Subsurface Methane Leakage in Unconventional Shale Gas Reservoirs: A Review of Leakage Pathways and Current Sealing Techniques

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Abstract

Shale gas extraction is seen to be a bridge fuel to the future due to lower GHG emissions compared to oil. However, it is also one of the most controversial topics due to the involvement of fracking in their production. Based on the analysis performed in this review we found that despite hydraulic fracture propagation being a possible conduit of methane leakage, the major cause of gas leakage is through leaking wells within the vicinity of fracturing sites. Remedial attempts have revealed promising yet inconsistent results, with no concrete method established for the methane leakage mitigation from shale gas wells.

Keywords

Methane leakage, Shale gas, Greenhouse gas emission, Hydraulic fracturing, Aquifer contamination, Unconventional Reservoirs

Introduction

Production from conventional fossil fuel resources is decreasing as these reserves continue to deplete, on the other hand the demand for energy is ever increasing. Natural gas has recently gained significant interest as a “bridge fuel” to the future that will develop energy security and reduce dependence on conventional oil and coal resources [1]. With further prospect of a cleaner burning fuel, natural gas has the potential to provide immediate climatic benefits. Shale gas reserves have been termed the energy of the future, due to the fact that the combustion of gas releases significantly less carbon dioxide (CO_2) compared to oil and coal [2]. On the other side, there are concerns associated with the release of natural gas such as methane to the atmosphere and contamination of ground water through leakage process during its production. It is important to understand how critical such environmental concerns are, and what would be the overall impact of production and utilising natural gas on our health and environment. In this study we summarised the studies conducted on the concept of methane leakage through fracking process and concluded how possible sources of methane leakage can be controlled. Therefore, despite its advantages, the extraction from shale gas reservoirs remains to be an

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ongoing environmental debate on risks and advantages associated with its production. Opposing arguments are mainly based on the environmental concerns and health risks, posed by the uncontrolled release of gases such as methane (CH_4) through fracking process [2]. The cause of methane leakage from oil and gas exploration have been directly attributed to unconventional extraction of shale gas via hydraulic fracturing stimulations. With uncertainties in the extraction process, pro-fracking groups emphasize on the safety of hydraulic fracturing, whereas opposing parties base their arguments on the uncontrolled nature of fracture propagation resulting from hydraulic fracturing. In theory, hydraulic fracturing has the potential to provide methane migration pathways via the intersection of naturally present geological faults in the subsurface, also leakage may happen via inadequately abandoned oil and gas wells [3]. The latter refers to current well abandonment practices which involve setting a series of cement plugs deep inside wells to restrict flow of hydrocarbons [4]. The cement commonly used for this process (Portland cement) readily undergo chemical degradation with time in the presence of various substances such as carbon dioxide (CO_2) [5-11]. The presence of CO_2 can be from naturally occurring geological sources or from the injected carbon dioxide during carbon capture and storage (CCS) process in depleted oil and gas reservoirs. Therefore, in cement based well abandonment procedures, CO_2, degrades cement and forms conduits for gas escape. Carey et al. in 2007 found that CO_2 leakage through casing-cement and casing-shale formation happened during CO_2 sequestration process, and they concluded cement in contact with CO2 was heavily carbonated and created a pathway for CO_2 migration [6]. In terms of shale gas extraction, instances of propagating fractures intersecting wells with reduced integrity may lead to migration of methane towards leakage pathways. Furthermore, for economic reasons, abandoned wells are regularly used to extract groundwater which is fed directly to domestic and commercial water supply lines that create a direct link for methane to invade groundwater reserves and its escape into the atmosphere [1].

Thus, the extraction of shale gas remains debatable. Some of the pros and cons of shale gas resources as a source of fuel are summarised in Table 1.

Table 1: The advantages and disadvantages of shale gas production and extraction [1], [5]

Advantages	**Disadvantages**
Burns cleaner leading to low CO_2 emissions compared to oil and coal Vast global reserves waiting to be tapped Cheaper fuel alternative to coal Creates more jobs as new reservoirs being tapped Provides energy security and aids the advancements of developing countries Enables the high CO_2 emitting countries to reduce emissions	Leakage of methane leads to environmental benefits being nullified Hydraulic fracturing process generates high levels of waste water Leakage pathways allow subsurface aquifer contamination Hydraulic fracturing is claimed to increase regional tectonic activities (earthquakes) Provides hindrance in advancements of renewable energy sector

Despite the reduced CO_2 emissions and numerous economic advantages of shale gas extraction, the possibility of methane leakage with already growing concerns of global warming remains to be a hindrance in widespread shale gas development. Methane is a highly potent greenhouse gas (GHG) with a global warming potential (GWP) 72 times more than CO_2 [6]. In April 2011, Howarth et al. stated in their report that the footprint of GHG from shale gas resources was approximately 20% greater than that of coal, and the sheer amount of emissions to date in 2011 suggests that the climatic benefits of using natural gas have already been eliminated [7]. Similarly, in 2015 Howarth [1] further argued that due to the potency of methane as an environmentally detrimental substance, the benefits of shale gas resources, both commercial and economic, are quashed by the amounts of methane leakage from unconventional wells [1]. In addition to contamination concerns, the complications of shale gas extraction and the associated problems, stem back to a lack of understanding of the leakage mechanisms and complex geological systems. Limited number of published documentation is available [1, 4, 5, 7, 12-17] and some of them provide contradictory information. High costs and difficult data collection methods have further limited the reliability of collected data and as a result, all national estimates of methane leakage quantities come from the extrapolation of regional data. Furthermore, any advancements in shale gas extraction have predominantly been in the US [7, 18-20], thus further reducing the area of study to a single region.

The aim of this review is to provide an understanding of the concerns of methane leakage from shale gas extraction. The review outlines the sources and quantities of methane leakage as a contaminant gas from the exploitation of shale gas reserves and further explores the current methods to record and mitigate the leakage of subsurface gases. However, with limited information available for the remediation of subsurface methane leakage, some references will be made to the leakage of CO_2 from CCS (Carbon Capture and Storage) studies.

Sources of methane leakage

The major sources of methane leakage can be split into two categories. The first category is the propagation of hydraulic fractures and how they interact with naturally occurring geological features, and with man-made subsurface features, such as conventional wells. The other sources of methane emissions are related to venting and flaring activities of gas well operators [6, 19].

A study conducted by Zoback et al. [13] in 2010 stated that the major concern surrounding shale gas production was the possibility that the subsurface fracturing operations may extend beyond the target formation and form a link to shallow aquifers. Despite being considered theoretically possible, the presence of geological layering in the overburden strata suggests that the unaided propagation of fractures thousands of feet upwards is highly unlikely. This statement was analysed by Zhang et al [2] in 2015, who came to similar conclusions, stating that a more realistic means of leakage may occur from induced fractures extending to natural faults in the subsurface. The viability of natural features providing a means of contaminants migration can be seen from the occurrence of thermal springs. With the consideration of a long geological timescale, deeply seated circulation of steam shows that the communications between the subsurface and surface are realistically possible. One such documented case is the Canadian Rocky Mountains. A study in the mountain area was undertaken by Grasby et al [14] in 2016 to assess the occurrence of methane within spring waters. The study stated that the temperature of each spring was directly correlated to the circulation depths, with temperatures ranging from 30^{o}C to 118^{o}C in the region, and methane quantities fluctuating between 0.00480-0.361% of total gases. However, one case, the Toad river spring, showed up to 23.3% methane is present in samples, with max temperatures of 118 ^{o}C and circulation depths of 3.8km. An isotopic analysis of the water from the Toad spring showed high levels of carbon isotope 13 based methane, $\delta^{13}C_{CH4}$, suggesting the presence of the gas was mainly from thermogenic sources, resulting from the decay of organic matter [14].

The high level of methane and deep circulation depths of the spring channels suggests that the circulation path may have intersected a dense network of naturally occurring fractures in the subsurface. Springs showing trace amounts of methane also demonstrate shallower circulation depths, while deeper circulation channels display higher methane percentages. This suggests that the geological features intersecting subsurface spring channels are mainly located at increased depths, such as the 3.8km deep channel circulation in the Toad spring. Furthermore, the scattered occurrence of naturally deformed basins in the region may have amplified the subsurface intersection of spring circulation paths with methane sources. It is important to note that the spring water samples that were collected, showed the presence of microorganisms in them, which are called methanotrophs. Methanotrophs are microorganisms which thrive in anaerobic, methane rich environments and oxidise methane to CO_2 by 10 – 90% [14]. Thus, the true amount of methane present in the water samples cannot be conclusively stated.

The study conducted in the Canadian mountains displays the theoretical possibility of induced and naturally occurring fractures interacting to form leakage channels. Thus, the presence of a fracture network within the vicinity of an induced fracturing site poses the risk of leakage pathway creation. Furthermore, many formations contain dormant natural fractures, filled with calcite or quartz composition cement that may act as planes of weakness and points of fracture propagation [3]. With concerns of regional stress redistributions caused by induced fracturing operations, the reopening of inactive fractures poses the concerns of pathways extending beyond intent. However, a study on the behaviour and response of fracture propagation in cement blocks by Deghan et al. [3], stated that the feasibility of fracture propagation via the interaction of induced fractures with natural fractures is only possible if the strike and dip angles (strike angle refers to the direction of a fault line with respect to magnetic North, Dip is the angle between the horizontal plane and the fault plane) of the natural fractures allow the stress to be transferred further into the formations. Despite the presence of required conditions for fracture interaction, the viability of the scenario is shown in the study conducted by Warner et al [15] on the Marcellus shale play of northeast Pennsylvania. The study found that shallow aquifer water had consistent geochemical properties with deep seated, high saline aquifers. This indicates that a naturally occurring communication path may already be present in the subsurface and the occurrence of even minimal fracture propagation may lead to the creation of leakage pathways extending to shallow aquifers [15]. However, with advancements in technology, operators today have the means to model induced fracture zones to some extent and thus control the lengths of fractures. In a 15-year simulation study on the Eagle Ford shale play, Brownlow et al. [16] stated that outside the intentionally Stimulated Rock Volume (SRV), the propagation of fractures was negligible. Investigations were conducted to understand the uncertainties in the migration of subsurface methane at Lawrence Berkley National Laboratories in the U.S [17, 21]. They found that the sources of methane migration are poorly designed hydraulic fractures, induced fractures intersecting natural fractures within the subsurface, and the occurrence of low integrity wells [2, 17, 21]. These scenarios are appropriately illustrated in Figures 1.

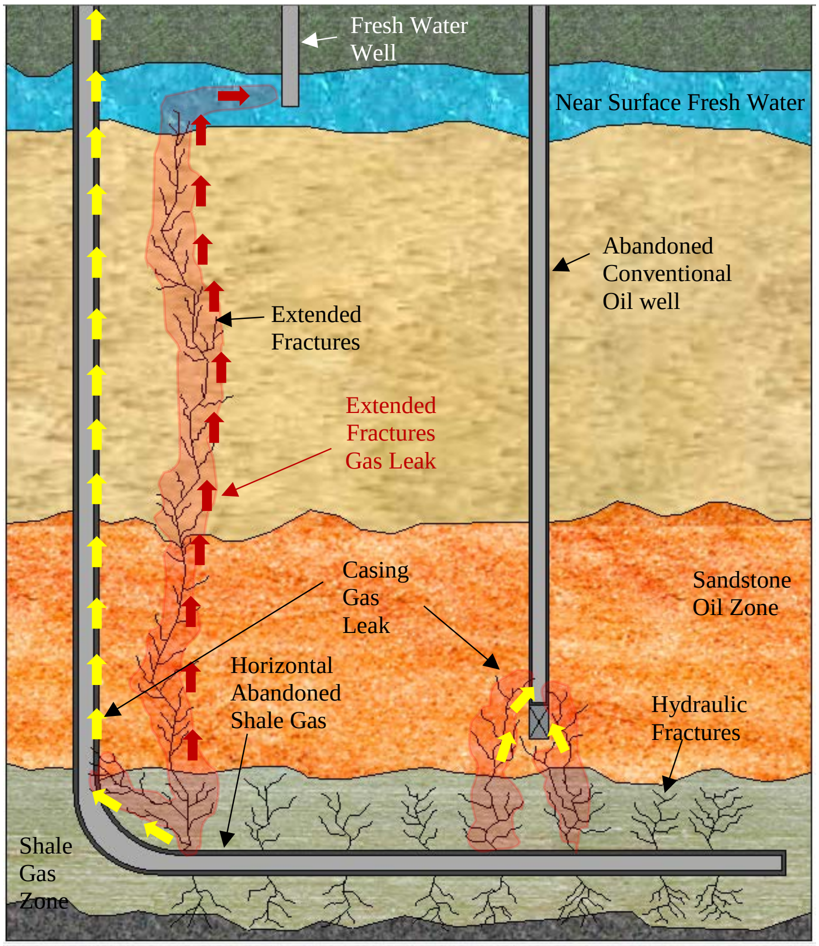

Figure 1: Poorly designed hydraulic fractures intersected an abandoned conventional oil well and formed direct links to the surface and near surface fresh water via fracture propagation

As shown in Figures 1, abandoned oil and gas wells with low structural integrity pose a higher risk of contaminants migration than naturally occurring leakage conduits. Jackson et al. [4] stated in their research that poor well integrity presents a more viable cause of methane migration compared to stress redistribution implications of hydraulic fracturing. Prior to advancements in oil and gas technology, the drilling of exploration wells was done profusely in search for hydrocarbons. In Texas alone, over 1.1 million wells were drilled before the first commercial well was commissioned. Other regions similarly exhibit over 75% of drilled wells in the area to be abandoned. Failure of well integrity can be attributed to leaking completion components, cement deterioration and the corrosion of steel casings [22]. Furthermore, for economic reasons, many abandoned wells are being commissioned for water extraction for domestic supply [16]. This poses a risk of freshwater contamination as in the UK alone, out of the 2152 wells drilled for oil production and exploration, 428 penetrate highly productive subsurface aquifers [22]. Figure 2 illustrates the possible leakage channels that may be created within cement and casing failures.

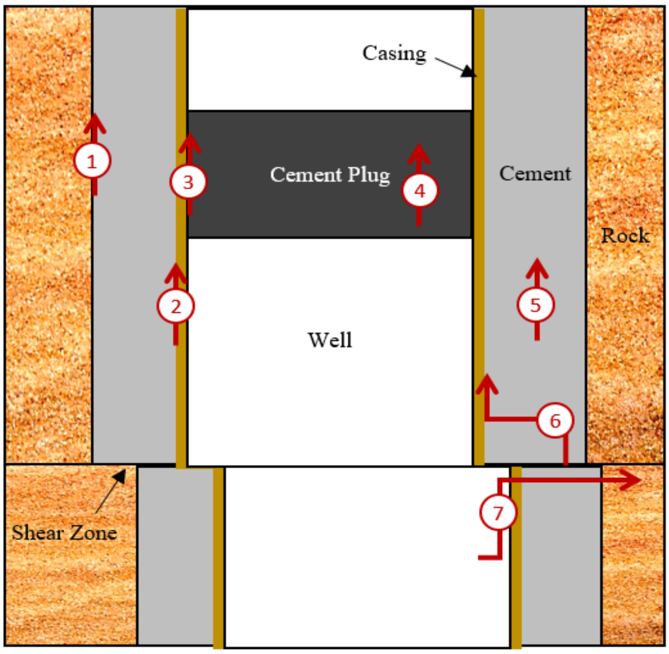

Figure 2: illustrates the leakage channels that may be present in cement. 1- between cement and formation, 2- cement and casing, 3- casing and cement plug, 4- through the cement plug, 5-through the integrity cement, 6- through the cement and in between cement and casing, 7- along features that damage the well (adopted from[22])

To understand the impact of leaky wells on the migration of methane and fracture fluids Schwartz in 2015, modelled the Damme 3 region in northern Germany [12]. The results of the study show that unconventional wells located 300m above hydraulic fractures are the major conduits of hydrocarbon and fracture fluids migration. The simulation displayed that the unaided migration of fracture fluids is highly unlikely, however in the presence of mobile methane, liquid may rise approximately 300 meters vertically [12].

Due to the nature of sedimentary beddings, layers of rocks with varying properties were formed geologically. This states that homogeneity of rock beds is more lateral as opposed to vertical, with horizontal permeability two orders of magnitude greater than vertical permeability [16]. In the context of leaky well, the occurrence of a "frac hit" may stimulate greater methane migration concerns due to these high lateral permeabilities. A frac hit is a fracture connection between a hydraulically stimulated well and an abandoned well. The occurrence of a frac hit may potentially allow communication of shale gas reservoirs with abandoned wells [16].

Despite research showing that frac hits are common, the likelihood of a frac hit occurring between a fracturing site and a well located outside the confines of the SRV is only possible given a long geological timescale [16]. Considering the properties of shale reservoirs, the lack of formation permeability would reduce chances of communication between wells in the absence of a frac hit. However, Brownlow in 2016, demonstrated that unconventional wells with hydraulic fracturing, compared to the no fracture cases, posed a high risk of well to well communication within the SRV, and since most conventional wells do not extend to similar depths as unconventional wells, the communication between two horizontal wells is more likely to present transport pathways for hydrocarbon migration [16]. Similarly, with the lack of monitoring of the abandoned well integrity, concerns about these wells as gas migration conduits are detrimental to the future of shale gas technology. Further enforcing data suggests that 6.3% of wells inspected in the Marcellus shale region in Pennsylvania between 2005 and 2013, had been reported with well integrity failure issues [22]. However, with the history of methane emissions and groundwater contamination from Marcellus shale play, lack of baseline data in the region does not allow a conclusive statement on the origin of the leaks to be solely attributed to the presence of leaky wells and induced fracturing. Table 2 provides a compilation of published reports to understand the migration pathways that may lead to the leakage of methane.

Table 2: Potential leakage pathways from different authors

Author	**Year**	**Pathways**
Zoback et al [13]	2010	Fractures that extend well beyond the target formation to water aquifers (uncontrolled hydraulic fracturing)
Stephen et al [23]	2011	Leakage from well casings; increased connectivity of fracture system due to hydraulic fracturing
Warner et al [15]	2012	Leakage through the naturally occurring pathways (fractures)
Jackson et al [4]	2013	Leakage due to poor well integrity
Darrah et al [24]	2014	Leakage from the target or intermediate-depth formations through a poorly cemented well annulus and leakage from target formation through faulty wells
Davies et al [22]	2014	Leakage due to poor well integrity
Zhang et al [2]	2015	Induced fractures extending to natural features in subsurface
Grasby et al [14]	2016	Leakage through the natural fracture systems that provides circulation pathway for natural spring water
Brownlow et al [16]	2016	Leakage through abandoned oil and gas wells converted into water wells

In summary, as can be seen from Table 2, Zoback et al [13] in 2010 first proposed that the uncontrolled hydraulic fractures can be extended from shale plays to near surface fresh water aquifers due to the fact that the fractures length and diameter will change by time due to geological stresses. Warner et al [15] in 2012 pointed out that not only hydraulic fractures increase the chance of shale gas leakage but also naturally occurring pathways such as faults and high permeable zones can provide an easy access for methane to leak to upper formations and contaminate subsurface fresh water aquifers.

In 2011, Stephen et al [23] proposed that the majority of the shale gas leakage comes from poor well integrity, bad cement bonding and near wellbore fractures. They concluded that the poor integrity of the casings, cement and near wellbore region increase the connectivity of fracture system which result in methane leakage from the shale gas reservoirs. In 2013, Jackson et al [4] also supported the idea of shale gas leakage due to failures in well integrity. They confirmed that poor well integrity presents a more viable cause of methane migration compared to stress redistribution implications of hydraulic fracturing. Darrah et al [24] and Davies et al [22] in 2014 concluded that methane leakage due to poor well integrity can be initiated from a target or intermediate-depth formations through a poorly cemented well annulus and faulty wells. They found that the methane leakage can be a complex connection between hydraulic fractures and poor well integrity.

Recently, Zhang et al [2] in 2015 provided an idea that the induced fractures can be extended to natural high permeable zones in the subsurface. They provided an example of gas leakage through thermal springs and concluded that the viability of natural rock features provide a means for contaminants migration from shale gas reservoirs to even surface water springs. Grasby et al [14] in 2016 confirmed that methane leakage through the natural fracture systems, creates a circulation pathway to natural spring waters.

Current methods used to quantify methane leakage

The common methods that are used to record the total or regional emissions of methane can be summarised in to top-down and bottom-up quantification approaches. Both refer to the measurements of atmospheric emissions, with the top-down dealing with regional methane activity, while bottom-up is focused on individual sources [25].

There are also a number of downhole and offshore gas detection methods such as fibre optic sensors [26], infrared cameras [27-28], combination of temperature and noise logging tools [29], ultrasonic detectors [30], remotely operated vehicle (ROV's) [31-32] and subsea leak detection systems consist of the chemical sensors along with current meters and acoustic sonars [33] are proposed for conventional oil and gas wells which can also be employed in unconventional shale gas wells (see Table 2).

In unconventional resources, the top-down method uses the total emissions of methane for a large region within a short period of time and usually implements a material balance approach. A good example is the work presented by Karion et al. [34], who conducted an investigation on the Barnett shale region, one of the major shale gas producing areas in the U.S. In their study, they used aircraft carriers to measure basin wide emissions of methane, with relatively consistent results over a period of 8 days. Another example of a top-down approach is the study presented by Peischl et al [35], in the Fayetteville, Haynesville and Marcellus shale plays, Pennsylvania which was conducted in less than two months. The top-down approach is effective in assessing the amount of methane being emitted in large scale area, however, the main uncertainty lies in the assumption that methane quantification is associated with all the oil and gas extraction activities. Furthermore, with the limitations in technology to distinguish the thermogenic and biogenic (produced by animals, bacteria, landfills, water treatment plants etc.) sources of emissions, the actual quantification of methane emissions specifically from oil and gas activities of a region is debatable. In addition, the top-down approach only considers the emissions during a specific period of time, which may overlook high emission activities, such as venting and flaring [34], [36].

The bottom-up approach records the emissions data from a single source as opposed to large regions covered in a top-down approach, thus it removes the need to calculate any background emissions. The bottom-up method uses direct well pad measurements and leakage inventory data to assess the atmospheric emissions. This approach is well documented in a compilation study conducted by Lyon et al [37], in the Barnett shale regions of Texas. The major limitations concerning the bottom-up approach are the cost of gathering emission data from many individual sites to form a representative average of the region. Thus, the common method of expanding the limited data is extrapolation methods which have been done on both regional and national scale in the U.S. [25].This method has been employed both by private and government agencies, such as the Environmental Protection Agency (EPA) of the U.S., and has come under scrutiny for the high uncertainties in under- and overestimated results by 2 to 3 folds [7], [37].

Furthermore, measurements using satellite based systems compared to aircraft based, have been regarded as the most reliable and robust method to record emissions quantities [7], [25], [38]. This is due to the fact that the data is recorded over a period of two years, thus it accounts for periods of high and low emissions. The limitation of this approach is that it only documents upstream activities as opposed to overall emissions. Since downstream emissions can comprise up to 2.5% of the total lifetime production of methane from a shale gas well, the estimations for emissions across the life of the well might be underestimated [5].

In contrast to the methods used to record the atmospheric emissions, the process of measuring groundwater contamination appears to be more robust. The method employed uses sampling from groundwater resources and conducting laboratory tests, including an isotope and compositional analysis, to determine the constituents of the samples. In the past, this method has been used by the EPA such as the studies by Molofsky et al. and Osborn et al. [23], [39]. To determine the origins of methane, an isotopic analysis is made where the presence of isotope $\delta^{13}C\text{-}CH_4$ with more negative than -64%, is indicative of biogenic sources, while less negative than -50% is related to thermogenic methane [23]. Many researchers have argued that the water sample data reported by Osborn et al. were based on the selective collection and do not encompass a wide enough data set to allow a representative conclusion to be drawn [23]. Furthermore, areas where aquifer potential has not been exploited, provide difficulties in obtaining water samples, thus leaving gaps in regional measurements.

The emission of thermogenic methane (resulting from the decay of organic matter) to the atmosphere and leakage to groundwater reserves, is an important factor in determining the successful sustainability of shale gas production in the future. Emissions data from shale gas wells is important in determining the impact of shale gas extraction on the environment. Due to limited research on the rate of methane emissions and amount of contamination, the data collected by numerous sources all show significant variability and thus decrease the reliability of the estimations. The major factor contributing to the spectrum of results obtained, is the extreme values of emissions that have been observed at individual sources that, in some cases, tend to exceed the average emission rates from numerous sources at other locations [25]. Furthermore, the methodology used to record the data varies from study to study, with employment of data extrapolation and estimations based on a few sources of leakage.

Table 3: Summary of leakage detection in previous studies

Author	**Year**	**Method**	**Region**	**Subsurface Leak**	**Surface Leak**	**Monitoring Duration**	**Remarks**
Walker et al [26]	2003	Fibre optic	USA nationwide	-	√	Various	Mostly for leakage detection in pipelines
Etiope et al [32]	2005	Remotely operated vehicle	Italy	-	√	Various	Subsea leakage detection
Alkamali et al [27]	2008	Infrared cameras	Various	-	√	Various	Common tool for leakage detection in surface facilities
Khalil et al [29]	2012	Temperature noise logging	UAE	√	-	less than 1 day	A case study on two wells, an injector and a producer
Allen [25]	2014	Top-down and bottom-up	USA nationwide	√	√	Various	Large scale area
Sizeland et al [30]	2014	Ultrasonic detectors	UK	-	√	Various	-Cannot be affected by wind and dilution -Potential in downhole application
Wecht et al [59]	2014	Satellite	California (USA)	-	√	Two years	California: 329333 kg hr-1:[gas/oil (73697 kg hr^{-1})]
Karion et al. [34]	2015	Top-down (mass balance)	Barnett shale (USA)	-	√	8 days	Total emission: 76±13 × 10^{3} kg hr^{-1}, O& G: 60±13 × 10^{3} kg hr^{-1} (95%)
Peischl et al [35]	2015	Top-down	Fayetteville, Haynesville and Marcellus shale plays (USA)	-	√	1 day	(8.0±2.7)×10^{7}g hr^{-1} Haynesvilleregion, (3.9±1.8)×10^{7}g hr-1 Fayetteville and (1.5±0.6)×10^{7}g hr-1 from the Marcellus
Lyon et al [37]	2015	Bottom-up and Monte Carlo simulation	25-county Barnett shale (USA)	-	√	about 15 days	Total emission: 72,300 (63400−82400) kg hr^{-1}, O&G:46200 (40000−54100) kg hr^{-1}
Suwagul et al [28]	2016	Infrared cameras	Kamphaeng Phet, Thailand	-	√	about 3 years	Common tool for leakage detection in surface facilities
Grasso et al [31]	2016	Remotely operated vehicle	Italy	-	√	Various	Subsea leakage detection

Waarum et al [33]	2016	Subsea leak detection system	Norway (North Sea)	-	√	Various	Consists of chemical sensors, current meters and acoustic sonars

As can be seen in Table 3, most of the previous methods have been developed for methane leakage detection in surface facilities, and except few trials there is no established technique for downhole gas leakage detection. This could be due to the variety of gas leakage sources at different subsurface depths and geological complexity of the subsurface environments. It is also obvious that a combination of various techniques is required to estimate methane leakages from wellbore to atmosphere. For example, satellite and aircraft measurement techniques are mostly used for regional measurements in comparison with ROV's, Infrared cameras, ultrasonic, subsea leak systems and special well logging tools are used for single well or facility measurements.

However, most of the published methane emission data are from satellite and aircraft measurement techniques and very limited and sometimes contradictive information are available on subsurface methane leakage detection. Figure 3 shows the amount (kg/hr) of methane emission recorded from oil and gas activities in 4 different studies during 2014-2015 in the United States using satellite and aircraft measurement techniques.

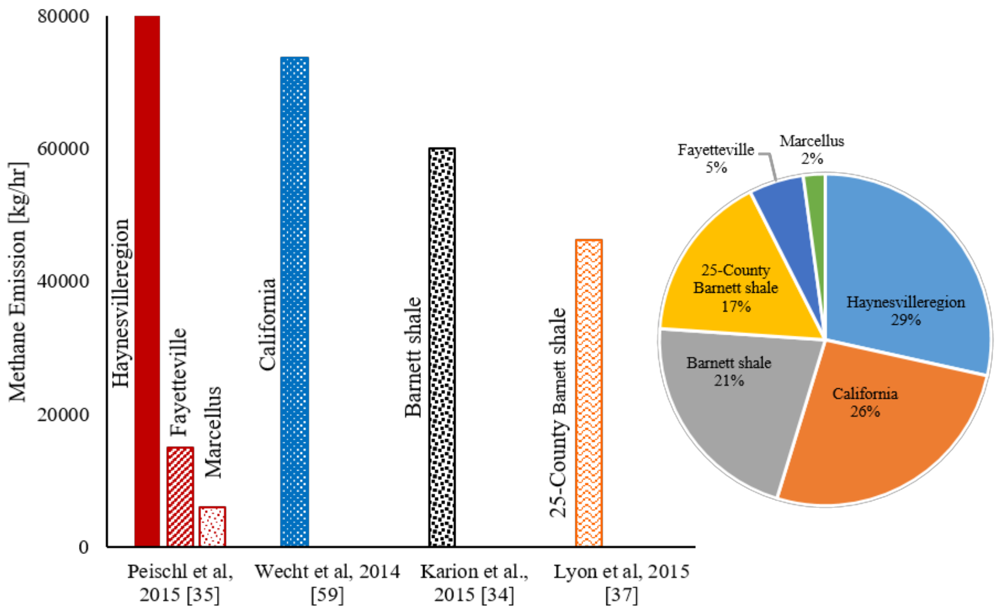

Figure 3: Methane emission recorded from oil and gas activities in the United Sates

As can be seen from the Figure 3, in four studies recorded the regional methane emission in the range of 46000 to 80000 kg/hour. However, it mostly depends on the amount of oil and gas production activities in each region with no specific data on each oil and gas production facility. These four studies confirmed that methane emissions from oil and gas activities are between 50 to 95 % of the total methane emission recorded in each region. All cases also shown that the regional methane emission is either equal or more than the standard level reported by the US environmental protection agency (EPA) except the study conducted by Peischl et al [35] where methane emission recordings were less than the standard level.

Lyon et al [37] further classified methane emission from various sources, and they found that a large portion (about 64%) of the total methane emission is from oil and gas production activities (Figure 4).

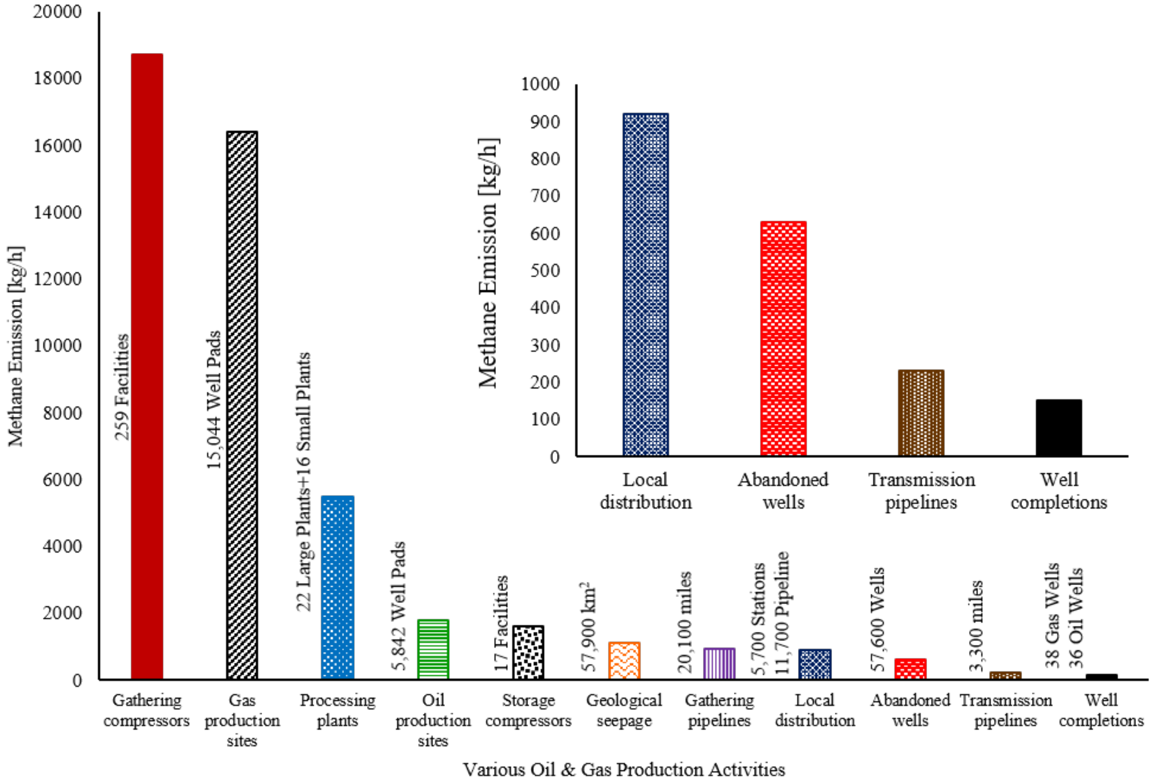

Figure 4: Methane emission from different oil and gas activities (adopted from [37])

As can be seen from Figure 4, the majority of methane emission reported by Lyon et al [37] is from the surface production facilities with the least amount from abandoned hydrocarbon wells and well completions. It can also be seen that the amount of gas emission from well pads of the gas well is approximately 9 times higher than the oil wells. However, the recorded data might not show the real impact of subsurface gas leakage because of low number of cases, lack of downhole gas leakage detections, complexity and uncertainties in the measurements.

Therefore, further investigations are required especially for shale gas reservoirs due to their complex nature and their methods of extractions such as hydraulic fracturing.

Groundwater contamination

The possibility of contamination of groundwater as a source of domestic water supply has raised public attention when dealing with shale gas extraction through hydraulic fracturing [6]. Although the dissolved methane in water is not detrimental to health upon consumption, elevated levels of the gas in regions underlying populated areas, present flammability risks and explosion hazards [40]. To assess the potential risks, the U.S. Department of the Interior recommends monitoring of water aquifers that have methane concentrations more that 10mg/L, and immediate actions to be taken if the concentrations exceed the 28mg/L threshold [23], [40].

In a study conducted by Stephen et al. [23] , the sampling of aquifer water in active and non-active gas extraction regions showed that on average the concentrations of dissolved methane were higher near the active regions, ranging between 19.2- 64 mg/L of methane, as compared to the non-active areas which showed an average methane concentration of 1.1mg/L. Similarly, groundwater studies in northeast Pennsylvania demonstrated that water samples that were collected less than 1 km from shale gas sites had elevated levels of dissolved methane, ethane and propane that showed composition proportions consistent with natural gas in the Marcellus shale play, suggesting a link to gas extraction activities. Stephen et al. [23] further stated that there is a direct relationship between the methane concentrations and distance to shale gas wells.

Conversely, samples that were collected across the Appalachian basin in Pennsylvania showed signs of naturally present thermogenic methane without the presences of hydrocarbon activity in the region [40]. Furthermore, a study on the water quality near the Fayetteville shale play, North Carolina, showed the traces of dissolved methane in 51 out of 127 wells that were used for sampling. However, only 32 wells had methane concentrations greater than 0.002mg/L, and only in 6 wells, methane concentrations were more than 0.5mg/L. Further analysis of the samples showed an isotopic presence of both biogenic and thermogenic methane, with some wells predominantly biogenic. Moreover, the concentration analysis showed no trace of longer chain hydrocarbons which suggests that the presence of deeply seated leakages had not affected the region. The collected data showed no correlation between sampling distance and methane concentrations, stating that the contamination rates were not higher closer to shale gas sites, nor was any statistical evidence found to support the claim [41].

Hammond [39] conducted research to analyse the methane concentrations in water wells of Dimock region in Pennsylvania before and after setting cement plugs in nearby gas wells. Initial samples used to represent levels of methane concentrations before cementing were acquired from inventories published by the Pennsylvania Department of Environmental Protection in 2009. The data provided was inconclusive as the levels of methane concentrations fluctuated after remediation attempts. In 2009, the methane concentration in water well A, had an average value of 6.8mg/L, however, after remediation in 2010, the level dropped to 0.6 mg/L. This illustrates successful mitigation of groundwater contamination. The same methodology applied to water well D, located in the same region, where initial methane concentration was around 39 mg/L in 2010. Following a cement squeeze job of the reference gas well, the methane concentration in water well D gradually declined to zero within three months. However further monitoring of the well showed concentrations fluctuate from 0 mg/L to 31.8 mg/L thereon. Furthermore, the study was continued on a different location within the same area, on gas wells 3 and 4. Initial methane levels in nearby water wells (Well H and I) displayed methane concentrations of 30-48 mg/L, and after plugging, the values were initially reduced, however, given a longer timescale, the concentrations increased and fluctuations varied between 29 and 31 mg/L. Therefore, cement plugging is unreliable and subject to deterioration which displays only partial success [39].

Atmospheric emissions

Limited atmospheric emissions data associated with shale gas production makes the exact quantification of methane emissions from unconventional activities highly difficult. Attempts have been made to quantify methane leakage, however detailed studies using different methods, conditions and locations to conduct the analysis, have shown limited consistency in estimated values. Furthermore, these studies were involved some levels of unavoidable uncertainties for the collected data, which decrease the accuracy of the results [1]. The concerns of methane leakage are based on the significant effect it has on the global warming. Due to the higher GWP of methane compared to $CO_{2,}$ shale gas presents a higher detrimental effect on the climate than coal or oil. With the scenario of continual leakage, the potency of methane presents higher environmental implications than CO_2. However, in order to consider the effect of methane compared to carbon dioxide, the time that each gas remains in the atmosphere needs to be taken in to account. Carbon dioxide remains in the atmosphere significantly longer than methane, thus having a prolonged effect [1].

Areas of unconventional well activities are likely to contain conventional wells that are used for either production or exploration purposes. Therefore, to accurately analyse emissions of methane to the atmosphere, the consideration of both conventional and unconventional wells is required. Table 4 is a compilation of methane emissions data from different locations. The data has been collected using various methods and conditions, however, all measurements aim to achieve similar outcomes and the quantities presented here are percentage emissions of methane as compared to the overall natural gas production in the specified region.

Table 4: A compilation of quantification data of combined conventional and unconventional methane emissions in shale gas extraction areas. The data is presented in percentages of total natural gas produced in the regions within the U.S.

Authors	**Year**	**Location**	**Regional Quantities (%)**	**CH_4 in sample (%)**
Petron et al. [42]	2012	Denver-Julesburg, Colorado	4.2±1.1	-
Petron et al. [43]	2014	Denver-Julesburg, Colorado	4.1±1.5	-
Karion et al. [34]	2015	Uinta Basin, Utah	9±2.8	89
Peischl et al. [35]	2015	Los Angeles Basin, California	17±5	-
Caulton et al. [39]	2014	Marcellus shale, Pennsylvania	10.2±7.1	-
Schneising et al. [44]	2014	Eagle Ford shale, Texas	9.1±6.2	93
Schneising et al. [44]	2014	Bakken shale, Central North USA	10.1±7.3	93
Peischl et al. [35]	2015	Marcellus shale, Pennsylvania	1.5±0.6	96±3
Peischl et al. [35]	2015	Haynesville, Louisiana	8.30±2.7	90±7
Peischl et al. [35]	2015	Western Arkoma, Oklahoma	3.3±1.5	95±5
Peischl et al. [35]	2015	Fayetteville, North Carolina	3.9±1.5	94±5

In each of the reported cases in Table 4, the estimations of the methane emissions are significantly higher than the reported concentration by the EPA, 3% emissions over the life of the well, expressed as the national production of natural gas (shown in Figure 5) [1]. This might be due to the fact that the EPA values come under the assumption that emissions are consistent throughout the oil and gas industry [34]. This assumption is nullified by the data presented in Table 4, which proves that the distribution of methane emissions varies from region to region. However, since the percentage of emissions is based on the total production in one specific region, collectively the data does not provide a base for comparison as the total natural gas production of the regions differ significantly, ranging from 0.05% of national gas production in the Los Angeles basin, to 2.7% of national gas production in the Marcellus shale region in 2010 [35]. In some cases, this information was not recorded, thus the data is only effective in a regional representation as compared to a proportionate national outlook of emissions data. The values presented in Table 4 raise a concern whether the benefits of using shale gas to reduce CO_2 emissions and protect the environment have been negated by the rates of methane leakage. Howarth et al. in 2011,reported that on a national scale, the overall emissions of natural gas from commercial extraction above 2-3% of total gas production, would invalidate the incentives of considering the use of shale gas as a source of energy [7]. Similar values of methane emissions; 3.2% and 2% of total gas production, have been suggested by both Alvarez et al. [45] and Wigley [46] respectively. Furthermore, both latter studies stated that the emission percentages below the reported values would provide an immediate climatic benefit, while Wigley [46] suggested that the loss rates of above 10% may still prove beneficial in the long run as overall CO_2 and black carbon emissions from coal would subsequently decrease with the introduction of widespread natural gas fired power plants. In order to assess the overall impact of methane leakages, national estimates need to be considered for an average emission over the life of a well. Figure 5 is a compilation of atmospheric emissions data [1].

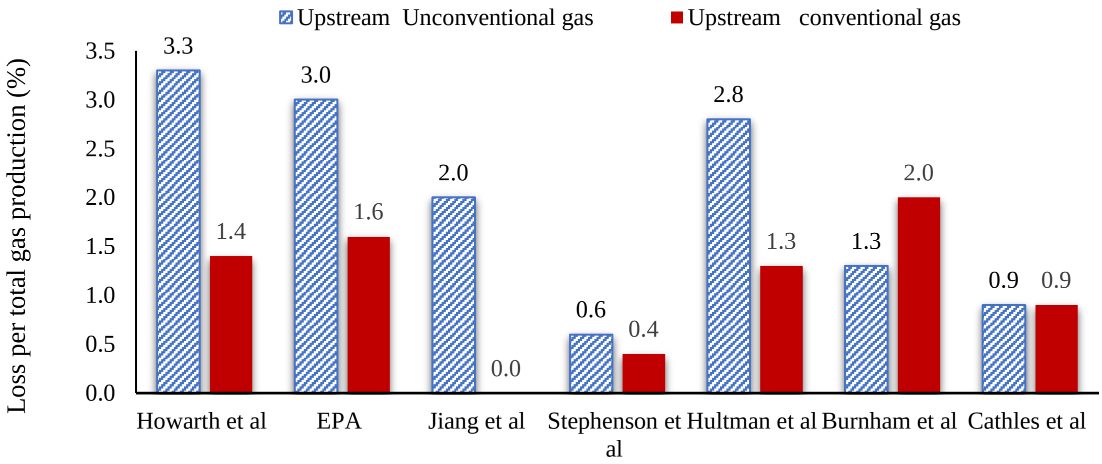

Figure 5: Represents the collected data from different studies on the overall leakage of methane over the life of a well, expressed as a percentage of total natural gas production [4]

In considering downstream production, Brandt et al. in 2016 stated that the overall emissions of natural gas are estimated at 5.4% (± 1.8%) [47]. Furthermore, the values presented in Figure 5 show that leakage percentages are mainly greater than the allowable emission values that would provide a climatic benefit for the widespread use of natural gas.

Current solutions to the methane leakage problem and their limitations

Very limited number of studies, if any, have been conducted so far to address the leakage of the subsurface, thermogenic methane as a stray, and contaminant gas. Studies on reducing leakage of gases from a subsurface environment have been predominantly focused on CO_2 leakage via carbon capture and storage (CCS) operations and enhanced oil recovery processes [48-53]. Thus, research conducted on the reduction of CO_2 leakage will be documented and used as a base to design a suitable experimental analysis addressing methane leakage.

Solutions for remediation and mitigation of CO_2 leakage have been developed with the focus on CO_2 sensitive chemicals and temperature activated systems that can react and form a plugging material, either as a solid or gel structure, in the presence of gas. Laboratory experiments have displayed promising results, with successful reductions in core permeabilities and high strength of generated blocking materials.

Studies conducted by Dexiang et al [54] focused on precipitation of various chemicals under acidic conditions in the presence of carbonic acid. Chemicals considered in their study were polyacrylamide (at 70 and 80^{o}C), sodium aluminate (70, 80 and 90 ^{o}C) and phenolic resin. All materials showed 82.5-99% plugging rates with good scouring resistance, while phenolic resin gave fluctuating results of 31-100% plugging rates with poor scouring resistance. Brydie et al [55] followed a similar approach, using CO_2 enriched brine with sodium silicate to induce the formation of precipitates. The found that the feasibility of sodium silicate to form a blocking agent was high, with good predicted stability under reservoir conditions. However, it was noted that the plugging agent was prone to degradation, and it might lead to premature gelling at extreme conditions. Furthermore, they stated that the use of sodium silicate is promising for downhole injections.

Another promising substance considered in many studies is calcium carbonate (calcite) as shown in Figure 6. Calcite precipitates showed a good performance as a blocking agent that effectively reduce the permeability of core samples in lab experiments [55], [56], [57]. Cunningham et al. conducted research on the microbially induced precipitation of crystalline calcium carbonate to form calcite as a blocking agent [46]. The bacteria used in their study, adopts biofilm growth patterns and in the presence of urea, urease and CO_2 induces permeability reductions.

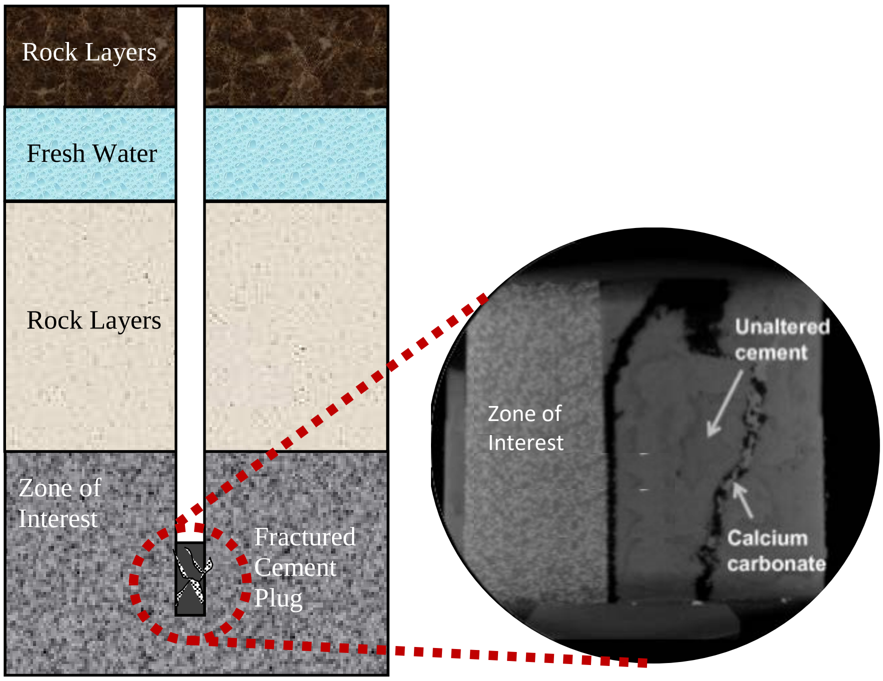

Figure 6: Blocking fractures by calcite precipitation in the cement plug after microbial treatment, (XMT image of calcite deposition adopted from [57])

Recognising the degrading effect that CO_2 can have on cement, Jelena et al. considered the use of polymer resin injections to fill leakage channels and increase cement integrity [58-59]. The laboratory experiments displayed an immediate success on a small scale, showing a reduction in permeability of cement plugs with an initial value of 47mD to approximately 0mD. However, when larger channels were made in cement plugs, polymer resin injections showed that despite significant reductions in permeability from 1717mD to 41mD, the resin could not completely fill the entire channel.

Each method showed its own limitations for mitigation of gas leakage. Most of the experiments did not assess the effects of temperature and pressure variations on the performance of such remedial solutions for gas leakage, and many did not consider the effects of concentration changes of injectants. As the experiments were mainly conducted in laboratory scales, the susceptibility of reservoir conditions was not observed.

Conclusions

This review showed that the major sources of methane leakage related to shale gas activities are the intersections of hydraulic fractures with abandoned oil and gas wells which have a reduced mechanical well integrity due to cement degradation. As a result the stress redistributions caused by hydraulic fracturing and the deterioration of cement in abandoned wells allows migration pathways to be created easily, leading to both groundwater contamination and atmospheric emissions. Some sources highlighted the influence of natural fracture networks on gas leakage; however, the reports demonstrate that unless specific conditions of regional deformation, stress, orientation, strike and dip angles of natural fractures are present, the interaction of induced fracture is limited.

Furthermore, the methods used to quantify leakages are based on extrapolations from short time periods, thus they are not representative of the problem. The occurrence of multiple leakage sources and methanotrophs in groundwater similarly does not allow accurate evaluation of methane quantities. The quantification values that were presented, are not consistent with each other, therefore requires the use of consistent methodology, considering the combination of a top-down and bottom-up approaches. Quantifications that were made in regions, were expressed as percentages of regional production as opposed to the national production of shale gas, therefore they are not proportionate and do not allow comparisons to be made. Similarly, the limited data that was expressed in terms of national production do not complement each other and are littered with assumptions and extrapolations. Based on the collected data on methane emissions from conventional and unconventional wells in shale gas extraction areas, quantities of emissions (typically between 3-10% of the total gas production) are significantly higher than the reported concentration by the EPA (3% emissions over the life of the well). Also, methane concentration in aquifers around the fracking areas have shown to exceed the 28 mg/L threshold recommended by the U.S. Department of the Interior where even the cement treatment of leaky wells could just temporarily reduce the leakage. Therefore, the main issue associated with the methane leakage to the atmosphere and ground water, is well integrity which require further investigation and studies.

In terms of mitigation of gas leakage, different methods that have been tested to remediate subsurface gas leakage were based on CCS operations, and practically no novel work has been done to address the subsurface leakage of methane. However, through the remedial solutions for CO_2 leakage it was found that polymer and calcium carbonate precipitations can considerably plug subsurface fractures and pathways for gas leakage.

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