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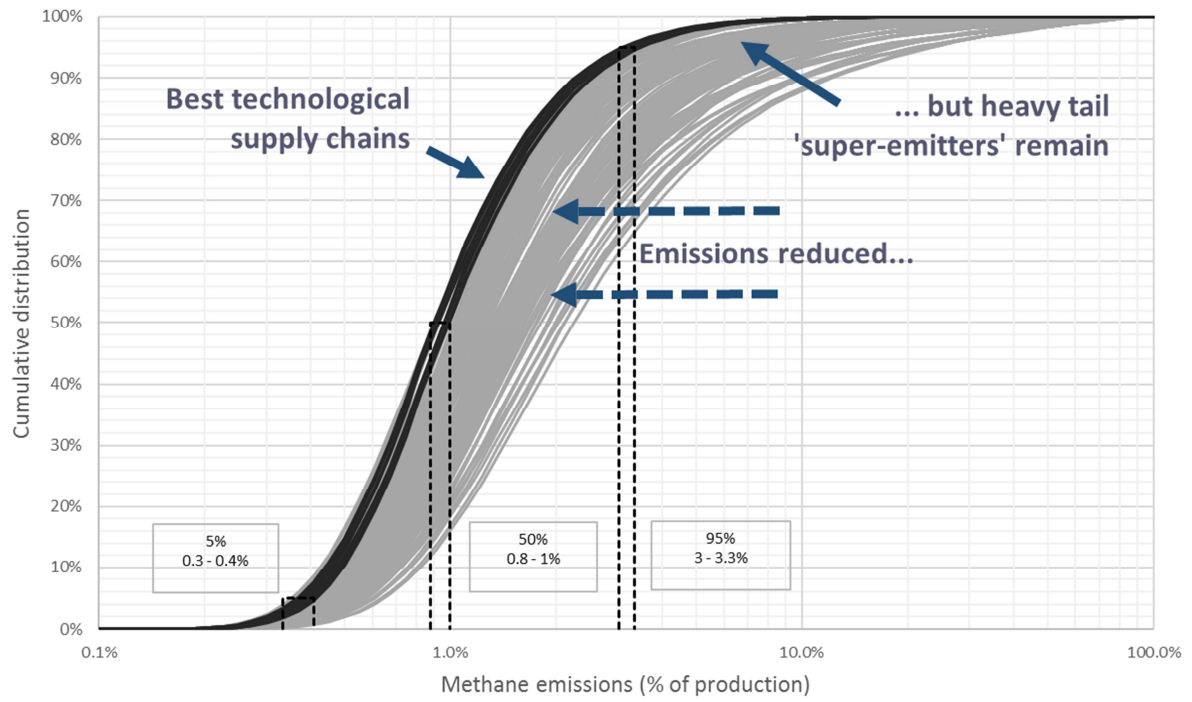
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# Characterising the distribution of methane and carbon dioxide emissions from the natural gas supply chain

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## Abstract

Methane and CO<sub>2</sub> emissions from the natural gas supply chain have been shown to vary widely but there is little understanding about the distribution of emissions across supply chain routes, processes, regions and operational practises. This study defines the distribution of total methane and CO<sub>2</sub> emissions from the natural gas supply chain, identifying the contribution from each stage and quantifying the effect of key parameters on emissions. The study uses recent high-resolution emissions measurements with estimates of parameter distributions to build a probabilistic emissions model for a variety of technological supply chain scenarios. The distribution of emissions resembles a log-log-logistic distribution for most supply chain scenarios, indicating an extremely heavy tailed skew: median estimates which represent typical facilities are modest at 18 – 24 g CO<sub>2</sub> eq./ MJ HHV, but mean estimates which account for the heavy tail are 22 – 107 g CO<sub>2</sub> eq./ MJ HHV. To place these values into context, emissions associated with natural gas combustion (e.g. for heat) are approximately 55 g CO<sub>2</sub>/ MJ HHV. Thus, some supply chain scenarios are major contributors to total greenhouse gas emissions from natural gas. For methane-only emissions, median estimates are 0.8 – 2.2% of total methane production, with mean emissions of 1.6 - 5.5%. The heavy tail distribution is the signature of the disproportionately large emitting equipment known as super-emitters, which appear at all stages of the supply chain. The study analyses the impact of different technological options and identifies a set of best technological option (BTO) scenarios. This suggests that emissions-minimising technology can reduce supply chain emissions significantly, with this study estimating median emissions of 0.9% of production. However, even with the emissions-minimising technologies, evidence suggests that the influence of the super-emitters remains. Therefore, emissions-minimising technology is only part of the solution: reducing the impact of super emitters requires more effective detection and rectification, as well as pre-emptive maintenance processes.

## Keywords

Natural gas; supply chain; greenhouse gas emissions; methane emissions; super emitters; heavy tail distribution

## 1. Introduction

As the drive to reduce climate change to 1.5 to 2 °C gathers pace, the production of natural gas is increasing and is considered to be the lowest carbon fossil fuel (IEA, 2014; UNFCCC, 2015). However, whilst CO<sub>2</sub> emissions from combustion are lower than those from coal or oil, emissions of CO<sub>2</sub> and

methane across the gas supply chain have been shown to be significant and vary widely, often by several orders of magnitude (Balcombe et al., 2017). The natural gas supply chain includes all processes and equipment from pre-production drilling through to delivery to the consumer. Given the projected increases in natural gas production, the impact of supply chain emissions becomes even more important. In particular emissions of methane, which is the main constituent of natural gas and a very strong greenhouse gas (GHG) over short timescales, are highly variable (Allen, 2016; Alvarez et al., 2012; Brandt et al., 2014).

Many factors affect supply chain emissions of methane and carbon dioxide, such as the reservoir type, supply chain route, equipment and operational practises, or regional regulation. There also exists a small proportion of equipment and facilities all across the supply chain that contribute disproportionately highly to supply chain emitters, known as super emitters (Brandt et al., 2014). Additionally, different estimates from industry, academia and government are affected by their methods of estimation, assessment boundaries and assumptions (Balcombe et al., 2017).

All these factors combine to result in large variations in emissions estimates, but there remains a lack of understanding of the contribution of each factor (Balcombe et al., 2015). The extremely large range and the multitude of dependent factors make the prediction of emissions from different regions or in different scenarios very difficult. An understanding of the distribution of supply chain emissions would greatly help to predict the impact of abatement technologies and guide emissions-monitoring regulation (Brandt et al., 2016). A number of studies have attempted to define distributions of certain portions of the gas supply chain, e.g. from gathering and processing plants (Marchese et al., 2015), distribution metering stations and pipeline leaks (Lamb et al., 2015). However, none have determined the distributions of both methane and carbon dioxide and none have done so for whole supply chains. A raft of new emissions measurements data and estimates (Allen et al., 2014a; Allen et al., 2014b; Brantley et al., 2014; Lamb et al., 2015; Lyon et al., 2015; Marchese et al., 2015; McKain et al., 2015; Mitchell et al., 2015; Rella et al., 2015; Subramanian et al., 2015; Townsend-Small et al., 2015; Yacovitch et al., 2015; Zimmerle et al., 2015) provides a unique opportunity for such a comprehensive analysis in this study.

This paper synthesises and analyses the recent series of emissions data aggregated in two recent studies (Balcombe et al., 2015, 2017) to define the distribution of methane and CO<sub>2</sub> emissions from each stage of the supply chain. The key questions addressed within this study are the following:

- What is the distribution of methane and carbon dioxide emissions from the natural gas supply chain and what is the contribution from each stage?
- How is the distribution affected by factors such as reservoir type and size, equipment, or operational practises?
- By how much could the magnitude and distribution of these emissions be minimised by the use of different equipment or processes?

The work consists of the characterisation of emissions from each supply chain stage and emission source, an emissions and mass balance, and a Monte Carlo simulation of a variety of supply chain scenarios. This probabilistic assessment quantifies the variation in emissions and the causes of these variations, as well as the impact of emissions-reducing equipment and monitoring.

This greater understanding of natural gas emissions profiles is important in informing engineering and investment decisions in the gas supply chain, and policy decisions regarding the best ways to reduce and/or regulate supply chain emissions. The results could be used to provide detailed and up-to-date inventories for environmental life cycle assessments. Defining the difference in emissions

across different gas assets could also help to inform industrial investment decisions under higher carbon price scenarios.

## 2. Methodology

The goal of the study is to determine the distribution profile of methane and carbon dioxide emissions from the natural gas supply chain and the key factors that govern the distributions. The scope includes the whole supply chain from pre-production, production from conventional, shale or tight reservoirs, to gathering and processing, transmission, storage and distribution ending at the customer meter. Only direct methane and carbon dioxide emissions are included, e.g. vents, leaks, fuel usage and flaring.

Exclusions from the study are emissions from offshore gas production, due to the relative lack of transparent and granular data. The study also excludes emissions from end-use combustion, as well as those associated with abandoned wells. It is acknowledged that emissions from a small number of abandoned wells may be significant but generally represent less than 1% of supply chain emissions (Boothroyd et al., 2016; Kang et al., 2014; Townsend-Small et al., 2016; Vielstädte et al., 2015).

The study uses emissions estimates from across the globe, but there has not been enough similar or transparent data to make direct comparison between geographical regions. Thus, whilst it is recognised that there are likely to be large regional variations, this is not included here. However, the causes of regional variation are in part due to differences in reservoir type, equipment and operational practises, which are investigated.

This paper expresses emissions estimates in two functional units: methane-only emissions are expressed as a percentage of total production from a well, or estimated ultimate recovery (EUR); total GHG emissions are expressed as grams of CO<sub>2</sub> equivalent per MJ of energy delivered on a higher heating value (HHV) basis. This allows a comparison of emissions from methane and carbon dioxide using a global warming potential (GWP), which compares the relative potency of different greenhouse gases in terms of climate forcing. It is defined as the average relative climate forcing of a pulse emission over a certain time period, compared to a similar emission of CO<sub>2</sub>. This study adopts the 100 year time horizon value of 34 g CO<sub>2</sub> eq./ g CH<sub>4</sub> for methane, but also investigates the impact of using different climate characterisation factors on the total emissions result. Note that GHG emissions other than methane and CO<sub>2</sub> (e.g. N<sub>2</sub>O) are excluded from this study as they are typically considered to contribute a very small part of supply chain emissions {Zammerilli, 2014 #424}. The methodology of this study splits into three stages, which are described below.

1. Building the emissions inventory
2. Formulating the supply chain emissions model
3. Applying the model in Monte Carlo simulations to produce distribution curves

### 2.1 Emissions and parameter inventory

A summary of the sources of direct emissions from each supply chain stage is given in Table 1, including whether there is an associated CO<sub>2</sub> or methane (CH<sub>4</sub>) emission and how it is included within this inventory. For each emissions source listed in Table 1, datasets were created using the reviews by Balcombe et al. (Balcombe et al., 2015, 2017). Datasets for each emission source were further split into subsets where there were discernible differences between categorical factors such as reservoir type, material of construction, or equipment type. Table 2 lists these categorical factors used within the model alongside their associated emission source.

Table 1. List of emission sources by supply chain stage, with an indication of the inclusion within the emissions model.

Stage	Source	CO <sub>2</sub>	CH <sub>4</sub>
1 Pre-production	1.1 Site preparation	✓	✗
	1.2 Drilling	✓	✗
	1.3 Hydraulic fracturing	✓	✓
	1.4 Well completion	✓	✓
2 Extraction	2.1 Fugitives	Total vents	
	2.2 Pneumatics venting	and leaks	
	2.3 Liquid tank venting		
	2.4 Flared gas	✓	
	2.5 Liquids unloading	✓	✓
	2.6 Workovers	Same as completions	
	2.7 Abandoned well leakage	✗	✗
3 Gathering	3.1 Compressor station venting and leaks	✓	✓
	3.2 Compressor fuel	✗	✗
4 Processing	4.1 Compressor venting and leaks	Total vents	
	4.2 Pneumatics venting	and leaks	
	4.3 Vented gas		
	4.4 Fuel		
	i. Compression	Processing fuel	
	ii. H <sub>2</sub> S scrubbing	usage	
	iii. CO <sub>2</sub> scrubbing		
	iv. Dehydration		
	v. Hydrocarbon liquids separation		
	4.5 Vented CO <sub>2</sub>	✓	✗
4.6 Flared gas	✓	✗	
5 Transmission and Storage	5.1 Compressor fuel	✓	✗
	5.2 Compressor venting and leaks	Transmission and storage vents and leaks	
	5.3 Pipework leaks		
	5.4 Storage site venting and leaks		
6 Distribution	6.1 Pipework leaks	✓	✓
	6.2 Metering and regulating station leaks and vents	✓	✓
	6.3 Customer meter leaks and vents	✓	✓

Table 2. Summary of discrete categories used within the emissions model alongside respective emission sources. RECs = Reduced emissions completions.

Emission source	Parameter	Categories
Well completions	Well type	(2) Conventional; Tight; Shale
Well completions	Equipment	(2) RECs; NonRECs
Well completions	Vent or flare	(2) Vent; Flare
Liquids unloading	Equipment	(4) Blowdown; Manual plunger; Automated plunger; No unloading
Distribution pipeline leaks	Material	(4) Cast iron; Bare steel; Protected steel; Plastic
<b>Total scenarios*</b>		<b>(2 x 2 x 2 + 2) x 4 x 4 = 160</b>

\* Conventional wells do not typically require completion flowback equipment, so there is no 'Conventional with RECs/NonRECs' combinations.

Different combinations of these factors result in a set of distinct supply chain route scenarios, for which the model estimates a distinct emissions profile. The total number of theoretical supply chain route scenarios that result from the combination of factors shown in Table 2 is 160. For example, one supply chain route may be the gas production from a tight gas well, using reduced emissions completions (RECs) but venting residual emissions, requiring no liquids unloading and distribution via plastic pipework.

## 2.2 Emissions model

The inventories provide the input data for the emissions model, which is formulated in Matlab and provides a mass and emissions balance accounting for venting, flaring, fugitives and fuel requirements. Note, it is assumed that flaring and fuel combustion are 100% efficient and thus do not emit methane.

The mass and emissions balance is created by synthesising the parameters for each dataset. The units of measurement are different for different parameters, as shown in Table 3: for discrete event emissions, these are typically measured as a mass emission per event, combined with an event frequency. Both are assigned distribution curves. Continuous emissions are typically measured as a percentage of throughput. The emissions from each source may be correlated with one or more parameters. Where these parameters are numerical variables, probabilistic distributions were assigned. For each emission dataset, histograms and Kernel Density plots were generated to investigate the appropriate probabilistic distribution curve. Different distribution curve types were tested for each dataset within Matlab, such as normal, log-normal, logistic, log-logistic, Weibull and gamma distributions. The most appropriate distribution type was selected based on both the quantification of best fit (i.e. the maximum log-likelihood) as well as visualisation of the fit using probability plots. These are summarised in the Supplementary Information Table SI-2.

Table 3. Emissions and parameter inventory for the model. Each emission source is listed, with affecting parameters and key statistics: mean values, number of data points and distribution information.

Stage	Emission source	Parameter 1	Parameter 2	Mean	Units	Data points	Distribution			
							Type	Mean	Standard deviation	
EUR	-	-	-	184.9	Mm <sup>3</sup> gas	53	Log-logistic	4.03	0.69	
Pre-production	Site preparation	-	-	390.6	t CO <sub>2</sub> / well	10	Log-normal	5.73	0.72	
	Drilling	-	-	511.2	t CO <sub>2</sub> / well	13	Weibull	501.55	0.96	
	Hydraulic fracturing	-	-	514.1	t CO <sub>2</sub> / well	11	Log-normal	6.10	0.54	
	Well completion: Conventional	Conventional non-fracked wells	Vent		0.8	t CH <sub>4</sub> / well	887	Log-normal	-2.62	-1.98
			Flare		0.7	t CH <sub>4</sub> / well	20	Log-normal	-3.66	-0.09
	Well completion: Shale	REC	Vent		5.8	t CH <sub>4</sub> / well	86	Log-normal	-0.02	-0.84
			Flare		12.3	t CH <sub>4</sub> / well	231	Log-normal	-0.15	-0.72
		NonREC	Vent		157.1	t CH <sub>4</sub> / well	164	Log-normal	2.49	-0.61
			Flare		5.4	t CH <sub>4</sub> / well	117	Log-normal	-0.21	-0.95
	Well completion: Tight	REC	Vent		17.6	t CH <sub>4</sub> / well	86	Log-normal	1.32	-1.00
			Flare		3.0	t CH <sub>4</sub> / well	82	Log-normal	-0.91	-0.86
		NonREC	Vent		75.2	t CH <sub>4</sub> / well	174	Log-normal	2.24	-0.81
			Flare		5.5	t CH <sub>4</sub> / well	48	Log-normal	-0.65	-0.72
	Production	Equipment and well head fugitive and vented	-	-	0.18%	of throughput		Log-logistic	-7.88	1.03
Flared		-	-	0.94%	of throughput	2	Flat	-	-	
Liquids unloading per event		No plunger	-	-	1,286.3	m <sup>3</sup> CH <sub>4</sub> / event	289	Weibull	428.42	0.43
		Plunger	Auto		34.4	m <sup>3</sup> CH <sub>4</sub> / event	25	Log-normal	2.55	1.45
			Manual		274.2	m <sup>3</sup> CH <sub>4</sub> / event	49	Weibull	276.15	1.01
Liquids unloading events/yr		No plunger	-	-	117.3	event/yr	289	Log-normal	1.91	2.10
		Plunger	Auto		2,444.9	event/yr	25	Weibull	2650.36	1.29
			Manual		13.2	event/yr	49	Log-normal	1.96	1.18
Workovers		-	-	0.2	event/yr	10	Log-normal	-3.18	1.68	



Stage	Emission source	Parameter 1	Parameter 2	Mean	Units	Data points	Distribution			
							Type	Mean	Standard deviation	
Gathering	Fugitive and vented	-	-	1.9%	of throughput	105	Log-normal	-6.70	1.29	
Processing	Fugitive and vented	-	-	0.136%	of throughput	16	Weibull	0.00075	1.08	
	Flaring (CO <sub>2</sub> )	-	-	0.91%	of throughput	7	Log-normal	-5.22	1.17	
	Fuel (CO <sub>2</sub> )	-	-	3.73%	of throughput	9	Log-normal	-3.63	0.96	
	Vented and separated CO <sub>2</sub>	Composition	-	4%	of throughput	-	-	-	-	
Transmission	Fugitive and vented	-	-	0.63%	of throughput	19	Log-normal	-5.49	1.11	
	Fuel CO <sub>2</sub>	-	-	2.61%	of throughput	10	Log-normal	-4.10	1.04	
Storage	-	-	-	0.10%	of throughput	1	-	-	-	
Distribution	Cast iron	-	-	0.062	m <sup>3</sup> CH <sub>4</sub> / hr	16	Log-normal	-0.80	1.92	
				4.4E-06	leaks/m <sup>3</sup>	-	-	-	-	
	Pipeline vents and leaks	Unprotected steel	-	-	0.058	m <sup>3</sup> CH <sub>4</sub> / hr	94	Log-normal	-1.24	1.64
					3.5E-06	leaks/m <sup>3</sup>	-	-	-	-
		Protected steel	-	-	0.075	m <sup>3</sup> CH <sub>4</sub> / hr	44	Log-normal	-1.33	1.93
					2E-07	leaks/m <sup>3</sup>	-	-	-	-
	Plastic	-	-	0.015	m <sup>3</sup> CH <sub>4</sub> / hr	62	Log-normal	-1.65	1.68	
				9.2E-08	leaks/m <sup>3</sup>	-	-	-	-	
	M+R station (HP) >300 psi	-	-	0.008%	of throughput	25	Log-normal	-11.26	2.04	
	M+R station (MP) 100 - 300 psi	-	-	0.001%	of throughput	10	Log-normal	-13.78	3.32	
	M+R station (LP) <100 psi	-	-	0.001%	of throughput	10	Log-normal	-13.78	3.32	
	Customer meter	-	-	0.025%	of throughput	-	-	-	-	

Most emission sources were logarithmically distributed, as shown in Table 3. A summary of each emission source parameter and its fitted distribution curve is given in the inventory in Table 3 and a description of the derivation and data sources for each emission source is detailed in the remainder of the methodology section.

The total production from a well, the estimated ultimate recovery (EUR), was given a log-logistic distribution, with a mean of 185 Mm<sup>3</sup> and a median of 57 Mm<sup>3</sup> (Bond et al., 2014; Burnham et al., 2012; Cooper et al., 2014; Dale et al., 2013; Heath et al., 2014; Jiang et al., 2011; MacKay and Stone, 2013; O'Donoghue et al., 2014; Santoro et al., 2011; Shahriar et al., 2014; Skone et al., 2014; Stamford and Azapagic, 2014; Stephenson et al., 2011; Weber and Clavin, 2012). It is likely that EUR is different across reservoir types (e.g. conventional onshore, shale, tight) and whilst we investigate the impact of varying EUR, the correlation between EUR and well type is not included within this model. The raw gas composition was assumed to be 80% vol/vol methane and 5% vol/vol CO<sub>2</sub>, based on typical compositions (Ferrera and Zandiri, 2010). Processed gas methane content is assumed to be 95% vol/vol. Gas composition is likely to have a significant impact on processing CO<sub>2</sub> emissions from fuel duty, but this is outside of the scope of this study. The impact of natural gas composition on supply chain emissions should be the subject of further research.

### 2.2.1 Pre-production

Emissions associated with site preparation (Bond et al., 2014; Jiang et al., 2011; MacKay and Stone, 2013; Santoro et al., 2011; Weber and Clavin, 2012), drilling (Broderick et al., 2011; Chang et al., 2014; Jiang et al., 2011; MacKay and Stone, 2013; Santoro et al., 2011; Weber and Clavin, 2012) and hydraulic fracturing (Broderick et al., 2011; Jiang et al., 2011; MacKay and Stone, 2013; Weber and Clavin, 2012) are generally CO<sub>2</sub> emissions from equipment fuel usage, based on engineering calculations. The distributions for site preparation, drilling and hydraulic fracturing were estimated as log-normal, log-normal and Weibull, respectively.

Well completions are a collection of activities carried out after the well has been drilled but prior to production. Such activities are inserting, cementing and perforating the well casing, as well as hydraulic fracturing, where flowback of the fracturing fluid is the main source of gas emissions. Well completions emissions estimates (Allen et al., 2013; EPA, 2016; Harrison, 2012) are highly variable and have been categorised to reflect the variation, depending on: well type (conventional, shale or tight); completion equipment (using reduced emissions completions (RECs) or not); and whether any emissions are vented or flared. Methane emissions from each category followed a skewed distribution, some log-normal, log-logistic and Weibull. For this category, a log-normal distribution was selected for all based on the fit associated with the heavy tail portion of the distribution: a Weibull in particular appeared to underestimate. There was no data that correlated the completion emissions with the EUR or initial production rate, thus it was assumed to be independent. Carbon dioxide emissions estimates were available from the US EPA Greenhouse Gas Reporting Program data (EPA, 2016) and were highly correlated with methane emissions but the correlation varies by category (i.e. the use of equipment and whether the emissions were flared). A log-log regression of CO<sub>2</sub> emissions against methane emissions yielded an R<sup>2</sup> between 0.3 and 0.99 for the different categories (i.e. well-type or equipment categories). Whilst some correlations were poor, the residual variation between the modelled results and the data was also incorporated into the estimate by using a random draw from a normal distribution with a mean of zero. The following equation was used to estimate CO<sub>2</sub> emissions:

$$\ln(CO_2) = \beta \cdot \ln(CH_4) - c + \varepsilon(0, \gamma)$$

where  $CO_2$  is the mass of  $CO_2$  emissions in t/ event,  $CH_4$  is the mass of methane emissions in t  $CO_2$  eq./ event (where the data source used a Global Warming Potential of 25 g  $CO_2$ / g  $CH_4$ ),  $\beta$  and  $c$  are regression coefficient and constants respectively, and  $\epsilon$  is the random draw from a normal distribution with a mean of zero and a standard deviation of  $\gamma$ . Values for  $\beta$ ,  $c$  and  $\gamma$  are given in Table 4 for each completion category.

Table 4. Correlation characteristics between the natural logarithms of  $CO_2$  and methane emissions.  $R^2$  is the measurement of goodness of fit,  $\beta$  is the proportional coefficient,  $c$  is the intercept coefficient and  $\gamma$  is the standard deviation of the error term.

Well type	RECs	Vent/flare	$R^2$	$\beta$	$c$	$\gamma$
Conventional	NA	Vent	0.38	0.52	4.8	1.4
Conventional	NA	Flare	0.99	1.0	2.0	0.21
Shale	RECs	Vent	0.28	0.51	-3.8	1.4
Shale	RECs	Flare	0.98	0.99	2.1	0.32
Shale	NonRECs	Vent	0.47	0.63	-3.9	1.4
Shale	NonRECs	Flare	0.92	0.98	2.1	0.67
Tight	RECs	Vent	0.66	1.1	-7.1	0.99
Tight	RECs	Flare	0.78	1.00	1.9	1.3
Tight	NonRECs	Vent	0.69	0.94	-5.9	1.1
Tight	NonRECs	Flare	0.99	1.0	2.0	0.26

### 2.2.2 Production

Well-head production-phase vents and leaks (Allen et al., 2014a; Omara et al., 2016; Rella et al., 2015) and flaring emissions (Burnham et al., 2012) were estimated as a percentage of throughput. Vents and leaks from Omara et al. (Omara et al., 2016) were again highly distributed and fit a log-logistic distribution. The emissions were shown to be highly correlated with the station throughput, with larger producers exhibiting far lower emissions. This study normalised the emissions distribution on facility throughput data, to base emissions on gas throughput rather than station count. This prevents an unfair weighting of several very small but highly emitting stations. Only two estimates for flaring emissions were found, consequently the model only accounted for a flat distribution between the two estimates.

Liquids unloading is a process by which liquids are removed from more mature wells in order to maintain gas flow and can be carried out using various types of equipment. Liquids unloading emissions (Allen et al., 2014b; Allen et al., 2013; EPA, 2016) were shown to vary depending on the equipment used. This study categorises these by the use of blowdowns, manual plunger lifts and automated plunger lifts. For each equipment type, the model uses a distribution for the emissions per event and the event frequency. Weibull distributions are attributed to blowdowns and manual plunger lifts, whereas a log-normal distribution was fitted to auto plunger lifts, due to the much heavier tail (Allen et al., 2014b). An additional scenario is used where no unloading emissions occur, as it is recognised that many, or even most, supply chain routes do not exhibit liquids unloading emissions (Shires and Lev-On, 2012). No data was found on the distribution of years that wells require unloading, so a flat distribution between zero and 10 years was assumed.  $CO_2$  emissions from liquids unloading were highly correlated with methane emissions from analysis of the GHGRP data. A log-log regression of methane emissions on  $CO_2$  emissions give an  $R^2$  of 0.51. The model consequently estimated the  $CO_2$  emissions based on the methane emissions, accounting for the poor correlation/ high variability as per the following equation:

$$\ln(CO_2) = 0.9651 \cdot \ln(CH_4) - 2.576 + \varepsilon(0, 2.21)$$

Workovers are a set of maintenance activities that may occur during a well's lifetime, where gas production is typically halted and operations similar to those associated with well completions are carried out. Workovers (Bond et al., 2014; EPA, 2014; Heath et al., 2014; Shires and Lev-On, 2012; Skone, 2011; Venkatesh et al., 2011) were assumed to exhibit the same event emissions as for well completions, but estimates for the number of workovers required in a well lifetime were log-normally distributed.

### 2.2.3 Gathering and processing

The gathering stage is where flows from a number of wells are gathered together to be sent to a processing plant. Compression is often necessary, as well as dehydration units. One recent study (Mitchell et al., 2015) estimated methane emissions from 115 gathering stations, again exhibiting a log-normal distribution. The high-resolution of information given in the study allowed a throughput normalisation of the emission rates, as described in Section 2.2.2 for production venting and leaks. Equipment fuel emissions will also occur at this stage, but was not included within the model because no data was found.

Processing methane vents and leaks were typically estimated to be small (Clearstone Engineering Ltd, 2002; Mitchell et al., 2015; NGML, 2006) and this study estimates a Weibull distribution from the study by Mitchell et al. (Mitchell et al., 2015), again throughput normalised. CO<sub>2</sub> emissions estimates from flared gas (Sevenster and Croezen, 2006; Skone et al., 2014; Weber and Clavin, 2012) and fuel for the processing equipment (Sevenster and Croezen, 2006; Skone et al., 2014; Weber and Clavin, 2012) were taken from a variety of primary and secondary data sources, due to the relative lack of granularity, where log-normal distributions fit the data. Emissions associated with venting the separated CO<sub>2</sub> were estimated based on the difference in CO<sub>2</sub> concentration between the extracted and delivered gas.

### 2.2.4 Transmission, storage and distribution

Transmission stage vents and leaks from compressor stations and pipework (Bouman et al., 2015; Ishkov et al., 2011; Lechtenboehmer and Dienst, 2010; Leliveld et al., 2005; Logan et al., 2012; Skone, 2011; Stephenson et al., 2011; Weber and Clavin, 2012; Weisser, 2007; Zimmerle et al., 2015), and fuel emissions (Lechtenboehmer and Dienst, 2010; Logan et al., 2012; Skone, 2011; Stephenson et al., 2011; Weber and Clavin, 2012; Weisser, 2007) are both log-normally distributed and are taken from a selection of primary and secondary sources. Very little data is available on storage emissions. Methane loss was estimated from two studies (Lechtenboehmer and Dienst, 2010; Zimmerle et al., 2015) to be approximately 0.1% of throughput, which is assumed here but with no distribution due to the lack of data.

Distribution emissions arise from a number of sources and were characterised in this paper as: pipeline leaks; metering and regulating (M+R) station vents and leaks, including high (>300 psi), medium (100 - 300 psi) and low pressure (<100 psi) stations; and customer meter leaks (Lamb et al., 2015). Pipeline leak data were taken from the most comprehensive recent measurement campaign of 13 distribution systems across the US, published by Lamb et al. (Lamb et al., 2015). Significantly different distributions were noted for different pipeline materials and were categorised into cast iron, bare steel, protected steel and plastic. The study presents emissions as a volume per leak, using the number of equivalent leaks per year and the volumetric throughput to estimate a normalised emissions rate.

It was assumed that the supply chain includes flow through a high, medium and low pressure M+R station, as well as a customer meter. Low pressure stations were assumed to exhibit the same emissions profile as medium pressure stations, due to the lack of alternative data. M+R station emissions were also estimated from Lamb et al. (Lamb et al., 2015), where measurements were expressed as a volumetric annual emission for each site studied. These measurements were normalised by an average US throughput per station: 2,119,700 Mcf/yr for high pressure stations; and 686,809 Mcf/yr for medium pressure stations. This represents a significant assumption, as it is likely that normalised emissions are correlated with throughput (larger stations are likely to emit proportionally less). However, not enough data was available to further analyse this and this was considered to be a conservative assumption.

### 2.3 Monte Carlo simulation

The emissions model and supply chain mass balance was then used to perform a Monte Carlo simulation (Rubinstein and Kroese, 2011). For each of the 160 supply chain scenarios, total emissions were estimated 10,000 times with random draws from the distributions classified in the previous section. The 10,000 estimates were then used to generate the distributions of methane, CO<sub>2</sub> and total GHG emissions for the 160 different supply chain scenarios, as well as the contributions from both CO<sub>2</sub> and methane.

## 3. Results

### 3.1 Total supply chain emissions

The cumulative distribution of total combined levelised GHG emissions across the 160 scenarios are shown in Figure 1, where median emissions are 18.2 - 24.5 g CO<sub>2</sub> eq./ MJ HHV, with a 5<sup>th</sup> percentile range of 10.0 - 12.2 g CO<sub>2</sub> eq./ MJ HHV and a 95<sup>th</sup> percentile range of 40.4 - 181.3 g CO<sub>2</sub> eq./ MJ HHV. Each curve describes the cumulative distribution of emissions for a single theoretical supply chain scenario as detailed in the methodological section 2.1. Whilst each curve cannot be distinguished within this graph, the range of estimates and averages are indicated. Further information and results for each individual supply chain scenario is given in Supplementary Information Table SI-1. These results are broadly in line with other estimates of total supply chain emissions, typically 2 - 42 g CO<sub>2</sub> eq./ MJ HHV (Balcombe et al., 2015), although clearly the highest emitters are not accounted for. Overall, the results showed an extremely wide variation in distributions, as expected considering that many of the distributions of individual parameters were log-normal in shape. The distributions shown in Figure 1 most closely resemble log-log-logistic distributions, resulting from the high degree of skew.

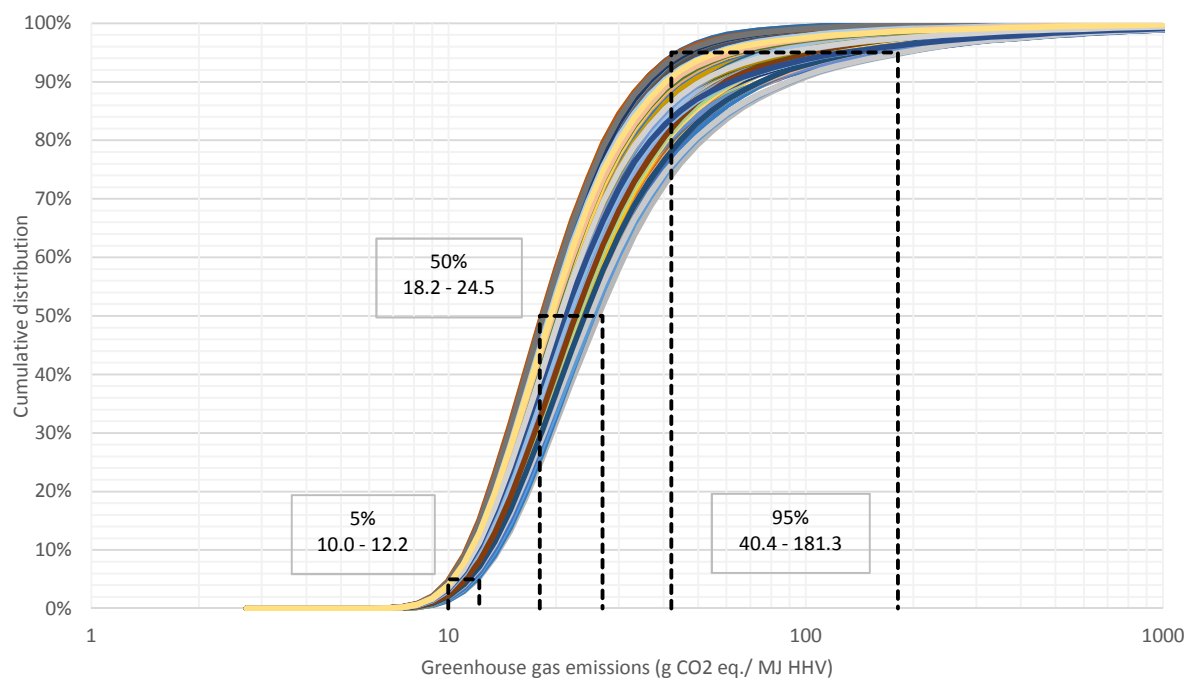


Figure 1. Cumulative distribution of total supply chain greenhouse gas emissions for each scenario. The range of 5th, 50th and 95th percentile estimates are also shown as dotted black lines.

Another demonstration of the degree of skew within the distributions is the ratio between the mean and median estimates from each scenario. A highly right-skewed distribution will result in higher estimates of mean compared to median. Mean estimates are from 21.6 - 107 g CO<sub>2</sub> eq./ MJ and the ratios of mean:median are 1.2 - 4.6 across scenarios, with an average of 2.3. A mean estimate that is 2.3 times greater than the median indicates that the higher emitters have a large impact on total GHG emissions. This is discussed further in Section 3.5.

The cumulative distribution of methane-only emissions is shown in Figure 2 for each scenario as a percentage of total produced methane. Median estimates of total supply chain emissions ranged from 0.8% to 2.2% across the scenarios, which is in line with other literature estimates (Balcombe et al., 2015). Whilst the low and median estimates were relatively constrained, the highest estimates varied widely: the 5<sup>th</sup> percentile emissions were between 0.3% and 0.6%, whereas the 95<sup>th</sup> percentile emissions ranged from 3.0% to 22.3% across all scenarios. To compare with the total greenhouse emissions described above, these emissions are equivalent to a 5<sup>th</sup> percentile estimate of 1.6 - 3.2 g CO<sub>2</sub> eq./ MJ HHV, a median estimate of 4.3 - 11.9 g CO<sub>2</sub> eq./ MJ HHV, and a 95<sup>th</sup> percentile estimate of 16.2 - 120.2 g CO<sub>2</sub> eq./ MJ HHV, using a global warming potential (GWP) of 34 g CO<sub>2</sub>/ g CH<sub>4</sub>.

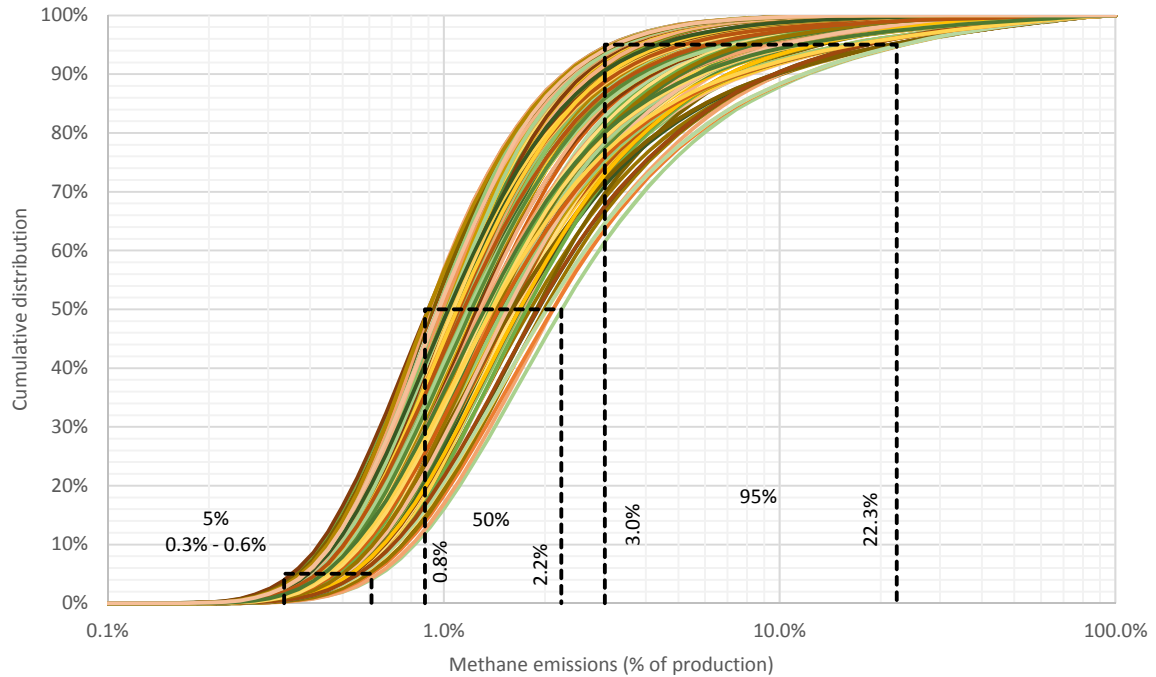


Figure 2. Cumulative distribution of total supply chain methane emissions for the 160 scenarios described in the methodology, expressed as a percentage of total methane production. The range of 5th, 50th and 95th percentile estimates are also shown as dotted black lines.

Carbon dioxide emissions from the supply chain are also variable, although the difference across scenarios is much less than seen for methane emissions. Figure 3 shows the cumulative distribution of supply chain emissions, showing a 5<sup>th</sup> percentile estimate of 6 - 6.7 g CO<sub>2</sub> / MJ HHV, a median range of 10 - 12.2 g CO<sub>2</sub> / MJ HHV, and a 95<sup>th</sup> percentile estimate of 24.5 - 66.7 g CO<sub>2</sub> / MJ HHV.

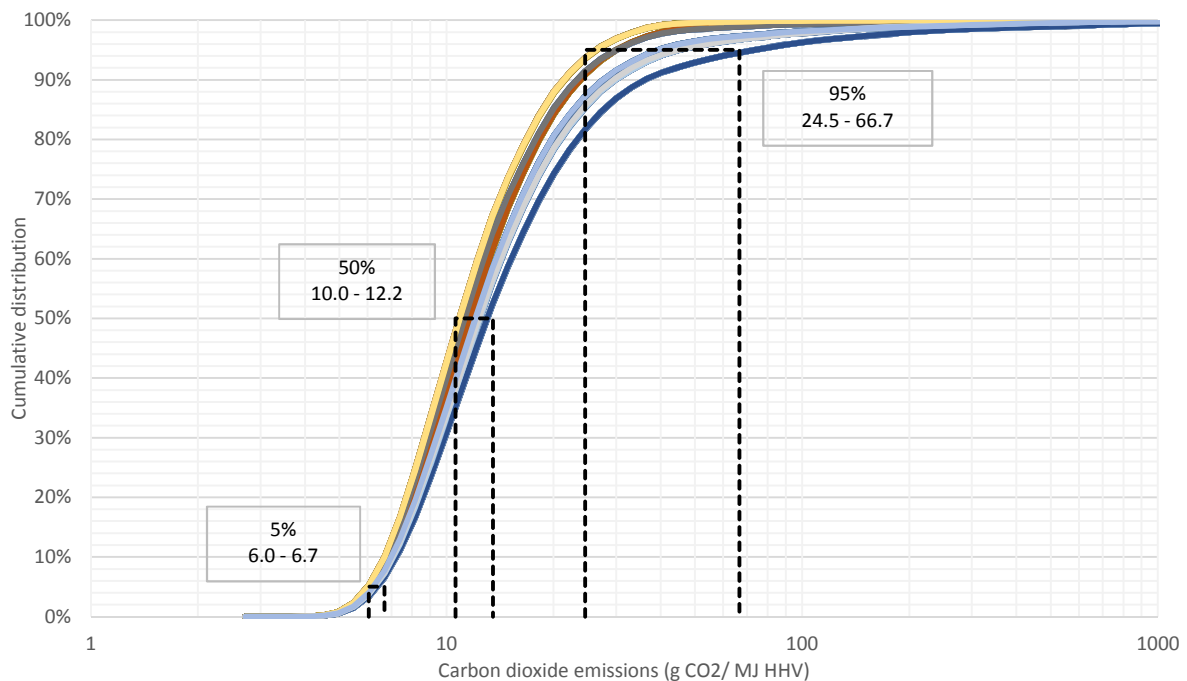


Figure 3. Cumulative distribution of total supply chain carbon dioxide emissions for the 160 scenarios described in the methodology, expressed per MJ of energy delivered on a higher heating value basis. The range of 5th, 50th and 95th percentile estimates are also shown as dotted black lines.



### 3.2 Contributions from supply chain stages

The contribution of methane emissions to total GHG emissions is generally lower than CO<sub>2</sub>, with median estimates ranging between 29% and 54% of total GHG emissions using a GWP of 34 (CO<sub>2</sub> makes up the remainder). Figure 4 shows the contribution of each supply chain stage to methane (4a) and CO<sub>2</sub> (4b) emissions. In terms of methane emissions, it is notable that emissions from the production, gathering and processing phases are generally very low (less than 0.1%) but with extremely high variances. Dominant emissions are from the transmission and storage phases (median of 0.4%), albeit with a significantly lower variation than the aforementioned stages. In terms of CO<sub>2</sub> emissions, the largest contributor by far is processing fuel requirement (a median estimate of 7 g CO<sub>2</sub> eq./ MJ HHV), typically representing over half (57%) CO<sub>2</sub> emissions.

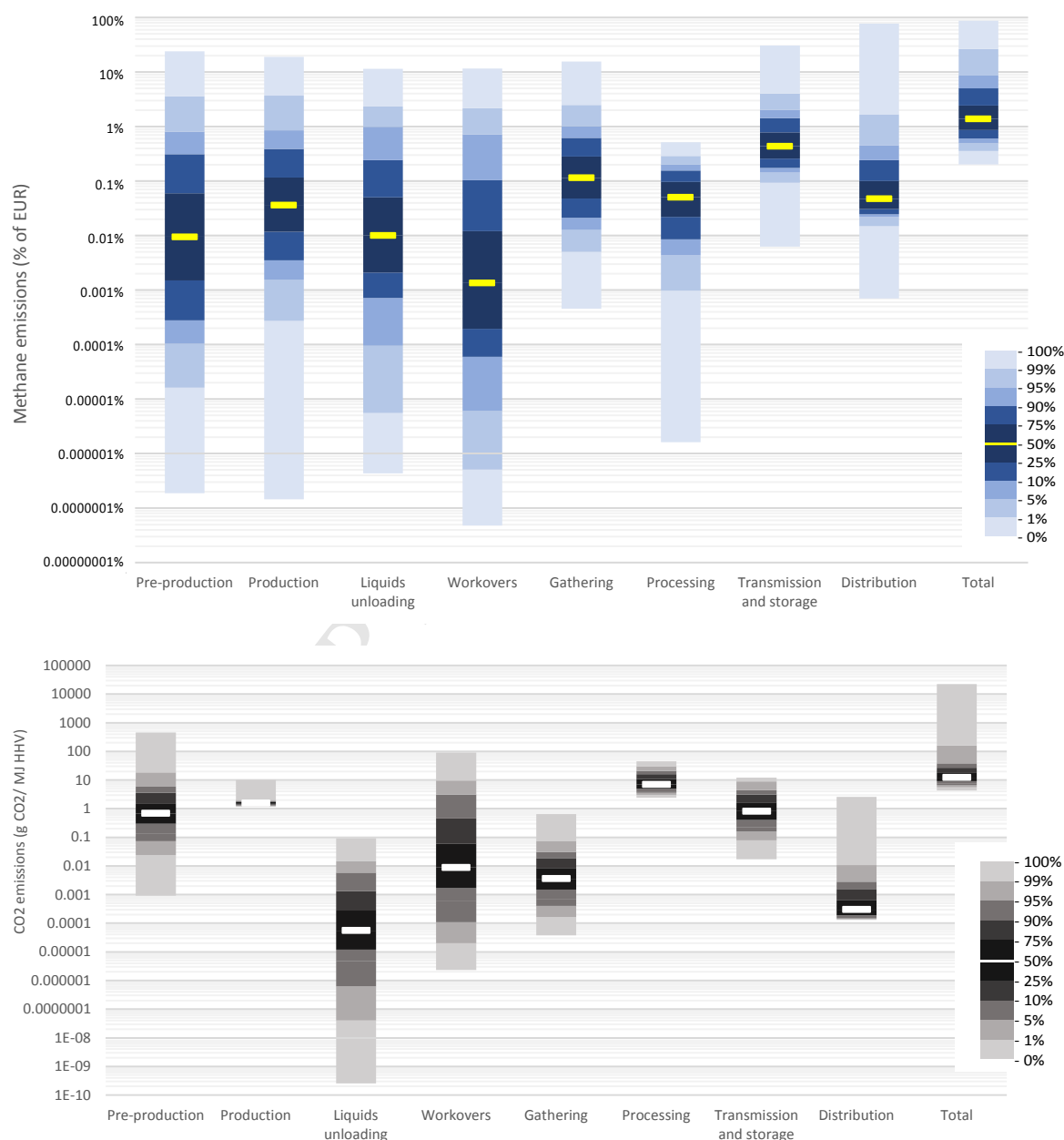


Figure 4a and b. Distribution of methane (a) and carbon dioxide (b) emissions associated with each supply chain stage. Shaded regions describe percentile estimates as shown in the legend.



### 3.3 The impact of well-type and technologies

Overall, lower GHG emissions are exhibited by: conventional wells or unconventional wells using RECs equipment and flaring residual emissions; wells with no liquids unloading emissions or manually unloaded via a plunger lift; and supply chain routes using plastic distribution pipework. These supply chain route scenarios are referred to in this study as the best technological option (BTO) in the further analysis. For those scenarios with lower average emissions, processing fuel and venting CO<sub>2</sub> emissions dominate, contributing between 31 and 36% of total supply chain emissions for the 10 lowest-emitting scenarios. The second highest contributor for lower emitting scenarios is transmission methane emissions, resulting from fugitives and vents from compressor stations, as well as pipeline leakage.

Scenarios with higher total GHG emissions become dominated by methane emissions, in particular from the event emissions such as well completions, liquids unloading and workovers. Specifically, high emissions are seen for scenarios with: liquids unloading with automated plunger lifts; completions and workovers that do not utilise RECs equipment; or even higher emissions associated with nonREC vented (as opposed to flared) scenarios.

To assess the contribution of parameter considered (well type; well completion type; liquids unloading equipment; distribution pipework), average 5<sup>th</sup>, 50<sup>th</sup> and 95<sup>th</sup> percentiles were estimated for each parameter across the supply chain scenarios. The results given in Table 5 show that individually, the different technological parameters exhibit a modest variation. The 5<sup>th</sup> percentile estimates do not vary more than 10% across categories, the median estimates by approximately 20%, whereas the 95<sup>th</sup> percentile estimates vary by around 100%. The largest variation in emissions was seen when comparing completion equipment options and liquids unloading equipment.

Table 5. Average total greenhouse gas supply chain emissions for various parameter categories, given in g CO<sub>2</sub> eq./ MJ HHV.

Parameters	5th percentile	Median	95th percentile
Well type			
Conv	11.3	21.0	60.4
Shale	11.6	22.3	97.8
Tight	11.7	22.7	100.1
Well completions			
Conv flare	11.2	20.8	61.4
Conv vent	11.4	21.1	59.4
REC flare	11.7	23.1	114.1
REC vent	11.4	21.6	70.1
NonREC flare	11.4	21.6	71.4
NonREC vent	11.8	23.8	140.2
Liquids unloading			
No unload	11.2	20.9	72.4
Man plunger	11.4	21.2	74.2
Auto plunger	12.1	24.5	122.4
No plunger	11.5	22.2	95.9
Distribution pipework			
Plastic	11.3	21.7	89.9
Protected steel	11.3	21.8	90.0
Bare steel	11.6	22.2	90.7
cast iron	11.9	23.1	94.4

However, when the different combinations of parameters are assessed in aggregate, the difference is stark. Figure 5 shows the same methane emissions distributions as in Figure 2 but highlights the options which indicate the best technological option (BTO) from the categories considered:

completions using RECs and flaring any residual vents; liquids unloading using manual plunger lifts where required; plastic distribution pipework composition. Whereas the median estimates were between 0.8% and 2% emission, the best technological option indicates a median of 0.8% - 1%, with a 95<sup>th</sup> percentile range of 3% - 3.3%. This is discussed further in Section 4.

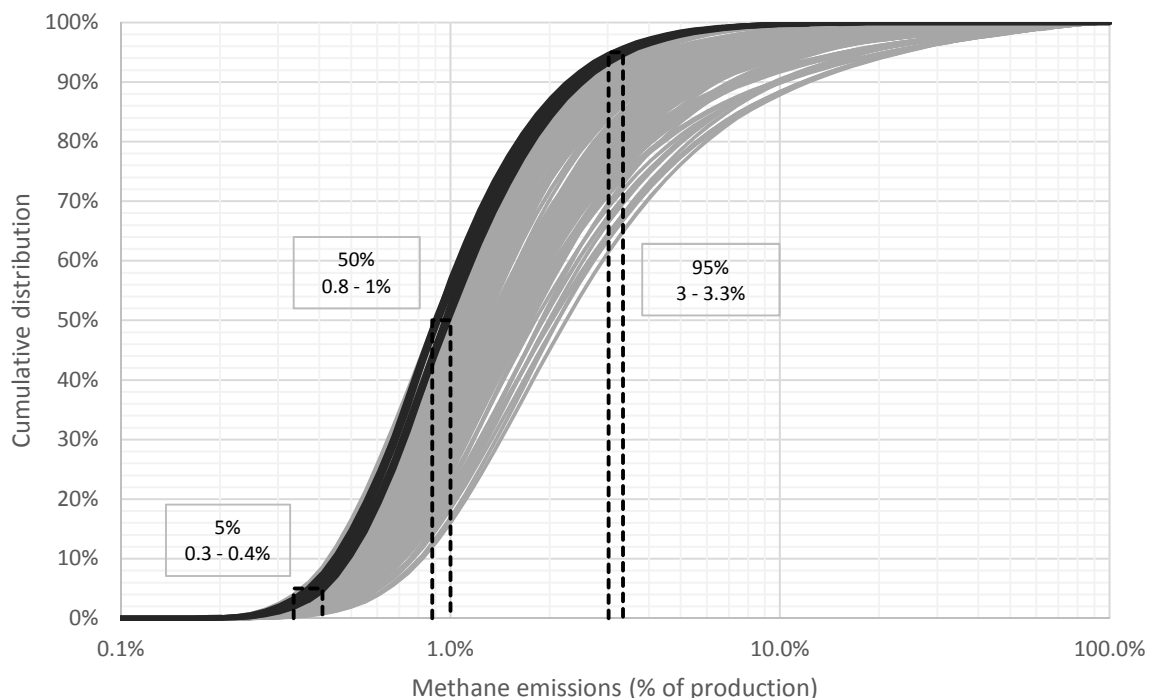


Figure 5. Cumulative distribution of methane emissions, with the best technological scenarios highlighted. 5<sup>th</sup>, 50<sup>th</sup> and 95<sup>th</sup> percentile ranges are given for these scenarios.

### 3.4 The impact of global warming potential

As has been previously noted, the assumption regarding the global warming potential (GWP) of methane has a significant impact on the estimation of combined greenhouse gas emissions. This is shown clearly in Figure 6a and b, showing the average percentile estimates of total greenhouse gas emissions for two supply chain scenarios using different GWP values from zero to 120. The two scenarios considered are: a conventional BTO scenario (ID 152 from Table SI-1); and an unconventional non-BTO scenario (ID 18 from Table SI-1). The effect of changing GWP is to proportionally change the methane contribution, with the median estimate ranging from 11 - 34.8 g CO<sub>2</sub> eq./ MJ HHV and 12.5 - 53.3 g CO<sub>2</sub> eq./ MJ HHV when GWP is changed from zero to 120 g CO<sub>2</sub>/ g CH<sub>4</sub>. The graph also indicates the mean emissions, which accounts for the heavy tail emissions. For the BTO scenario (Figure 6a), mean emissions rise from 15 to 56 g CO<sub>2</sub> eq./ MJ across the GWP values, but the second scenario (Figure 6b) exhibits mean estimates from 23 to 130.6 g CO<sub>2</sub> eq./ MJ. Given that emissions arising from combustion of gas and coal are approximately 50 and 90 g CO<sub>2</sub>/ MJ HHV respectively, the graph indicates that the supply chain is a major contributor to total emissions under certain scenarios.

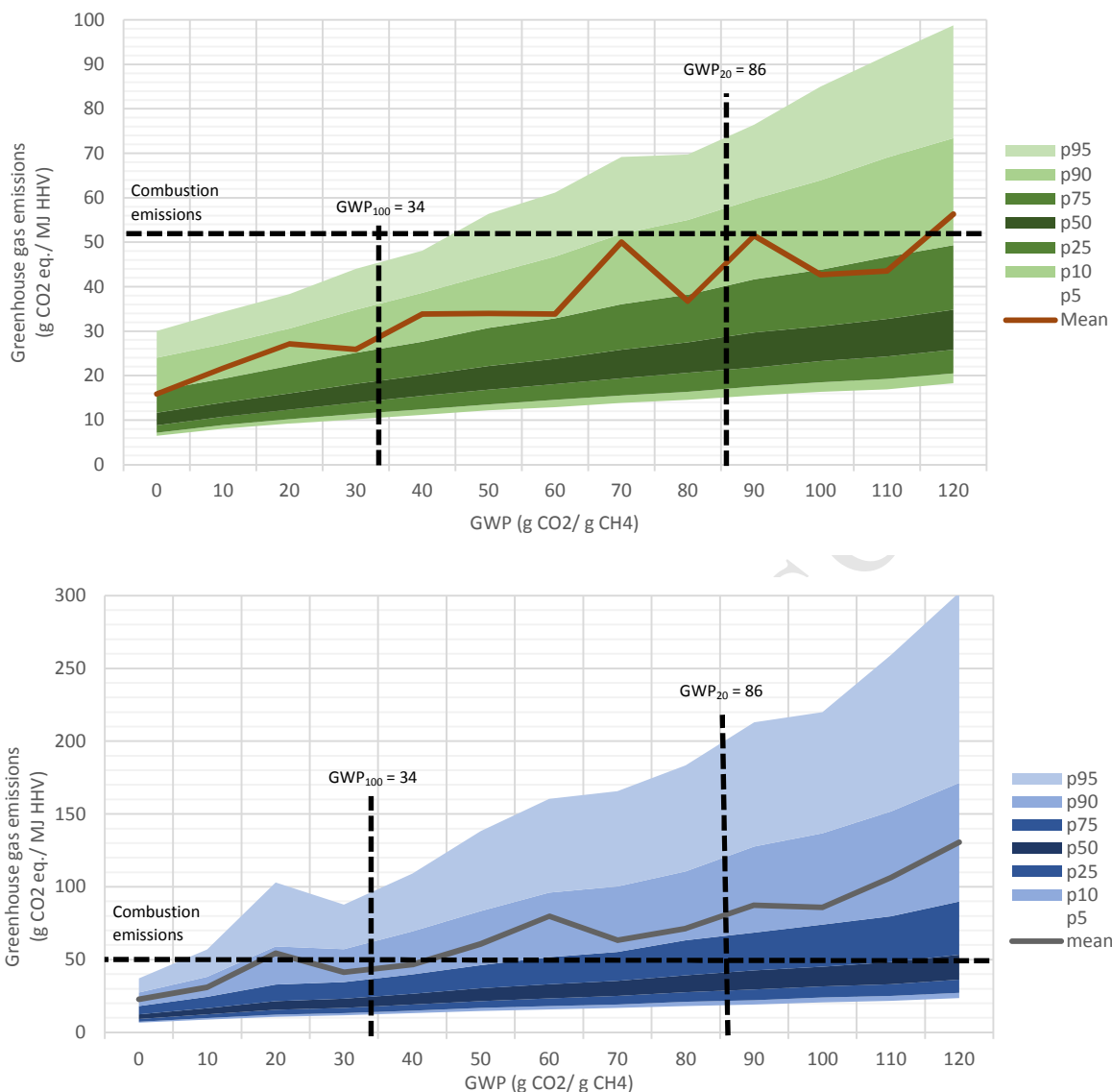


Figure 6a and b. The impact of methane global warming potential characterisation factor on total greenhouse gas emissions for two supply chain scenarios: a) conventional well, not requiring unloading, with plastic distribution pipework, and b) tight gas well, using RECs and flaring residual emissions, using an automated plunger lift and with cast iron distribution pipework. Dotted lines represent 100 and 20 year GWP characterisation factors, as well as an approximate value of gas combustion emissions.

### 3.5 The impact of EUR

The impact of the well size on levelised emissions has been discussed previously (Balcombe et al., 2015; Stamford and Azapagic, 2014) and is shown in relation to this model in Figure 7. An extremely low EUR tends to result in higher emissions as the relative impact of the intermittent events such as liquids unloading or workovers is higher. Previously it had been noted that the EUR had a significantly large and inversely proportional impact on emissions (Balcombe et al., 2015; Stamford and Azapagic, 2014). As total GHG emissions are levelised, i.e. divided by total production, an increased EUR reduces emissions on this basis. However, many continuous emissions are directly correlated to throughput, eliminating the impact of EUR. This study suggests the impact of EUR is very low for all well-sites besides the smallest. An inverse proportional relationship may be true at lower EUR values (up to approximately 30 Mm<sup>3</sup>) but as EUR increases single 'event' emissions play a smaller role than the continuous throughput-normalised emissions.

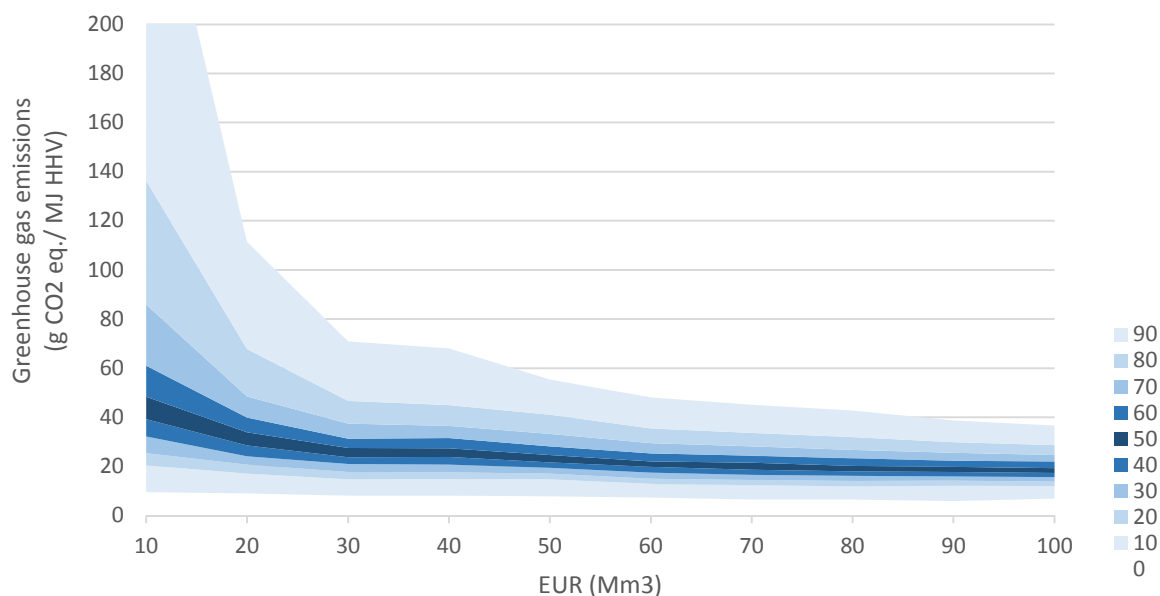


Figure 7. Percentile estimates of total GHG emissions for different estimates of estimated ultimate recovery (EUR). Different percentiles are averaged across all scenarios for the purpose of illustrating the impact of EUR.

#### 4. Discussion

The study demonstrates that emissions from the supply chain are significant in magnitude. Median emissions which represent typical facilities are relatively modest at 18.2 - 24.5 g CO<sub>2</sub> eq./ MJ HHV, but mean estimates which account for the heavy tail range from 22 – 107 g CO<sub>2</sub> eq./ MJ HHV. To place these values into context, emissions associated with natural gas combustion (e.g. for heat) are approximately 55 g CO<sub>2</sub>/ MJ HHV (assuming 100% efficiency). Thus, some supply chain scenarios are certainly significant in terms of the overall GHG emissions of natural gas.

Overall GHG emissions demonstrate a highly-skewed distribution, even more than log-normal. This corroborates recent analysis by Brandt et al. (Brandt et al., 2016), who warn that attributing log-normal distributions to methane emissions underestimates the contribution from the heavy tail. The shape of the heavy tail is different for each supply chain scenario detailed in this study.

The theoretical supply chain scenarios in this study demonstrate the effects of different technologies and routes on total emissions, but the emissions associated with specific regions, for example in the UK or the US, must be determined based on their specific emissions profiles, supply chain routes and processes. This is the subject of further work, to apply this model to specific case study regions.

There are several opportunities for emissions reduction. Firstly, technological improvement will deliver reduced emissions, as per the options investigated across the different technological supply chain scenarios (see Table 2). This study identifies certain supply chain routes that utilise the best technological option (BTO) and their improvement in terms of emissions reduction are indicated in Table 6. Notably, median estimates of emissions are reduced significantly, from 0.8 – 2.2% to 0.8 – 0.9% in terms of methane emissions. However, whilst median estimates may represent a supply chain with typical facility emissions, mean estimates account for the heavy tail. Whilst mean emissions are significantly lower, 1.7% from 2.7%, there is still a significantly heavy tail associated with the BTO scenarios, as indicated by the average skew of 1.9 (ratio of mean to median estimate).

Table 6. A comparison of emission statistics across all scenarios compared to scenarios representing the best technological option (BTO). Minimum, average and maximum figures for emissions are given for both groups, listing the median, mean and skew (ratio of mean to median estimate) for methane-only emissions and total GHG emissions.

	Methane only (% of EUR)			Total GHG emissions (g CO <sub>2</sub> eq./ MJ HHV)		
	Mean	Median	Skew (mean/ median)	Mean	Median	Skew (mean/ median)
All scenarios						
Min	1.63%	0.82%	1.7	21.6	18.2	1.2
Mean	2.65%	1.23%	2.1	48.0	20.2	2.3
Max	5.49%	2.24%	3.1	106.9	24.5	4.6
BTO						
Min	1.63%	0.82%	1.8	23.5	18.2	1.3
Mean	1.67%	0.88%	1.9	41.8	18.8	2.2
Max	1.73%	0.91%	2.0	60.2	20.1	3.0

Those scenarios utilising a BTO exhibit the smallest heavy tail but they are still significantly skewed. This demonstrates both the potential and the limits of relying on technology in decreasing overall emissions. A decrease in average methane emissions by 1% of EUR, such as the Table 6 figures show, represents a large climate and economic benefit.

For example, a GHG emissions reduction from 48 to 41 g CO<sub>2</sub> eq./ MJ HHV (as per the mean GHG figures from all scenarios and from the BTO scenarios, respectively) would result in avoided carbon cost and increased product revenue (an additional 1% of methane product). With carbon prices of 30 - 120\$/ t CO<sub>2</sub>, the avoided cost is 0.02 – 0.08 cents/ MJ, equivalent to 7% - 30% of the Henry Hub gas spot price of 0.28 cents/ MJ as at August 2016. Note, this assumes that methane emissions are incorporated into carbon pricing, which is typically not the case.

Even so, much greater emissions reduction could be achieved by tackling the heavy tail which prevails regardless of supply chain route (Brandt et al., 2016). The heavy tail is typically caused by equipment and facilities known as super-emitters, those that produce disproportionately high emissions. A number of quantitative definitions have been suggested (Brandt et al., 2016; Zavala-Araiza et al., 2015), but it is the authors' opinion that there should be no quantitative definition with respect to emissions as the population of super-emitters is continually changing, dependent on maintenance, operational and detection procedures. Super-emitters may arise for several reasons, be it equipment failure due to abnormal process conditions, age or improper maintenance, and/or operational error (Allen et al., 2014b; Zavala-Araiza et al., 2017). Such is the case with any chemical process plant, vents and leaks occur and once detected are addressed within a timeframe. Thus, super-emitters are not a discrete set of faulty equipment, but a continually changing set. A piece of equipment is only a super-emitter until the issue is addressed. Consequently, the identification and rectification processes and timelines are critical in minimising the impact of super-emitters.

This study estimates that the top 5<sup>th</sup> percentile of the supply chain population typically contributes between 40% and 60% of total emissions (with an average of 49%). It must be recognised that super-emitters cannot be simply eliminated. However, a reduction in prevalence by pre-emptive maintenance, or a reduction in the lifespan of each super-emitter by effective leak detection and repair (LDAR) processes is certainly achievable. The costs of such measures are likely to be significant, but the potential emissions reductions are also large. As a simple example, consider a theoretical measure that reduced super-emitter emissions by half (either by prevention or quicker rectification). If these super emitters were the top 5% mentioned above, this would reduce total emissions by 24% (9 - 37% across all scenarios). The significant cost benefit lies in improved economic return and reduction of environmental costs.

## 5. Limitations of the study: data and distributions

A number of limitations associated with the supply chain emissions model should be highlighted, relating to the data resolution and availability, as well as the distribution curve-fitting. Firstly, input datasets are taken from multiple sources, where data is either in the form of detailed measurement campaigns, or average figures. Aggregation of average emission factors to form a distribution will inherently underestimate the spread of emission data, given that the data is an average in the first instance. For those supply chain stages where this is applicable (e.g. transmission fugitives and vents, flaring from production, fuel use during processing), it is likely that there is greater variation that attributed here. Greater detail clearly adds to defining the distribution, but it must be acknowledged that datasets are region-specific and there are likely to be significant differences across different regions. Most of the detailed measurement campaigns have been conducted in a variety of regions across the US, implying that this study is most reflective of the US case. More high-resolution measurement data across different regions will shed further light on the variation across the globe and will help to further parameterise regional differences.

The method of fitting the distributions to the data is another source of uncertainty within the model. As described in the methodology, fitting distributions to the data was based on both quantitative and qualitative assessment, and summarised in the Supplementary Information Table SI-2 which includes an indication of the goodness of fit. Thus, the judgement from the authors plays a role in the selection of the distribution types and it has been shown that in other studies, different distribution types are selected (Brandt et al., 2016; Marchese et al., 2015; Zavala-Araiza et al., 2015). Given that the impact of the heavy tails is so significant, particular consideration was given to appropriately fitting the right-hand side of the distributions.

The availability of data for certain supply chain stages and sources has been described in Section 2.2, but it should be noted that there are gaps in data with respect to storage facilities, offshore gas sites and transmission pipeline leaks. Additionally, there are still very few high-resolution data sources for supply chains outside of the US. Key regions must face further transparent scrutiny in order to understand global natural gas impacts and the embodied emissions associated with production, processing and transport from regions such as Russia, Qatar, Australia, UK and Europe.

## 6. Conclusions

This study describes a new probabilistic model of methane and carbon dioxide emissions from the natural gas supply chain, using recent detailed measurement campaigns and data. A variety of theoretical supply chain route technological scenarios were created from the data to assess the differences across well-types and technologies. The study provides insight for industry and academia in identifying the opportunities for engineering development and emission reduction, as well as for regulators and policy-makers in illustrating the potential benefits in effectively tackling the heavy tail super-emitters.

Firstly, the distribution of GHG emissions (measured in g CO<sub>2</sub> eq./ MJ HHV) resemble a log-log-logistic distribution for most technological scenarios, indicating an extremely heavy tailed skew. Across all supply chain scenarios, median estimates were 18 – 24 g CO<sub>2</sub> eq./ MJ HHV, 5<sup>th</sup> percentile estimates were 10.0 - 12.2 g CO<sub>2</sub> eq./ MJ HHV and 95<sup>th</sup> percentile estimates were 40.4 - 181.3 g CO<sub>2</sub> eq./ MJ HHV. Methane-only estimates are expressed as a percentage of the total production, with median estimates of 0.8 – 2.2%, 5<sup>th</sup> percentile estimates of 0.3 – 0.6% and 95<sup>th</sup> percentile estimates of 3.0 – 22.3% across all scenarios. Whilst emissions from carbon dioxide are reasonably constrained within the literature, venting and fugitive methane emissions varied widely.

Given the large distributions associated with many of the individual emission sources, there are many potential emissions 'hotspots' across the supply chain. Typically, processing CO<sub>2</sub> emissions from fuel usage and transmission methane emissions from vents and leaks from compressor stations and pipelines are major contributors for all scenarios, in particular those with lower total emissions. However, scenarios with large total emissions are dominated by emissions associated with completions, liquids unloading and workovers.

Whilst the majority of the emissions are relatively low, as indicated by the median estimates, total emissions are heavily affected by the large heavy tail: arithmetic mean emissions were typically a factor of 1.5 – 5 times larger than the median estimates. The heavy tail emissions distribution is the signature of the disproportionately large emitting equipment known as super-emitters. It is widely acknowledged that super-emitters appear at each stage of the natural gas supply chain and may arise for a number of reasons, such equipment failure due to abnormal process conditions, age or improper maintenance, or be it operational error.

The study analyses the impact of different technological options across the supply chain and identifies a series of options as best technological option (BTO) scenarios. The analysis suggests that technology can reduce emissions significantly: 5<sup>th</sup>, median and 95<sup>th</sup> percentile estimates of methane emissions are 0.3%, 0.8 - 1% and 3.3 - 3.6% respectively. Median GHG emissions are 18 - 20 g CO<sub>2</sub> eq./ MJ HHV, with 5<sup>th</sup> and 95<sup>th</sup> percentile estimates of 10 and 40 - 90 g CO<sub>2</sub> eq./ MJ HHV. However, the skewed heavy tail emissions distribution still prevails: mean estimates were approximately 2 times higher than the median emissions due to the influence of the super-emitters. The BTO supply chain scenarios considered in this study reduce supply chain emissions by approximately 20%, but it is clear that a higher reduction would be achieved by reducing the heavy tail.

Pre-emptive maintenance and a faster response to high-emission detection are methods for reducing the impact of super-emitters. Identifying a cost-effective solution is imperative and much attention is being given to developing lower cost emission monitoring and detection equipment. As Brandt et al. (Brandt et al., 2016) point out, identifying larger leaks from the highest emitters may be carried out using less sensitive, and consequently cheaper, detectors in areas at the highest risk.

Further work associated with this model will be to adapt the theoretical supply chain route emissions profiles to a series of case study regions, to determine the variability and uncertainty associated with specific countries and to determine the most environmentally and cost effective routes to minimise supply chain impacts.

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## References

- Allen, D.T., 2016. Emissions from oil and gas operations in the United States and their air quality implications. *Journal of the Air & Waste Management Association* 66(6), 549-575.
- Allen, D.T., Pacsi, A.P., Sullivan, D.W., Zavala-Araiza, D., Harrison, M., Keen, K., Fraser, M.P., Daniel Hill, A., Sawyer, R.F., Seinfeld, J.H., 2014a. Methane Emissions from Process Equipment at Natural



- Gas Production Sites in the United States: Pneumatic Controllers. *Environmental Science & Technology* 49(1), 633-640.
- Allen, D.T., Sullivan, D.W., Zavala-Araiza, D., Pacsi, A.P., Harrison, M., Keen, K., Fraser, M.P., Daniel Hill, A., Lamb, B.K., Sawyer, R.F., Seinfeld, J.H., 2014b. Methane Emissions from Process Equipment at Natural Gas Production Sites in the United States: Liquid Unloadings. *Environmental Science & Technology* 49(1), 641-648.
- Allen, D.T., Torres, V.M., Thomas, J., Sullivan, D.W., Harrison, M., Hendler, A., Herndon, S.C., Kolb, C.E., Fraser, M.P., Hill, A.D., Lamb, B.K., Miskimins, J., Sawyer, R.F., Seinfeld, J.H., 2013. Measurements of methane emissions at natural gas production sites in the United States. *Proceedings of the National Academy of Sciences of the United States of America* 110(44), 17768-17773.
- Alvarez, R.A., Pacala, S.W., Winebrake, J.J., Chameides, W.L., Hamburg, S.P., 2012. Greater focus needed on methane leakage from natural gas infrastructure. *Proceedings of the National Academy of Sciences of the United States of America* 109(17), 6435-6440.
- Balcombe, P., Anderson, K., Speirs, J., Brandon, N., Hawkes, A., 2015. Methane and CO<sub>2</sub> emissions from the natural gas supply chain: an evidence assessment, in: Sustainable Gas Institute (Ed.) White Paper Series. Imperial College London, [www.sustainablegasinstitute.org/publications/white-paper-1](http://www.sustainablegasinstitute.org/publications/white-paper-1).
- Balcombe, P., Anderson, K., Speirs, J., Brandon, N., Hawkes, A., 2017. The Natural Gas Supply Chain: The Importance of Methane and Carbon Dioxide Emissions. *ACS Sustainable Chemistry & Engineering* 5(1), 3-20.
- Bond, C., Roberts, J., Hastings, A., Shipton, Z., João, E., Tabyldy Kyzy, J., Stephenson, M., 2014. Life-cycle Assessment of Greenhouse Gas Emissions from Unconventional Gas in Scotland. *ClimateXchange*, Edinburgh, UK, p. 94.
- Boothroyd, I.M., Almond, S., Qassim, S.M., Worrall, F., Davies, R.J., 2016. Fugitive emissions of methane from abandoned, decommissioned oil and gas wells. *Science of The Total Environment* 547, 461-469.
- Bouman, E.A., Ramirez, A., Hertwich, E.G., 2015. Multiregional environmental comparison of fossil fuel power generation—Assessment of the contribution of fugitive emissions from conventional and unconventional fossil resources. *International Journal of Greenhouse Gas Control* 33, 1-9.
- Brandt, A.R., Heath, G.A., Cooley, D., 2016. Methane Leaks from Natural Gas Systems Follow Extreme Distributions. *Environmental Science & Technology* 50(22), 12512-12520.
- Brandt, A.R., Heath, G.A., Kort, E.A., O'Sullivan, F., Pétron, G., Jordaan, S.M., Tans, P., Wilcox, J., Gopstein, A.M., Arent, D., Wofsy, S., Brown, N.J., Bradley, R., Stucky, G.D., Eardley, D., Harriss, R., 2014. Methane Leaks from North American Natural Gas Systems. *Science* 343(6172), 733-735.
- Brantley, H.L., Thoma, E.D., Squier, W.C., Guven, B.B., Lyon, D., 2014. Assessment of Methane Emissions from Oil and Gas Production Pads using Mobile Measurements. *Environmental Science & Technology* 48(24), 14508-14515.
- Broderick, J., Anderson, K., Wood, R., Gilbert, P., Sharmina, M., Footitt, A., Glynn, S., Nicholls, F., 2011. Shale gas: an updated assessment of environmental and climate change impacts, A report commissions by the Co-operative and undertaken by researchers at the Tyndall Centre, University of Manchester.
- Burnham, A., Han, J., Clark, C.E., Wang, M., Dunn, J.B., Palou-Rivera, I., 2012. Life-Cycle Greenhouse Gas Emissions of Shale Gas, Natural Gas, Coal, and Petroleum. *Environmental Science & Technology* 46(2), 619-627.
- Chang, Y., Huang, R., Ries, R.J., Masanet, E., 2014. Shale-to-well energy use and air pollutant emissions of shale gas production in China. *Applied Energy* 125, 147-157.
- Clearstone Engineering Ltd, 2002. Identification and evaluation of opportunities to reduce methane losses at four gas processing plants, in: US Environmental Protection Agency (Ed.). *Gas Technology Institute*, Calgary.
- Cooper, J., Stamford, L., Azapagic, A., 2014. Environmental Impacts of Shale Gas in the UK: Current Situation and Future Scenarios. *Energy Technology* 2(12), 1012-1026.



- Dale, A.T., Khanna, V., Vidic, R.D., Bilec, M.M., 2013. Process Based Life-Cycle Assessment of Natural Gas from the Marcellus Shale. *Environmental Science & Technology* 47(10), 5459-5466.
- EPA, 2014. Inventory of US Greenhouse Gas Emissions and Sinks: 1990 - 2012, in: US Environmental Protection Agency (Ed.). Washington, DC.
- EPA, 2016. Greenhouse Gas Reporting Program.
- Ferrera, M., Zandiri, S., 2010. Report on gas composition range in Europe, Integrated GAS powertrain – Low emissions, CO<sub>2</sub> optimised and efficient CNG engines for passengers cars (PC) and light duty vehicles (LDV). CRF, E.ON Ruhrgas, GDF SUEZ.
- Harrison, M., 2012. Revised Attachment 3: Gas Well Completion Emissions Data, in: URS corporation (Ed.). US EPA, Austin, Texas.
- Heath, G., Meldrum, J., Fisher, N., Arent, D., Bazilian, M., 2014. Life cycle greenhouse gas emissions from Barnett Shale gas used to generate electricity. *Journal of Unconventional Oil and Gas Resources* 8(0), 46-55.
- IEA, 2014. Key World Energy Statistics 2015, in: International Energy Agency (Ed.). Paris.
- Ishkov, A., Akopova, G., Evans, M., Yulkin, G., Roshchanka, V., Waltzer, S., Romanov, K., Picard, D., Stepanenko, O., Neretin, D., 2011. Understanding Methane Emission Sources and Viable Mitigation Measures in the Natural Gas Transmission Systems: Russian and US Experience, International Gas Union Research Conference 2011. Seoul, South Korea.
- Jiang, M., Griffin, W.M., Hendrickson, C., Jaramillo, P., VanBriesen, J., Venkatesh, A., 2011. Life cycle greenhouse gas emissions of Marcellus shale gas. *Environmental Research Letters* 6(3).
- Kang, M., Kanno, C.M., Reid, M.C., Zhang, X., Mauzerall, D.L., Celia, M.A., Chen, Y., Onstott, T.C., 2014. Direct measurements of methane emissions from abandoned oil and gas wells in Pennsylvania. *Proceedings of the National Academy of Sciences* 111(51), 18173-18177.
- Lamb, B.K., Edburg, S.L., Ferrara, T.W., Howard, T., Harrison, M.R., Kolb, C.E., Townsend-Small, A., Dyck, W., Possolo, A., Whetstone, J.R., 2015. Direct Measurements Show Decreasing Methane Emissions from Natural Gas Local Distribution Systems in the United States. *Environmental Science & Technology* 49(8), 5161-5169.
- Lechtenboehmer, S., Dienst, C., 2010. Future development of the upstream greenhouse gas emissions from natural gas industry, focussing on Russian gas fields and export pipelines. *Journal of Integrative Environmental Sciences* 7, 39-48.
- Leliveld, J., Lechtenbolumer, S., Assonov, S.S., Brenninkmeijer, C.A.M., Dienst, C., Fishedick, M., Hanke, T., 2005. Greenhouse gases: Low methane leakage from gas pipelines, *Nature*. pp. 841-842.
- Logan, J.A., Heath, G., Macknick, J., Paranhos, E., Boyd, W., Carlson, K., 2012. Natural Gas and the Transformation of the U.S. Energy Sector: Electricity, in: Joint Institute for Strategic Energy Analysis, National Renewable Energy Laboratory (NREL), University of Colorado Law School, Colorado State University (Eds.). JISEA & NREL.
- Lyon, D.R., Zavala-Araiza, D., Alvarez, R.A., Harriss, R., Palacios, V., Lan, X., Talbot, R., Lavoie, T., Shepson, P., Yacovitch, T.I., Herndon, S.C., Marchese, A.J., Zimmerle, D., Robinson, A.L., Hamburg, S.P., 2015. Constructing a Spatially Resolved Methane Emission Inventory for the Barnett Shale Region. *Environmental Science & Technology* 49(13), 8147-8157.
- MacKay, D.J.C., Stone, T.J., 2013. Potential Greenhouse Gas Emissions Associated with Shale Gas Extraction and Use. Department of Energy & Climate Change, London.
- Marchese, A.J., Vaughn, T.L., Zimmerle, D.J., Martinez, D.M., Williams, L.L., Robinson, A.L., Mitchell, A.L., Subramanian, R., Tkacik, D.S., Roscioli, J.R., Herndon, S.C., 2015. Methane Emissions from United States Natural Gas Gathering and Processing. *Environmental Science & Technology*.
- McKain, K., Down, A., Raciti, S.M., Budney, J., Hutyra, L.R., Floerchinger, C., Herndon, S.C., Nehr Korn, T., Zahniser, M.S., Jackson, R.B., Phillips, N., Wofsy, S.C., 2015. Methane emissions from natural gas infrastructure and use in the urban region of Boston, Massachusetts. *Proceedings of the National Academy of Sciences* 112(7), 1941-1946.
- Mitchell, A.L., Tkacik, D.S., Roscioli, J.R., Herndon, S.C., Yacovitch, T.I., Martinez, D.M., Vaughn, T.L., Williams, L.L., Sullivan, M.R., Floerchinger, C., Omara, M., Subramanian, R., Zimmerle, D., Marchese,

- A.J., Robinson, A.L., 2015. Measurements of Methane Emissions from Natural Gas Gathering Facilities and Processing Plants: Measurement Results. *Environmental Science & Technology* 49(5), 3219-3227.
- NGML, 2006. Cost-Effective Directed Inspection and Maintenance Control Opportunities at Five Gas Processing Plants and Upstream Gattling Compressor Stations and Well Sites, in: National Gas Machinery Laboratory, Clearstone Engineering Ltd, Innovative Environmental Solutions (Eds.), EPA Phase II Aggregate Site Report. US Environmental Protection Agency.
- O'Donoghue, P.R., Heath, G.A., Dolan, S.L., Vorum, M., 2014. Life Cycle Greenhouse Gas Emissions of Electricity Generated from Conventionally Produced Natural Gas. *Journal of Industrial Ecology* 18(1), 125-144.
- Omara, M., Sullivan, M.R., Li, X., Subramanian, R., Robinson, A.L., Presto, A.A., 2016. Methane Emissions from Conventional and Unconventional Natural Gas Production Sites in the Marcellus Shale Basin. *Environmental Science & Technology* 50(4), 2099-2107.
- Rella, C.W., Tsai, T.R., Botkin, C.G., Crosson, E.R., Steele, D., 2015. Measuring Emissions from Oil and Natural Gas Well Pads Using the Mobile Flux Plane Technique. *Environmental Science & Technology* 49(7), 4742-4748.
- Rubinstein, R.Y., Kroese, D.P., 2011. Simulation and the Monte Carlo method. John Wiley & Sons.
- Santoro, R.L., Howarth, R.W., Ingraffea, A.R., 2011. Indirect Emissions of Carbon Dioxide from Marcellus Shale Gas Development, A Technical Report from the Agriculture, Energy, & Environment Program at Cornell University.
- Sevenster, M., Croezen, H., 2006. The natural gas chain: Toward a global life cycle assessment, in: CE (Ed.). Delft.
- Shahriar, A., Sadiq, R., Tesfamariam, S., 2014. Life cycle greenhouse gas footprint of shale gas: a probabilistic approach. *Stochastic Environmental Research and Risk Assessment* 28(8), 2185-2204.
- Shires, T., Lev-On, M., 2012. Characterizing Pivotal Sources of Methane Emissions from Natural Gas Production, in: API and ANGA (Ed.). URS Corporation and The LEVON Group, .
- Skone, T.J., 2011. Life Cycle Greenhouse Gas Inventory of Natural Gas Extraction, Delivery and Electricity Production in: NETL (Ed.). National Energy Technology Laboratory.
- Skone, T.J., Marriott, J., Cooney, G., Jamieson, M., Hakian, J., Schivley, G., 2014. Life Cycle Analysis of Natural Gas Extraction and Power Generation. US Department of Energy, National Energy Technology Laboratory.
- Stamford, L., Azapagic, A., 2014. Life cycle environmental impacts of UK shale gas. *Applied Energy* 134, 506-518.
- Stephenson, T., Valle, J.E., Riera-Palou, X., 2011. Modeling the Relative GHG Emissions of Conventional and Shale Gas Production. *Environmental Science & Technology* 45(24), 10757-10764.
- Subramanian, R., Williams, L.L., Vaughn, T.L., Zimmerle, D., Roscioli, J.R., Herndon, S.C., Yacovitch, T.I., Floerchinger, C., Tkacik, D.S., Mitchell, A.L., Sullivan, M.R., Dallmann, T.R., Robinson, A.L., 2015. Methane Emissions from Natural Gas Compressor Stations in the Transmission and Storage Sector: Measurements and Comparisons with the EPA Greenhouse Gas Reporting Program Protocol. *Environmental Science & Technology* 49(5), 3252-3261.
- Townsend-Small, A., Ferrara, T.W., Lyon, D.R., Fries, A.E., Lamb, B.K., 2016. Emissions of coalbed and natural gas methane from abandoned oil and gas wells in the United States. *Geophysical Research Letters* 43(5), 2283-2290.
- Townsend-Small, A., Marrero, J.E., Lyon, D.R., Simpson, I.J., Meinardi, S., Blake, D.R., 2015. Integrating Source Apportionment Tracers into a Bottom-up Inventory of Methane Emissions in the Barnett Shale Hydraulic Fracturing Region. *Environmental Science & Technology* 49(13), 8175-8182.
- UNFCCC, 2015. Adoption of the Paris Agreement, in: United Nations Framework Convention on Climate Change (Ed.) 21st Conference of the Parties. United Nations, Paris.
- Venkatesh, A., Jaramillo, P., Griffin, W.M., Matthews, H.S., 2011. Uncertainty in Life Cycle Greenhouse Gas Emissions from United States Natural Gas End-Uses and its Effects on Policy. *Environmental Science & Technology* 45(19), 8182-8189.

- Vielstädte, L., Karstens, J., Haeckel, M., Schmidt, M., Linke, P., Reimann, S., Liebetrau, V., McGinnis, D.F., Wallmann, K., 2015. Quantification of methane emissions at abandoned gas wells in the Central North Sea. *Marine and Petroleum Geology* 68, Part B, 848-860.
- Weber, C.L., Clavin, C., 2012. Life Cycle Carbon Footprint of Shale Gas: Review of Evidence and Implications. *Environmental Science & Technology* 46(11), 5688-5695.
- Weisser, D., 2007. A guide to life-cycle greenhouse gas (GHG) emissions from electric supply technologies. *Energy* 32(9), 1543-1559.
- Yacovitch, T.I., Herndon, S.C., Pétron, G., Kofler, J., Lyon, D., Zahniser, M.S., Kolb, C.E., 2015. Mobile Laboratory Observations of Methane Emissions in the Barnett Shale Region. *Environmental Science & Technology* 49(13), 7889-7895.
- Zavala-Araiza, D., Alvarez, R.A., Lyon, D.R., Allen, D.T., Marchese, A.J., Zimmerle, D.J., Hamburg, S.P., 2017. Super-emitters in natural gas infrastructure are caused by abnormal process conditions. *Nature Communications* 8, 14012.
- Zavala-Araiza, D., Lyon, D.R., Alvarez, R.A., Davis, K.J., Harriss, R., Herndon, S.C., Karion, A., Kort, E.A., Lamb, B.K., Lan, X., Marchese, A.J., Pacala, S.W., Robinson, A.L., Shepson, P.B., Sweeney, C., Talbot, R., Townsend-Small, A., Yacovitch, T.I., Zimmerle, D.J., Hamburg, S.P., 2015. Reconciling divergent estimates of oil and gas methane emissions. *Proceedings of the National Academy of Sciences* 112(51), 15597-15602.
- Zimmerle, D.J., Williams, L.L., Vaughn, T.L., Quinn, C., Subramanian, R., Duggan, G.P., Willson, B., Opsomer, J.D., Marchese, A.J., Martinez, D.M., Robinson, A.L., 2015. Methane Emissions from the Natural Gas Transmission and Storage System in the United States. *Environmental Science & Technology* 49(15), 9374-9383.

## Highlights

- Probabilistic model of methane and CO<sub>2</sub> emissions from the natural gas supply chain
- Distribution of emissions is greater than log-log-logistic indicating a heavy tail
- Median methane emissions are 0.8 - 2.2% of total production
- Technologies can reduce emissions significantly but do not eliminate super emitters
- Preventative maintenance and effective detection will reduce super emitters