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Air quality concerns of unconventional oil and natural gas production

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Increased use of hydraulic fracturing ("fracking") in unconventional oil and natural gas (O & NG) development from coal, sandstone, and shale deposits in the United States (US) has created environmental concerns over water and air quality impacts. In this perspective we focus on how the production of unconventional O & NG affects air quality. We pay particular attention to shale gas as this type of development has transformed natural gas production in the US and is set to become important in the rest of the world. A variety of potential emission sources can be spread over tens of thousands of acres of a production area and this complicates assessment of local and regional air quality impacts. We outline upstream activities including drilling, completion and production. After contrasting the context for development activities in the US and Europe we explore the use of inventories for determining air emissions. Location and scale of analysis is important, as O & NG production emissions in some US basins account for nearly 100% of the pollution burden, whereas in other basins these activities make up less than 10% of total air emissions. While emission inventories are beneficial to quantifying air emissions from a particular source category, they do have limitations when determining air quality impacts from a large area. Air monitoring is essential, not only to validate inventories, but also to measure impacts. We describe the use of measurements, including ground-based mobile monitoring, network stations, airborne, and satellite platforms for measuring air quality impacts. We identify nitrogen oxides, volatile organic compounds (VOC), ozone, hazardous air pollutants (HAP), and methane as pollutants of concern related to O & NG activities. These pollutants can contribute to air quality concerns and they may be regulated in ambient air, due to human health or climate forcing concerns. Close to well pads, emissions are concentrated and exposure to a wide range of pollutants is possible. Public health protection is improved when emissions are controlled and facilities are located away from where people live. Based on lessons learned in the US we outline an approach for future unconventional O & NG development that includes regulation, assessment and monitoring.

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Environmental impact

Increased use of hydraulic fracturing ("fracking") in unconventional oil and natural gas (O & NG) development from coal, sandstone, and shale deposits in the United States has created environmental concerns. Production of O & NG affects local and regional air quality. Nitrogen oxides, volatile organic compounds, ozone, hazardous air pollutants, and methane are pollutants of concern related to O & NG activities. Close to well pads, emissions are concentrated and exposure to a wide range of pollutants is possible. Public health protection is improved when emissions are controlled and facilities are located away from where people live. The replacement of coal with natural gas for electricity generations has air quality benefits.

1. Unconventional oil and gas development

Unconventional oil and natural gas (O & NG) is an umbrella term for development that does not meet the criteria of conventional production.¹ The US Energy Information Administration (EIA) defines conventional O & NG production as "crude oil and natural gas that is produced by a well drilled into a geologic formation in which the reservoir and fluid characteristics permit the oil and natural gas to readily flow to the wellbore".¹ The O & NG in unconventional reservoirs are usually distributed throughout the pore spaces of the reservoir rock, making it more difficult to extract the hydrocarbons. The term unconventional changes over time as techniques develop and then become common practice. Here we consider unconventional as the development of O & NG resources from tight sandstone, coal and shale through the application of hydraulic fracturing and directional drilling techniques. Together these techniques have transformed the energy landscape of the United States (US). The use of these techniques has allowed for a

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dramatic expansion of unconventional O & NG production.² Hydraulic fracturing involves the pumping of water, sand, and various chemicals, at high pressure to generate fractures in the target rock formation (Fig. 1).^{2,3} As opposed to traditional vertical drilling, directional drilling provides the control required to locate pockets of gas within a geologic formation by drilling horizontally. This technique has enabled upwards of 50 wells to be drilled from one well pad location.³

The US EIA predicts that the US will become a net exporter of petroleum liquids by 2030.⁴ The EIA also estimates an increase in shale gas production from 5.0 trillion cubic feet (tcf) per year in 2010 to 13.6 tcf per year in 2035, when shale gas is expected to account for 49% of total US gas production (Fig. 2).⁴ The increased supply and coincident price decline of natural gas has led to the rapid development of proposals for liquefied natural gas terminals for export of natural gas to other nations. In 2013 the Henry Hub benchmark average wholesale price of natural gas of \$3.73 per Mcf (thousand cubic feet) was, besides 2012, the lowest since 2002.⁴ Low natural gas prices have caused the O & NG industry to focus resources on exploiting shale oil deposits. As natural gas prices continue to remain low, drilling

Marcellus Shale to crack. These lisures are held open the sand particles so that natural gas from the shale can flow up the well. Well turns Marcellus Shale Marcellus Shale

hic by Al Granberg

Fig. 1 Hydraulic fracturing and direction drilling is graphically illustrated to show subsurface and surface processes that are associated with the delivery of natural gas to market.⁵ Courtesy of the Propublica.

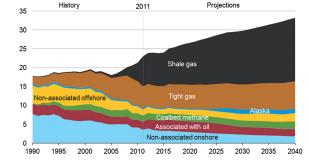


Fig. 2 Natural gas production by source 1990 to 2040 (trillion cubic feet). Courtesy of the US Energy Information Agency.³

rigs in the US have migrated away from natural gas toward the more lucrative oil-rich plays.⁵ In 2013 natural gas annual production growth rate was the lowest since 2005, by contrast oil production increased to the highest level in more than a decade. Europe and the rest of the world are looking at lessons learned in the US, where the benefits from an improved energy supply have come with a wide range of environmental costs. While advocates promote natural gas as a clean, low carbon energy supply, detractors point to irreversible damage due to land use change, potential groundwater contamination, and increased air pollution.

Unconventional O & NG production can include hydrocarbon recovery from coal, tight sandstone, and shale deposits resulting in a spectrum of products – from methane (natural gas) to heavy hydrocarbons (oil) with produced water as a byproduct. Wells are defined as gas or oil based on which product is predominant. A gas well produces more than 100 Mcf of gas per barrel (bbl) of oil, whereas an oil well produces less than 100 Mcf per 1 bbl of oil. Product composition is determined by the geology of the target formation. At the basin scale a number of formations may be developed simultaneously, each recovering product with different chemical makeup.

Environmental concerns regarding unconventional O & NG production have been focused on subsurface issues, such as aquifer contamination,⁶⁻¹¹ downhole communication ("frac hits"),¹² seismic events;¹³ and wastewater issues (salinity, toxicity, and radioactivity).¹⁴ Trade secret claims surrounding the chemical composition of hydraulic fracturing fluids have enhanced these concerns. Researchers have identified a large number of chemicals used in hydraulic fracturing fluids¹⁵ and have classified a wide range of possible human health hazards¹⁶ including cardiovascular, respiratory, nervous, immune and endocrine disorders.¹⁷ While subsurface issues are important, the handling of hydraulic fracturing fluids, produced water, and the extracted products above ground determines air quality impacts.

Determining the air pollutants that will be of most concern for an unconventional O & NG field begins with knowing the chemical composition of the product. In addition, wet natural gas, dry natural gas, and oil wells will have different product handling approaches and therefore different associated air emission sources. Natural gas that contains more than 0.1 gallon of condensate per 1000 cubic feet of gas may be labelled "wet gas".18 Alternatively wet gas is defined as a natural gas with less than 85% methane,19 or more simply as "containing sufficient quantities of hydrocarbons heavier than methane to allow their commercial extraction or to require their removal in order to render the gas suitable for fuel use".20 Wet gas contains complex hydrocarbons from natural gas liquids (ethane, propane, butanes) and condensate, which is a mix of longer chain (C_5^+) and cyclic hydrocarbons (cycloalkanes and aromatics), whereas dry natural gas is predominately methane. Dry gas will contain other hydrocarbons albeit at lower proportions that preclude commercial extraction or the need for removal in order to render the gas suitable for fuel use.20 Because wet gas usually contains a more complex mixture of hydrocarbons than dry gas, the variety of VOC from wet gas is greater. Furthermore, wells producing crude oil, a complex mixture of hydrocarbons that generally have

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from five to 40 carbon atoms per molecule, have different air emissions of concern. In broad terms, hydrocarbons with lower volatility (a greater number of carbon atoms) are less likely to be emitted to the atmosphere during production.

1.1. Unconventional O & NG air pollution emission sources

As soon as subsurface hydrocarbons become mobile there is potential for an eventual air emission. These emissions should follow the fracture zone up the wellbore to the wellhead at the surface, with the possibility of some transmission outside of the well casing. Notwithstanding seepage,²¹ potential emissions start at the wellhead and continue until consumer end use. Emissions from unconventional O & NG sources are classified by air regulatory agencies as point sources (from a stack or pipe), mobile sources (from trucks, trains, drill rigs), fugitive sources (from equipment leaks, or external forces such as wind, or natural or man-made faults or fractures in the earth's surface) and area sources. In the US, point sources, also known as stationary sources, are termed as major if emissions exceed certain levels, for VOC a level of 10 tons per year is needed. Area sources are aggregate emissions from small point sources of a particular category that are not considered to be major point sources.

Fugitive emissions are difficult to quantify because they can be hard to detect and may be intermittent. Elevated methane in soil surrounding some active wells in Utah has been reported.²² Considerable leakage from natural gas pipelines for urban delivery networks has also been reported.²³ Fugitive emissions occur at many points and magnitudes, and are estimated for specific processes. Leakage rates for the natural gas sector may be determined for upstream development activities or by lifecycle analyses that include transmission and supply chains. In this perspective, leakage rates are those predicted or calculated with reference to upstream activities that are performed within O & NG developments, namely, drilling, completion, and production.

Point source, mobile, fugitive, and areas source emissions will vary with the type of O & NG activity. There will be emissions of oxides of nitrogen (NO_x), particulate matter (PM) and VOC associated with combustion from drill rig, compressor, and generator engines, as well as from heaters and pumps. Mobile sources, in particular truck traffic, also contribute to NO_x, PM and VOC emissions as well as producing fugitive dust from unpaved roads. Fugitive emissions from product handling, including produced water, and equipment leaks represent another source of methane and VOC emissions. Exposure to fine silica dust associated with sand used in the hydraulic fracturing process has also been recognized as impacting air quality at the well pad.²⁴ There are also VOC emissions from the use and handling of drilling waste and fluids.

An important stage of hydraulic fracturing is the process of flowback. During flowback, fluids and solids used during hydraulic fracturing return to the surface along with produced water and reservoir O & NG. Flowback typically takes place for three to 10 days, and may include significant emissions of methane, HAP and VOC. Once flowback is complete, with solids absent from the flow, air emissions occur from various production activities. In this regard, unconventional and conventional wells are no different, and both connect to the same midstream and downstream distribution network.

At the well pad, the main O & NG field processing activities are the use of pressure and heat to separate gas, produced water, and liquid hydrocarbons from each other. The gas fraction is then typically routed to a dehydrator that removes any excess water from the gas. The produced water and liquid hydrocarbon fractions are stored in separate tanks. Fugitive emissions from tanks are an important source of methane, HAP, and VOC. These emissions are enhanced during tanker truck transfers for processing elsewhere.

Dehydration of produced gas is required to reduce condensation of water and heavier hydrocarbons that reduce the efficiency of gas flow in high pressure pipelines. Most dehydration systems employ glycol, which absorbs water and VOC, in a closed loop system. Aromatic hydrocarbons, including benzene, toluene, ethylbenzene and xylenes (BTEX), are more easily absorbed by the glycol than other hydrocarbons found in natural gas. When glycol is heated in a reboiler for regeneration, BTEX and other VOC are released. BTEX emissions, a subset of VOC, are also classified as hazardous air pollutants (HAP) and are sometimes referred to as air toxics, meaning they are particularly harmful to human health. Because of the hazardous nature of BTEX emissions, control of pollutants from dehydration units through combustion is preferable to venting. Despite control by combustion, BTEX and other VOC emissions are significant. Emission controls on dehydration units should not be confused with natural gas flaring that takes place in oil rich developments that do not have established natural gas distribution networks. Combustion units and flares have different designs and purposes. Combustion units often have control efficiencies close to 100%; however, flaring under optimum conditions yields combustion efficiencies of \sim 70% that decline to less than 15% at wind speeds above 6 meters per second.25

Produced water is an important but uncertain emission source. Like flowback fluids and drilling fluids, there are a number of possible handling approaches. In some locations, produced water management has included deposition of this waste byproduct into evaporation ponds or pits. Because ponds are not enclosed, evaporation leads to downwind emissions. The use of containment, whether restricted to tanks at a pad, or liquid gathering systems to pipe water offsite, is part of the development plan and regulatory oversight. Developments in remote areas are suitable for systems that incorporate dedicated water treatment and recycling for reuse in subsequent operations. In urban and rural areas, water handling may be confined to tank storage with removal by trucks. Water handling is important, as water contains VOC stripped during wet gas dehydration. Emissions from open pond storage methods are more difficult to quantify than emissions from storage tanks. Research has identified highly variable emissions of VOC from produced water ponds,26 with VOC emissions highest from recently filled ponds during warmer months.27 Water treatment facilities have also been identified as significant, difficult to quantify, emission sources.28 The lack of measurement methods and emissions data for fugitive emissions from Well workovers and refracturing, together with maintenance operations add to normal production emissions. Workovers repair or stimulate a well to restore, prolong or enhance production of hydrocarbons.²⁰ Another potentially significant source of emissions for some wells is liquids unloadings, where a well is vented to atmosphere to remove liquids that had gathered and slowed gas production from the well. In addition, there will be combustion emissions from compression of natural gas for delivery into high-pressure pipelines. Compression takes place at the well pad and at dedicated compressor stations that link a number of well pads.

As drilling has encroached on residential and urban areas, attention has turned to the impact of O & NG emissions on ambient air quality and human health (Fig. 3).^{29,30} The US Government Accountability Office has reported that cumulative air quality impacts are difficult to determine as the extent and severity of risks vary significantly within and between developments due to location and process driven factors.³¹ As such, one size does not fit all, and the importance of location specific assessment has emerged.

1.2. Regulatory approach in the United States

In the US there is a complex interwoven set of relationships between the public, regulators, and developers with respect to O & NG activities. Development may take place on private, county, state or federal land with permitting of anywhere from a few to many thousands of wells. For significant actions on federal land, the National Environmental Policy Act (NEPA) usually requires an Environmental Impact Statement (EIS). Air quality is a primary consideration and potential impacts are estimated by the modeled influence of predicted emissions on ambient air quality. Regulatory agencies will only permit activities so long as they are able to predict that National Ambient Air Quality Standards (NAAQS), regulated through the Clean Air Act, will be achieved.



Fig. 3 A well pad in the Marcellus shale in Hopewell, Washington County, Pennsylvania shows the close proximity between development activities and where people live. Pennsylvania is a region with large human populations. Photo © Scott Goldsmith.

Compliance with NAAQS is defined by data collected at air quality monitoring stations. When an area is designated as "nonattainment", more stringent permit conditions and increased scrutiny of emission inventories are generally imposed. A nonattainment area is a locality where air pollution levels persistently exceed one or more NAAQS. With respect to the O & NG industry, this scrutiny is placed on emissions that affect ozone and nitrogen dioxide.³² Given that ozone is a secondary pollutant (not emitted directly but formed from atmospheric reactions), the emission and distribution of precursors such as VOC and NO_x are important to know before one can understand the influence of O & NG emissions on air quality.

In 2012, US EPA issued important amendments to existing air quality regulations that apply to the O & NG industry. The Clean Air Act requires US EPA to develop new source performance standards (NSPS) for industrial categories that cause, or significantly contribute to, air pollution that may endanger public health or welfare.²⁹ Another regulatory program specified by the Clean Air Act, the National Emission Standards for Hazardous Air Pollutants (NESHAP), was reviewed at the same time. Together, the revised rules and regulations are set for full implementation in 2015.

The 2012 revision regulates a number of upstream processes not addressed previously, including: well completions and recompletions, compressors, pneumatic controllers, and storage containers. Notably, the rule requires Reduced Emissions Completions (REC) or, alternatively, flaring for most fractured wells. REC are predicted to greatly decrease emissions compared to pit storage of liquids and venting of gas to the atmosphere. While emissions from well completions are known to vary, REC are highly effective. At the US national scale, these control measures are predicted to reduce annual methane emissions by 1.0 to 1.7 million short tons, HAP emissions by 12 000 to 20 000 tons and VOC emissions by 190 000 to 290 000 tons. If fully implemented, VOC emissions from newly fractured wells should be reduced by 95%. Reduced methane emissions, although not directly addressed by the rule, are viewed as a "co-benefit". REC are now required in the US for both new wells and those that are recompleted or worked over to improve production.

VOC emission reductions from REC, combined with reductions from other equipment have both economic and environmental benefits.²⁹ Considerable variation among US states remains for permitting guidelines, reporting requirements, leak testing and leak detection and repair programs. Some states are acting to strengthen US EPA regulations. Colorado regulations proposed in November 2013 and recently finalized by the Colorado Public Health and Environment Department reduce risks to human health. The comprehensive set of rules set stringent requirements to monitor, control, and reduce methane and VOC emissions. Particular attention is given to uncontrolled emissions of VOC, including BTEX, from glycol dehydration units. New units have limits on emission rates and distances from buildings and designated outside activity areas. The draft rules (regulation 7) should further reduce air pollution over and above mandatory NSPS and NESHAP rules.33

1.3. Shale gas development in other countries

In the global context, most interest for shale gas development has been expressed within Europe. To date only Poland and the United Kingdom (UK) have performed exploratory shale gas extraction. Preliminary indications are that extensive shale gas resources are present in Europe. The most promising resources have been identified within France (180 tcf), Poland (187 tcf), and Norway (83 tcf). Technically recoverable reserves of 20 tcf were also identified within the UK. These reserves are smaller than the 1275 tcf and 862 tcf estimated for China and the US, respectively, but are still very significant.³⁴

In 2012, the European Commission published three reports that addressed the potential risks and benefits of O & NG development using hydraulic fracturing, with a focus on shale gas.³⁵⁻³⁷ These reviews provided supporting information for development of a European union (EU) regulatory policy for shale gas development. This policy was published in January 2014.³⁸ Prior to publication the regulatory framework for shale gas development was seen as critical for determining if investment will proceed and whether the environment and public health will be protected.³⁹

Rather than propose new legislation, the European Commission published a non-binding recommendation, communication, and impact assessment on the exploration and production of hydrocarbons using hydraulic fracturing in the EU. A review of the effectiveness of this approach is planned in 2015. The adoption of a framework directive setting specific requirements for exploration and extraction remains a possibility. An EU wide regulatory framework would ensure a consistent regulatory environment for developers throughout the EU, while reassuring the public that it has clear and transparent environmental protection.

Although some EU member states are interested in developing their shale gas resources, others are not. Most notably, France has banned hydraulic fracturing. However widespread activity is pending for the UK, with a strategic environmental assessment that aims to provide guidance in advance of additional onshore O & NG licensing.⁴⁰ While proposed new licensing areas cover ~50% of the UK (Fig. 4), the proposed shale gas development for the entire UK is predicted at an upper limit of only 2880 wells. Experience in the US has shown initial estimates can be low compared to actual number of wells drilled.⁴¹

Three recent reports in the UK have, to varying degrees, considered the air quality impacts of unconventional O & NG.^{40,42,43} The possibility of significant negative impacts were noted for greenhouse gas emissions. However, the UK's strategic environmental assessment⁴⁰ did not directly address or even refer to air pollutants such as VOC, ozone or benzene. Human health impacts were not expected to be significant.

In the US, policy is starting to catch up with development activity.²⁹ The accelerated pace of drilling across the US has led to a thorough review of regulations related to O & NG activities. A mandatory, clear, predictable, and coherent approach for the EU was not adopted in January 2014.³⁵ Reporting protocols, assessment methods, and regulatory programs developed in the US could still be applied in the EU and elsewehere.^{36,44} O & NG

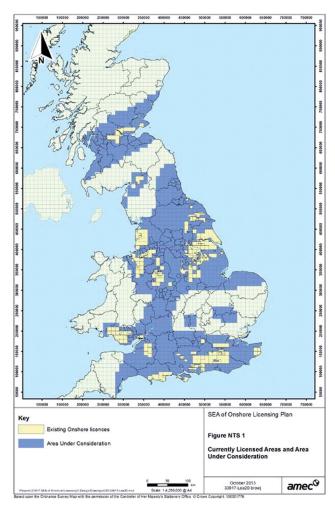


Fig. 4 Shale gas development has the potential to occur throughout the United Kingdom as show by the area under consideration for onshore licensing.³⁷ Courtesy of UK Department of Energy and Climate Change.

products enter national and international supply chains that connect wells to end-users. Emissions happen at all points along this supply chain, starting at the point of extraction. Emissions are largely determined by how efficient the system is – that is, how open or closed the system is to the surrounding environment.

2. Estimating air pollutant emissions

The increased scrutiny placed upon the O & NG industry in the US due to hydraulic fracturing has highlighted the need for a review of current approaches to understanding and quantifying air quality impacts from O & NG emissions.

2.1. Quantifying O & NG emissions through US emission inventories

In the US, emission inventory (EI) based calculations may be used for permitting decisions, enforcement actions, and health risk assessments related to air quality. EI represent a long-term average (usually a year) for a particular source category and may not reflect short-term behavior or extremes. They offer the possibility of estimating current and future emissions for the geographic area and economic sector that they define. Three US emission inventories report emissions from O & NG production: the National Emission Inventory (NEI): the Inventory of US Greenhouse Gas (GHG) Emissions and Sinks (GHG inventory), and the latest, under review, Nonpoint Emissions from the O & NG Production Sector Inventory (Nonpoint O & NG EI).

At face value, it appears easy to add up all the bits and pieces and calculate a bottom-up EI. However, significant issues have emerged for established EI. The US Office of the Inspector General⁴⁵ noted that data in the 2008 NEI were incomplete, with many states simply not reporting key data, leading to underestimation of O & NG emissions. For example, only nine out of 35 states with O & NG development reported any well pad production activities. The deficiency of the 2008 NEI for well completions was noted during NSPS and NESHAP rule making.²⁹ Because of this, an additional 480 000 tons of VOC emissions were added to the original estimate of 21 000 tons. In its 2011 GHG inventory, the US EPA used an average emission factor of 7700 Mcf (1 McF = 1000 ft³) of methane per well completion, more than doubling the previous emission factor.⁴⁶

Reporting requirements and data collection for small point sources that make up area sources pose difficulties due to the overwhelming number that are below permitting thresholds and/or unregulated. Area sources accounted for over 90% of all O & NG VOC emissions for the 2008 NEI.⁴⁷ With recognition that upstream emissions are the dominant O & NG sources and the realization that area sources were not properly accounted, the US EPA⁴⁷ developed the Nonpoint O & NG EI. The new inventory considers emissions at the basin scale while allowing for revisions that consider local conditions. The application of basin-specific emission factors improves the chances that inventory predictions will represent actual emissions.

Emission source categories in the Nonpoint O & NG EI are broadly categorized as follows:

- Engines (including pumps, heaters and drill rigs).
- Tanks (including loading and unloading).
- Dehydration units.
- Pneumatic devices.
- Venting.
- Fugitive emissions.
- Flaring.

Emissions from the listed categories are calculated by combining activity inputs, basin factors and emission factors. The following activity parameter categories are needed to estimate emissions:

- Produced volumes (oil, gas, condensate and water).
- Well counts.
- Well completions.
- Spud counts (the number of wells drilled).
- Feet drilled.

With reliable information for the parameters noted above, calculations can be used to estimate the relative impact of air emissions from different aspects of unconventional O & NG development. Table 1 shows 2011 statewide O & NG production for three US states together with estimated emissions.

There is considerable variation in both absolute and relative amounts of production and emissions among these states. North Dakota production is dominated by the oil-rich Bakken Shale formation, and the relatively low methane emissions reflect reduced gas handling with flaring at production sites. Pennsylvania production is dominated by dry natural gas from the Marcellus Shale formation, whereas Wyoming production has a greater contribution from wet gas. Table 1 shows higher methane and NO_x emissions for Pennsylvania compared to Wyoming for given production rates. Increased NO_x is due to additional compression emissions, while higher methane emissions are likely due to more fugitive emissions.

On a state-by-state basis the level of emissions is a function of the level of production with differences due to basin specific characteristics that influence contributing sources. However, some key processes are known to be responsible for the bulk of emissions, notwithstanding the issues associated with quantifying emissions from completion activities and produced water handling. Compressor engines are the most significant source of NO_x accounting for 70% of emissions.⁴⁷ Up to 75% of VOC emissions are estimated to be from well pad storage tanks and pneumatic devices at production units.44 While a number of fugitive sources emit methane, over 50% of methane emissions are from pneumatic devices.⁴⁷ Pneumatic device losses are often not part of the financial accounting chain and as such they have received less attention than midstream fugitive losses. Dehydration units account for 40% of HAP emissions at well pad production sites.⁴⁷ The US EPA Nonpoint O & NG EI gives a clear indication of the relative magnitude of air emissions for different sources and thereby helps identify air quality concerns.

An EI is a construct that has great value as a predictive tool. As they are utilized to tabulate annual average emissions they may be less effective over short time frames. While consistency of emission estimation is important, there are two fundamental problems with understanding air quality impacts from emerging O & NG developments – timeframe and accuracy. The timeframe of an inventory will not represent the pace of development in a given area, the emergence of air quality issues, or the complexity of real world operations.

Table 1 Selected state-wide O & NG production and pollutant emissions for 2011^{*a*}

	State		
Parameter	North Dakota	Pennsylvania	Wyoming
Oil (bbl)	151 156 326	2 108 613	49 985 043
Casinghead gas ^b (Mcf)	133 023 400	10 333 040	156 430 973
Gas well gas (Mcf)	23 636 077	1 307 030 772	2 218 414 912
Condensate (bbl)	1 275 296	1 201 918	11 951 745
$NO_x (tpy)^c$	13 578	104 068	58 434
VOC (tpy)	95 642	191 893	186 141
Methane (tpy)	41 392	708 303	462 056
Total HAP (tpy)	2168	8571	10 622

 a Abridged from US EPA Nonpoint O & NG EI. ^4 $^b\,$ The gas produced from an oil well. c Tons per year. The ability of EIs to accurately estimate emissions from the O & NG sector has been questioned by independent, measurement-based approaches that have identified higher methane leakage rates than predicted by inventories for developments throughout the US. Using methane measurements from aircraft, towers and ground networks, combined with wind field data, it appears that the US is emitting more methane than predicted by EPA emission inventories, and the Emissions Database for Global Atmospheric Research (EDGAR).^{48,49} While fossil fuel extraction and processing was the most important methane source ($45 \pm 13\%$), it was calculated to be 4.9 ± 2.6 times larger than EDGAR estimates. This discrepancy contradicts the recent 25-30% downward revision of methane emissions in the EPA NEI.⁴⁹ Underestimation of US O & NG emissions of methane was noted as early as 2002.⁵⁰

Inaccurate data can lead to incomplete analysis of human health risks and inappropriate emission control strategies. The US EPA uses the NEI to assess risks from HAP. States use the NEI to inform their plans for compliance with NAAQS. States are compelled to use published emission factors for new source review decisions and calculated emission inventories for permitting of major sources.

2.2. Validation of emission inventories through independent analysis

Given the reliance on bottom-up EIs for air quality management, regulators, O & NG developers, environmental organizations, academic intuitions and research groups have started to collaborate to improve published emission factors and determine uncertainties.⁵¹ A perfect EI would account for all sources within the area it represents and the factors used would accurately reflect the activities and processes considered. Notwithstanding the issue of inventory timescale and reporting approach, factors will not be representative of damaged or poorly maintained equipment, or accidents that contribute to emissions. Air quality measurements can serve as important validation methods⁴⁸ and define actual air quality impacts. The use of onsite,⁵² offsite,⁵³ airborne⁵⁴ and satellite monitoring methods⁵⁵ are creating new data that can be used to improve existing inventories.

2.2.1. Onsite measurements. Onsite measurements require a strategy that captures a representative sample of the targeted process. Field measurements of operating equipment at well pads have revealed that some emission factors are close to inventory values while others are not. Enclosure sampling has shown pneumatic devices operating at much higher than anticipated emission rates.⁵² Direct sampling techniques are time consuming and costly, and while data reflects actual operational behavior, sample sizes are limited and operator cooperation is required. It has been recognized that additional methods for determining emissions should be developed.⁴⁵ Mobile monitoring or surveying has emerged as a complementary approach to onsite studies that have numerous difficulties including site access, safety and cost.

2.2.2. Offsite mobile surveying and plume quantification. Offsite tracer studies are an established method for estimating

downwind emissions from natural gas operations.⁵⁶ While tracer release studies are effective at quantifying emissions, access to emission location is required.57-59 The development of robust, fast-response methane analyzers has led to mobile monitoring that has revealed emission plumes throughout O & NG developments. Several recent studies have reported high concentrations of methane and VOC in basins with intense O & NG extraction. Mobile monitoring is now widespread in the US including projects in the Pinedale Anticline in Wyoming,60 Barnett shale in Texas,⁶¹ San Juan Basin in New Mexico,⁶² Marcellus shale in Pennsylvania,63 Denver-Julesburg Basin in Colorado48 and the Uintah Basin in Utah.64 But, often, mobile surveying studies have been unable to make quantitative estimates of which sources are the largest emitters based on concentration alone. This has led to renewed interest in quantifying emission plumes using remote measurements. The US EPA recognized the need for plume assessment and developed a remote emission quantification methodology.53 The EPA approach allows for estimation without the necessity of tracer releases from the target source. As such, site access is not necessary and data collection can be conducted independent of developer oversight. This methodology can provide ground truth evaluation of different processes to determine emission rates for comparison to EI emission factors. These methods can also characterize pollution hotspots and evaluate emission variability among similar well pad operations.

2.2.3. Airborne surveying. With appropriate meteorology, aircraft measurements provide the possibility of measuring basin wide emission fluxes. A mass balance method determines the total flux of methane in a given basin⁵⁴ that is directly comparable to EI data. The first published assessment of methane emissions for the Unitah Basin in Utah revealed an emission rate of between 6.2% and 11.7% of total gas production.⁵⁴ This entire range is significantly higher than the US EPA EI estimate of 2.4%.65 Additional airborne campaigns led by the National Atmospheric Oceanic and Atmospheric Administration (NOAA) have been performed to determine basin-wide methane fluxes across a number of US locations These basins have many similarities, including product compositions and management approaches. Flux measurements will be compared to EI predictions and are awaiting publication.66,67 These airborne assessments include the Haynesville shale in eastern Texas and western Louisiana, the Fayetteville shale in northern Arkansas, the Marcellus shale in western Pennsylvania, the Denver-Julesburg Basin in Colorado and the Barnett shale in Texas. Basinspecific characteristics for VOC have been reported, reinforcing the importance of location specific assessments.68,69

2.2.4. Space-based surveying. The utility of space-based sensors to inform near-ground concentration and spatial distribution of trace atmospheric constituents is limited by spatial resolution and lack of vertical sensitivity. Nonetheless, satellite data is now informing understanding of emissions from O & NG emission sources. Preliminary analysis of the Tropospheric Emission Spectrometer data on the Aura satellite from 2006 to 2012 has shown evidence of increasing methane concentrations over the Marcellus shale in Pennsylvania.⁵⁵ As satellite data is able to provide rapid regional scale information

it has utility for identifying areas with elevated concentrations. The NASA DISCOVER-AQ (Deriving Information on Surface Conditions from Column and Vertically resolved Observations Relevant to Air Quality) project is validating the use of satellite data. In the summer of 2014, ground, airborne and space-based sensors will be combined in an assessment of the Denver Julesburg Basin in Colorado.⁷⁰ The Franco-German MERLIN (Methane Remote Sensing Lidar Mission) is planned for launch in the 2016 and will provide measurement quality suitable to assess ground based anthropogenic emissions, including O & NG developments.⁷¹

3. Air quality concerns

While estimation of the magnitude of pollution from O & NG activities through EI validation and analysis is important as a tool, it is a "means to an end". Even if perfectly constructed, an EI is idealized and cannot account for the complexity encountered in the physical world. For that we need measurements of pollution in the environment. While there are also complications associated with monitoring we get closer to understanding pollution when we measure the air we breathe.

Air quality impacts are complex, as emissions from O & NG sources vary in composition, magnitude, and duration. Emissions are dependent upon raw product composition, extraction, and handling approaches. Emissions from O & NG development are inevitable. While drilling and completion activities for a particular well will result in a pulse of emissions for weeks or months, production activities can last from years to decades. The impact of these emissions on air quality depends on the geographic context. At the regional scale, emissions from a single well are insignificant, but at the local scale they could dominate air quality. As with any air pollution issue, transport and dilution is critical to determine the downwind impacts that depend on the distribution and behavior of contributing emissions.

Emission inventories can predict the contribution of O & NG sources, but without measurements, actual impacts may remain unknown. While it is impractical to locate air quality monitoring stations next to every well pad, it is reasonable to perform additional monitoring to produce statistically sound predictions of the influence of O & NG emissions upon air pollutant concentrations.72 Air quality networks are sited with respect to population density and, while expensive, additional stations can be installed at locations influenced by O & NG sources to determine any impact on air quality. Along with stationary air quality monitoring networks, it is possible that additional ambient measurements can be acquired through mobile monitoring techniques to address potential concerns at specific locations. This approach can identify specific areas of concern in context with longer-term established network monitoring locations. Additionally, personal exposure monitoring can be conducted to determine actual inhaled doses as part of a strategy that includes a health impact assessment. The primary concerns for those interested in air quality are for air pollutants that are either regulated, hazardous, or act as climate forcing agents. These include ozone, BTEX, and methane, respectively.

3.1. Regulated pollutants

O & NG emissions can contribute additional concentrations of the following pollutants, all of which have set ambient air quality standards:

- Ozone.
- Nitrogen dioxide.
- Particulate matter.
- Benzene (EU not US).
- Carbon monoxide.
- Sulfur dioxide.
- Benzo[*a*]pyrene (EU not US).

These pollutants are regulated due to established health impacts and are controlled to ensure protection of public health.⁷³ By virtue of being regulated by ambient air quality standards, measurements are required at quality-assured air quality monitoring stations. As a result of the Clean Air Act, a general decline of ambient concentrations of regulated pollutants has been observed in the US.73 However, it has been noted that in some areas of Pennsylvania this trend is being reversed for nitrogen dioxide (NO₂) downwind of shale gas development areas.74 Trend analysis of pollutant concentrations can enable understanding of the behaviour of particular emission sources, in particular when they are the dominant source.⁷⁵ Ozone (O_3) and NO₂ pollution is of particular relevance given the predicted high levels of precursor emissions from O & NG sources. Significant NO_x emissions come from drill rigs, pumps and engines at the well pad, and also from associated mobile sources, in particular truck traffic. Emissions of VOC, including benzene, have a contribution from engines but well-pad emissions are most often dominated by various production activities. Ratio analysis of various VOC and methane has been used to identify the contribution of emissions from O & NG sources.72,76,77 Certain VOC are markers for O & NG activity (e.g. propane) while others are associated with traffic sources (e.g. ethyne). Analysis of the composition of measured air, in particular when compared to known emission profiles or signatures, can reveal the contribution of different sources to ambient air. Such analysis is complicated when there are many contributing emission sources but the importance of O & NG emissions can be determined. In northeastern Colorado O & NG sources were estimated to account for over 50% of the ozone production potential from VOC.72

The phenomenon of wintertime O_3 has been directly linked to emissions from O & NG sources in remote locations in Wyoming⁷⁸⁻⁸⁰ and Utah.^{81,82} In the Upper Green River Basin of southwest Wyoming, air quality degradation from O & NG developments was not predicted through modeling. The discovery, through air quality monitoring was a surprise. High O_3 levels have traditionally been thought to only be a summertime problem. In the Upper Green River Basin in Wyoming, O & NG emissions can contribute close to 100% of the O_3 precursor pollutant burden (Fig. 5).⁷⁸ Rural communities unaccustomed to air pollution have produced a new generation of environmental activists motivated by exposure to elevated ozone produced from O & NG emissions. Close to the town of Pinedale, Wyoming (population around 2000), during the winter of 2011 O_3 levels rose to levels higher

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than those reported during the summer of the same year in the Los Angeles Basin. While a reduction of precursor concentrations in the Pinedale Anticline is coincident with a decline of ozone pollution in Wyoming during 2012 and 2013, severe winter episodes persist in the Uintah Basin in Utah. During 2013, O_3 concentrations in the Uintah basin reached 165 parts per billion (ppb) due to the presence of a strong temperature inversion.⁸³ These situations are somewhat unique due to geography, weather patterns and chemistry. O & NG emissions associated with strong temperature inversions lead to the accumulation of high levels of VOC and NO_{x^*} . With snow cover accentuating solar radiation and providing the basis for unique nitrogen chemistry,^{80,84} rapid production of O_3 can result in extremely elevated concentrations.^{85,86}

At the regional scale, the influence of O & NG emissions on summertime O₃ formation is more significant. In more populated areas O & NG emissions may comprise a small contribution to the overall pollution burden, *e.g.* Marcellus shale activities are estimated to account for 10% of regional O₃ precursor emissions,⁸⁷ but relatively small additions of precursors can significantly impact downwind ozone concentrations. Higher peak O₃ levels have been predicted from the Haynesville shale⁸⁸ and the Barnett shale in Texas.⁸⁹ Recent EI revisions suggest impacts, showing the importance of the balance and magnitude of precursor emissions.⁹⁰ Emissions from the Eagle Ford shale contribute to peak O₃ concentrations at the downwind cities of Austin and San Antonio, Texas.⁹¹

3.2. Hazardous air pollutants

Hazardous air pollutants (HAP) are not regulated in ambient air in the US pursuant to a NAAQS. They are regulated in the US through emission standards (section 112 of the Clean Air Act). While US EPA lists 187 different regulated HAP, the following are known to be associated with O & NG emissions:

- BTEX.
- Formaldehyde.
- Hydrogen sulfide.

• Others including certain VOC (*e.g. n*-hexane) and polyaromatic hydrocarbons.



Fig. 5 The Jonah field development in the Upper Green River Basin, Sublette County, Wyoming is expected to produce gas for up to 50 years from 1500 wells. Photo courtesy of Skytruth.

BTEX emissions are often the most prevalent HAP from O & NG activities. While there is an extensive body of knowledge relating to the impact of HAP, there have been few health impact assessments of population exposure to O & NG activities. In Colorado, a health risk assessment has shown significant differences of risks from exposure to O & NG emissions for people living within half a mile of well pad completion and production activities, compared to those living farther away.92 Exposure to aromatic and aliphatic hydrocarbons from natural gas operations, in particular well completions, were contributors to non-cancer hazard indices (neurological and respiratory impacts). Benzene was the most important compound for estimated cancer risks.92 In contrast, other researchers have reported no health concerns related to HAP associated with shale gas development based on annual average concentrations from monitoring stations around the Barnett shale.93 This difference relates to a number of factors but perhaps most importantly, reflects the difference between considering exposures from the "fence line" with "background" concentrations. Fence line exposure refers to pollution levels in the area of O & NG well pads not accessible to the general public. In addition to drilling and completions, exposures from proximity to key well infrastructure (e.g. dehydrators and pneumatic values) and during well maintenance processes can be also be expected.⁵² A need for better information regarding actual human exposure has been noted,94 in particular for situations close to drilling and completion operations.95 Quantification of health impacts is fraught with difficulties and epidemiological studies require extensive time periods and sample sizes. In the future any health impacts are most likely to be detected within the most exposed groups,96 for example O & NG field workers and people living in close proximity to development. Surveys of self-reported health-related symptoms in Pennsylvania have been used to develop an understanding of potential associations with O & NG activities. Symptoms were most prevalent for respondents living within 1500 feet of a natural gas facility.97 Emission control regulation in the U.S is predicted to be highly effective at reducing HAP emissions from O & NG development. Adoption of US EPA's NSPS and NESHAP emission control regulations for the O & NG sector should not only reduce product losses and improve revenue, but also reduce impacts for those living near O & NG production.29 Recently updated regulations in the state of Colorado recognized the likely significance of population exposure from O & NG facilities. To protect public health the minimum distance, or setback, between O & NG facilities and occupied buildings to a distance of 500 feet, with this value increased to 1000 feet for high occupancy buildings, including schools.98

Exposure to HAP is difficult to predict, in particular for acute exposures from intermittent sources. Localized HAP exposure assessments may be needed to support forthcoming health impact assessments. In the EU, the regulation of benzene in ambient air is already in place. Because EU air quality networks already exist, expansion to address air quality concerns from new O & NG development should be relatively straightforward. Health impact assessments benefit from measurement-based approaches that more accurately define actual exposures.

3.3. Potential improvements in air quality

While there are numerous negative impacts on air quality from O & NG development, there are also significant positive impacts if the produced natural gas is used to replace coal for power generation. It has also been postulated that the increased use of natural gas will reduce regional O₃ in Texas due to reduced power plant emissions as natural gas replaces coal, despite increased emissions associated with O & NG production.99 Reduction of NOx, particulate matter, black carbon, sulfur dioxide, mercury and other heavy metal emissions by using natural gas compared to coal for electricity generation is also significant at the regional scale.¹⁰⁰ The use of natural gas in power generation with combined cycle technology has been calculated to emit 44% less carbon dioxide than coal-fired power plants.¹⁰¹ For 2012, carbon dioxide emissions from all fossil fuel fired power plants were 23% lower than the carbon dioxide emissions estimated from coal-fired power plants in 2007. The trend of the replacement of coal with natural gas continues in the US.4

3.4. Climate forcing pollutants

There is an on-going debate about the costs of methane leakage and the benefits of replacing of coal with less carbon-intensive natural gas for generating electricity.^{102,103} This is important when considering the end use of natural gas, as the benefits over the use of coal are reduced and perhaps nullified by high leakage rates, as methane is a strong climate forcing agent.¹⁰⁴

Given the significance of climate change, this debate is critical. To varying degrees, the following climate forcing compounds are associated with O & NG emissions:

- Methane.
- Carbon dioxide.
- Black carbon.
- O₃

In 2009, a methane loss rate of 2.4% was estimated for the O & NG industry by the US EPA.66 This amounts to nearly 40% of US methane emissions, with natural gas operations contributing nearly 90% of the emissions from the O & NG sector. However, noting the significance of emissions from completions, when production volumes are at a peak, researchers have postulated much higher emissions for unconventional compared to conventional wells.105 The loss rate of methane from venting and leaks over the course of the lifetime of a shale gas well was estimated to range from 3.6% to 7.9% of total production from a typical well. This rate proposed in 2011 has been described by others106 as "unreasonably large and misleading". However, it is now generally accepted that larger emissions of VOC and methane are associated with unconventional natural gas development.¹⁰⁷ It has become evident that the fugitive emissions predicted by applying US EPA emission factors does not, to date, match estimates using ground level and airborne measurements48,54 or inverse box modeling.108 Comparing different methods for estimating natural gas emissions is difficult due to inherent methodological differences, including processes addressed, timescales considered and geographic boundaries. Terminology is also a problem, in particular "leakage rate". This

term is not the same as fugitive emissions and is critically dependent upon whether upstream, midstream and downstream emissions are considered. National scale EI factors may also better represent general rather than specific basin scale behavior. Production emissions are variable between developments due to geology and developer practices. It is important that the appropriate contextual information accompanies emissions estimates as well as clear definition of what is considered as being losses and leakage. Metering of lost and unaccounted for natural gas can add further confusion. The importance of reconciliation between the results gained from different approaches for estimating emissions is now widely recognized.¹⁰⁹ Better estimates will require collaboration between different stakeholders, in particular regulators, developers and scientists. The development of common strategies is proceeding with a number of collaborative projects. The need for validation and improvement of emission inventories and as well as closure between emission estimates for the O & NG sector has recently been noted.¹⁰⁹

Despite uncertainty regarding fugitive and leakage emission rates, natural gas is viewed as a clean-burning, bridge fuel that can reduce greenhouse gas impacts.¹¹⁰ Natural gas has a smaller carbon footprint compared to coal when end use is considered.¹¹¹ A popular argument is that natural gas use allows for continued use of fossil fuels while reducing greenhouse gas emissions as part of a transition towards a low carbon economy.¹¹² Of course the benefits of switching from coal to natural gas are negated if replaced coal does not stay in the ground.¹¹³ Without a global climate policy that aims to reduce greenhouse gas emissions, unconventional O & NG exploration and production is adding to potential cumulative carbon emissions and thereby the risk of climate change.⁴³

4. Discussion

Air quality concerns exist from local to global scales. At the local scale exposure to hazardous air pollutants associated with drilling, completion, and production activities is of most concern. If wet gas is produced, then higher BTEX emissions are likely. Local considerations are connected with regional ones. The replacement of coal with natural gas in power stations can improve regional air quality. It is also evident that the production and use of natural gas can have negative impacts on air quality. At the regional scale O & NG emissions contribute to secondary pollutants, notably NO₂ and O₃. At the global scale O & NG emissions contribute to methane, a climate-forcing agent, concentrations.

The end-use benefits of replacing of coal with natural gas should not be taken to mean acceptance of exploration and production emissions that could be controlled. Scientists, engineers and regulators, not to mention politicians and various members of the public including industry and environmental interest groups have different perspectives on the influence of unconventional O & NG upon air quality. Each group has different motives, approaches and understanding of air quality impacts.¹¹⁴ Consensus can be hard to find when considering the complexity of understanding the influence of O & NG emission sources on air quality. A member of the

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general public may be concerned about the influence of a specific well pad upon the air that they breathe. One well pad, while important, is a small part of the wider responsibility of both developers and regulators. Development activities and regulatory oversight are constantly evolving and common practice in one area may be unknown elsewhere. Regulation will never guarantee zero emissions so the issue becomes what is an acceptable level and what impact will that have. As noted previously, recent US O & NG regulations are predicted to be highly effective. However, some sources are known to be highly variable and others will have high emissions despite the use of best available controls.

Defining impacts can also be an area of confusion. Even if air quality for an area is in compliance with standards, people in the vicinity of development may be exposed to levels of pollution that impact health. Some O & NG developments in the US are situated in relatively unpopulated locales. A recent epidemiological study of birth outcomes in rural Colorado found that congenital heart defects, and possibly neural tube defects, were more likely with births from mothers who live in areas with a high density of natural gas wells compared to those that live in areas with no natural gas wells.115 This study reinforces the need for conducting additional research to gain better estimates of actual exposures and potential associated health effects. When O & NG developments are located nearer to urban populations more people may experience health impacts from pollution exposure. As any pollution exposure issue is also a social justice issue, the question becomes what level of protection is afforded to the impacted population by regulatory agencies. While population exposures have been estimated we are unaware of any exposure studies using personal sampling methods to measure inhaled doses. A recent review of risks to public health confirmed that to date no comprehensive population-based studies of the public health effects of unconventional natural gas operations exist.¹¹⁶ The frequency and duration of human exposure to O & NG pollutants for workers and communities was noted as a major uncertainty.

Scientists have a wide range of interests, from concerns about the validity of emission estimates to understanding health impacts from unconventional O & NG activities. The latter would be improved with personal exposure monitoring. Discrepancies between air quality measurements and inventory-based emissions calculations need to be rectified. Since 2007 the rate of increase of methane concentrations in the global atmosphere has accelerated and some see unconventional O & NG development as one of the possible causes.117 There is more reason than ever to reduce emissions and quantify impacts. There is a strong need for implementation and enforcement of new and more effective regulations, supported by monitoring programs, to reduce emissions and protect public health. The approach in Colorado with enhanced leak detection and repair is leading the way for other US states. As with other air quality regulations, economic and environmental benefits are significant for GDP and public health.¹¹⁸ When impacts are likely, as shown by some developments in the US, a precautionary approach based on existing knowledge may be preferable to a laissez-faire stance, with action occurring only later if problems are identified.

5. Translating the US experience

For those looking to the US experience with unconventional O & NG development there is a wealth of information available. A few steps could be applied to effectively assess and mitigate potential air quality impacts from unconventional O & NG development.

• Define regulations. Consider regulations based on those published in the US, in particular US EPA NSPS and NESHAP and the state of Colorado's O & NG rules.

• Calculate potential emissions. US EPA inventory tools can be used to estimate the magnitude of pollution attributable to unconventional O & NG sources.

• Develop location-specific case studies. Estimate the contribution of unconventional O & NG sources to the total pollution burden.

• Evaluate air quality monitoring network coverage. Assess the applicability of established sites for determining potential air quality impacts of unconventional O & NG gas sources.

• Add equipment and/or network stations to determine air quality impacts from development activities. This may require targeted air quality measurements including innovative mobile techniques.

• Require the best emission controls and instigate programs of leak detection and repair.

• If necessary, undertake health impact risk assessments, especially in populated areas. Such work could include population exposure evaluations.

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