distribution are mainly limited to fugitive CH_4 (and, to a lesser extent, C_2H_6) emissions from an aging natural gas pipeline infrastructure and venting during pipeline and compressor station maintenance. A few studies have focused on methane leakage from the natural gas distribution network across cities such as Los Angeles, California,[47] Boston, Massachusetts,[48] and Washington, DC.[49] For example, Phillips et al.[48] mapped ~3400 natural gas distribution pipeline leaks across Boston's 800 road miles in 2011. An example of these leaks is shown in Figure 3 where concentrations of methane as high as 28.5 ppm (compared to a

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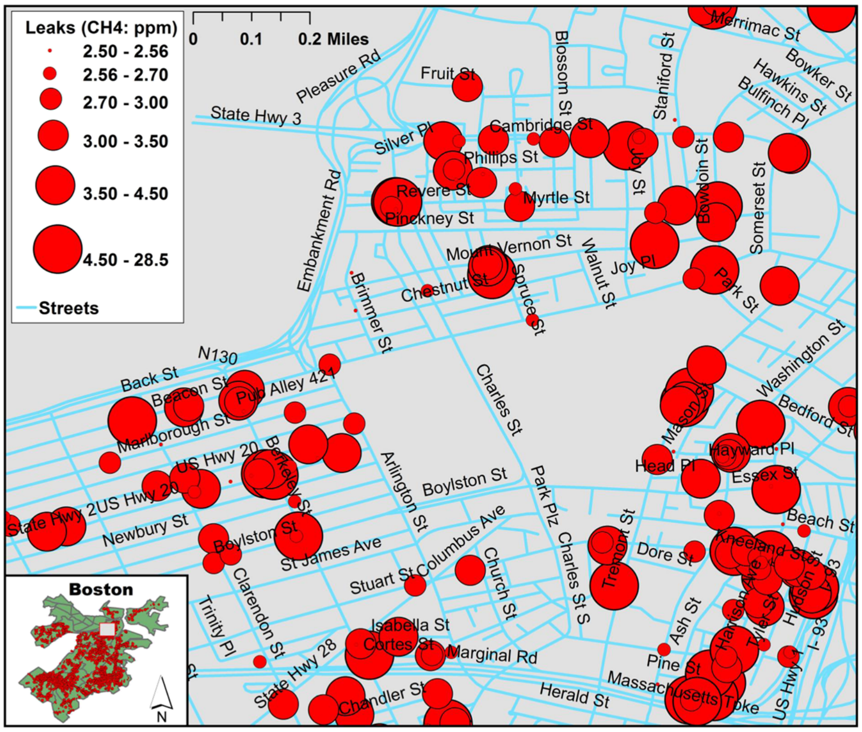

Figure 3. Locations of elevated methane concentrations in the Beacon Hill area of Boston, Massachusetts, associated with natural gas distribution pipeline leaks. Data from Phillips et al.[48]

global background of 1.8 ppm[28]) were measured. The presence of older cast-iron distribution mains was the strongest predictor for the leaks that they observed ($r^2 = 0.79$, $P < 0.001$[48]).

The US EPA CH_4 leakage rates for distribution alone are in the range 0.35−0.70%.[50,51] Lelieveld et al.[52] combined loss estimates for storage and distribution together to suggest an overall average loss rate of 1.4% (with a range from 1.0% to 2.5%). Based on additional data from Texas and elsewhere, Howarth et al.[53] assumed a higher range of values, from 1.4% to 3.6% leakage of CH_4 during transmission, storage, and distribution; but these estimates have been disputed.[54] Cathles et al.[54] suggested that Howarth et al.[53] "significantly overestimated" fugitive emissions and undervalued the emission reduction from the use of "green technologies". Other authors[54,55] have criticized Howarth et al.[53] for use of "heat rather than electricity generation" for their life cycle assessment, and a 20 year time frame that overemphasized the shorter-term impact of CH_4 on radiative forcing. However, with the current lack of representative and recently measured emissions, we are left to wonder just what the actual leakage rates are at the regional and national scales, emphasizing the difficulty with elucidating existing interpretations.

A review of 20 years of literature on CH_4 leaks[42] has found that the extent of leakages from North American natural gas systems may be larger than anticipated, yet best management practices and regulation for technologically achievable emissions reduction and effective leak detection and repair programs can significantly reduce the climate footprint of natural gas.[56] The large recent changes in US EPA methodology and annual emission estimates and disparities in site level and regional level emission measurements highlight the need for additional research to better understand emissions across the natural gas life cycle (see above) and to reconcile emissions measured at different spatiotemporal scales.[15,22,40,52,53,57,58]

■ AIR QUALITY IMPACTS OF THE FIRST TWO LIFE CYCLE STAGES

Preproduction. In addition to CH_4, activities in the first two of the five natural gas life cycle stages emit other compounds than can impact local and regional air quality. The preproduction stage includes everything from exploration, site clearing, and road construction to drilling, hydraulic fracturing, and well completion. For a single well, preproduction is usually completed within a few weeks; but these operations may be carried out for a dozen or more wells on a pad and at multiple sites in the field, typically lasting for months.[16] Several pollutants with environmental and human health impacts[59] have been linked to this stage[60−64] and a few monitoring efforts are underway to document actual atmospheric exposures.[65−67]

Air quality impacts begin with the use of large diesel-powered equipment during site preparation,[62] including the construction of roads and holding ponds as well as clearing of the well pad.[68,69] Emissions from on and off-road diesel use continue throughout drilling and hydraulic fracturing as millions of gallons of water, sand, and hydraulic fracturing chemicals are transported to and from the well pads.[70] Diesel emissions are known to include airborne fine particulate matter (2.5 μm and smaller in diameter; $PM_{2.5}$)[71−74] as well as ozone precursors such as NO_x and NMVOCs.[75,76] Long-term exposure to $PM_{2.5}$ can lead to decreased lung function, asthma, and increased respiratory symptoms such as coughing and difficulty breathing.[77] Truck traffic also generates coarse particulate matter ≤10 μm in diameter (PM_{10}),[62]emitted from tire wear, brake wear, and resuspended road dust. However, Litovitz et al.[62] found that emissions from oil and gas operation related transportation in Pennsylvania were small compared to other emissions from natural gas activities statewide, contributing only 0.5−1.2% of VOCs, 3.2−3.5% of NO_x, and 2.1−3.5% of $PM_{2.5}$ emitted from natural gas activities.

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Emissions can continue into the drilling and hydraulic fracturing procedures. During the process of drilling, pockets of CH_4, and potentially C_2H_6 and propane, through which the drill passes, can be released into the atmosphere.[57] However, little information exists on the frequency and volume of emissions from these releases, which is currently a major uncertainty in emissions inventories. Emissions measurements are strongly needed during this section of preproduction.

After drilling is completed, water, hydraulic fracturing fluid, and proppant (e.g., silica sand or man-made ceramic beads) are pumped underground at pressures of ∼10 000 to 20 000 psi to fracture the low permeability reservoir rock to allow the natural gas to flow.[7,78] Emissions during drilling and hydraulic fracturing include exhaust from diesel[63] and natural-gas powered engines for drilling rigs and pumps.[79] Bar-Ilan et al.[63] estimated that 12 to 27% of NO_x emissions from natural gas activities in three areas of Wyoming originate from drilling rigs alone. Litovitz et al.[62] estimated that well drilling and hydraulic fracturing in Pennsylvania accounted for 2.6−10% of VOC, 29−39% of NO_x, 16−33% of $PM_{2.5}$, and 35−55% of SO_x emissions from natural gas activities. The fluid used during hydraulic fracturing can contain hundreds of chemicals, including acids, ethylene glycol, and isopropanol.[7,80−82] However, the detailed constituents of the hydraulic fracturing fluid mix are often proprietary, meaning that reporting of the constituents is voluntary by the industry[82] and often incomplete. Also, no information exists on the interactions of the chemicals in the fracturing fluid with naturally occurring chemicals down the well and what potential problems this might cause. Many of the constituents are volatile under atmospheric conditions. A portion of the fracturing fluid mix returns to the surface during the flowback stage and is stored in holding ponds or flowback tanks and later disposed at industrial waste or deep injection facilities. A full classification of all emissions during drilling and hydraulic fracturing does not to our knowledge exist.

Another area where little information exists is on the emission of (and exposure to) respirable silica (crystalline silica "small enough to enter the gas-exchange regions of the lungs[83]"; 10 μm and smaller[83]) from the proppant injected during hydraulic fracturing. The U.S. National Institute for Occupational Safety and Health conducted field studies at 11 sites in five states between 2010 and 2011 and found that workers were exposed to high levels of respirable silica in 31% of sampled cases (N = 111).[84] The high values observed were ten or more times the recommended exposure limit and above the filtration capabilities of half-face respirators worn by the workers.[84] This exposure can occur during transportation of the sand by truck or conveyor belt and also can occur upstream, at the site where the silica is extracted.[83] Exposure to respirable silica can decrease lung function, increase respiratory symptoms such as coughing, result in difficulty breathing, and cause asthma and silicosis.[83] The impacts of respirable silica are greatest for workers on site, but broader studies are needed for people living near well pads and production staging areas.

Once drilling and hydraulic fracturing operations have been finished, the well is completed and prepared to produce natural gas. Emissions during the well completion process, particularly during venting and flaring of initial natural gas before the well is connected to a transmission pipeline, can include CH_4 and BTEX.[60,85] These emissions can also other nonmethane hydrocarbons, along with hydrogen sulfide H_2S,[63] NO_x, and if there is incomplete combustion of natural gas formaldehyde,[86] at concentrations in the air that have the potential to affect residents living within <800 m of wells.[60] Most of these emissions, however, are scheduled to be mostly eliminated by 2015[87] when the US EPA will require use of "green completions" or "reduced emission completions" when technically feasible. During these processes, flowback fluid, oil and gas are separated as soon as possible in well completion, and the gas and oil are routed for sale. Green completions reduce overall emission of CH_4 and air pollutants that traditionally would have been vented.[15,88,89]

Allen et al.[15] describe four different completion flowback configurations at hydraulically fractured gas wells and present direct measurements of CH_4 emissions at 27 sites in four different regions of the U.S. On average, the sites sampled by Allen et al.[15] had lower emissions than what is assumed by the 2013 US EPA GHG NI for 2011. Methane emissions measured during 27 well completion flowbacks, for instance, averaged only 1.7 Mg CH_4[15] compared to an average of 81 Mg per event used in the 2013 US EPA GHG NI.[15] Measured emissions during a flowback event, however, varied by two orders of magnitude within a basin.[15] The distribution of emissions from completion flowback measured by Allen et al.[15] is not Gaussian, and therefore, a simple set of uniform average emission factors at the regional and national levels for an average green completion configuration will most likely not capture the actual aggregated emission magnitude.

Production. Several atmospheric pollutants have been linked to the production stage of the natural gas life cycle and have been studied in a few areas.[65−67,90−97] As mentioned earlier, the natural gas that flows directly from the well often contains other associated NMVOCs, water vapor, carbon dioxide, hydrogen sulfide, or natural gas liquids[14] and needs processing in order to meet purity standards for addition to the pipeline infrastructure, known as "pipeline quality natural gas".[14,98] Processing occurs near the well and/or at a centralized processing plant and includes compression of the processed natural gas to be transported through pipelines to consumers. Once production at a well has begun, emission sources can include well-head compressors or pumps that bring the produced gas up to the surface or up to pipeline pressure (engines are often fired with raw or processed natural gas), well pad equipment bleeding and leaks, flare emissions, maintenance emissions, and compressor station emissions. Litovitz et al.[62] estimated that production sites and compressor stations in Pennsylvania accounted for 91−97% of VOCs, 59−68% of NO_x, 64−84% of $PM_{2.5}$, and 40−64% of SO_x emissions from natural gas activities.

Other sources of CH_4 and NMVOCs (including BTEX) emissions during the production stage can include dehydrator regeneration vents, venting from pneumatic pumps and devices that are actuated by natural gas, leaks through faulty casing, incomplete emissions capture, or burning in flaring systems. Some of these emissions can be continuous or intermittent but will be ongoing during the entire lifetime of the well unless direct emissions capture and destruction or recovery are put into place. Emissions from crude oil and liquid condensate (light crude oil) storage tanks were estimated to be responsible for 66% of total NMVOCs emitted by oil and gas operations in Denver-Julesburg Basin in the northeast Colorado Front Range.[99] Other emissions related to maintenance or production stimulation, for example, will be episodic such as during liquid unloadings and during workovers. Due to the diffuse nature of emissions from hundreds of thousands of well pads, variations

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in composition of the raw gas itself, and varying degrees of emissions controls and reduction requirements, conclusions on the overall air quality impact of this stage span from highly detrimental[8,43,44,100,101] to little or no impact at all.[65,90,92,95,102−105] This level of discrepancy indicates that more work needs to be done at the basin scale on the emissions from the production stage of the life cycle and their impacts.

Oil and gas emissions of ozone (O_3) precursors (NMVOCs)[44,62,63,99,101,106−112] have been linked to regional exceedances of the 8-h national ambient air quality standard for O_3 (75 ppb for fourth highest daily maximum concentration averaged for three consecutive years). O_3 precursors[44,62,63,99,101,106−112] emitted from the natural gas and oil production stage can make attainment of US EPA O_3 exposure limits difficult even in winter for some areas.[99,106−108,113,114] High surface level O_3 concentrations, produced by increased NO_x and VOC abundance,[86,115] can lead to respiratory problems, particularly in children and older adults.[116] The US EPA nonattainment designation for the O_3 standard has been a driving force behind state-level regulation of O_3 precursor emissions from oil and gas operations and increased ambient air monitoring programs in Wyoming and Colorado,[89] two states with the most stringent air regulations in the U.S. for their affected areas. Air monitoring before and during oil and gas development can help regulators and air quality managers keep track of the air impacts of different air pollution sources and how they may change over time. To date, most US EPA and state air monitoring (especially for O_3) is done in urban areas, leaving entire industrialized rural and suburban communities without baseline and routine air quality measurements.

Other Stages. Much less information exists on the non-CH_4 emissions from two of the three other natural gas life cycle stages. Since pipeline quality natural gas is predominantly CH_4, few other pollutants have been reported to be emitted from the transmission, storage and distribution stage (Figure 1). On the other hand, some emissions (e.g., NO_x, SO_2, CO_2, and CH_4) from the use of natural gas are estimated each year by the US EPA,[117] particularly what is emitted during use for power generation (discussed in more detail below), and researchers have attributed some formaldehyde emissions to natural gas combustion. In particular, Zhang et al.[118] attributed 10−30% of the primary formaldehyde concentrations to natural gas combustion in the Houston, Texas area during the 2006 Texas Air Quality Study (TexAQS). Other studies have indicated that O_3 concentration criteria exceedances in Texas cities are attributed to natural gas combustion.[118,119]

At the end of the well production life (well production end-of-life), the well is "plugged" (if not just abandoned). What information is available on the potential for gas leakage is derived primarily from historical studies of conventional wells. In Alberta, for instance, 4% of abandoned wellbores leaked, including many which were plugged before abandonment.[120] In Pennsylvania, an estimated 325 000 oil and gas wells were drilled between 1860 and 2000, but the Pennsylvania Department of Environmental Protection only has records for 88 300 regulated operating wells, 44 700 plugged wells, and 8000 abandoned wells, leaving the status of 184 000 wells unknown.[121] Other states have similar issues, for instance, New York plugged 323 (mostly old/abandoned) wells in 2012 with many more still needing to be plugged.[122] Until the number of orphaned/abandoned wells is known, we cannot even begin to estimate the air quality impacts from this portion of the natural gas life cycle.

POTENTIAL AIR QUALITY BENEFITS OF INCREASED NATURAL GAS USE

The interest in increasing production and use of natural gas in the U.S. during the past decade is due, in part, to the fact that natural gas emits less CO_2, sulfur dioxide (SO_2), NO_x, and mercury (Hg) compared to coal and oil when burned to produce heat or electricity.[23,36,95,123,124] Natural gas use for electricity generation emits roughly half the CO_2 of coal per kWh produced, potentially improving air quality and reducing GHG emissions compared to coal. An immediate benefit from an increased share of natural gas for electricity generation in the U.S. (from 14% in 2000 to 29% in 2012[125]) is a reduction in the carbon intensity of U.S. electricity generation in 2011 and 2012.[25,126,127] The controversy, however, arises in attempting to estimate the total methane leakage associated with natural gas production, distribution, and use, and, to a lesser extent, the methane leakage associated with coal mining.[55,57] Most life cycle comparison studies have relied on leakage estimates derived from the US EPA GHG NI for natural gas systems. Venkatesh et al.[128] estimated that approximately 1−3 kg of NO_x per MWh and 2−10 kg of SO_2 per MWh are the typical emissions from coal-fired power plants likely to be retired or replaced by combined cycle natural gas plants. Alternatively, emissions of SO_2 and Hg from natural-gas-fired power plants are negligible; and emissions of NO_x are substantially lower than for coal-fired power plants.

Another potential use for natural gas (conventional or unconventional) includes replacing petroleum in products such as liquid fuels and olefins.[129] Olefins are used to produce plastics (polyethylene, polyester, polyvinyl chloride (PVC), and polystyrene) that are, in turn, used to produce millions of consumer goods. Access to CH_4, C_2H_6, propane, and butane through unconventional natural gas development, may increase their use in the production of high-value chemicals. The benefits of a potentially "new" source of materials for making these products is clear, but new process chemistry will be needed to replace petroleum with natural gas,[129] and these uses will need to be included in new life cycle assessments for unconventional natural gas.

Until the efficiency of compressed natural gas (CNG) vehicles increases, and CH_4 leakage rates from natural gas production decrease further, the GHG benefits of substituting natural gas for gasoline in vehicles are small[22] or negligible.[40,130,131] Alvarez et al.[40] estimated that converting a fleet of gasoline cars to CNG would increase radiative forcing for at least 80 years before modest net climate benefits would be achieved; the comparable crossover point for heavy-duty diesel vehicles would be nearly 300 years. In fact, Alvarez et al.[40] estimated that CNG conversion would result in more rapid climate change for decades, attributable to the greater radiative forcing in the early years after conversion. In contrast, converting vehicles to natural gas would have immediate (nonclimate) air quality benefits compared to gasoline because of the cleaner burning properties of natural gas and reduced non-methane air pollution.

REGULATIONS

Until recently, air regulation of oil and gas production operations was done at the state level. The US EPA attempts to quantify and minimize the air quality impacts of industrial activities, including oil and natural gas operations. In 2012, the agency released a set of new source performance standards

(NSPS).[87] The NSPS take effect in 2015 and rely heavily on self-reporting from the industry of emissions to the US EPA.[132] The standards attempt to limit VOC emissions during well completion by requiring the use of green completion technologies, which the US EPA estimates will result in a 95% reduction of VOC emissions and a 99.9% reduction in SO_2 emissions.[87,89] Further requirements of the rule include limiting emissions of VOCs from a new single oil or condensate tank to four tons per year[133] and limiting BTEX from a single dehydrator to one ton per year.[87] The rule focuses on two types of compressors: centrifugal compressors with wet seals must reduce VOC emissions by 95% and reciprocating compressors must have regular maintenance to keep them from leaking VOCs.[87] Also, pneumatic controllers are required to vent less than six standard cubic feet per hour. Other air toxics are not specifically regulated under this new rule and are limited to major sources that emit 10 or more tons of a single air toxic or 25 or more tons of a combination of toxics.[87]

The US EPA also has adopted multiple tiers of emission standards for on-road[134] and off-road[135] diesel engines that may influence overall air impacts from the natural gas life cycle. These standards apply to criteria pollutants including NO_x, non-methane hydrocarbons, CO, and PM. Manufacturers must currently ensure that each new engine, vehicle, or equipment meets the latest emission standards. If diesel engines were built before US EPA emission standards came into effect, however, they are generally not affected by the standards or other regulatory requirements. Although the latest tiers of diesel engine emission standards are very stringent, heavy-duty diesel engines are long lasting. Thus, many older trucks and off-road equipment are still being used.

Many states have also taken separate, individual actions to regulate the overall environmental impacts of the oil and natural gas industries, and some states are developing public disclosure laws for hydraulic fracturing fluids.[82] Colorado passed regulations from 2007 to 2009 requiring operators to (1) use no-bleed or low bleed pneumatic devices at oil and gas production sites in the northeastern Front Range O_3 non-attainment area, (2) use green completion technologies at oil and gas wells when technically feasible, and (3) control flashing emissions from condensate and oil storage tanks. The Colorado system-wide emissions reduction requirements for NMVOCs from tanks are 90% in the summertime and 70% otherwise; the state, however, estimates that the actual annual average reduction in emissions has been 53% (compared to having no controls in place).[136,137] Wyoming has required green completions since 2004 and requires 98% reduction of emissions (instead of 95% for the NSPS) for newly installed tanks.[89] Montana requires the control of emissions from the well immediately upon completion and has specific regulations regarding compression devices, pneumatic controllers, condensate/crude oil storage tanks, and glycol dehydrators.[89] New York has issued a moratorium on high-volume hydraulic fracturing.

Other states have taken fewer additional regulatory steps and will rely largely on the NSPS that will begin January 1, 2015.[89] These include Alaska, North Dakota, New Mexico, and West Virginia. Texas has been tracking emissions data from the oil and gas industry for years, but often limits regulation of emissions to the Houston and Dallas−Fort Worth federal O_3 standard non-attainment areas.[138] Utah has regulations that limit emissions from hydrocarbon storage tanks; however, these regulations only apply to Salt Lake City and Davis County.[89] These areas are not near the Uintah Basin where oil and gas operations exist, and therefore do nothing to improve the high wintertime O_3 concentrations observed during strong temperature inversions.[89,139] Pennsylvania has recently reevaluated and limited the oil and natural gas facilities that were previously exempt from regulations.[140] The wide variety of regulations and practices by different states indicates that much more attention should be focused on systematically assessing the air emissions from oil and gas operations and their air impacts in those states with substantial levels of unconventional natural gas activities and production.

RECOMMENDATIONS

Based on our examination of the literature on the air quality impacts of unconventional gas extraction and distribution, we have determined that actual measurement data on various individual segments of the natural gas life cycle are sparse or critically lacking. To maximize the true benefits and minimize the negative impacts of this resource, we recommend that the following steps be taken to fill critical knowledge gaps:

- Air quality measurements need to be made prior to oil and gas development, including during drilling and hydraulic fracturing, to more clearly understand the direct impacts of these activities. Air monitoring during these operations can help ensure emissions management strategies are effective and exposure to air pollutants, including silica, are kept to a minimum.
- A full chemical classification of emissions, including air toxics, during all life cycle stages needs to be obtained to properly perform source apportionment modeling and to understand all potential air quality and health impacts.
- Independent scientific data on the true nationwide extent of methane leaks from the production, processing, transmission, storage, and distribution infrastructure, including measurements of flows and fluxes, should be acquired.
- An inventory of abandoned/orphaned wells should be collected so that emissions can be properly estimated.
- Measurements on the variation of air emission composition and magnitude by natural gas and oil plays need to be made.
- Collaborations between independent scientists, regulators, and operators need to be increased to gain access to areas where measurements should be made and to inform effective emissions detection, reduction, and monitoring strategies.

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Notes

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