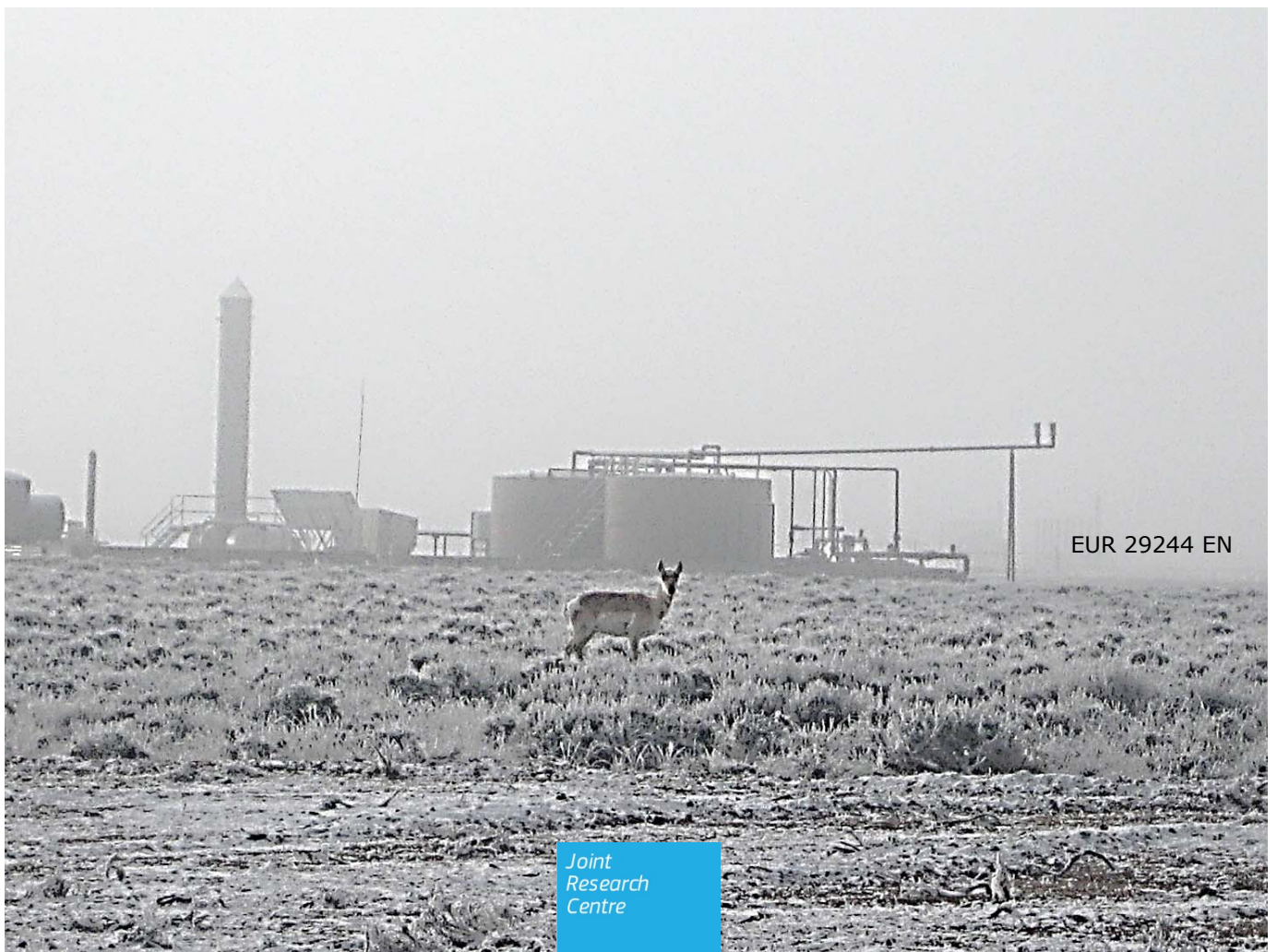# JRC TECHNICAL REPORTS

Unconventional oil and gas development: Evaluation of selected hydrocarbons in the ambient air of three basins in the United States by means of diffusive sampling measurements

Pérez Ballesta P., Field R.A., Cabrerizo A. and Edie R.

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Acknowledgements

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Abstract

The impact of emissions associated with the extraction of crude oil and natural gas upon air quality in the United States (US) is widely recognised to have an impact on climate change, human health and ground-level ozone formation. A number of measurement approaches are being applied to evaluate the environmental impact of the oil and gas (O&G) sector, including satellite, airborne and ground-based platforms. Measurement based studies, in particular those that estimate flux rates, are critical for the validation of emission inventories that often under-report actual emissions of methane and volatile organic compounds (VOC) from the O&G sector. On-going research projects in the US are investigating the consistency of emission rates from O&G emission sources associated with extraction, transmission and distribution activities. The leakage rates of methane, as related to production levels, in the US for O&G developments varies from less than 1% (e.g. Upper Green River Basin, Wyoming) to over 6% (Uintah Basin, Utah). European research and policy approaches can learn from efforts in the US that are improving the accuracy of reporting emissions from O&G sources, enhancing our understanding of air quality impacts, and reducing emissions through regulatory controls.

The Joint Research Centre (JRC) of the European Commission performed a diffusive sampling project, with the collaboration of the University of Wyoming, in conjunction with the SONGNEX (Studying the Atmospheric Effects of Changing Energy Use in the US at the Nexus of Air Quality and Climate Change) project led by the US National Oceanic and Atmospheric Administration. The SONGNEX project is an airborne measurement campaign supported by a number of associated ground based studies. The applicability of the Pocket Diffusive (PoD) sampler, for measurement of VOC (C_4-C_{10}), heavy hydrocarbons and volatile polycyclic aromatic hydrocarbons (PAHs) in areas heavily influenced by O&G development, is evaluated. Three sampling surveys were performed to assess three basins (Upper Green River, Uintah and North Platte) characterised by different management regimes, meteorology and hydrocarbon products.

This first extensive field deployment of the PoD sampler demonstrates the effectiveness of the sampler for time-integrated measurements of targeted pollutants over wide spatial areas. The ambient air at these basins reveals different compositional profiles of hydrocarbons (C_4-C_{10}). Analysis of aromatics supports a finding of relatively elevated levels in the Pinedale Anticline (Upper Green River). From an evaluation of the behaviour of alkanes, it is evident that there is a relatively high leakage rate in the Uintah Basin. Heavy hydrocarbons (C_{11}-C_{22}) and PAHs are measured at relatively low levels. Despite low concentrations, analysis of these compounds improves the accuracy of source identification. A comparison of ground based PoD data and airborne SONGNEX data showed good agreement for commonly reported VOCs. The utility of the PoD sampler for analysis of emission sources was enhanced with reporting of a wide range of compounds. Spatial Positive Matrix Factorisation analysis showed the possibility of using PoD samplers for differentiating emission sources, characterising different areas and estimating the relative contribution of different emission sources.

1 INTRODUCTION

While unconventional Oil and Gas (O&G) development within Europe is in its infancy, the EU is looking to the US not only for guidance for preparation of regulatory frameworks, but also for designing impact assessment approaches. During recent years, a number of different measurement, modelling and analysis approaches have been applied in the US to understand the actual impact of O&G development. Annex 7 of this report provides a detailed description of the progression of policy and research studies related to the O&G sector carried out in US up to the end of 2016.

This project tests the ability of diffusive sampling to act as a scoping tool for measuring potential impacts from O&G development activities. Compared to Europe, emission control policies and the study of air quality impacts from the extraction of unconventional O&G resources in the US is more advanced. Therefore, the European Commission collaborated with the University of Wyoming and the US National Oceanic and Atmospheric Administration (NOAA) with sampler deployment in the assessment of three US basins with significant unconventional O&G development. This diffusive sampling campaign was conducted in conjunction with the SONGNEX (Studying the Atmospheric Effects of Changing Energy Use in the US at the Nexus of Air Quality and Climate Change) project led by NOAA. The primary aim of the airborne SONGNEX project was to determine basin-scale emission rates from O&G developments. **Figure 1** shows the extent of the flights performed by NOAA for the assessment of the O&G sector through a variety of projects, including SONGNEX.
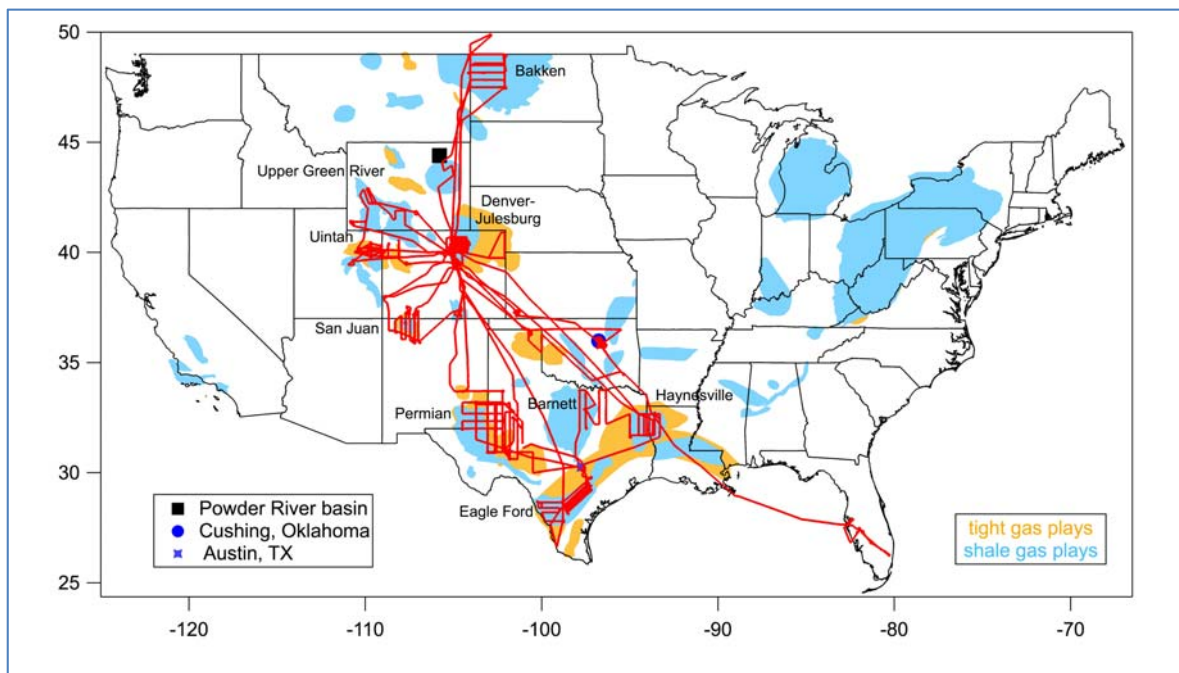
(Source EIA, courtesy of Joost De Gouw)

Figure 1 - SONGNEX Flights 2015 and NOAA Oil & Gas Research areas

While data validation is complete for the SONGNEX study (NOAA 2015) at the writing of this report, analysis is on-going for the airborne measurements as well as supporting projects, e.g. JAGUAR (Joint Air and Ground based Uintah Basin Air emissions Reconciliation project). Preliminary results for the SONGNEX project were discussed at a workshop at Boulder (CO) in October 2015 and at dedicated sessions at the AGU conference in December of 2015 and 2016. Through 2016 and into 2018 a number of academic papers related to the SONGNEX project are now under preparation, submitted or published. Papers related to the analysis of O&G emissions from SONGNEX, and

through other US projects that were published in 2017 and 2018 are not included in this report (ANNEX II provides a comprehensive literature review up to 2016).

SONGNEX was designed to address three key issues (NOAA, 2015): Climate, Air Quality, and Air Toxics (also known as Hazardous Air Pollutants, HAPs). A key driver for the project is the evaluation of emission inventories for the O&G sector, through analysis of atmospheric observations. Miller et al. (2013) report that regional methane emissions due to fossil fuel extraction and processing could be 4.9 ± 2.6 times larger than estimates reported in EDGAR (Emission Database Global Atmospheric Research).

(Courtesy of Joost De Gouw)

Figure 2 - SONGNEX P3 aircraft

For the diffusive sampling project, undertaken by the JRC of the European Commission and the University of Wyoming, surveys were performed in three basins with contrasting situations in terms of production type (water, oil and gas balance), production methods, regulations, geography, meteorology and anticipated O&G emissions. The Upper Green River Basin (UGRB) contains three distinct developments, namely the La Barge/Big Piney area; the Jonah Field; and the Pinedale Anticline. The La Barge/Big Piney area is an older development with a higher proportion of conventional oil wells compared to the Jonah Field or Pinedale Anticline. This area has potential for future unconventional development. The Pinedale Anticline is an active field that has experienced considerable development in the past 20 years. The Pinedale Anticline produces high volumes of gas, condensate (lower density than crude oil) and water. The Jonah Field was developed earlier than the Pinedale Anticline and while production is similar, albeit with relatively less water production, the field differs by having less directional drilling, more well pads and on-pad liquids storage in tanks. The area surrounding the Jonah Field is under review for further extraction activities (Normally Pressurized Lance project). The western side of the Uintah Basin is dominated by oil production; many wells are old and inactive.

The eastern edge of the Uintah Basin is dominated by wet gas production. Sites were selected in the eastern area due to the relative similarity of hydrocarbon product with the Pinedale Anticline and Jonah Field. The Converse County region of the North Platte Basin is an oil rich area with many old and inactive wells. However, this area has more new wells being drilled than other regions in Wyoming. These three basins were selected due to the similarities and differences that they presented in terms production of oil, gas and water (see **Table 1**). Production data was accessed from Wyoming Oil and Gas Conservation Commission (2015) and the Utah Oil and Gas Commission (2015). In this report we refer to gas, this term is synonymous with natural gas, however natural gas may be defined as dry (predominately methane) or wet (greater levels of water and associated hydrocarbons than dry gas). Operations at well pads handling wet gas are associated with separation of production into water, gas and condensate oil. As for gas, oil is also a broad definition. Condensate and heavier crude oil are both reported by developers and regulators as oil, without distinction. But these two categories of oil have very different composition, density and volatility. As such, similar handling approaches may be associated with different emission profiles.

The similarities and differences between the three basins are evident when considering the monthly production volumes for 2014 (Table 1). The highest level of the production of gas, oil, and water was around sampling sites in the Pinedale Anticline. The lowest level of production of oil and water is associated with the samplers in the La Barge/Big Piney area. Converse County has the lowest production of gas, likely accentuated by flaring of hydrocarbons in this region due to a lack of gas supply transmission pipelines. When considering the ratios of gas to water to oil there are clear differences between the well bore production from these fields. The Pinedale Anticline and Jonah Field have the highest gas to water ratios. The gas to water ratio at the Uintah Basin, La Barge/Big Piney area and Converse County are lower and of comparable magnitude to each other. These basins use similar extraction techniques with a mix of traditional Derek style oil pumps and gas separators. Oil to water ratios have a narrower range with the lowest ratios evident for the Pinedale Anticline and La Barge/Big Piney area.

Table 1 - Monthly oil, gas, water production and ratios in the studied areas during 2014

Basin	Oil (barrels)	Gas (Mcf)	Water (barrels)	Gas/Oil	Gas/Water	Oil/Water
Uintah Basin	2,744,675	30,252,764	9,253,200[*]	11.0	3.3	0.30
Pinedale Anticline	300,038	40,122,047	1,403,306	133.7	28.6	0.21
La Barge/Big Piney	17,076	823,350	87,543	48.2	9.4	0.20
Jonah Field	152,817	15,726,339	330,876	102.9	47.5	0.46
Converse County	1,068,339	3,049,573	983,906	2.9	3.1	1.09

*estimated using 2015 data

The main objective of this diffusive sampling pilot study was to show the potential use of the diffusive sampling technique for assessment of the impact of emissions from O&G development areas upon air quality. This potential was initially considered by determining if samplers could clearly differentiate the quality of background air and air influenced by emission sources. As a next step, the ability of samplers to accurately characterise the hydrocarbon profile of areas with different combinations of development activities and emission sources was assessed. As such, the applicability of the sampler as a tool for spatial source apportionment technique was determined. Finally, through a comparison with results from the SONGNEX project a further level of field validation was performed.

1.1 Upper Green River Basin

The UGRB is located in Sublette County Wyoming, a remote area of the US, with significant O&G development (**Figure 3**). The UGRB is enclosed by the Wyoming mountain range to the west, the Gros Ventre range to the north, and the Wind River Mountains to the northeast.
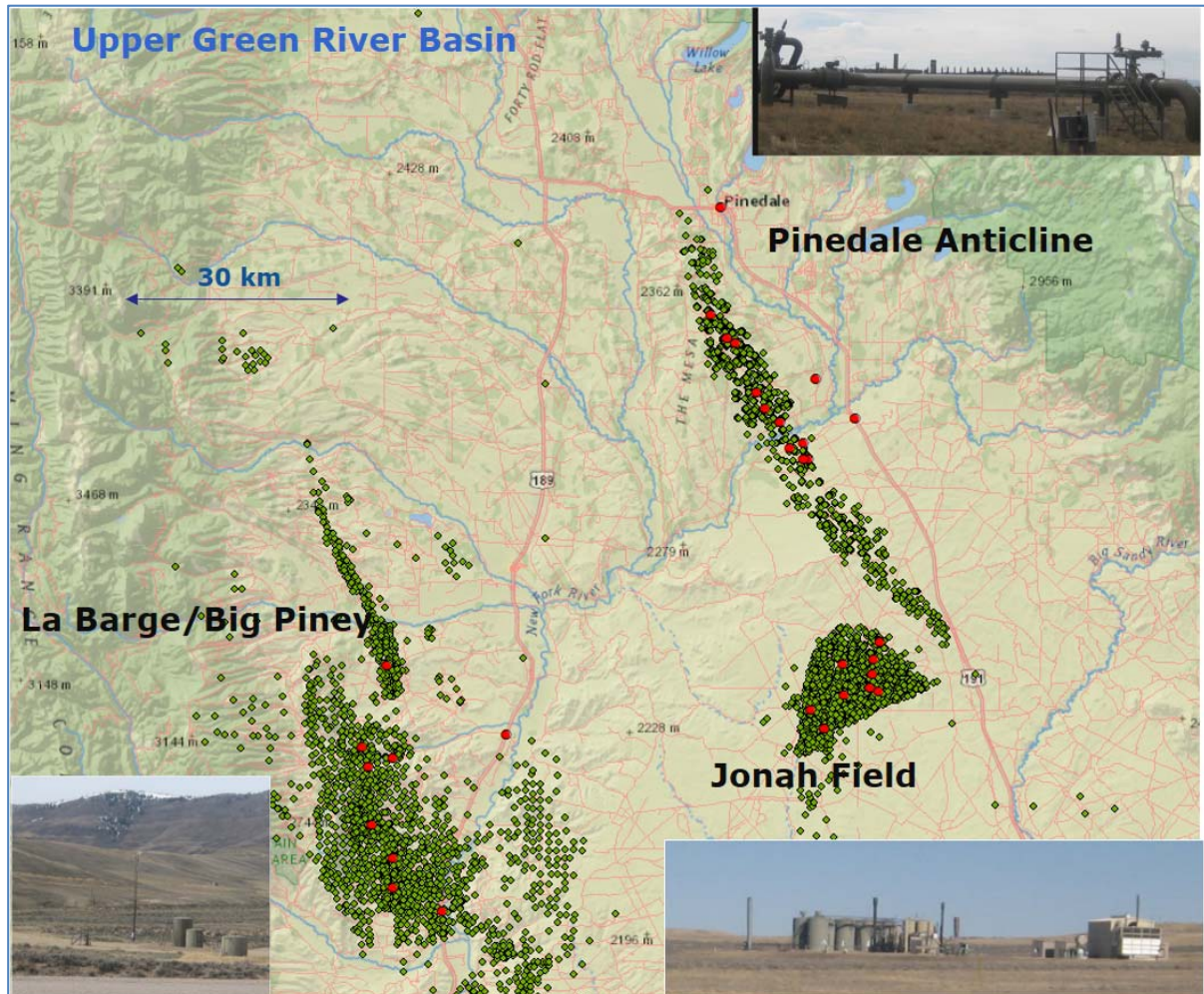
Figure 3 - Wells (green) and diffusive sampling locations (red) in the Upper Green River Basin

The UGRB contains three important O&G developments: the Big Piney/La Barge area, the Jonah Field and the Pinedale Anticline. The latter two are both ranked in the top 10 of US onshore natural gas fields. The Jonah Field and Pinedale Anticline are also ranked in the top 100 of onshore oil fields in the US (US EIA, 2015b). By contrast, while the Jonah Field and Pinedale Anticline are both hydraulically fractured unconventional developments with oil produced as condensate, the La Barge/Big Piney area is predominately a conventional crude oil development. Of the ~ 6000 wells in the UGRB, ~3000 are in the Pinedale Anticline ~2000 are in Jonah, and ~1000 are in La Barge/Big Piney area. **Figure 3** shows the distribution of wells in the UGRB.

The phenomenon of wintertime ozone (O_3) was first reported in the scientific literature in the UGRB by Schnell et al. (2009). They reported that wintertime O_3 episodes measured at Wyoming Department of Environmental (WDEQ) monitoring stations in 2005 and 2008 were a result of precursor emissions from the Pinedale Anticline and Jonah Field. Pollutant emissions in the UGRB are almost entirely from O&G activities with minimal contributions from other sources (WDEQ, 2012).

Since 2005, air quality and meteorological monitoring stations have been deployed throughout the UGRB to determine the extent of O_3 episodes and their characteristics. Modelling studies of 2011 wintertime O_3 in the UGRB (Carter and Seinfeld 2012; and Rappenglück et al. 2014) have demonstrated that O_3 production is critically dependent upon non methane hydrocarbons (NMHC). Additional research has focused upon the distribution and specific identity of NMHC compounds that have the highest O_3 production potential (Field et al. 2015a). These monitoring and modelling studies have led to significant progress in understanding the mechanisms and critical reaction pathways for wintertime O_3 formation. Field et al. (2015a) reinforced the importance of fugitive emissions of natural gas and showed the influence of emissions from a water treatment facility designed to recycle and treat water from the Pinedale Anticline.

1.2 Uintah Basin

The Uintah Basin is located in north-eastern Utah, and as for the UGRB in Wyoming, has experienced wintertime O_3 that is driven by emissions from production of O&G activities (**Figure 4**). High wintertime O_3 concentrations were reported in the winters 2010 and 2011 and 2013. Significant scientific progress with respect to understanding the importance of winter O_3 production was made through three winter campaigns led by NOAA (Edwards et al 2014; Ahmadov et al. 2015; Schnell et al. 2016). These field intensives known as the Uintah Basin Winter O_3 Study (UBWOS) were conducted in the winters of 2012, 2013 and 2014.
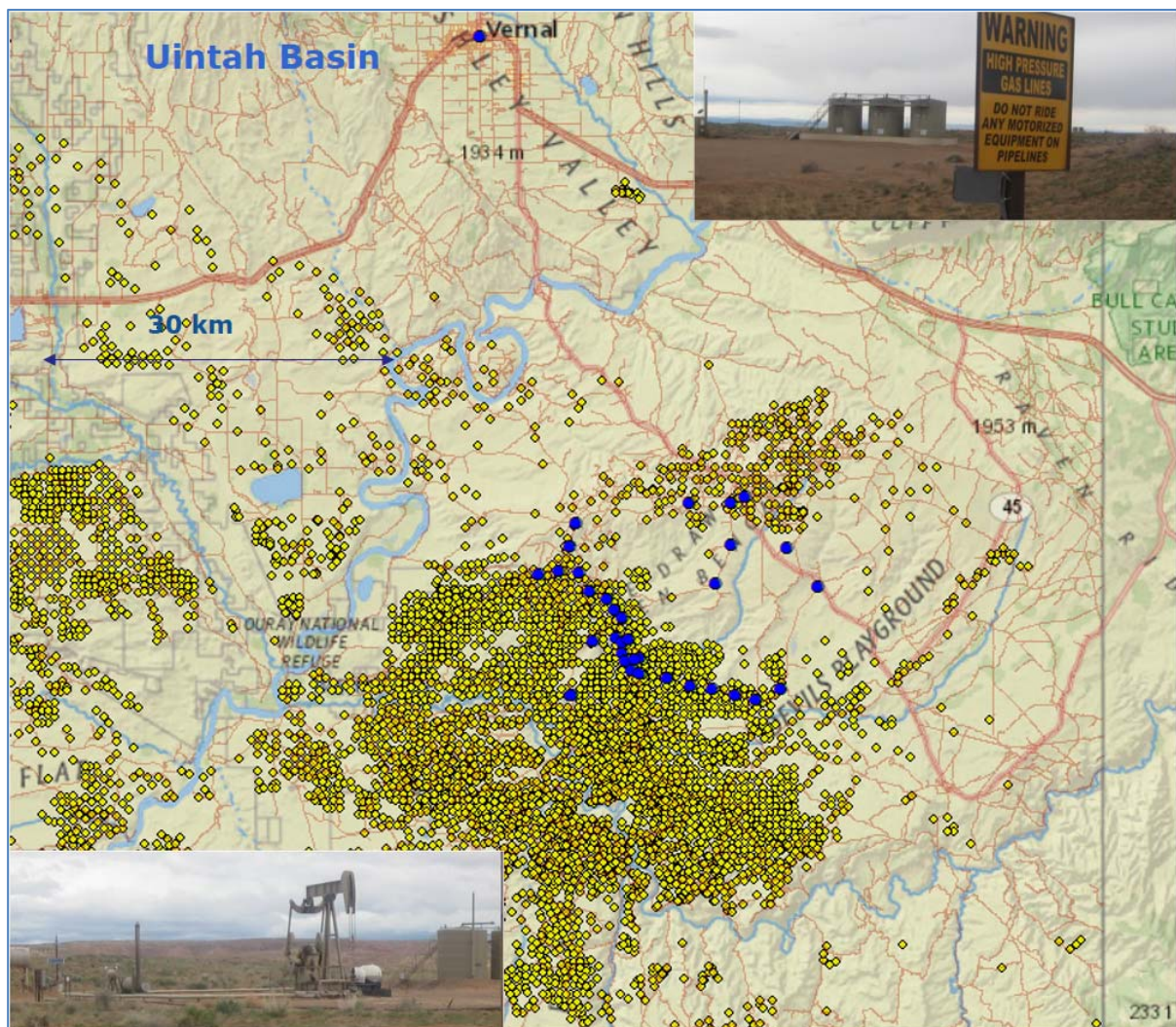
Figure 4 - Wells (yellow) and diffusive sampling locations (blue) in the Uintah Basin

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An extensive suite of research instruments were deployed by a large group of scientists from different institutions at a site named Horse Pool (40.143◦N; 109.468◦W; 1530 m elevation), located on the northern edge of the gas field, 20 miles south of Vernal, UT. This work highlighted the importance of using measurement-based input for predictive modelling and revealed the importance of secondary carbonyl compounds as radical amplifiers. The Uintah Basin region is still being developed but has ~8000 natural gas wells and ~2000 oil wells in operation. **Figure 4** shows the distribution of wells in the Uintah Basin.

1.3 North Platte Basin (Converse County)

The Converse County portion of the North Platte Basin is an area with a long history of O&G development that continues to the present day with an environmental impact statement under consideration by the US Bureau of Land Management for an additional 5,000 wells (US BLM, 2014). Historically this region of the US has experienced considerable oil exploration.
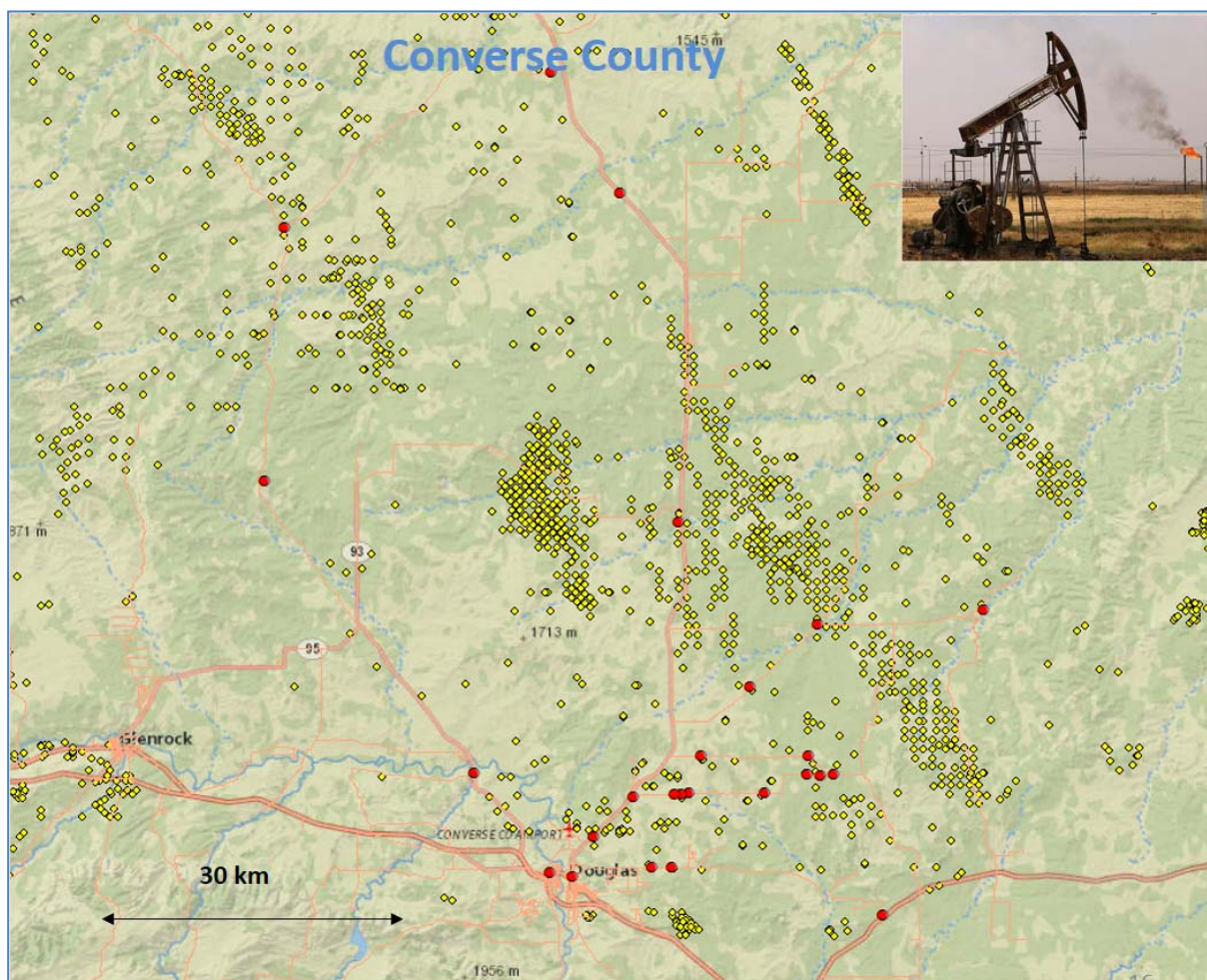
Figure 5 - Wells (yellow) and diffusive sampling locations (red) in the Converse County

There are ~1100 active O&G wells in Converse County with an additional 1300 inactive wells. Over 95% of wells in Converse County are classified as oil wells. **Figure 5** show the distribution of wells in Converse County. Recent development in the extensive Niobrara formation accounts for ~170 new wells. The Wyoming wind corridor with strong and persistent Westerly and North Westerly winds positively influences air quality in Converse County. The basin provides an opportunity to assess conditions with relatively low production levels at the present time.

2 METHODOLOGY

2.1 Sampling Approach

Diffusive sampling has been successfully applied to monitor contaminants in air for many decades (Kot-Wasik et al. 2007). Applications have included a wide range of environmental conditions including indoor, workplace and ambient air environments, with pollutant concentrations ranging from trace to highly contaminated levels (Walgraeve et al. 2011; Pérez Ballesta et al. 2006; Carrieri et al. 2014). While fast response continuous analysers have facilitated the emergence of mobile monitoring as an approach for understanding some air quality problems, diffusive sampler surveying still remains a cost-effective approach for mapping spatial distributions of pollutants. High-density simultaneous measurements with diffusive samplers have the advantage of providing a more time-integrated representation of the considered area.
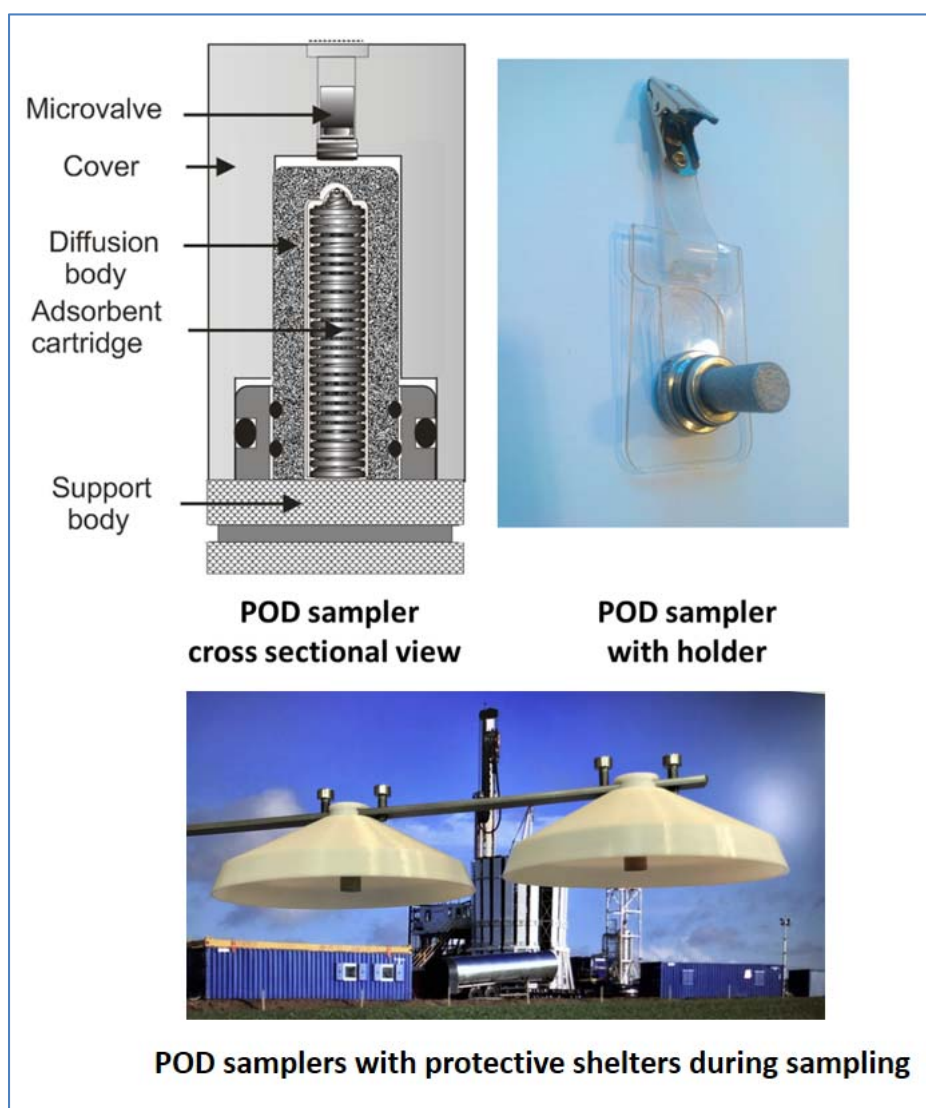
Figure 6 - PoD sampler design

In the field of ambient air quality, diffusive sampling is more often applied in Europe (Pérez Ballesta et al. 2008; Cocheo et al. 2000) than in the US (Mukerjee et al. 2009, Smith et al. 2011), in part due to the use of this method as indicative technique for determining compliance with air quality standards in the European Union. In the US, diffusive sampling has received renewed interest due to new EPA regulations requiring

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fence line sampling at petrochemical refineries using EPA Method 325A/B (US EPA 2015a; US EPA 2015b). A number of pilot studies have been performed to ensure the suitability of diffusive samplers for this new monitoring requirement (Thoma et al. 2011; Mukerjee et al 2016, Thoma et al. 2016). More recently diffusive samplers have been applied to directly assess O&G emissions in the US (Field et al., 2015b; Eisele et al. 2016; Paulik et al. 2016).

This project employs a prototype diffusive sampler known as the PoD sampler that has been developed by DG JRC (European Commission). The driving force of the diffusive sampling technique is the gradient of analytes' concentration from the external environment to the sampler adsorbent. The PoD samplers are designed to ensure a constant diffusion rate during the defined sampling period. The project extends upon one field validation campaign and extensive laboratory testing of key variables including temperature, wind velocity and humidity. A patent (13792334.8-1553 EPO) has been awarded for the PoD sampler (Pérez Ballesta, 2017). **Figure 6** shows details of the PoD sampler. The PoD sampler has been validated for use as an ambient air sampler (Pérez Ballesta et al. 2016).

The importance of environmental variables and their influence upon the operation of diffusive samplers is well known (Bartkow et al. 2005; Tolnai et al. 2001). The benefits and limitations of measuring air pollution using diffusive sampling are noted elsewhere (See Thapathy et al. 2008). Diffusive sampling is an effective tool for environmental and personal exposure monitoring, as there are few limitations for deployment locations. While diffusive samplers do not provide temporal detail, as provided by continuous monitors, when sufficient numbers are also deployed they can provide cost effective determinations of spatial distributions of measured pollutants. Diffusive samplers are well suited for use in air quality networks and for defining the extent of air pollution problems (Cocheo et al. 2008).

The performance of diffusive samplers is strongly influenced by the physical and chemical characteristics of the adsorbent (Martin et al. 2003) and the analytical sensitivity of the detection method. Here we introduce a novel diffusive sampler, operating with a zero-equilibrium interface concentration and patented as PoD sampler (Pérez Ballesta 2014) that has been designed to exceed the performance of existing marketplace products. In this study, two types of PoD sampler were used (**Figure 7**): (i) a PoD sampler with stainless steel diffusion body of 20 µm porosity for the analysis of VOCs; and (ii) a PoD sampler with a bronze diffusion body of 60 µm porosity for the analysis of volatile polycyclic aromatic hydrocarbons (PAHs) and heavy hydrocarbons (HHCs).

Figure 7 - PoD sampler with different diffusion bodies for PAHs and VOCs sampling

2.2 Analytical Method

Analysis of VOCs was carried out by thermal desorption using a Perkin Elmer turbomatrix thermal desorption system (AT50) coupled with an Agilent gas chromatograph CG 6890 with a double column (DB1-DB624) and an Agilent mass spectrometer 5890. Cylinders of certified reference materials supplied by the UK National Physical Laboratory (NPL) for 33 hydrocarbons at ~4 ppb were used for calibration of the gas chromatograph. Known amounts of these standard mixtures were loaded onto Perkin Elmer glass tubes containing from 35 mg to 300 mg of Carbopack-X 60-80 mesh. Loaded volumes from 100 ml to 20 litres, depending on the concentration of the standard, allow calibration of the system from 0 to 2 µg for individual compounds. Breakthrough volumes were checked with a backup tube in series during loading of the standard mixture. A more detailed description of the analytical technique can be found elsewhere (Pérez Ballesta et al. 2016). Analysis of PAHs was carried out by thermal desorption using a TDU injector from Gerstel coupled with a Agilent gas chromatograph GC6890 and a Mass Spectrometer MS 6975C inter MSD from Agilent Technologies with a capillary column RXI-17Sil MS 50 m, 0.250 mm i.d. and 0.150 µm. The system was calibrated by means of certified reference solutions containing the target and corresponding deuterated compound. An internal standard technique was applied for the quantification of target compounds in the samples. Sampling rates were previously determined within an environmental exposure chamber by simulating real ambient air conditions. (Cabrerizo et al. 2014). Samplers were used to collect VOCs (alkanes, cyclo-alkanes, alkenes and aromatic compounds), PAH (volatile phase C_{10}-C_{16}) and HHCs (C_{11}-C_{22}), respectively. **Table 2** shows the list of considered compounds. It should be noted that for this project we report only hydrocarbons within the VOC category.

Table 2 - Compounds and PoD sampling rates

VOC PoD SAMPLER				
Alkanes	**Methyl-Alkanes**	**Cyclo-Alkanes**	**Alkenes**	**Aromatics**
n-butane (4.94)	**2-methyl-propane (1.28)**	methyl-cyclo-pentane (7.89)	**1-butene (3.01)**	**benzene (8.89)**
n-pentane (8.59)	2-methyl-butane (8.56)	cyclo-hexane (6.84)	**1, 3-butadiene (4.29)**	**toluene (8.1)**
n-hexane (8.10)	2,2-dimethyl-butane (6.76)	methyl-cyclo-hexane (6.79)	**trans-2-butene (5.97)**	**ethyl-benzene (6.92)**
n-heptane (7.13)	2,3-dimethyl-butane (6.76)	1,3-dimethyl-cyclo-hexane (6.80)	**cis-2-butene (4.13)**	**m,p-xylene (5.93)**
n-octane (6.29)	2-methyl-pentane (9.90)	1,4-dimethyl-cyclo-hexane (6.78)	1-pentene (9.55)	**o-Xylene (5.69)**
n-decane (3.73)	2-methyl - hexane (5.77)		trans-2-pentene (8.83)	cumene (5.29)
	3-methyl- hexane (5.77)		isoprene (9.89)	1,3,5-trimethyl-benzene (5.5)
	2,2,4-trimethyl-pentane (6.08)			1,2,4-trimethyl-benzene (5.85)
				1,2,3-trimethyl-benzene (5.85)

PAH PoD SAMPLER			
Poly-Aromatics	**Heavy Hydrocarbons**	**HCB**	**DBTP**
naphthalene (6.1)	C_{11} (3.29)	hexaclorobenzene (9.3)	dibenzo-thiophene (6.4)
1-methyl-napthalene (6.1)	C_{12} (2.91)		
acenapthylene (5.8)	C_{13} (2.54)		
acenapthene (5.0)	C_{14} (2.31)		
fluorene (6.3)	C_{15} (1.98)		
anthracene (6.5)	C_{16} (1.75)		
1+2methyl-phenanthrene (6.4)	C_{17} (1.55)		
3+4methyl-phenanthrene (6.4)	C_{18} (1.36)		
phenanthrene (6.4)	C_{19} (1.19)		
fluoranthene (8.7)	C_{20} (1.04)		
pyrene (5.7)	C_{21} (0.90)		
	C_{22} (0.77)		

Sampling rates in cm^3/min are written between brackets.

Experimental sampling rates were determined for compounds written in bold. The uncertainty for the reported concentrations ranged between 10 % and 40 %, whilst for the others, sampling rates were estimated from diffusivities and their uncertainties varied between 40 % and 100 % depending on the compound considered (Pérez Ballesta, et. al. 2016).

2.3 Site Selection

Approximately 100 PoD samplers of each type (VOCs and PAHs) were co-located in the three studied basins. Site selection was based upon existing knowledge of the air quality in the given region. For the UGRB extensive measurements have been performed since 2005. For the Uintah Basin intensive measurements started in 2009 upon realisation that wintertime O_3 is also an air quality problem in Utah. For the North Platte Basin measurements started in 2012 as part of State and Federal efforts to better understand baseline conditions in advance of significant O&G development in the area.

2.3.1 UGRB Pinedale Anticline

Sites for the Pinedale Anticline shown in **Figure 8** were selected based upon experienced gained from previous research in the area (Field et al. 2015a).

Figure 8 - Wells outside (yellow) and inside (violet) sampling buffer zone, diffusive sampling locations (blue) and 3 km sampling buffers (pink) for Pinedale Anticline study area

A 3 km diameter violet circle around the sampling point, designated as the "sampling buffer", represents an area of influence on the sampling point when levels of production inside of this area are considered. The level of production inside the buffer area defines the characteristics of the sampling point. The selection of this buffer distance for data evaluation is discussed in section 3.1. Samplers were placed in two hotspot areas identified previously (Field et al. 2015b) as the Pinedale Mesa (UGPA01-UPGA-06) and the Boulder area (UGPA09-UGPA11).

2.3.2 UGRB Jonah Field

Sites for the Jonah Field shown in **Figure 9** were selected to provide a good coverage of the entire field. Samplers were evenly distributed (UGJF01-09) with some inside and others at the boundary of the development.

Figure 9 - Wells outside (yellow) and inside (violet) sampling buffer zone, diffusive sampling locations (blue) and 3 km sampling buffers (pink) for Jonah Field study area

2.3.3 UGRB La Barge/Big Piney Area

Sites for the La Barge/Big Piney area were selected to provide a good coverage of the northern part of the field (**Figure 10**). Samplers were evenly distributed (UGLB01-UGLB09) with eight inside and one at the boundary of the development.

Figure 10 - Wells outside (yellow) and inside (violet) sampling buffer zone, diffusive sampling locations (blue) and 3 km sampling buffers (pink) for La Barge/Big Piney study area

2.3.4 Uintah Basin

Sites for the Uintah Basin shown in **Figure 11** were selected to provide a good coverage of the north-eastern part of the basin. Samplers were relatively evenly distributed (UB01-UB35) with most inside and some at the boundary of the development (UB12-UB14, UB02, UB04, UB31, UB33). Samplers were deployed within the area associated with wet gas production, as this area has been the focus of research related to wintertime O_3.
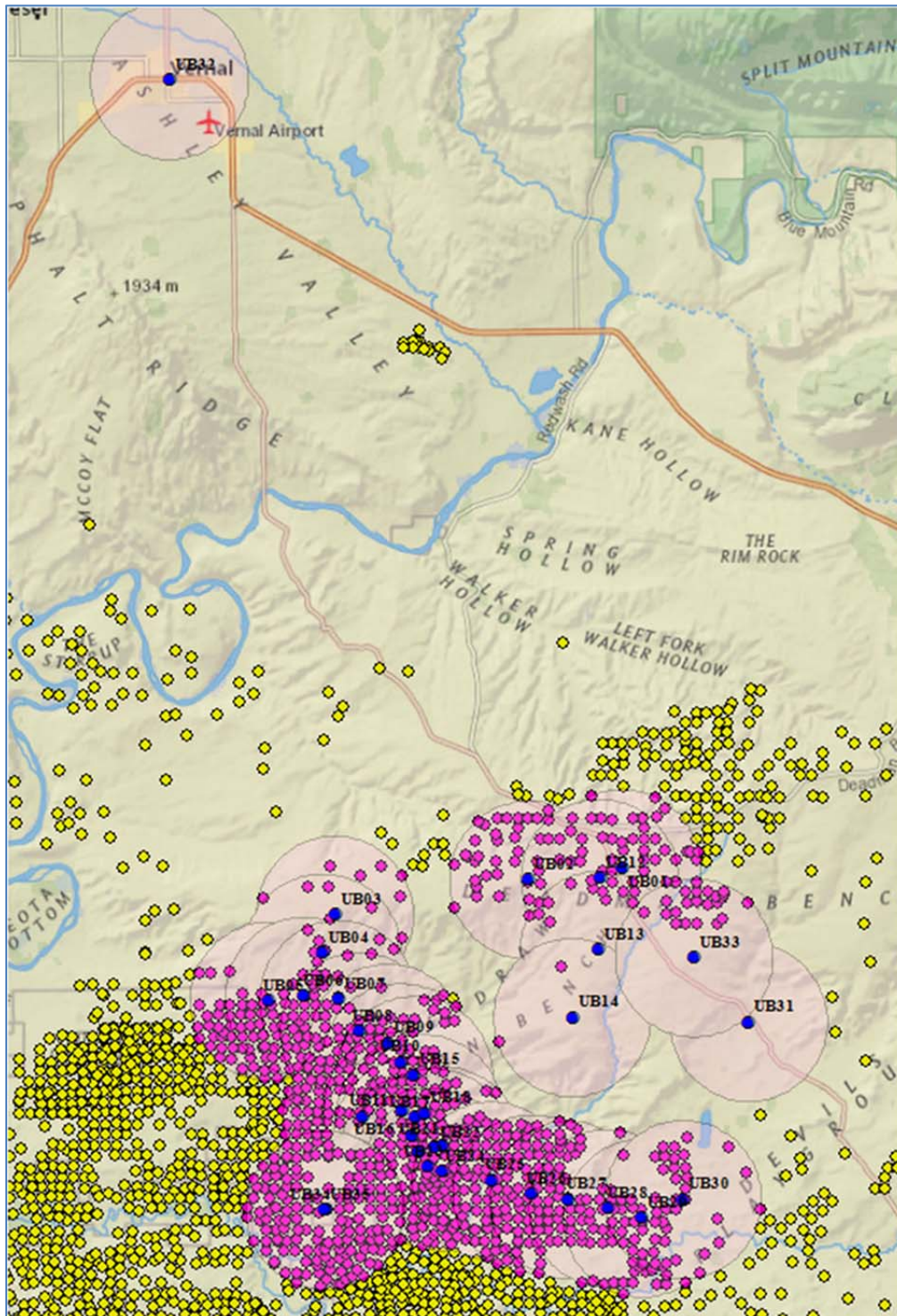
Figure 11 - Wells outside (yellow) and inside (violet) sampling buffer zone, diffusive sampling locations (blue) and 3 km sampling buffers (pink) for study area in the Uintah Basin

2.3.5 North Platte Basin (Converse County)

Sites for North Platte Basin shown in Figure 12 were selected to provide a good coverage of the Converse County section of this basin. Samplers were relatively evenly distributed (CC01-CC25) with most in areas with ongoing development activities, with some close to older existing wells (e.g. CC18, CC22 and CC23).
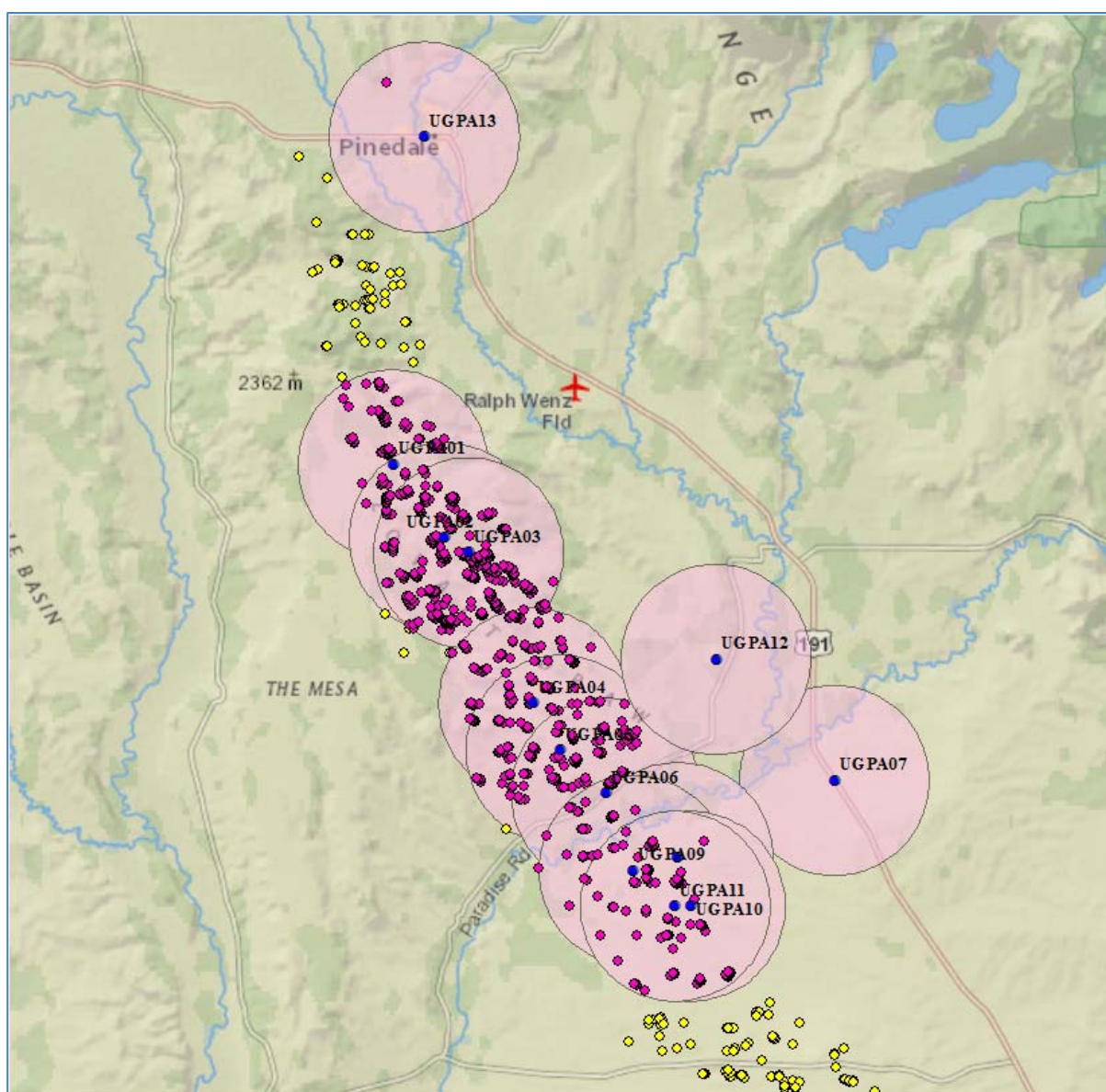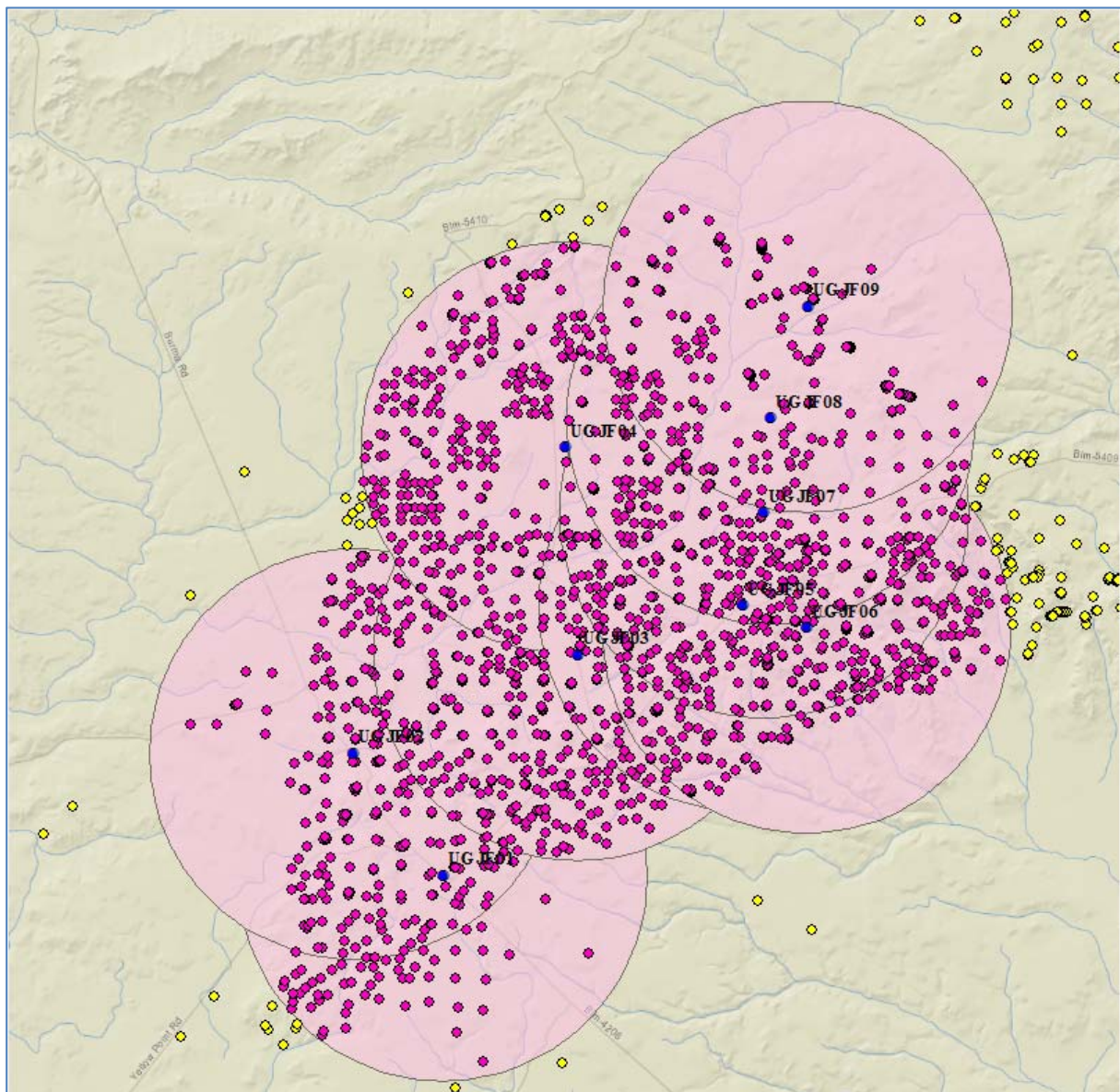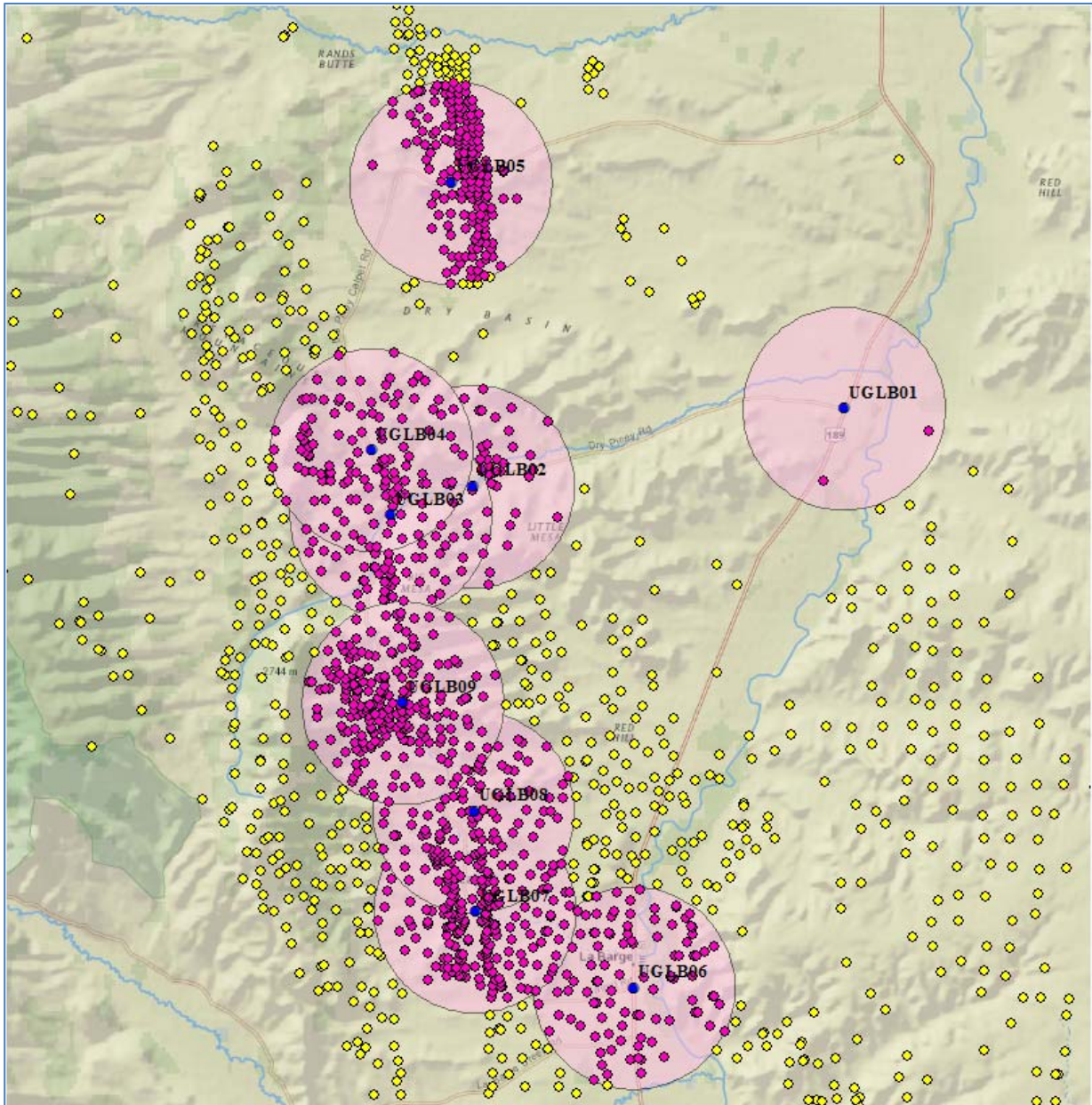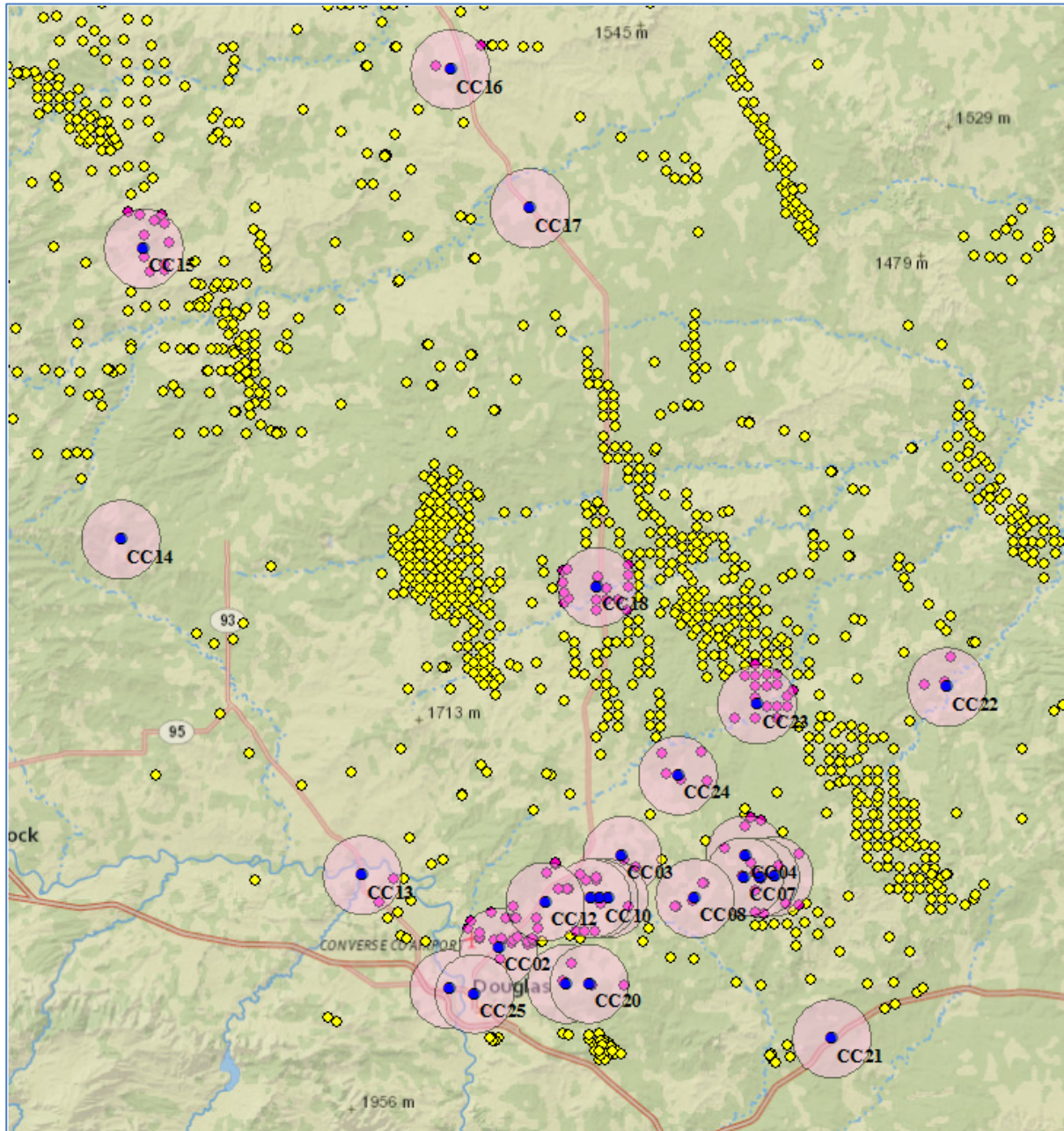

Figure 12 - Wells outside (yellow) and inside (violet) sampling buffer zone, diffusive sampling locations (blue) and 3 km sampling buffers (pink) for study area in the Converse County portion of the North Platte Basin

3 RESULTS AND DISCUSSION

Although the sampling durations were similar for each deployment (~three days), meteorological conditions differed considerably, despite each survey being performed within a 5-week time-frame. **Table 3** shows the average meteorological conditions for corresponding sampling periods for each surveyed basin.

Table 3 - Meteorological conditions for surveys of the selected basins

Basin =>	Uintah Basin	Pinedale Anticline La Barge /Big Piney Jonah Field	Converse County
Starting date*	21/04/2015	07/04/2015	31/03/2015
End date*	24/04/2015	10/04/2015	03/04/2015
Number of samples	70	62	50
Considered SONGNEX flight days	26/03/2015 27/04/2015	29/03/2015 27/04/2015	21/04/2015
Temperature, °C**	12.5	0.1	6.1
Relative Humidity, %**	42	76	39
Wind Velocity, m/s**	2.4	3.2	6
Atmospheric Pressure, mbars**	845	777	822

*   Sampling times were individually registered for each sampler. The average exposure time for all samplers were of 72 h. The use of deployment teams ensured a standard deviation between sampling times of circa 1 h with deployment and collection times matched to circa 9:00 am

** Averaged values for the considered sampling period

Uintah Basin Meteorological data provided by the University of Utah Mountain Meteorological Group; UGRB Meteorological data provided by the Wyoming Department of Environmental Quality; Converse County Meteorological data provided by the Wyoming Bureau of Land Management

Meteorological conditions will influence the dispersion of emissions and, as such, the measured ambient concentrations. The highest temperatures were observed during the survey of the Uintah Basin, while the lowest temperatures were observed for the UGRB. Temperature may influence certain emission sources, e.g. at higher temperatures evaporative losses may be enhanced. Temperature also affects mixing, for the UGRB night-time temperatures were below freezing and some stagnation of air is anticipated through the formation of temperature inversions. As such, measurements are likely affected by overnight pooling of emissions, in particular at areas with lower elevations. Converse County had the highest wind velocities with the subsequent enhanced dilution effect upon the emissions measured downwind.

3.1 Characterisation of sampling locations

ArcMap 10.3 statistical and geoprocessing tools were applied to derive production volumes of water, oil, and gas for different radius settings, namely: 1 km, 1 mile; 3 km; 3 miles; 5 km; and 5 miles. The relative increase of production with increasing radius is indicative of the density of development surrounding a particular sampler. The production levels at sites in a given area indicate the applicability of samplers for subsequent analysis.

A 3 km radius was selected as the buffer distance to relate diffusive sampler data with oil, gas, and water production data. Shorter distances would magnify the influence of hotspots or high emitters, which would mainly affect the closest sampling points. On the other hand, higher buffer distances may hide differences in the surrounding production between sampling points. The 3 km radius avoided production levels being too restrictive in coverage, i.e. 1 km, or too broad with excessive overlap, i.e. 5 miles. Similarities and differences between the three basins are evident when considering the production volumes associated with wells within a radius of 3 km from all sampling sites for April 2015 (**Table 4**).

Table 4 - Sum of oil, gas, water production and ratios in the studied areas during April 2015 for wells within a 3 km radius of each sampler

Basin	Oil (barrels)	Gas (Mcf)	Water (barrels)	Gas/Oil	Gas/Water	Oil/water
Uintah Basin	148,882	7,089,118	1,100,433	47.6	6.4	0.14
Pinedale Anticline	582,380	79,958,596	2,798,788	137.3	28.6	0.21
La Barge/Big Piney	38,883	1,807,637	145,006	46.5	12.5	0.27
Jonah Field	488,167	49,996,161	428,350	102.4	116.7	1.14
Converse County	375,174	2,884,693	239,062	7.7	12.1	1.57

The applicability of the selected sampling sites to represent the areas considered can be inferred by comparison with total production statistics for 2014 (Table 1). While production from some wells (**Table 4**) are counted multiple times as indicated by **Figure 8**, **Figure 9**, **Figure 10**, **Figure 11** and **Figure 12**, comparison of the amounts of oil, gas and water production reveal differences between survey areas. Using a 3 km buffer production is oversampled at the Pinedale Anticline, Jonah Field and La Barge/Big Piney area, while the reverse is evident for Converse County, and in particular the Uintah Basin.

A comparison of production ratios in **Table 1** and **Table 4** reveals that the sampling at Converse County and, to a greater extent, the Uintah Basin were, as designed, biased towards gas production. The Uintah Basin encompasses Uintah County (gas wells more prevalent) and Duchesne County (oil wells more prevalent). Ratios at Uintah County for the first quarter of 2015 were closer to those evident in **Table 4**. However, the Gas/Oil ratio for sampled sites (**Table 4**) of 47.6 was far higher than that for the entire basin (**Table 1**) of 11.0, confirming the selection of an area with more influence from gas wells than from oil wells. As noted earlier, site selection at Converse County was directed towards locations with newer wells whereas sites selected in the Uintah Basin were set within a gas rich region of the basin.

Figure 13 represents the median values of production, distance to closest well and number of wells around the buffer-area that characterise the set of sampling points in

each studied basin. Background sites are excluded and only production sites, as identified in ANNEX I, are considered for subsequent calculations. It is noted that Pinedale Anticline has the highest production levels for gas, oil, and water. Sampling sites in the Converse County survey have the greatest distance from the nearest well and the lowest number of wells in the 3 km radius. At Converse County, the production of oil is similar to Uintah Basin and La Barge/ Big Piney area, despite a lower number of contributing wells. The characterisation of the studied basins as a function of the production evident within the sampling buffer provides the basis of comparison with the determined concentrations.
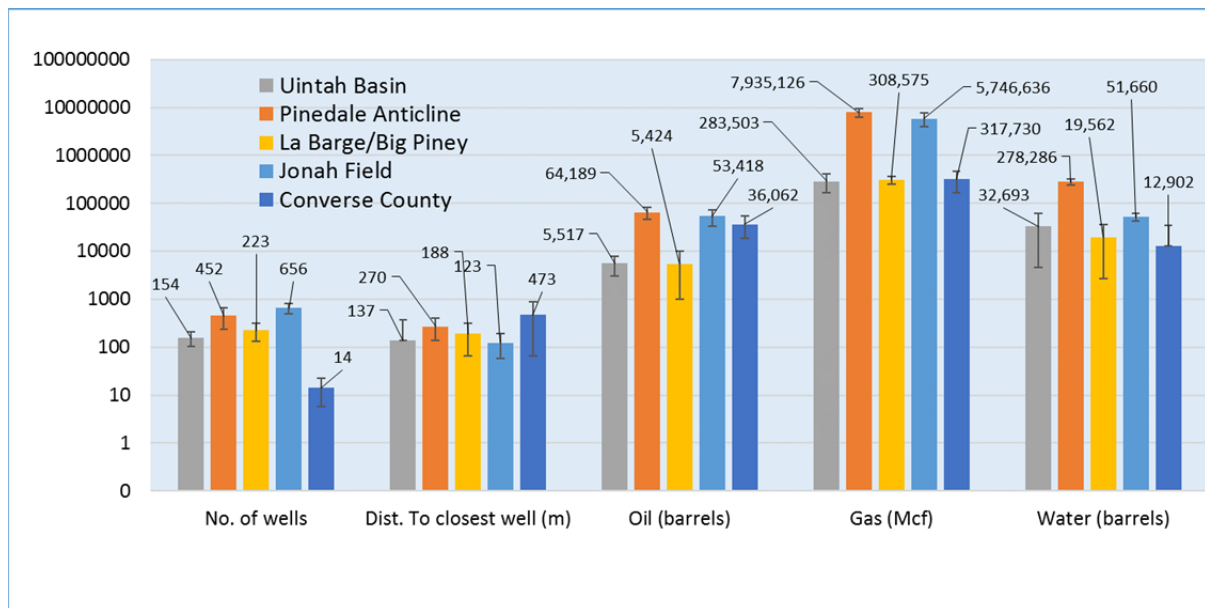
y-axis represents different units according to the referred value in the x-axis

Figure 13 - Median distance to the closest well, number of wells, production of oil, gas and water in 3 km distance around the sampling point

3.2 Basin scale comparisons

Each sampling point was defined according to production volumes samples within a 3 km buffer distance. A detailed description of the procedure applied to characterise and label sampling points is given in Annex 6.1. This analysis provides a clear identification of the sampling points in terms of the surrounding production (**Table 8**) based on oil, gas and water production amounts.

The concentrations of selected alkanes, alkenes, aromatics, PAHs and HHCs in production sites are reported in **Figure 14**, **Figure 15**, **Figure 16**, **Figure 17** and **Figure 18**, respectively. These figures represent the average concentration of the three-day sampling period.

Figure 14 shows that despite lower gas production mid-chain alkanes, strongly associated with gas production, are measured at far higher concentrations in the Uintah Basin compared to the Pinedale Anticline. This finding appears to support the hypothesis generated from basin wide flux measurements of CH_4 that indicate higher leakage rates in the Uintah Basin compared to the Pinedale Anticline (Robertson et al. 2017).
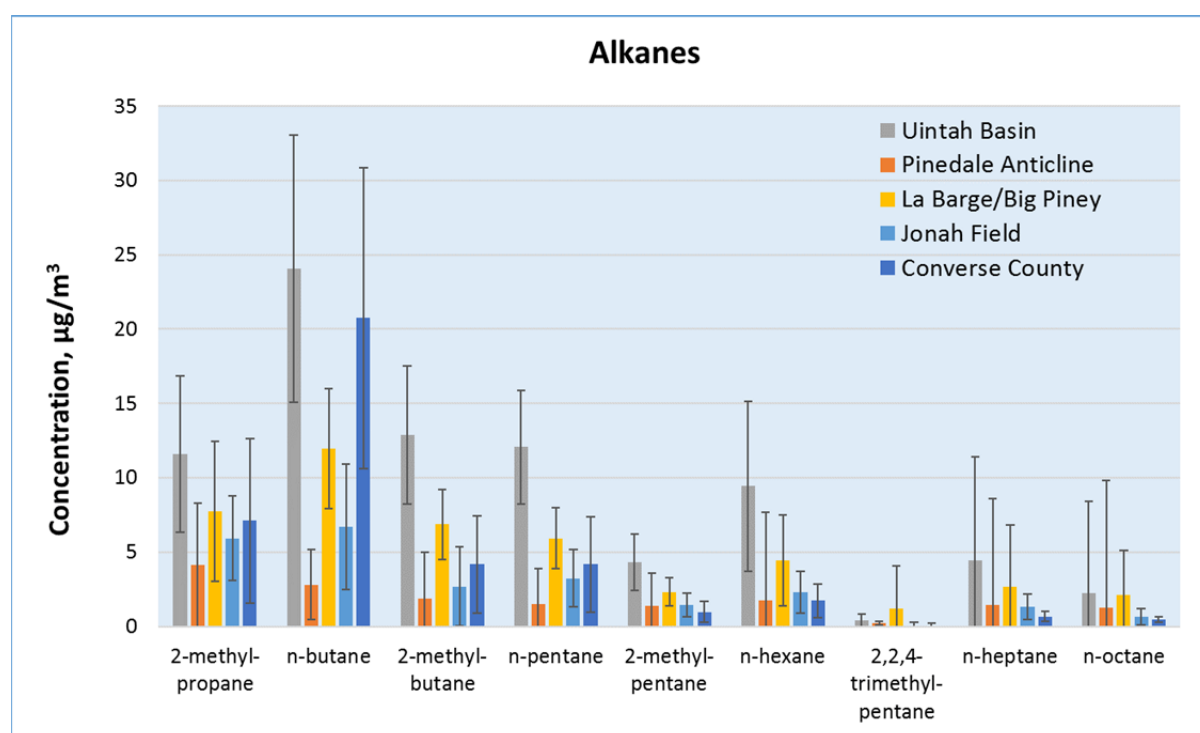
The errors bars on the graph represent the standard deviation of the sample's mean value

** Ethane and propane were not quantified because of the low sampling efficiency of the used adsorbent for C2-C3 hydrocarbons. However, these compounds are expected to be the highest concentrations.*

Figure 14 - Median concentration of alkanes in the production area of the studied basins

Compared to alkanes, alkenes are measured at very low concentrations (**Figure 15**). The order of magnitude difference of concentrations of alkanes and alkenes reported here is consistent with other measurements and analysis in the US. Alkane/alkene ratios are elevated at basins with significant O&G activity. As reported by Warneke et al. (2012) there is significant difference between VOC profiles measured in O&G influenced basins compared to those influenced by urban emissions, when traffic emissions are dominant far lower alkanes/alkenes ratios are evident.

While some of the sampling locations selected for the Pinedale Anticline were within areas previously reported as BTEX hotspots (see the high standard deviation associated with Pinedale Anticline values in **Figure 16**), the elevation of benzene, toluene and m,p-xylene compared to the Jonah Field and Uintah Basin is striking (Koss et al. 2017).
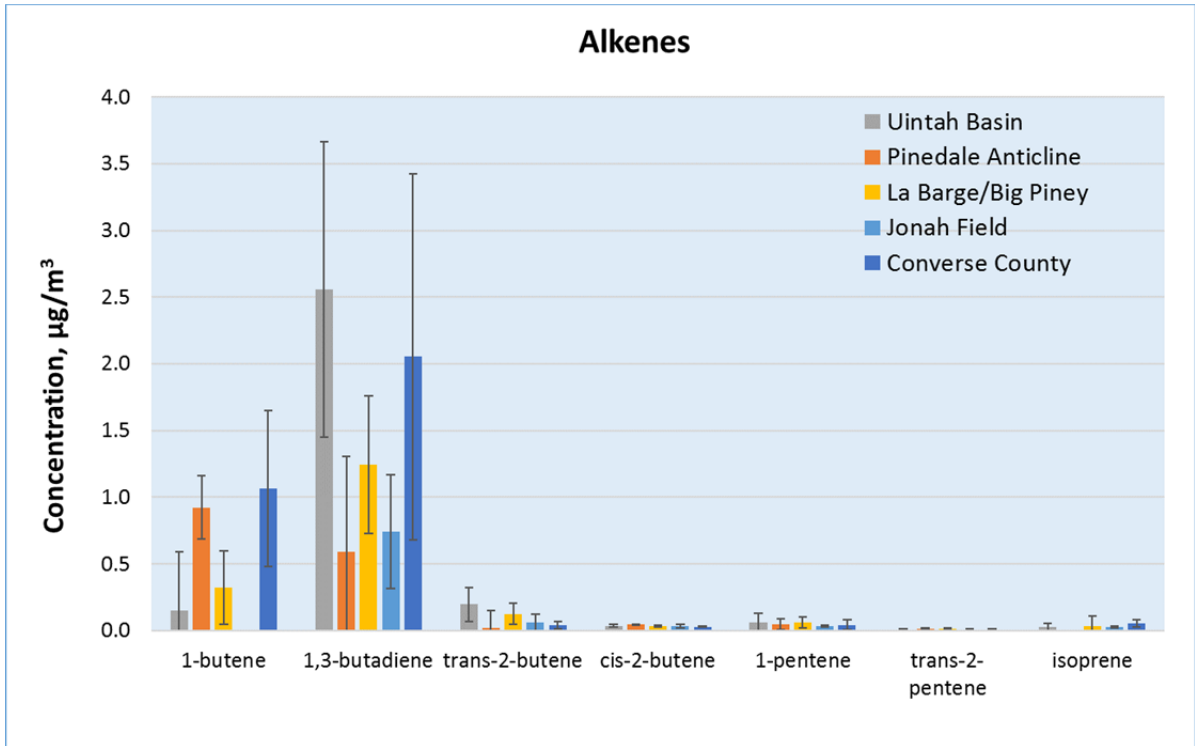
*The errors bars on the graph represent the standard deviation of the sample's mean value.

** C2-C3 hydrocarbons (ethane or propene) were not quantified because the low sampling efficiency of the used adsorbent form these compounds.

Figure 15 - Median concentration of alkenes in the production area of the studied basins
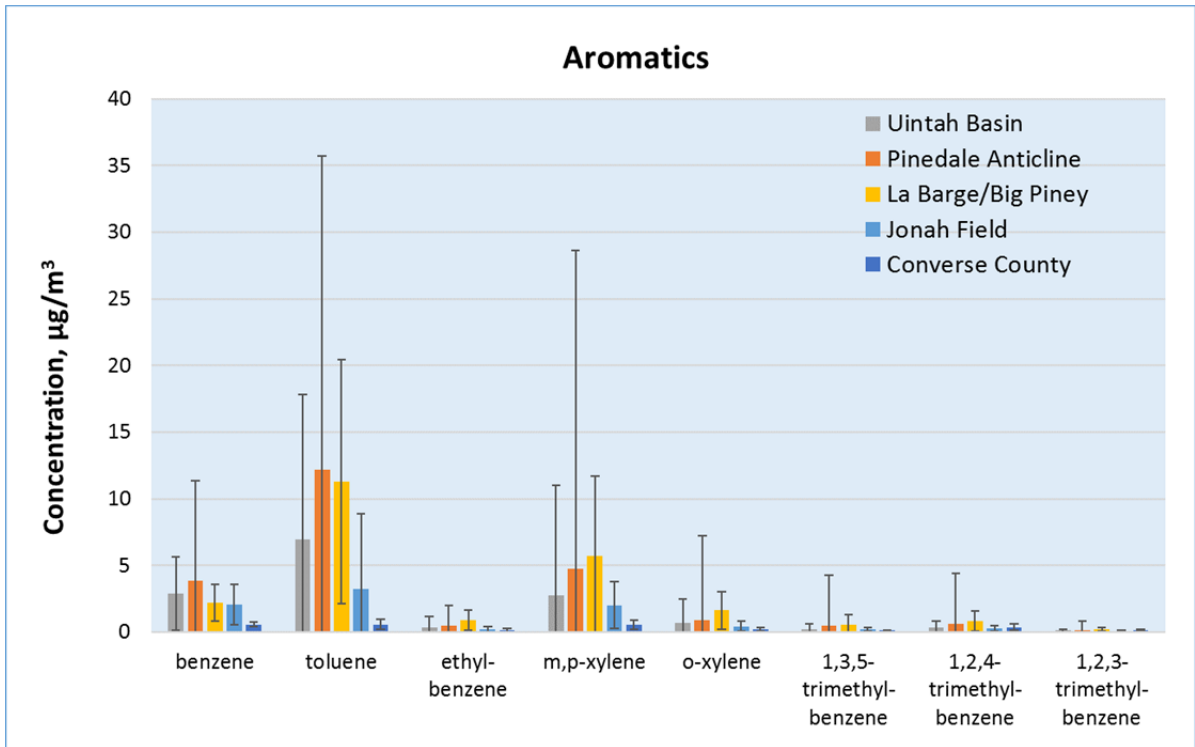
*The errors bars on the graph represent the standard deviation of the sample's mean value

Figure 16 - Median concentration of aromatics in the production area of the studied basins

The concentrations determined for selected PAHs and HHCs at each basin are reported in **Figure 17** and **Figure 18**, respectively. By contrast with VOC, PAHs and HHCs concentration levels were much lower, which, despite the influence of volatility, implies O&G sources have far lower emission factors for these compound classes. This is not unexpected as the compositional profiles of raw gas, from wet gas fields, are dominated by alkanes (C_2 to C_5). However, with dehydration of wet gas using glycol, emissions of aromatics are enhanced despite being present in relatively insignificant amounts within raw gas (Field et al. 2014). Levels of PAHs and HHCs are higher in basins with higher oil production (i.e. Pinedale Anticline) compared to those with lower oil production (i.e. La Barge/Big Piney).
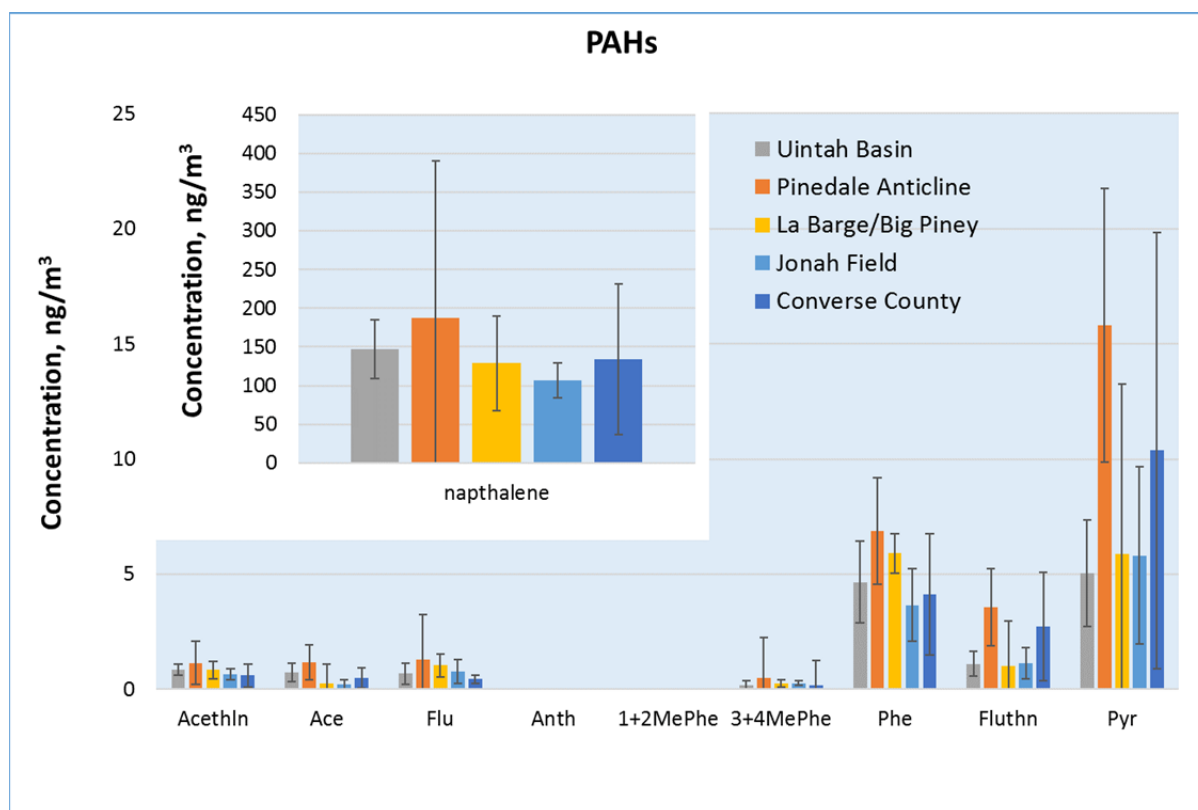
The errors bars on the graph represent the standard deviation of the sample's mean value

(AcethIn: acenapthylene: Ace: acenaphtene; Flu: fluorene; Anth: anthracene; 1+2MePhe: 1+2-methyl-phenanthrene; 1+2MePhe: 3+4-methyl-phenanthrene; Phe: phenanthrene; Fluthn: fluoranthene; Pyr:pyrene)

Figure 17 - Median concentration of PAHs in the production area of the studied basins

The concentration of reported pollutants differs at the surveyed basins. The highest concentrations of alkanes, and to lesser extent alkenes, are measured in the Uintah Basin. The Pinedale Anticline shows the highest concentration for aromatics, PAHs and HHCs. These differences are likely due to a number of factors including emission source behaviour and meteorological conditions. While the mean level of pollutant concentrations and ratios between measured compounds, within each basin are different, there is also variability within each basin, in particular when considering hotspot locations that have strong influence from proximate emissions.
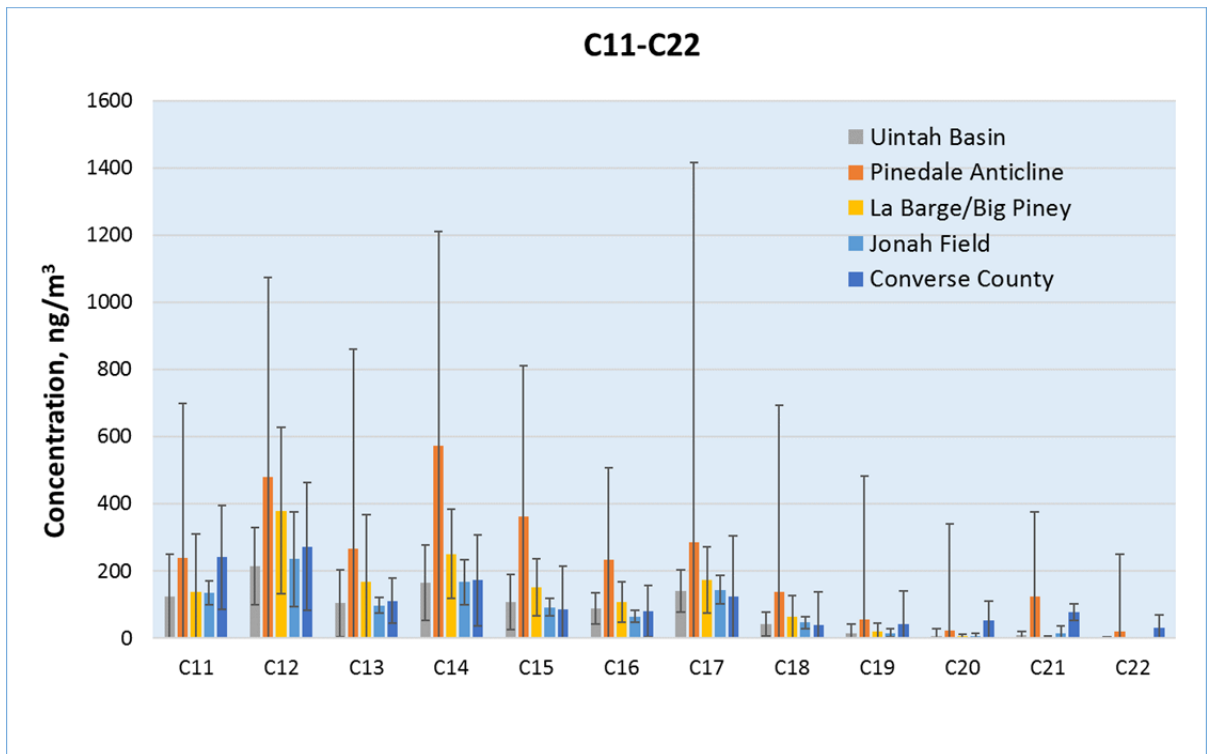
The errors bars on the graph represent the standard deviation of the sample's mean value

Figure 18 - Median concentration of heavy hydrocarbons in the production areas of the studied basins.

3.3 Correlation between oil, gas and water production and concentration of hydrocarbons in air

In section 3.2, it was noted that oil, gas and water production are basin/development specific. Raw gas and crude oil composition may differ between and within the basins evaluated here. Differences in production profiles, in particular the balance between oil, water and gas will define the preferred production handling approach and, consequently, emission source profiles. When emissions are dependent upon particular handling approaches, e.g. dehydration of raw natural gas production, analysis of the pollutant concentrations may reveal the impact of particular activities. It is, therefore, reasonable to anticipate that air pollutant distributions may be associated with specific locations within given developments.

In this section, correlations between oil, gas and water production with ambient pollutant concentrations are presented. These correlations represent a snapshot of the complex relationship between production, emissions and ambient pollutant concentrations. A strong correlation between production and hydrocarbon concentrations in air supports the idea that concentration ratios can characterise different production profiles or situations in particular areas. Consequently, a sensitivity analysis of concentrations of ambient hydrocarbons with respect to oil gas and water production is useful for our understanding of how production activities influence pollution profiles.

At an illustrative level, it is possible with all basins to correlate, by means of a log linked multivariable regression (Eq. 1), oil, gas and water production in the 3 km-distance buffer area (P_i) with their corresponding ambient hydrocarbon concentration profiles, C_j, at each sampling point:

$$P_i = e^{A_i + \sum_j B_{i,j} \cdot C_j}$$ **Eq. 1**

where A_i and $B_{i,j}$ are the corresponding regression parameters.
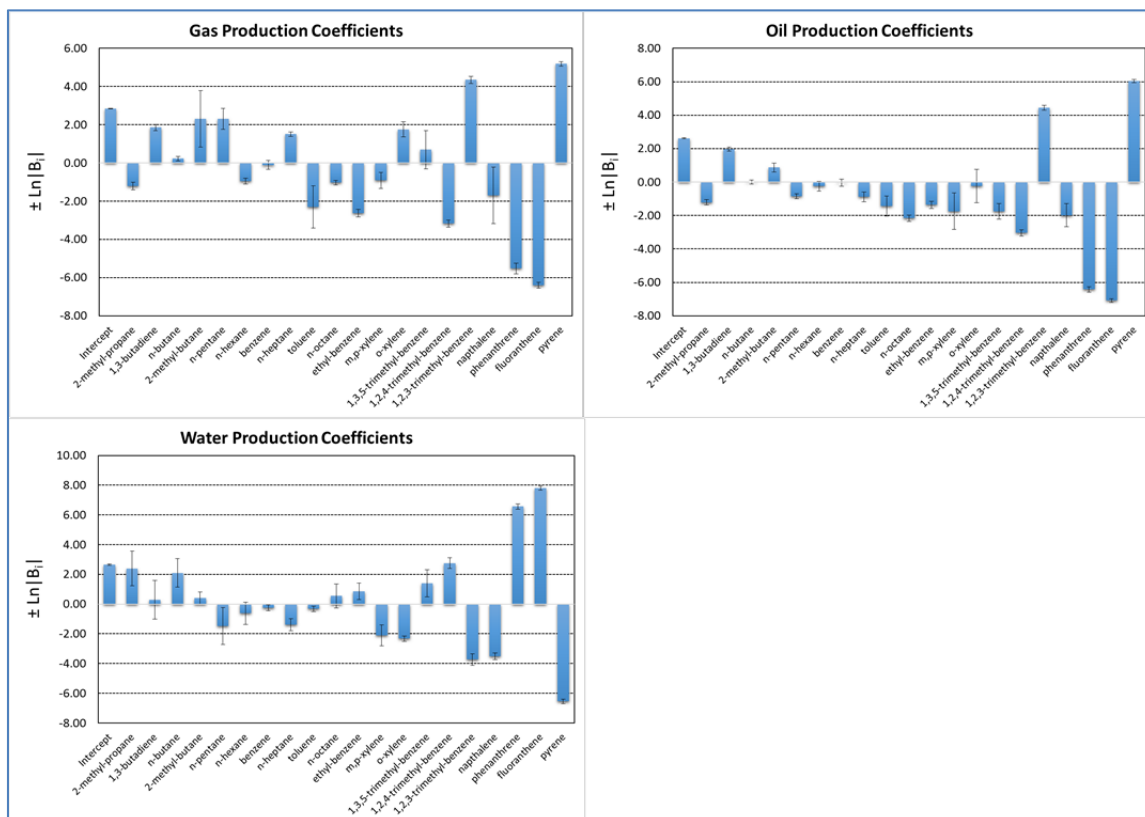
* $ln(A_i) = Intercept, \quad (\pm)ln(|B_j|) = Compound_j$

Figure 19 - Regression parameters of Eq. 1

Eq. 1 estimates production volumes as a function of the measured air concentration profiles for this project. The resulting regression parameters, expressed in a logarithmic format, for clearer visualisation, are shown in **Figure 19**. The different regression parameters associated with each hydrocarbon in relation to oil, gas and water production suggest that pollutant ratios may have a role in identifying the production regime of a particular area (section 3.4). But the concentrations of the hydrocarbons in Eq. 1 are not independent variables as concentration profiles are dependent upon the emission sources.

Figure 20 shows estimated production from the correlated Eq. 1 versus actual oil, gas and water production for the buffer areas around the sampling points. Highest uncertainties correspond with areas that report lower production levels; in these cases, the estimated emission "fingerprint" has the greatest influence from dilution processes.
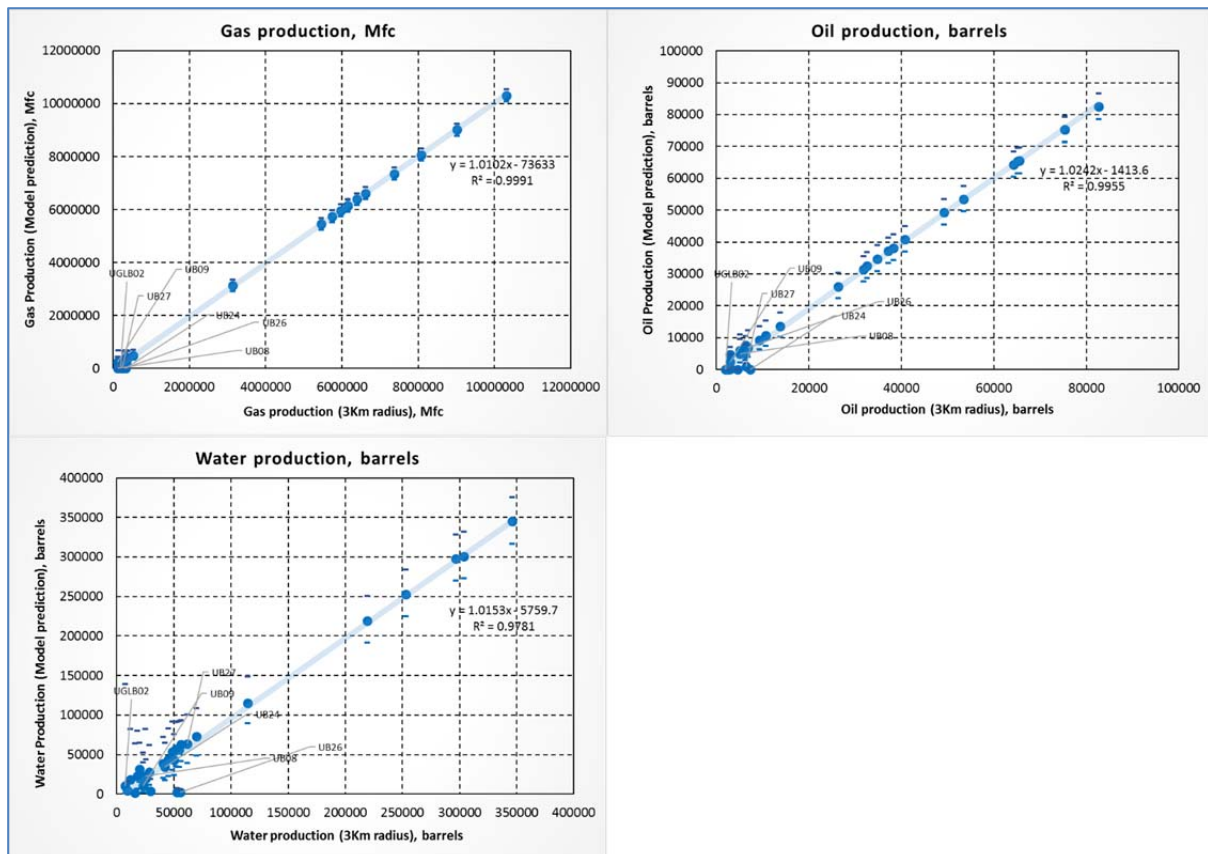
Figure 20 - Estimated vs actual production data

Figure 21 contrasts median values of actual production of oil, gas and water with those estimated from measured hydrocarbon concentrations. Despite differences in absolute terms, in particular with lower production volumes, estimated productions show a similar pattern to the corresponding actual production in each basin. The largest deviation from reported values is for estimated oil production at the Jonah Field. While this deviation is visually striking for the relative amount of oil production estimated from ambient data, there is considerable difference in absolute terms between reported and estimated for water, gas and oil production. The correlations and modelled estimates presented here are a first attempt at discerning possible relationships between measured pollutant ratios and production volumes. Further research will be required to determine if this model can be refined to improve the accuracy of predictions. Additional testing of this model, that relies upon correlations for estimation of production, is recommended under different environmental conditions.
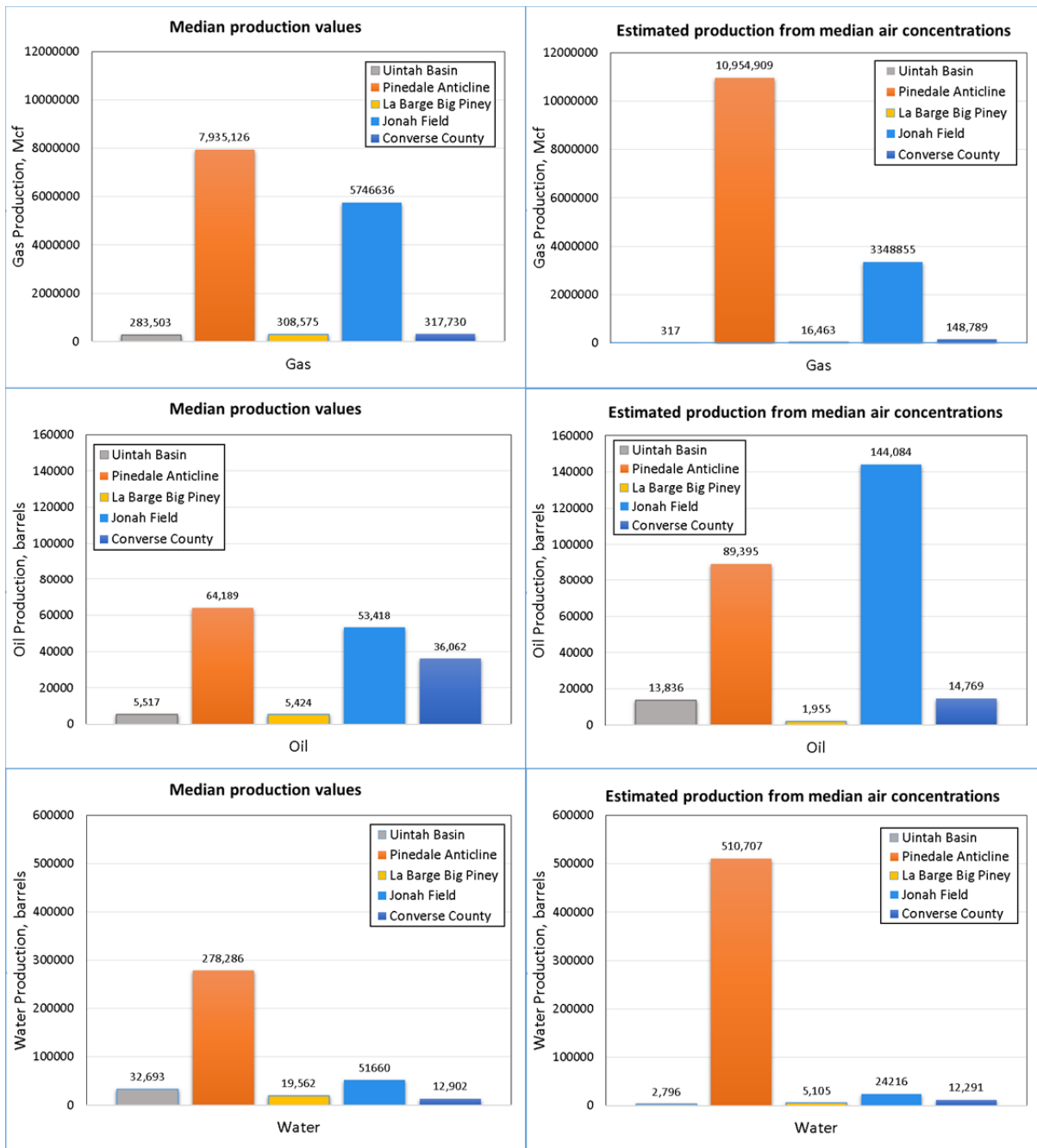
Figure 21 - Median of gas, oil and water production for the 3 km buffer distance and corresponding estimations from the median air pollutant concentration level in each basin

Furthermore, the correlations from Eq. 1 represent a snapshot of three day sampling at the studied areas. As such, these correlations should not be extrapolated to other sampling conditions, periods or zones. However, a sensitivity analysis can be performed by addressing variations of ambient concentrations in groups of compounds related to sources or production rates. Therefore, slight variations in the air concentration of groups of compounds in Eq. 1 should reflect parallel variations in production rates according to the characteristics of each basin. **Figure 22** shows changes of production rates and ratios when the concentrations of certain compounds in air increase with respect to the mean air concentration measured in each basin.

In the case of a modelled 5 % increase of lighter hydrocarbons (C4-C5) in ambient air, estimated oil and gas production increases in the Pinedale Anticline and Jonah Field and water production decreases for all basins. A 5% increase of the C6-C7 hydrocarbon

concentrations increases gas production in the Pinedale Anticline, oil production in all basins and water production (except in the case of the Converse County). Furthermore, in this case production ratios of gas/oil and gas/water decrease.

An increase of 5 % in air concentration of the heavier hydrocarbons (≥C8) predicts a decrease of oil and gas production and a general increase of the ratio gas/oil, while water production increases for the Pinedale Anticline and Jonah Field. In this case, gas/water and oil/water ratios show a general decrease. The relationship of heavier hydrocarbons with production is harder to explain, as these compounds can be also related to combustion processes and traffic in the area, but an increase in ambient air concentrations are positively correlated with water production in these developments.
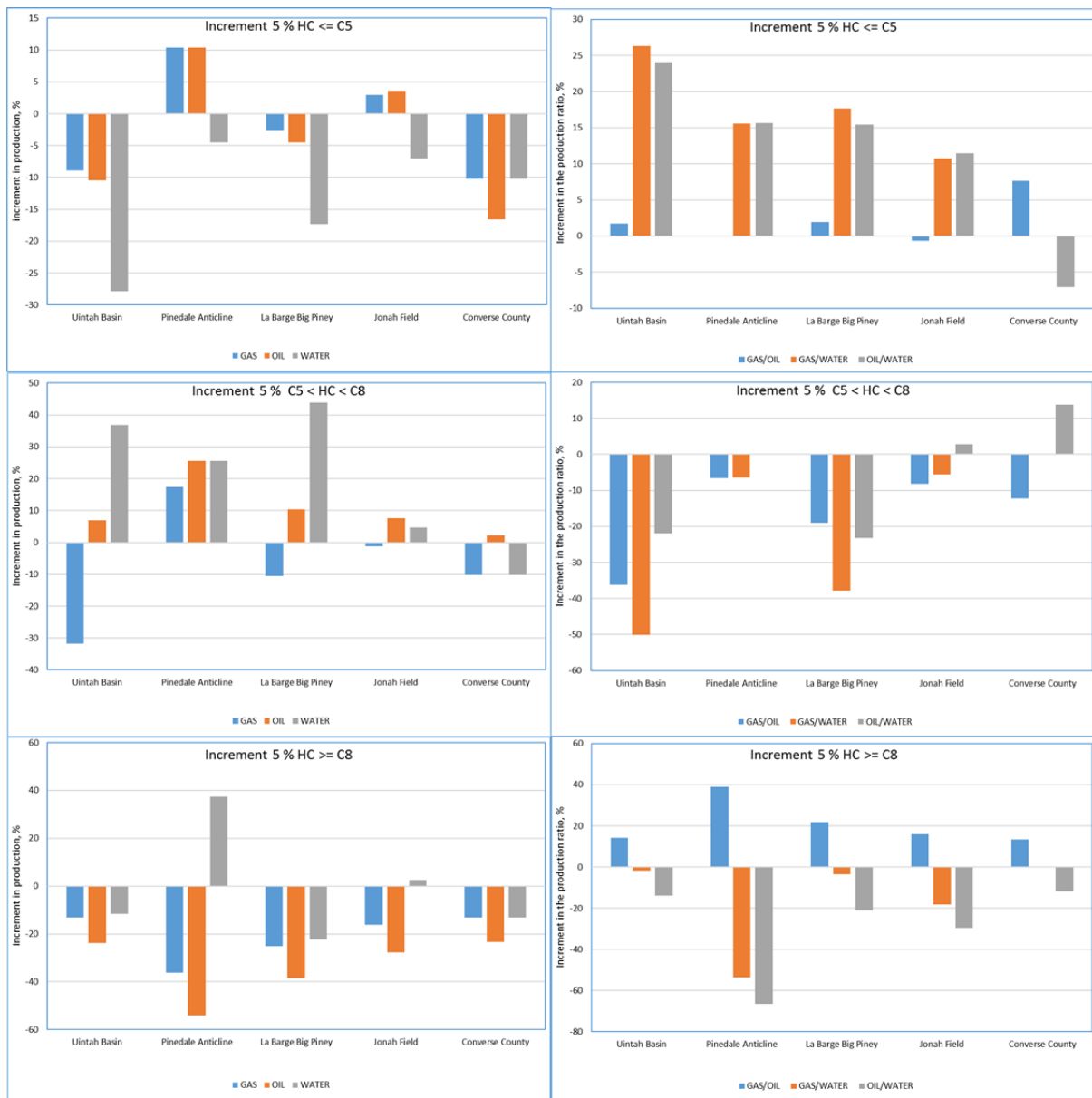
Figure 22 - Increment in percentage of gas, oil, water production and corresponding ratios for a partial 5 % increase of hydrocarbon concentrations

3.4 Comparison of PoD and SONGNEX measurement data

From the SONGNEX dataset, flights with dates and routes closest to the sampling times of this project were chosen for comparison with PoD samples. SONGNEX data corresponds to flights carried out on the 21st of April 2015 for Converse County, on the 26th of March and 27th April 2015 for Uintah Basin and the 29th of March and the 27th April 2015 for the Upper Green Basin. Data points selected for comparative analysis are shown in **Figure 23**.
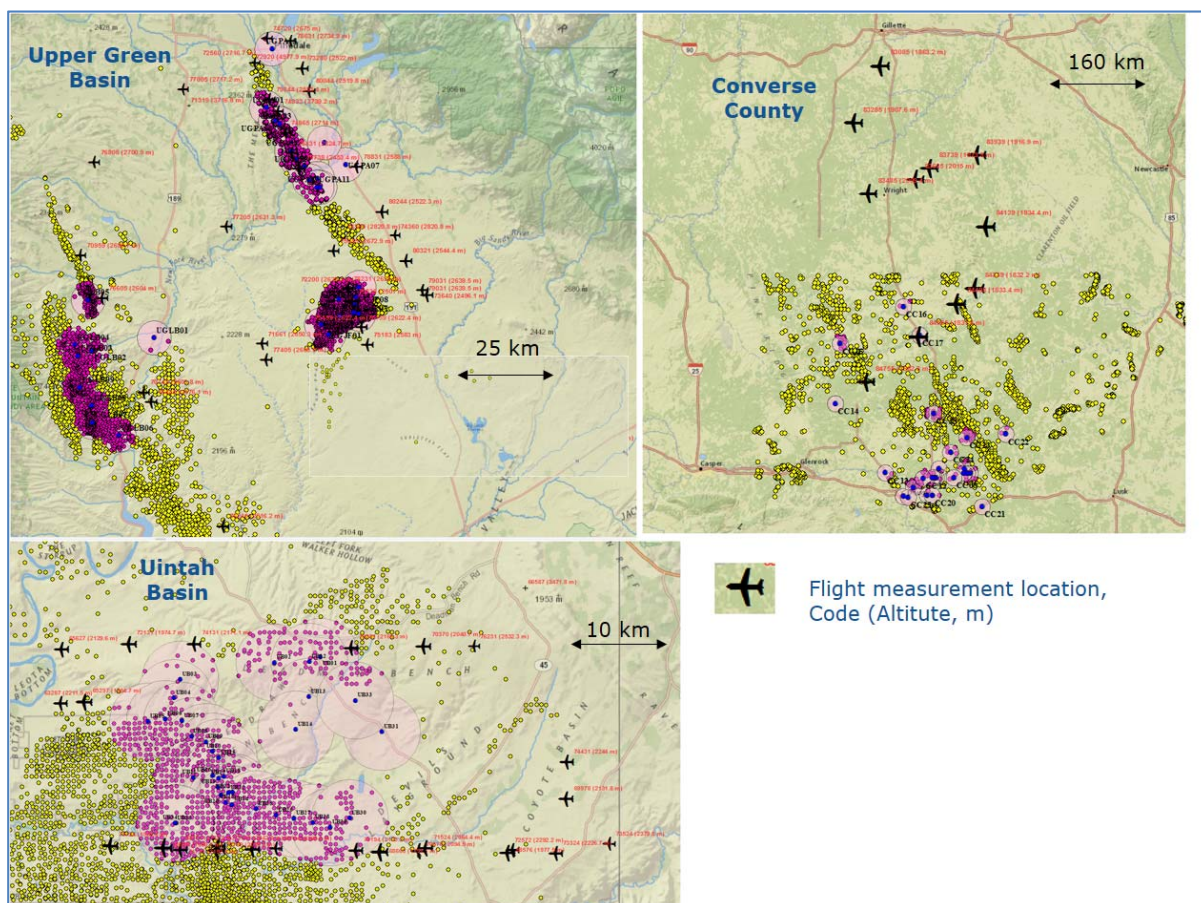
Figure 23 - Selected SONGNEX flight measurements for comparison with PoD samples in the different basins

While SONGNEX flights were designed to determine basin scale fluxes of methane and hydrocarbons, here we consider ratios between pollutants in selected samples. As aircraft measurements were taken between 1000 and 3000 m altitude above ground level, at different times and for different sampling durations, they are not directly matched or coupled with PoD measurements. Differences between reported concentrations within the two datasets are expected not only due to timing but also as a result of mixing of emissions, with elevated measurements also more homogenous than ground level ones through enhanced dilution. Measurements at ground level are more likely to have greater variation due to closer proximity to emission sources. Notwithstanding the processes noted above, if ground level samplers are measuring the same emission sources as aloft then comparison of the ratios of compounds can serve to corroborate the quality of the applied measurement techniques.

The interplay of various of emission and dispersion related factors are expected to characterise the measured pollutant concentrations at each sampling location. As anticipated in section 3.3, ratios between particular pollutants may reveal key characteristics of an assessed area; including applied technology, infrastructure design and production type (i.e. gas, water, oil ratios). Gilman et al. (2013) were the first to

employ pentane ratios to differentiate O&G emissions from vehicle exhaust sources in the era of hydraulic fracturing in the US. When applied to different datasets this approach enables comparison between different sampling methods. **Figure 24** represents the ratios: n-butane/2-methyl-pentane, n-pentane/2-methyl-butane, n-butane/n-pentane, and n-hexane/n-heptane determined by aircraft measurements and PoD samplers at ground level for the studied basins.
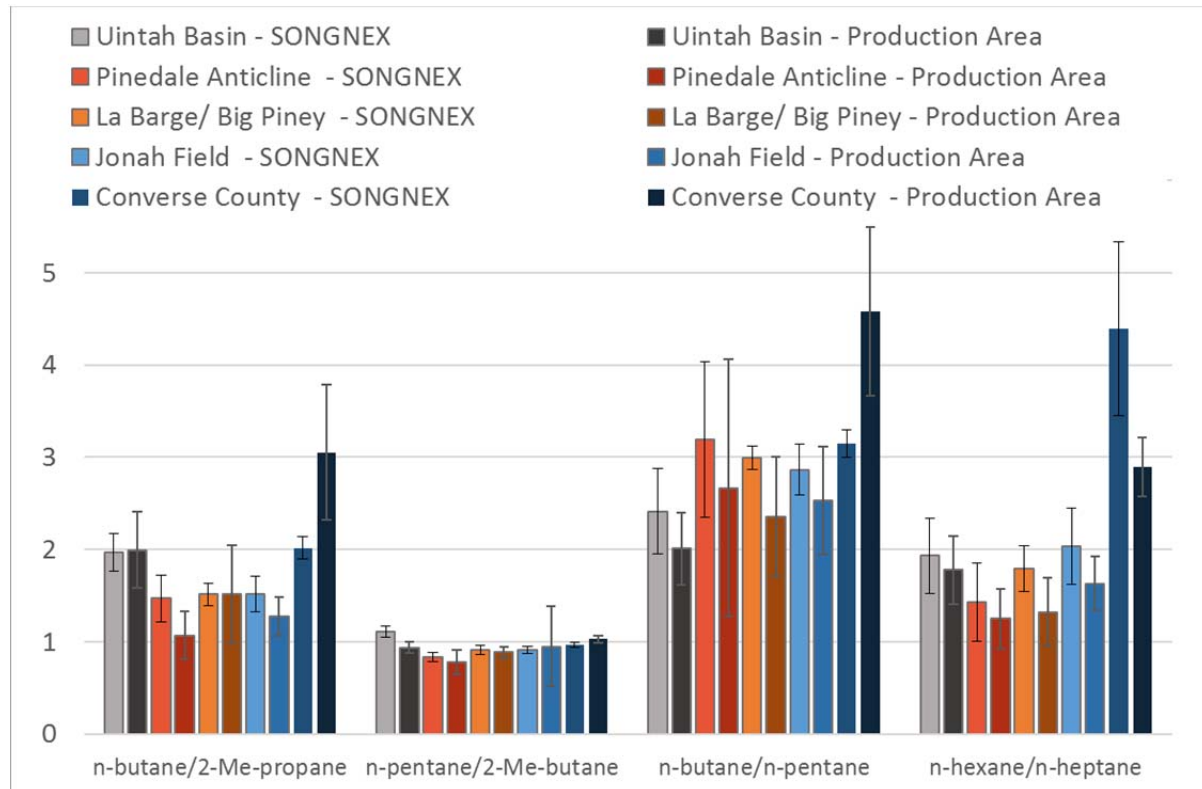
* SONGNEX data courtesy of Joost De Gouw

Figure 24 - Median compound ratios for SONGNEX aircraft measurements and PoD samplers in the production area

Ratios for both measurement approaches are shown in **Figure 24**. Each production area is different as conditioned by the reservoir(s) associated with the given development. The Pinedale Anticline produces wet gas and condensate oil with liquid gathering systems. The Jonah Field is also a wet gas and condensate oil production area, but has well pad liquids storage (tanks) and associated tanker traffic. The Uintah Basin produces wet gas with condensate oil, with well pad liquids storage, and crude oil. The La Barge/Big Piney is a crude oil field with some associated natural gas, generally with older equipment. Finally, Converse County is a crude oil field with some associated natural gas, where newer equipment is mixed with old.

The relationship between concentration and production ratios is visible in Figure 25, where n-butane/2-methyl-propane, n-pentane/2-methyl-butane and n-heptane/n-hexane ratios are represented versus Oil/Gas ratio for aircraft and ground measurements. Both data sets provide similar patterns. A relationship between concentration and production ratios is also found between compounds that are typically linked to gas (very volatile compounds) or oil (PAHs) production. See for instance the ratio between n-butane and acenapthylene concentrations versus the gas and oil production ratio determined by the PoD samplers provided by **Figure 30** in the ANNEX I.

While Gas/Oil and concentration ratios are well correlated, poorer correlations are obtained with the Gas/Water and Oil/Water ratios (see Annex 6.2). Nevertheless, it is still possible to differentiate between basins with very high and low Oil/Water or

Gas/Water production ratios. Despite this low-resolution ratio characterisation for water production, ratios of n-butane/2-methyl-propane, n-butane/n-pentane and n-heptane/n-hexane for aircraft and ground measurements again show consistent results. It is noted that the effect of temperature, wind velocity and/or differences in the well density may contribute, among other factors, to the uncertainty and variations of these ratios.
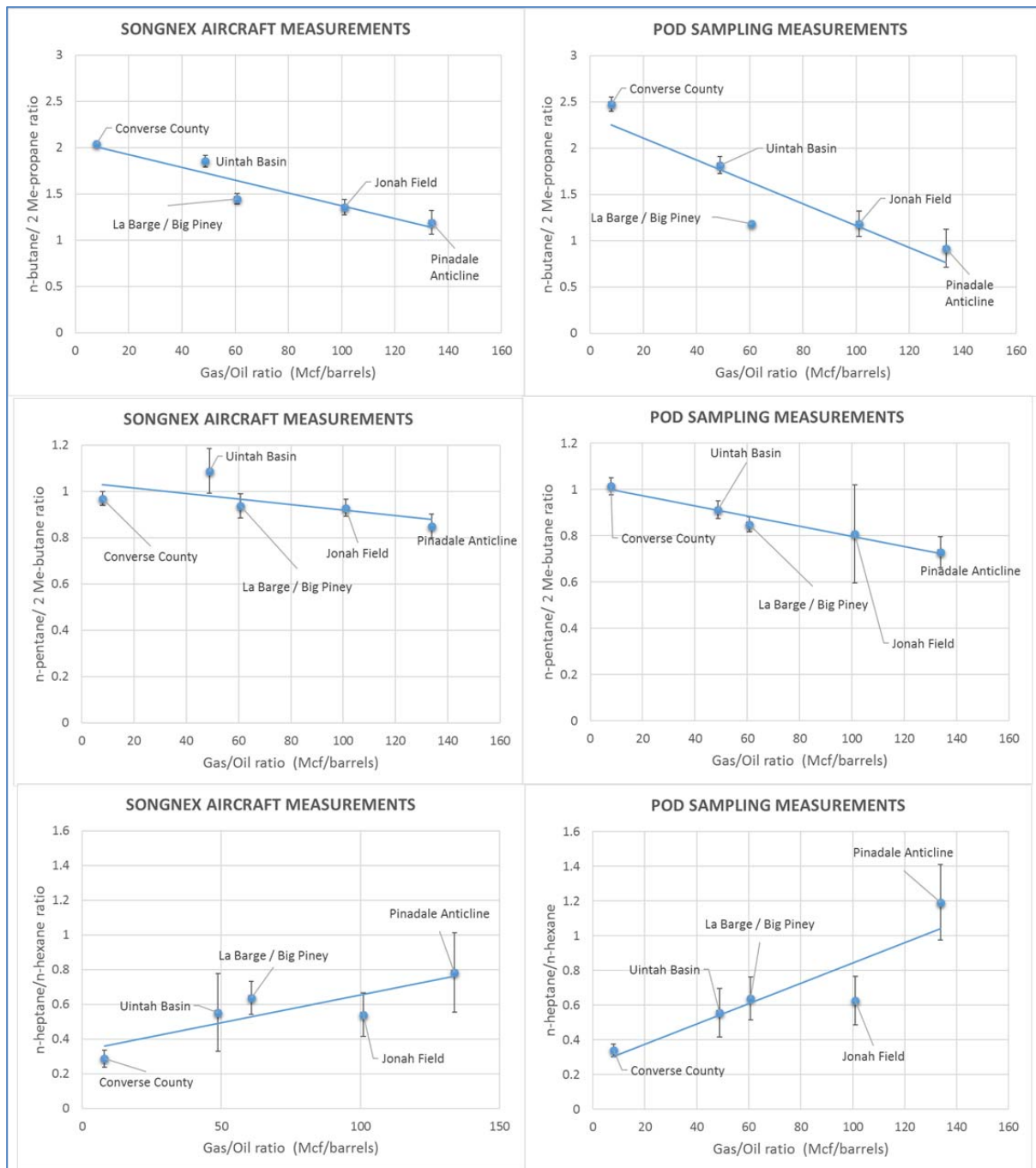
Figure 25 - Median compound ratios for SONGNEX aircraft and PoD sampler measurements versus ratios of gas and oil production in the studied basins

3.5 PMF analysis of VOCs measured with diffusive samplers

Factors of relevance to O&G emissions have been identified in different studies, in which the PMF analysis has been used as an assessment technique (Abeleira et al. 2017, Rutter et al. 2015, Field et al. 2015a). The compositional profiles, of O&G emission factors identified in these studies were conditioned by the species selected, the local distribution of sources, and other influences affecting measurements at selected sampling points. The only previously published PMF study within the areas under consideration here was carried out in the UGRB of Wyoming (Field et al. 2015a), in which three factors that characterised air quality at a site downwind of the Pinedale Anticline were identified, namely: Background (with influence from combustion), Fugitive Natural Gas and Fugitive Condensate.

In the current study, we apply EPA PMF software version 5.014.21735 to the whole PoD sampling dataset, and thus mix data from the three basins considered here. This analysis was limited to sampling points representing production areas (~60 % of the whole dataset). Uncertainty of the input data was associated with the uncertainty of the method quantification (ANNEX I., **Figure 33**). As shown in showed in **Figure 26** our PMF analysis produces three factors for the analysed hydrocarbons. Factor 1 is dominated by heavier VOC, in particular aromatic compounds with contributions increasing with greater molecular weight e.g. 1,3,5 trimethyl-benzene. Factor 2 has significant contributions from C5 to C7, with contributions declining with greater molecular weight. Factor 3 is dominated by light alkanes, e.g. n-butane (C4), with a small contribution from heavier aromatic hydrocarbons.

About 80 % of the considered species showed a high correlation coefficient R^2, with an overall median value of 0.94. Species with $R^2 < 0.90$ were considered as weak inputs, in particular PAHs and HHC, which due to their relatively high analytical uncertainty showed a poorer model correlation $R^2 < 0.6$. As a result, only a few PAHs or HHCs were considered in the model (**Figure 26**). A three-factor solution provides convergent results with factors appropriately mapped in the bootstrap analysis. This was not the case for trials with lower or higher numbers of factors. The input uncertainties and corresponding values for method detection limit were quite conservative and this is also reflected in the uncertainty of the PMF output (ANNEX I, Figure 34). Despite the relatively high uncertainties of some compounds in their contribution to the identified factors, the model provides a good interpretation of results, fitting with the production characteristics related to the different areas under consideration. In any case, the PMF analysis would provide as a possible interpretation of this data set. Given the availability of production data and understanding of exploration operations, further analysis of estimated factors can be performed on a basin specific basis.
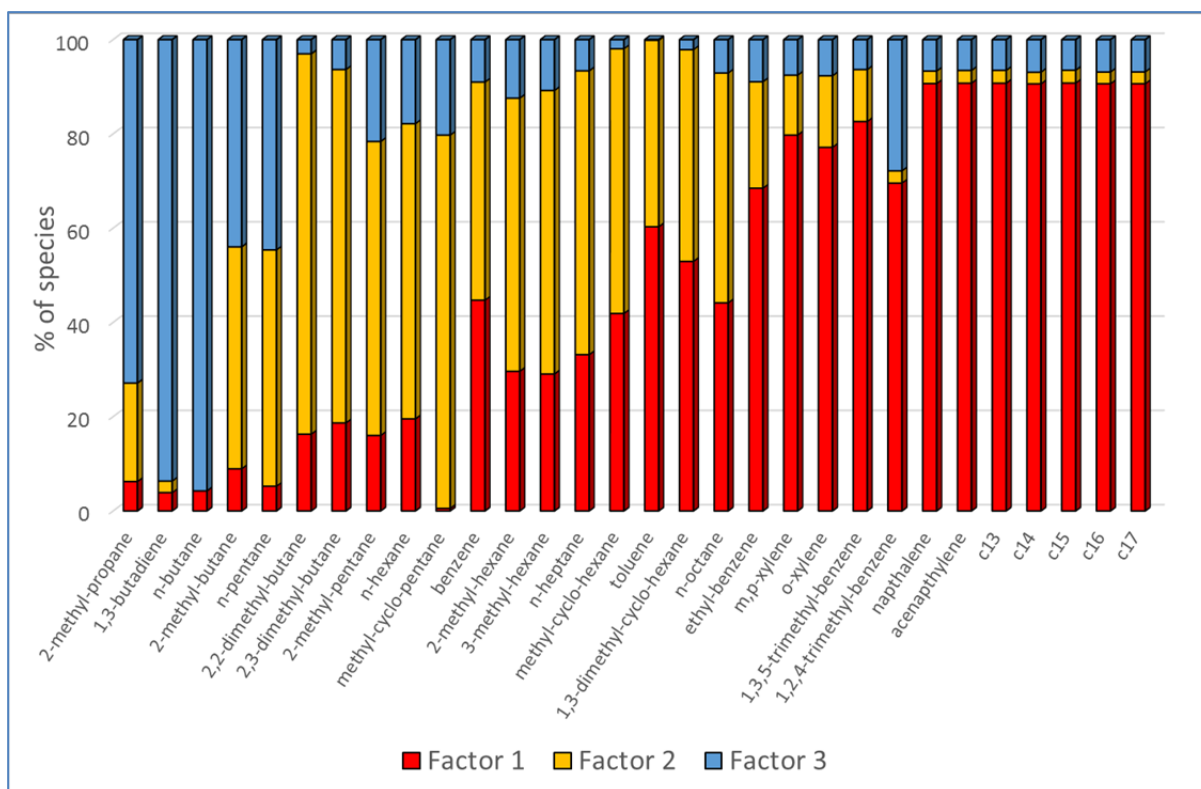
Figure 26 - Selected three-factor PMF analysis solution

shows the average profiles of the three derived factors. Factor 3 from this study is significantly correlated, R 0.71, with the Fugitive Natural Gas from Field et al (2015a). However, it should be noted that the factors reported by Field et al (2015a) were derived from continuous measurements that included methane and C2 to C3 VOC at one site downwind of the Pinedale Anticline, with known influence from a water treatment facility with emissions that contained relatively high levels of BTEX. Factor 3 in this study is also assigned as Fugitive Natural Gas due to the importance of lighter weight alkanes.

Factor 1 is similar to a combination of Combustion and Fugitive Condensate factors reported by Field et al. (2015a), with correlations of R 0.53 and R 0.84, respectively. This is not unexpected given the significant differences of input data, but strengthens the importance of these sources, that are highly likely to have different strengths depending upon the location considered. As such, we do not label this factor as a particular source and ascribe it as a Heavy VOC factor.

Factor 2 has a correlation of R 0.31 with the Fugitive Natural Gas factor reported by Field et al (2015a), probably due to the C4 missing contribution. These authors previously reported their factor included additional BTEX (important components in condensate) from dehydration emissions as these occur simultaneously on well pads together with fugitive emissions. It is not possible to label Factor 2 as a particular emission source but this factor is distinct and is ascribed as a Mid-range VOC factor. Smearing of factors and contributing emission sources is a known issue for interpretation of PMF analysis. The separation of measured VOC into distinct factors is significant given that this analysis is attempting spatial rather than temporal variations. Identification of behaviour patterns that separate Fugitive Natural Gas factors from those with heavier VOC, generally associated with combustion and condensate rich emissions, is important for subsequent air quality management, including potential emission control.
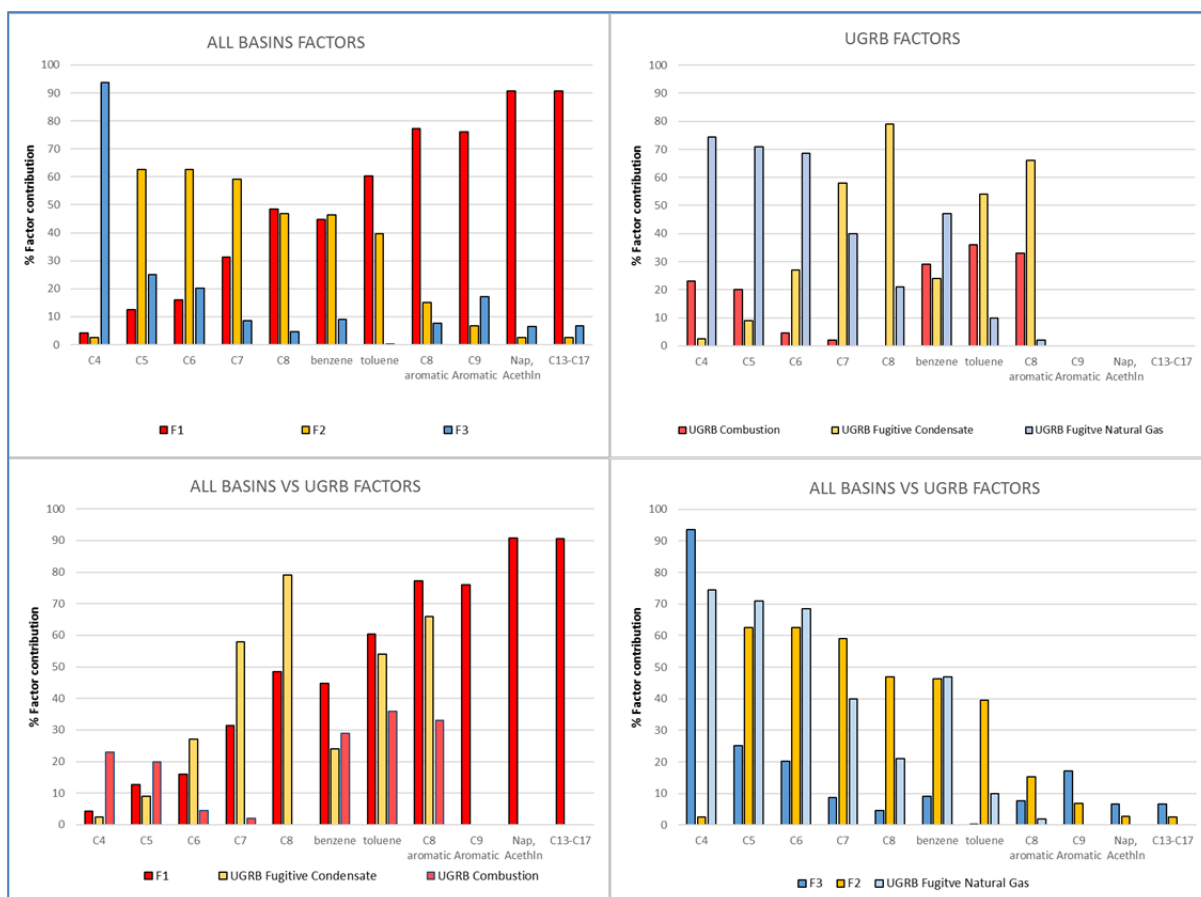
Figure 27 - Factor profiles with the PoD sampling assessment and the previous UGRB study

A number of factors may account for the differences we report; including the use of different input VOC pollutant profiles and the addition of different basins, some of which have contrasting production and management regimes. Emissions from particular O&G developments are also expected to shift over time as production increases, stabilises and declines. Furthermore, equipment management regimes and the application of emission controls may shift as ownership and regulatory frameworks change.

From Figure 26 it is possible to identify a series of compounds representative of certain emission factors. In this case, 1,3,5-trimethyl-benzene seems to be a good indicator for Factor 1 (Figure 28). Areas with a relatively high contribution of Factor 1 also show higher levels of PAHs and HHCs (Figure 17 and Figure 18). Factor 1 based on 1,3,5 trimethyl-benzene concentration could be underestimated for certain hotspots characterised by intensive O&G related activities (i.e. UPA1: drilling, UPA11, UGLB02: water treatment, UGLB04, CC11: compressor station, CC02: refinery). n-Hexane is a good indicator of Factor 2, which would characterised a mid-range VOC factor but again these hotspots are uncorrelated. Finally, n-butane is well correlated with Factor 3, which would represent fugitive natural gas emissions. Such a relation between n-butane and fugitive natural gas emissions has already been introduced in section 3.4.
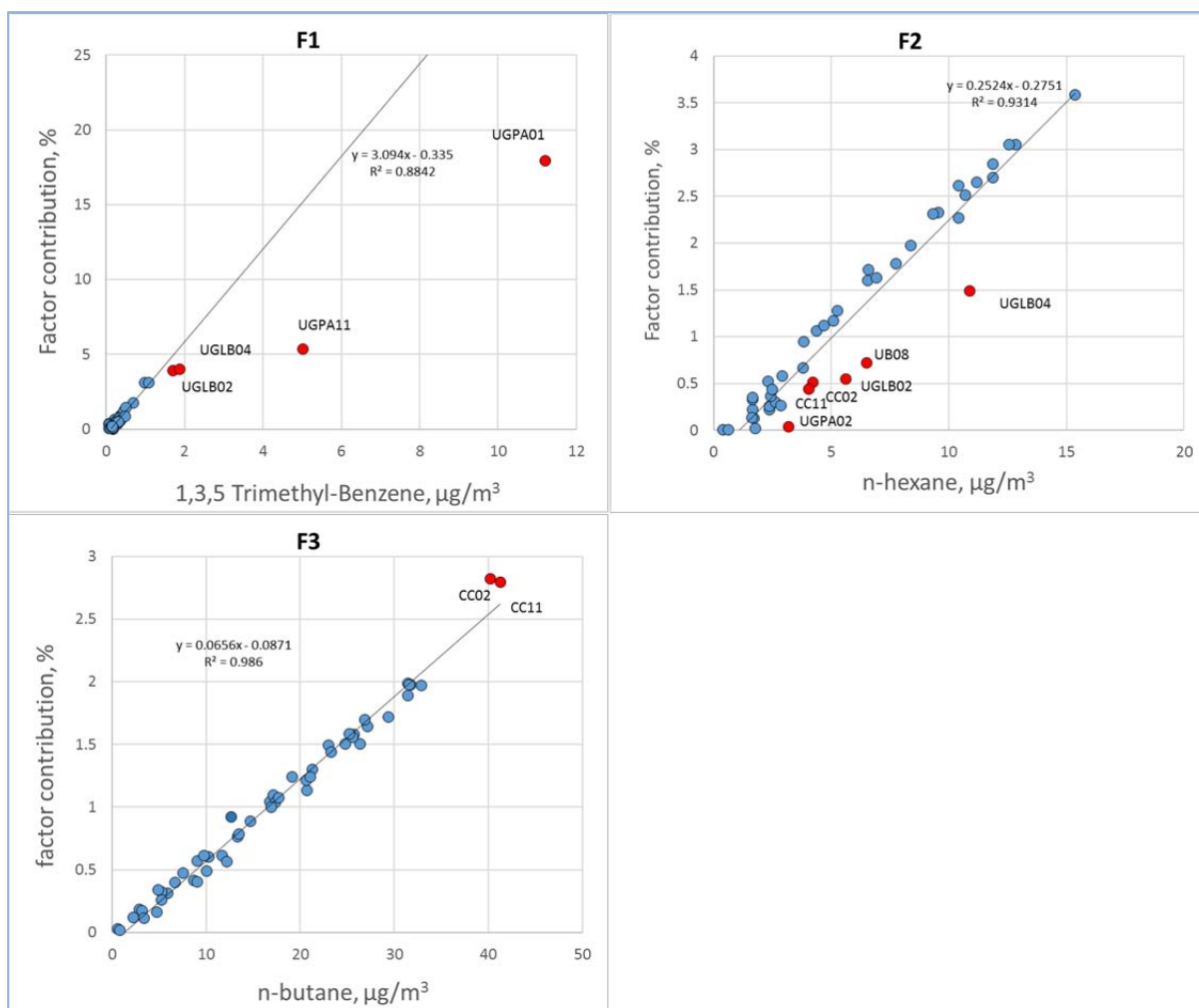
Figure 28 - Factors versus characteristic compounds

Figure 29 shows the partial contribution of each factor for the set of sampled points considered for analysis. The contribution of a given factor to each point varies according to the particular location that is considered. In general, each basin seems to be characterised by a different balance of contributing factors, and this is likely conditioned by the nature of the production and associated activities in the given area. Factor 1 (heavy VOC) is most prevalent in the UGRB, in particular the Pinedale Anticline area. Factor 2 is evident in all basins but is most important for the Uintah Basin. While Factor 3 (Fugitive Natural Gas) is present in all measured basins although it is of greater importance for Converse County and the Uintah Basin. The importance of the Fugitive Natural Gas for the Unitah Basin again confirms previous findings originally reported by Karion et al. (2013). The relatively high contribution for Fugitive Natural Gas factor at Converse County is interesting given the lack of natural gas infrastructure and the flaring of gas in this development area.

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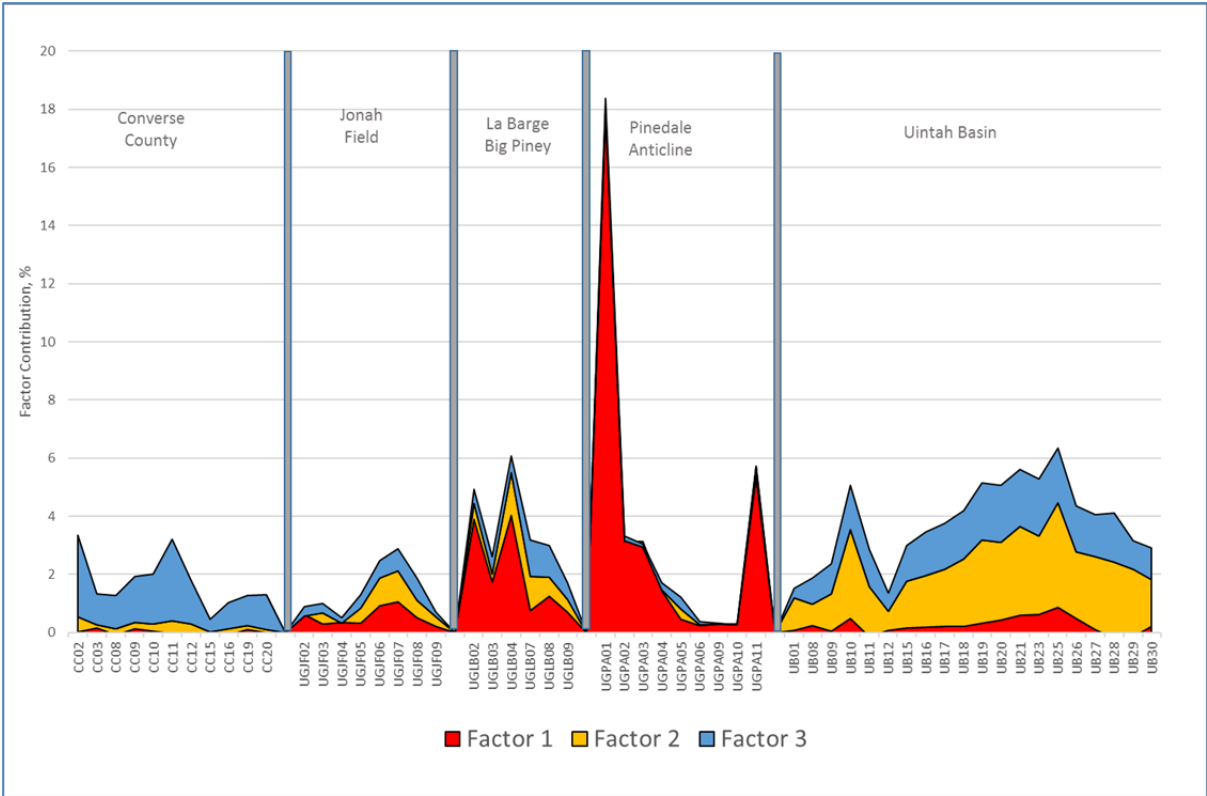
Figure 29 - Factors associated with the sampling points

4 CONCLUSIONS

- Diffusive sampling of VOCs, PAHs and HHCs at three different US basins with significant O&G development demonstrated that PoD samplers are effective for measuring air pollutant distributions over wide spatial areas where pollutant concentrations range from background to highly polluted levels.

- Diffusive samplers can be applied as a scoping and analysis tool to show the relative importance of O&G emission sources to ambient air quality.

- Our measurements support existing literature that has reported relatively high ambient concentrations of aromatic compounds in the Pinedale Anticline and relatively high ambient concentrations of alkanes in the Unitah basin.

- For our study (C_1-C_3 not measured), increased production are positively correlated with ambient concentrations of particular pollutant categories, namely: gas production and C_4 and C_5 alkanes; oil production and C_6 to C_8 hydrocarbons; water production and $>C_8$ hydrocarbons.

- Compound ratios of VOC, namely; n-butane/2-methyl-propane, n-pentane/2-methyl-butane and n-heptane/n-hexane, can serve as indicators of gas to oil production ratios.

- PMF analysis of the PoD sampler measurements enabled identification of different factors associated with O&G production activities. At each basin contributions of these factors can be related to the composition of production as defined by the relative contributions of gas, water and oil.

- Comparison of aircraft (SONGNEX) and ground level measurements showed good agreement, notwithstanding the influences of location, dilution and mixing.

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6 ANNEX I

6.1 Production around sampling sites

In order to systematically compare the areas considered each sampler site was characterised in terms of production levels. As described in **Table 5**, four levels of production are arbitrarily defined for the 3 km radius at each site for both O&G production. These levels are coded zero to marginal (Z), low (L), medium (M) and high (H).

Table 5 - Codes for levels of monthly production

Level	Code	Oil (barrels)	Gas (Mcf)	Water (barrels)
Zero to marginal	Z	>1,000	>10,000	>1,000
Low	L	1,001 to 9,999	10,001 to 99,999	1,001 to 9,999
Medium	M	10,000 to 29,999	100,000 to 999,999	10,000 to 29,999
High	H	<30,000	<1,000,000	<30,000

And in order to define a site as production or background the following rules were applied to the two codes evident for each site.

If there is zero production for either oil or gas: the site is a background site.

If there is high production for either oil or gas: the site is a production site.

If there is low production for both oil and gas: the site is a background site.

If there is one high/medium and one low production for either oil or gas: the site is a production site.

If there is a combination of medium and high production for either oil or gas: the site is a production site.

With application of these rules, the number of site that are defined as production varied between the areas considered. **Table 5** shows the number of production sites with medium high and low levels for O&G production. While the data in this table allows for the definition of production sites (i.e. medium and high production levels), it also shows clear differences between the basins. For Converse County 11 out of 25 sites were defined as production sites. For the Jonah Field 9 out of 9 sites were defined as production sites. For the La Barge/Big Piney development, 7 out of 9 sites were defined as production sites. For the Pinedale Anticline 10 out of 13 sites were defined as production sites. For the Uintah Basin 26 out of 35 sites were defined as production sites.

Therefore, in agreement with the afore-mentioned terms, the following designations could characterise each of the areas in terms of production:

- Uintah Basin: low oil and medium gas
- Pinedale Anticline: high oil and high gas
- La Barge/Big Piney: low oil and medium gas
- Jonah Field: high oil and high gas
- Converse County: high/medium and a medium/low gas

Table 6 - High, medium and low gas and oil production points in each area

Basin	Total sampling points	Gas-Oil Production sites	High Oil	Medium Oil	Low Oil	High Gas	Medium Gas	Low Gas
Uintah Basin	35	26	0	3	29	0	26	6
Pinedale Anticline	13	10	10	0	0	10	0	1
La Barge/Big Piney	9	7	0	1	6	0	7	1
Jonah Field	9	9	7	2	0	9	0	0
Converse County	25	11	7	4	7	0	8	9

In a similar way, according to the water production ranges established in **Table 4**, **Table 7** shows the distribution of water production around sampling sites of each basin. The relationship between water, gas and oil production is divergent, as indicated by the ratios shown in **Table 4**, for the assessed basins. **Table 7** highlights Pinedale Anticline, Johan Field and Uintah Basin as relatively high water producing areas compared to La Barge/ Big Piney and Converse County. The different production characteristics of the sampled areas provide a context for comparisons between basins.

Table 7 - High medium and low water production points in each area

Basin	Total sampling points	Water Production sites	High Water	Medium Water	Low Water	Zero Water
Uintah Basin	35	28	13	15	4	3
Pinedale Anticline	13	10	10	0	0	3
La Barge/Big Piney	9	4	2	2	4	1
Jonah Field	9	9	8	1	0	0
Converse County	25	7	3	4	6	12

Table 8 shows instead the codes associated with each sampling point according to the established criteria.

Table 8 - Characterisation of the sampling points (3 km surrounding production)

Converse County				La Barge / Big Piney				Uintah Basin			
Sampler	OIL	GAS	WATER	Sample	OIL	GAS	WATER	Sampler	OIL	GAS	WATER
CC01	Z	Z	Z	UGLB01	Z	Z	Z	UB01	M	M	H
CC02	H	M	M	UGLB02	L	M	L	UB02	M	M	H
CC03	H	M	H	UGLB03	L	M	L	UB03	L	L	L
CC04	L	L	Z	UGLB04	L	M	M	UB04	L	L	L
CC05	L	L	Z	UGLB05	L	L	L	UB05	L	L	L
CC06	L	L	Z	UGLB06	Z	M	L	UB06	L	L	L
CC07	L	L	Z	UGLB07	M	M	H	UB07	L	L	M
CC08	H	M	M	UGLB08	L	M	M	UB08	L	M	M
CC09	H	M	M	UGLB09	L	M	H	UB09	L	M	M
CC10	H	M	H					UB10	L	M	M
CC11	H	M	H	Pinedale Anticline				UB11	M	H	H
CC12	H	M	M	Sampler	OIL	GAS	WATER	UB12	M	M	H
CC13	Z	Z	Z	UGPA01	H	H	H	UB13	L	L	M
CC14	Z	Z	Z	UGPA02	H	H	H	UB14	Z	Z	Z
CC15	M	L	L	UGPA03	H	H	H	UB15	L	M	M
CC16	M	L	L	UGPA04	H	H	H	UB16	L	M	M
CC17	Z	Z	Z	UGPA05	H	H	H	UB17	L	M	M
CC18	L	Z	L	UGPA06	H	H	H	UB18	L	M	M
CC19	M	L	L	UGPA07	Z	Z	Z	UB19	L	M	M
CC20	M	M	L	UGPA08	H	H	H	UB20	L	M	M
CC21	Z	Z	Z	UGPA09	H	H	H	UB21	L	M	H
CC22	L	L	L	UGPA10	H	H	H	UB22	L	M	H
CC23	Z	Z	Z	UGPA11	H	H	H	UB23	L	M	M
CC24	L	L	Z	UGPA12	Z	Z	Z	UB24	L	M	H
CC25	Z	Z	Z	UGPA13	Z	L	Z	UB25	L	M	H
Jonah Field								UB26	M	H	H
Sampler	OIL	GAS	WATER					UB27	L	M	H
UGJF01	M	H	M					UB28	L	M	H
UGJF02	M	H	H					UB29	L	M	H
UGJF03	H	H	H					UB30	L	M	H
UGJF04	H	H	H					UB31	Z	Z	Z
UGJF05	H	H	H					UB32	Z	Z	Z
UGJF06	H	H	H					UB33	L	M	M
UGJF07	H	H	H					UB34	L	M	M
UGJF08	H	H	H					UB35	L	M	M
UGJF09	H	H	H								

H: High, M: Medium, L: Low, Z: Zero to marginal

6.2 Compound versus production ratios in aircraft and ground measurements
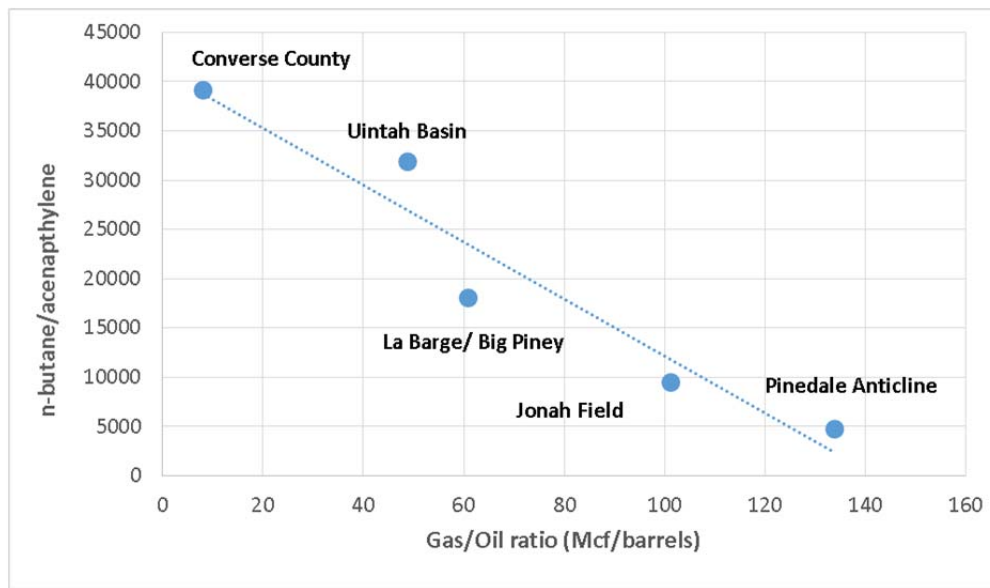
Figure 30 - n-butane/acenapthylene median ratio versus gas and oil production ratio in the studied basins
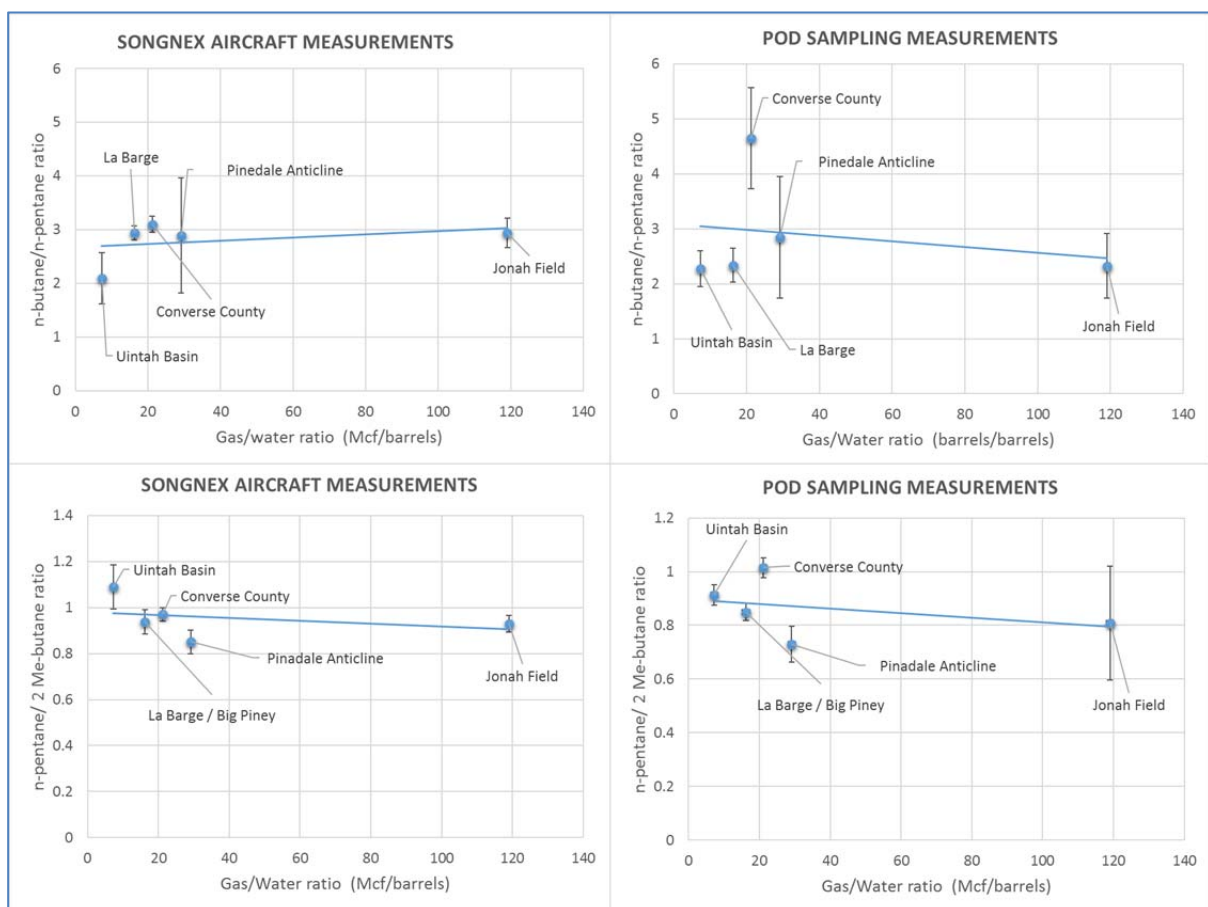
Figure 31 - Median compound ratios for SONGNEX aircraft and PoD sampler measurements versus ratios of gas and water production in the studied basins
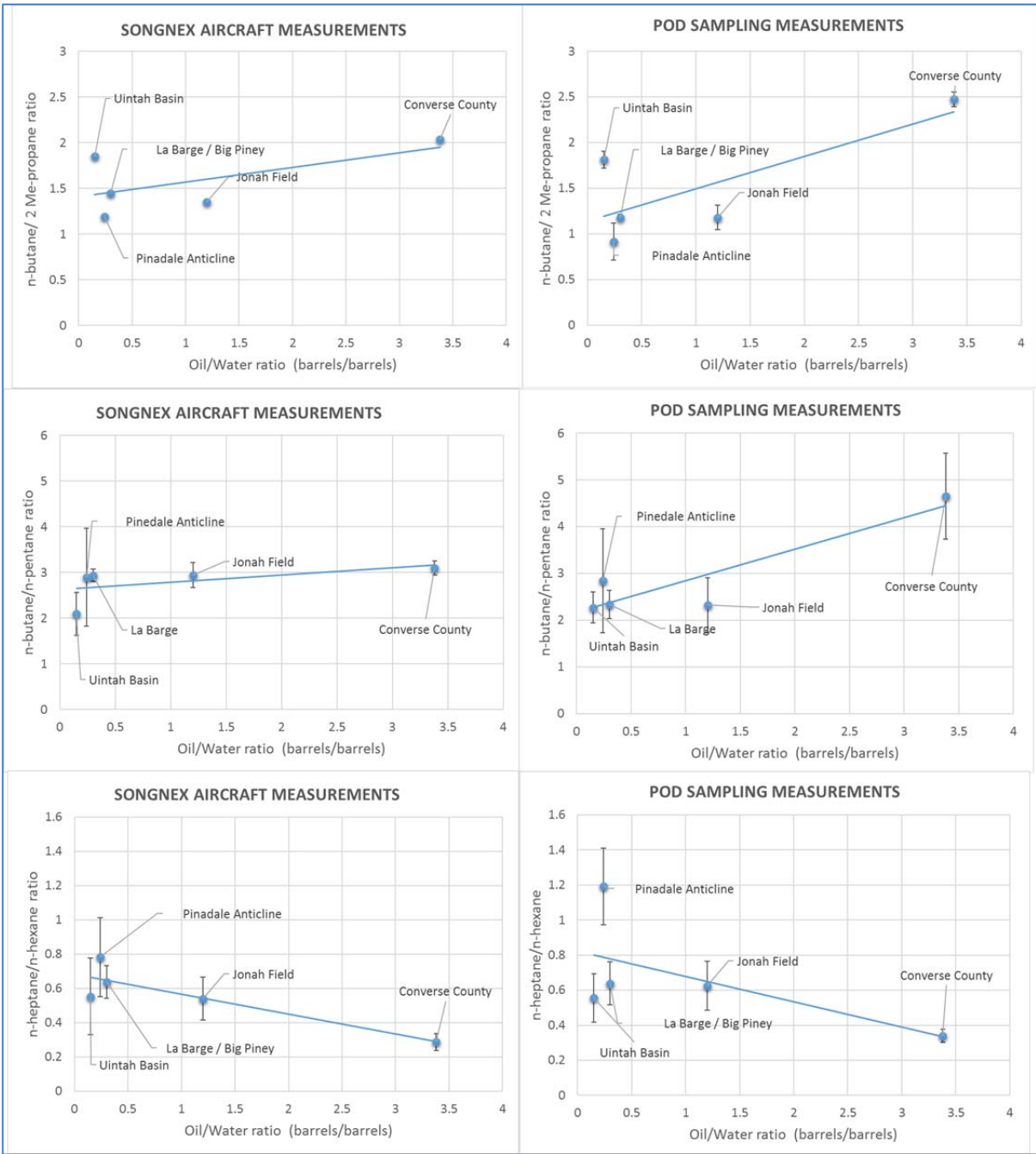
Figure 32 - Median compound ratios for SONGNEX aircraft and PoD sampler measurements versus ratios of oil and water production in the studied basins

6.3 Concentration range, method detection limit and uncertainty of the input data for the PMF analysis
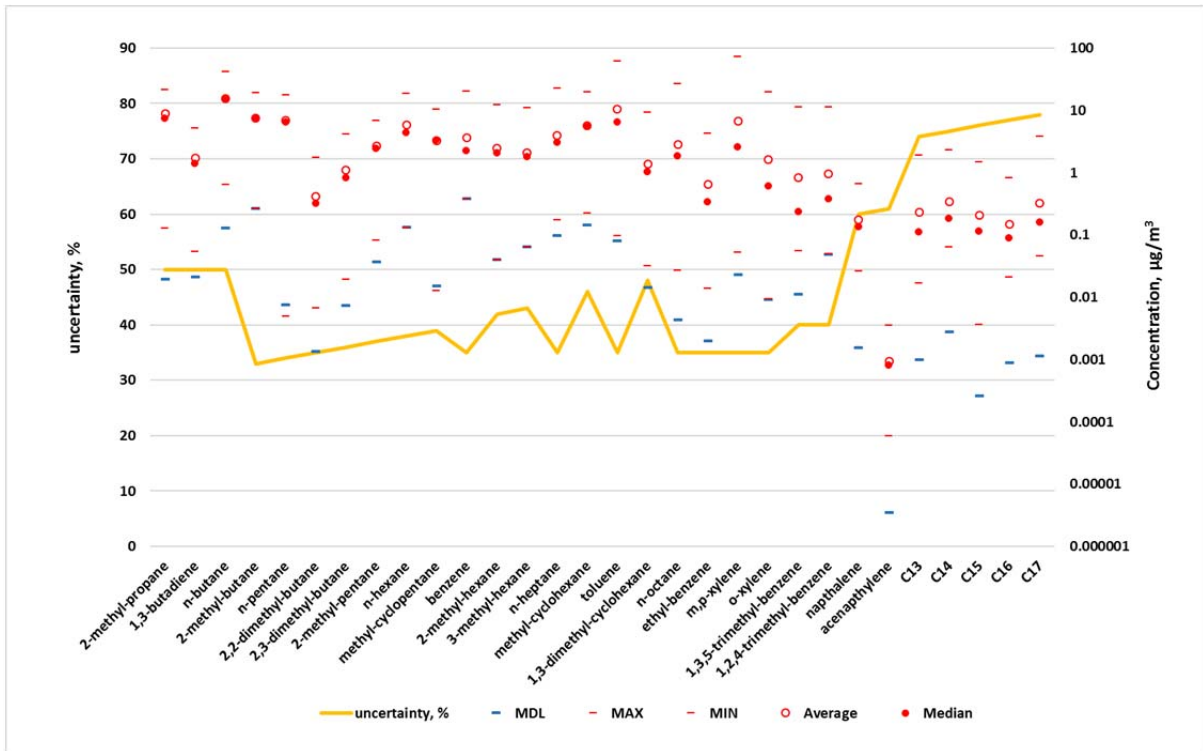
Figure 33 - Concentration range, method detection limit and uncertainty of the evaluated compounds

6.4 Bootstrap, Displacement and Bootstrap-Displacement errors
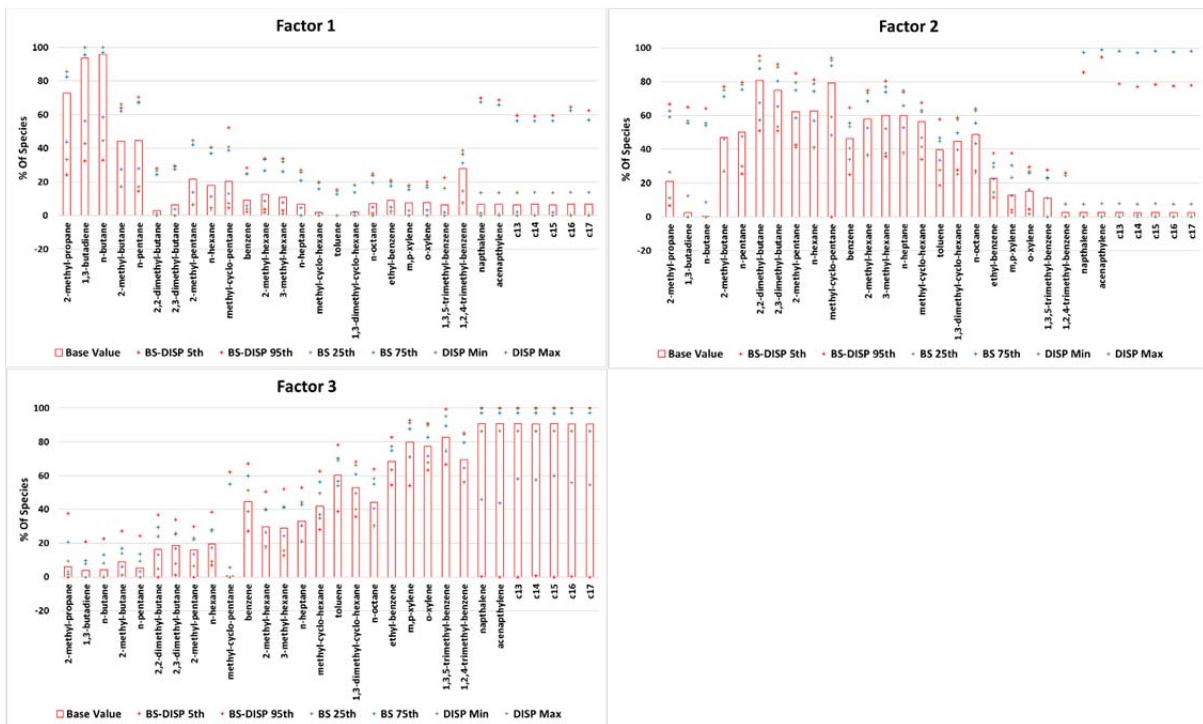
Figure 34 - PMF error estimation of % of species contribution to each factor

7 ANNEX II

State of the art of unconventional oil and natural gas development in the United States.

(Extracted from Expert Contract Report 392796: R. A. Field, October 2016)

7.1 BACKGROUND

7.1.1 Onshore unconventional oil and natural gas development in the United States

Driven by the development of hydraulic fracturing the oil and gas (O&G) energy sector in the United States (US) has undergone rapid expansion (Wang et al. 2014). The US Energy Information Administration (EIA) predicts that US energy imports and exports will come into balance for the first time since the 1950's sometime between 2020 and 2030 (US EIA, 2015a). US crude oil production increased from 5 million barrels per day (b/d) in 2008 to 9 million b/d in 2016 (US EIA 2016a). Low prices are predicted to drive production down through 2017; however, the US EIA (2016b) reference case predicts a rise in production to 11 million b/d by 2040. While predicted oil production scenarios show a wide range for 2040, 7 million b/d to 17 million b/d, tight onshore oil development is the primary driver for increased production in the US. Despite low prices natural gas production in the US continues to increase with dry natural gas production of 27.1 trillion cubic feet (Tcf) in 2015 exceeding production of 25.9 Tcf in 2014 (US EIA, 2016a). In 2015 shale gas and tight oil developments accounted for approximately half of natural gas production in the US. Projections by US EIA (2016b) predict that in 2040 dry natural gas production in the US will increase to over 40 Tcf with the contribution of shale gas and tight oil developments increasing to nearly 70%. While production from tight sandstone and carbonate formations is projected to increase, the market share is relatively stable. Contributions to total production from offshore, Alaska and coalbed methane are predicted to remain stable or decline (US EIA, 2016a).

While changing the energy balance of the US, increased use of hydraulic fracturing ("fracking") for the development of O&G resources from coal, carbonate, sandstone, and shale deposits has also created environmental concerns over water and air quality impacts (Gregory et al. 2011, Vengosh et al. 2014; Field, Soltis and Murphy, 2014; Moore et al. 2014). Impacts to air quality are predominately due to emissions of oxides of nitrogen (NO_x) and hydrocarbons. Besides direct impacts upon humans and the environment, these pollutants can also lead to the formation of secondary pollution including ozone (O_3) and fine particles (Liggio et al. 2016), which also deteriorate air quality. Besides motor vehicles, NO_x emissions are from a variety of combustion sources,

most often engines, employed during exploration (e.g. drill rigs and pumps), production (e.g. heaters, pumps and combustors) and gathering/transmission/distribution (compressors). Hydrocarbons are co-emitted with NO_x from combustion sources but are also released to the atmosphere through leakage as extracted products often reach the surface infrastructure at high pressure. Hydrocarbon emissions are of prime importance as methane (CH_4) is an important, and potent, greenhouse gas and non-methane hydrocarbons (NMHC) are O_3 precursors, some of which are hazardous air pollutants (HAPs). Emissions of HAPs can have direct health effects (Tustin et al. 2016). Concern regarding the influence of emissions of HAPs upon communities located in close proximity to O&G development has led to a number of measurement based assessments (Olaguer et al. 2016; Macey et al. 2014). While occupational exposure assessments have been performed for silica dust from fracking operations (Esswein et al. 2013), studies of exposure to VOC for either O&G workers or locally impacted populations are absent from the literature (Shonkoff et al. 2014; Zielinska et al. 2014; Webb et al. 2016). Epidemiological and survey approaches often report negative health effects from exposure to emissions (McKenzie et al. 2012; Steinzor et al. 2013; Brown et al. 2014; Colborn et al. 2014; Bamberger and Oswald, 2015; Brown et al. 2015; Tustin et al. 2016; Rasmussen et al. 2016). However, a review of heath based studies by Werner et al. (2015) notes a lack of long term studies, together with a need for improved methodological rigour. As such there is a scientific gap is evident between inferred and actual health outcomes due to exposure to emissions from O&G development (Marrero et al. 2016).

While pollutants from the O&G sector contribute to air quality concerns there are potential benefits when natural gas replaces coal, in particular within the power generation sector as the carbon dioxide (CO_2) emission rate is lower per megawatt of electricity produced (Alvarez et al. 2012). With a lower carbon footprint than coal, natural gas is often promoted as a bridge fuel for an energy budget that transitions from fossil fuels to renewable energy sources (Levi, 2013). The relative climate forcing impacts of coal and natural gas requires calculation of the influence of both CO_2 and CH_4 to the climate system. Replacing coal with natural gas also requires consideration of the emissions of other radiative forcing components including black carbon (NOAA, 2015). Furthermore, other air quality issues are impacted, including exposure to particulate matter and O_3. The environmental benefits of the replacement of coal by natural gas, in particular for power generation, has focused upon both ozone production and climate change (De Gouw et al. 2014; Pasci et al. 2015). Air quality challenges in China, in particular with respect to visibility and exposure to particulate matter are strongly tied to the relatively high use of coal. While complicated by the timeframe considered the potential climate benefits associated with increased use of natural gas are diminished

with greater leakage rates (Alvarez et al. 2012). While highlighting the importance of the timescale for estimating any costs or benefits Howarth (2014) concluded that natural gas had limited benefit as a bridge fuel. However, Melvin et al. (2016) report significant financial and climate benefits of US EPA programs and policies that have reduced methane emissions over the past twenty years. The most recent regulatory initiatives in the US for the O&G sector is noted in the Annex II Section 7.2.

Increased production of unconventional O&G resources in the US has led to greater scrutiny of CH_4 and NMHC emissions within the entire supply chain of natural gas and other petroleum products. Emissions can occur at any position along this supply chain, starting at the point of extraction. Emissions are largely determined by how efficient the system is – that is, how open or closed the system is to the surrounding environment. As soon as subsurface hydrocarbons become mobile there is potential for an eventual air emission. Emissions from O&G sources are classified by air regulatory agencies as point sources (emitted from a stack or pipe), mobile sources (from trucks, trains, drill rigs), and fugitive sources (emissions due to equipment leaks, or external forces such as wind, or natural or man-made faults or fractures in the earth's surface). A variety of measurement-based approaches have been applied to improve understanding of the complexity of O&G emissions. Mobile monitoring (Rella et al. 2015; Albertson et al. 2016), flux quantification (Allen et al. 2013; Omara et al. 2016; Robertson et al. 2017) tall tower (Pétron et al. 2012; Gilman et al. 2013) and network stations (Pekney et al. 2014; Zavala-Araiza et al. 2014; Schade and Roest 2016), tethered ballon (Helmig et al. 2014), aircraft (Pétron et al. 2012; Karion et al. 2015; Caulton et al. 2014; Frankenberg et al. 2016) and satellite platforms (Streets et al. 2013; Kort et al. 2014) each have utility for measuring air quality impacts from O&G emission sources. A number of studies have applied isotopic methane analysis to separate biogenic and thermogenic methane contributions (Townsend-Small et al. 2015). Correlation approaching using ethane as an indicator of the presence of thermogenic methane have also been successfully applied at a number of US basins (Wennberg et al. 2012; Peischl et al. 2013; Smith et al. 2015). A variety of analysis methods including Chemical Mass Balance (CMB) and Positive Matrix Factorisation (PMF) have also been applied to determine the significance of contributing emission sources. Such approaches benefit from the measurement of a wide range of chemical species. Field et al. (2015a) applied PMF analysis to demonstrate the dominance of fugitive emissions, and the influence of water treatment emissions in the Pinedale Anticline, Wyoming.

7.1.2 Air quality and onshore oil and natural gas energy sector

Determining the air pollutants that will be of most concern begins with understanding of the chemical and physical characteristics of the extracted product is of

particular importance. O&G production is a matrix of water, oil, and natural gas that characterises a particular reservoir. The temperature and pressure of products reach the surface is related to the depth of drilling. Dry natural gas, wet natural gas, and oil wells have different product handling approaches, and as such potentially divergent balance between contributing emission sources. Natural gas that contains more than 0.1 gallon of condensate per 1,000 cubic feet of gas (<95% CH_4) is termed "wet gas." Wet gas contains complex hydrocarbons from natural gas liquids (ethane, propane, butanes) and condensate, which is a mix of longer chain (C_{5+}) and cyclic hydrocarbons (cycloalkanes and aromatics), whereas dry natural gas is predominately CH_4. Because wet gas usually contains a more complex mixture of hydrocarbons than dry gas, the variety of VOC and other air emissions from wet gas is potentially greater. Furthermore, wells producing crude oil, a complex mixture of hydrocarbons that generally have from five to 40 carbon atoms per molecule, will have different air emissions compared to natural gas wells. The importance of properly discriminating between different types of wells and producing regions was noted by Zavala-Araiza et al. (2015a).

The chemical and physical characteristics of production also determine the activities and processes that act upon products that reach the well head. The onshore O&G industry encompasses a wide range of activities that are needed to extract raw hydrocarbon products from underground reservoirs, transform them into products, and transport them to end users, most importantly power generation and domestic consumption. The US EPA (2016a) identifies four important sectors namely: production; gathering and processing; transmission; and distribution. Emissions from production are due to the extraction of hydrocarbons from the subsurface and are classified as either conventional or unconventional. Production activities include drilling, fracking, completion, and initial separation of water, oil, and gas components. Production emissions are dependent upon a number of factors that are outlined further in this section under "Production emission sources". Gathering and processing activities are necessary to ensure that products are the required quality for transmission. These activities connect production with transmission and can occur at well pads and/or at locations removed from O&G developments. For supply of natural gas to consumers' removal of impurities, high molecular weight hydrocarbons, and fluids is necessary to ensure pipeline grade product (typically 95%-98% CH_4) enters national and international transmission pipelines. Emissions from transmission activities (see "Transmission emission sources" in this section) result from the delivery of natural gas through networks of high-pressure pipelines to city gate stations or industrial end users. Natural gas storage is within this sector and is within tanks, underground reservoirs, aquifers, and salt caverns (US EPA, 2016a). Emissions from distribution are from (see

"Distribution emission sources" in this section) networks of lower pressure pipelines that mainly deliver natural gas to domestic end users beyond city gate stations.

In the US considerable research attention has been given to the leakage of natural gas from basins with significant amounts of O&G development. Leakage is often calculated related to production volumes. A wide range of basin scale leakage rates are reported in the US, with the importance of super emitters evident in different US O&G developments (Brandt et al. 2014). Leakage rate estimates from aircraft flux approaches at the basin scale range from less than 1% (e.g. UGRB in Wyoming) to over 10% (e.g. Uintah Basin in Utah) (Murphy et al. 2014; Karion et al. 2013). Peischl et al. (2015) report loss rates of 1.0–2.1% for the Haynesville shale region (Texas), 1.0–2.8% for the Fayetteville shale region (Arkansas), and 0.18–0.41% for the Marcellus shale region (Pennsylvania). Peischl et al. (2015) note that these natural gas loss rates are within the range of 0.3–1.9% estimated by Howarth et al. (2011) and together the investigated regions represent over half of the U.S. shale gas, and 20% of total natural gas production for the US in 2013. Hence, extrapolations of basin wide fluxes of the Uintah basin to the national scale are inappropriate when considering unconventional production losses. Notwithstanding losses due to transmission, distribution and end use, such loss rates are far lower than the threshold set by Alvarez et al. (2012) of 3.2%, below which the climate impact of using natural gas as a fuel in power plants is estimated to be less than that of coal. A production-weighted loss rate of 1.1% reported by Peischl et al. (2013) is similar to a 1.0% loss rate derived from the 2012 EPA GHG emissions inventory for natural gas systems. On-going evaluations at O&G basins in the US (NOAA, 2015; RPSEA 2015) should reconcile apparent differences between inventory estimates and flux estimates (Pétron et al. 2014) along with the reasons behind regional differences of loss rates for different O&G developments in the US (Robertson et al., 2017). In some US basins O&G sources account for over 95% of the pollution burden for NO_x and VOC (UGRB as reported by Wyoming DEQ, 2012), whereas in other basins with significant urbanisation these activities can make up less than 15% of total air emissions (Roy et al. 2013; Swarthout et al. 2015). However, while the relative importance of O&G sources may be low, the impact upon attainment of the National Ambient Air Quality Standard for O_3 can be significant (Kemball-Cook et al. 2010; Olaguer 2012). While estimated emissions from US basins have a wide range (Brandt et al. 2014), CH_4 fluxes rates measured at well pads indicate that emissions from production are the most important at the basin scale (Robertson et al. 2017). As more US basins are evaluated the influence of long term leak detection and repair programs upon emission rates, and the importance of sites with relatively high emission sites will become clearer. In addition to producing wells attention is also being given to emissions from idle, orphaned and abandoned wells (Boothroyd et al. 2016).

Research into emissions of CH_4 and NMHC has focussed upon reconciliation between differences that have emerged between estimates of emissions derived from emission inventory calculations and atmospheric measurement based approaches (Lyon et al. 2015). Bottom up emission inventories, derived from emission measurements or engineering estimate, used for a various air quality regulatory efforts, are often far lower than top down estimates of emissions (Harriss et al. 2015). Such differences have important implications for air quality management (Omara et al. 2016). Emission inventories are critical for prediction of the potential significance of O&G emission sources for a particular area. Spatial inventories are required input for photochemical grid models that are required for air quality management. In Wyoming inaccurate inventories have led to the failure of models to replicate wintertime ozone episodes in the Upper Green River Basin (UGRB). Without a model that replicates actual behaviour, and defines the relative importance of different ozone precursors, any control policies are arbitrary and not based upon a scientific foundation. In the Unitah basin models successfully replicated wintertime ozone with simple adjustment of model inputs using ambient measurements. (Ahmadov et al. 2015; Edwards et al. 2014). A plethora of recent studies in the US have shown that emission inventories do not reflect actual emissions from the O&G production sector (Karion et al. 2013; Brandt et al. 2014; Miller et al. 2013). This divergence is related in part due to the use of inappropriate factors and the under reporting of activities. However, the importance of super emitters has emerged as a critical reason for the under reporting of top down emission inventories (Brandt et al. 2014). The concept and importance of super emitters was first noted for the motor vehicle fleet by Stedman et al. (1989). Allen (2016a) note that emission inventories would be improved with the additional of a factor for so called super emitters. However, as noted by Robertson et al (2017) different basins have different distributions of leakage rates from well pads. And while so called super emitters are evident in all basins the application a distribution curve may be basin specific. Lyon et al. (2016) performed helicopter-based infrared camera surveys of more than 8000 well pads in seven US basins, and report that 4% of site could be considered as high-emitting sources. However, this proportion ranged from 1% in the Powder River (Wyoming) to 14% in the Bakken (North Dakota). For the Four Corners region of the US Frankenburg et al. (2016) also report a heavy tail distribution for the 250 point sources that they measured, with the top 10% of emitters accounting for 50% of measured emissions of methane. Roy et al. (2013) noted the limitation of a single activity factor for emission inventory calculations and recommended the use of a distribution curve that would better reflect emissions from a hypothetical population. A comprehensive reconciliation assessment of the Barnett shale development in Texas by Zavala-Araiza et al. (2015b) showed that when emission inventories are complete and representative then top down

and bottom up estimates can be within uncertainty limits. However, the need for both improved reporting and validation is evident, as the revised inventory for the Barnett Shale development reports emissions of methane 90% higher than the base inventory. Zavala-Araiza et al. (2015c) provide a functional model for incorporation of super emitting sites into emission inventories.

While compositional profiles are often consistent within particular reservoirs they can vary widely between different reservoirs, even within the same basin. As such applying single factors within emission inventories, while reasonable as an approximation, can lead to high degrees of uncertainty. To support emission inventory validation efforts considerable research effort has been applied to source apportionment of contributions from different emission sources associated with natural gas production (Townsend-Small et al 2015; Allen 2016b). While different approaches may be applied, most often driven by characteristics of the extracted product, the basic steps and associated emissions, are consistent for natural gas development (Field et al. 2014).

The US Government Accountability Office (2012) has reported that cumulative air quality impacts are difficult to determine as the extent and severity of risks vary significantly within and between developments due to location and process driven factors. As drilling has encroached on residential and urban areas, attention has turned to the impact of O&G emissions on ambient air quality and human health (US GAO, 2012). As such, one size does not fit all, and the importance of location specific assessment has emerged. The following sections provide more detail for production, transmission and distribution of natural gas.

7.1.2.1 Production emission sources

A wide variety of potential emission sources can be spread over tens of thousands of acres of a production area and this complicates assessment of local and regional air quality impacts. Production emissions are predominately from drilling, fracking, completion, gathering and processing activities. Emissions of nitrogen oxides are from the use of engines to drive equipment including drill rigs, pumps for fracking operations and compressor engines. If fuel consumption and engine performance are known reporting of emissions for nitrogen oxides is fairly straightforward. Estimates of emissions of hydrocarbons are more complicated due to the number of associated processes and variability of the composition of production. A number of studies have attempted to survey production sites to determine distributions of emission flux rates from production activities on well pads using direct measurements (Allen et al. 2013) tracer release (Lamb et al. 1995; Roscioli et al 2015; Omara et al. 2016) and gaussian plume methods (Brantley et al. 2014; Foster-Wittig et al. 2015; Lan et al. 2015; Yacovitch et al. 2015; Robertson et al. 2017).

Close to well pads, emissions are concentrated and exposure to a wide range of pollutants is possible. In general, emissions are from combustion (engines), processing (separators) and fugitive (leakage) sources. Fugitive emissions are difficult to quantify because they can be difficult to detect and may be intermittent. Hydraulic fracturing, more specifically "flowback" is an important example. There are few measurements of flux rates from flow back operations and estimates of emissions remain controversial (O'Sullivan and Paltsev 2012). Air emissions occur from various production activities. In this regard, unconventional and conventional wells are no different upon completion after flow back is completed, and both connect to the same midstream and downstream distribution network. And while attention has focussed upon unconventional development Omara et al. (2016) note that emission rates from older conventional wells can exceed those of unconventional well, highlighting the importance of age of equipment and associated maintenance. At the well pad processing activities that separate gas, produced water, and liquid hydrocarbons from each other are important. Produced water is an important but uncertain emission source with a number of possible handling approaches. Aromatic hydrocarbons, including benzene, toluene, ethylbenzene and xylenes (BTEX) are of particular importance when considering the handling of wet gas that leads to the production of condensate. Because of the hazardous nature of BTEX emissions, control of pollutants from dehydration units through combustion is preferable to venting. Despite control by combustion, BTEX and other VOC emissions can be significant (Hildenbrand et al. 2016). Relatively high BTEX emissions from well pads storage tanks, again despite control mechanisms, is reported by Brantley et al. (2015). The application of different control measures also depends on the availability of infrastructure. In the oil rich Bakken development, South Dakota US, a lack of pipelines for natural gas lead to extensive flaring operations (Prenni et al. 2016).

7.1.2.2 Transmission emission sources

Transmission losses include those from gathering and processing operations, compression facilities (Johnson et al. 2015) and storage locations. Leakage rates from high pressure transmission pipeline networks, known as midstream, are reported by Lamb et al. 2016 as relatively low, compared to production, and close to EPA estimates of less than 1%. Transmission losses of less than 1% are also reported for extensive Russia networks (Dedikov et al. 1999; Lelieveld et al. 2005; Lechtenböhmer et al. 2007). By contrast Marchese et al. (2015) report that CH_4 emissions from US natural gathering and processing are approximately double EPA emission inventory estimates. They report an overall modelled loss rate of 0.47% for all U.S. gathering and processing. Loss rates from processing plants were relatively insignificant compared to gathering facilities. Methane emission rates ranged from 1 to 700 kg per hour (kg/h), with skewed

distributions, in particular for gathering facilities, with 30% of facilities contributing 80% of the total emissions (Mitchell et al 2015). The importance leaking equipment was noted, and facilities with substantial venting from liquids storage had emission rates four times that observed at similar facilities. A similar finding for compressor stations is reported by Subramanian et al. (2015) with the highest emitting 10% of surveyed sites accounting for 50% of the total emissions. With the exclusion of two super emitters, emission rates from compressor stations were similar to factors used in the EPA greenhouse gas inventory. There are few studies in the literature related to leakage rates from storage facilities. However, a catastrophic failure at the Aliso Canyon underground storage facility in Los Angeles US highlighted the importance, and magnitude, of the storage of natural gas in the US (Conley et al. 2016). This failure, caused by a blowout of a connected well, led to the loss of 97,100 metric tons of methane. At its peak, the leak from this facility contributed half of the total methane emissions for the entire Los Angeles Basin (Conley et al. 2016). Normally operating facilities are anticipated to have insignificant emission levels relative to gathering and processing infrastructure. However, there remains some uncertainty for emission rates from the hundreds of thousands of miles of gathering pipelines, in particular as EPA emission estimates are based outdated emission factors from the 1990's (Marchese et al. 2015).

7.1.2.3 Distribution emission sources

Underground distribution networks within cities are harder to evaluate than over ground O&G production sites. Distribution losses are generally calculated based upon engineering factors that rely upon a number of assumptions. However, as noted by Cambaliza et al. (2015) estimates of city scale methane emission fluxes can adopt a number of different approaches including eddy covariance, mass balance, stable isotopes, and correlation slopes. Leakage of natural gas in the urban environment has received limited attention in Europe (Gioli et al. 2012) compared to the US, where recent studies have determined the contribution of leakage from natural gas distribution to total methane emissions. (Phillips et al. 2013; Jackson et al. 2014; Gallagher et al. 2015).

For the US city of Indianapolis Cambaliza et al. (2015) report a methane flux of 7800 ± 3300 kg.hr^{-1} with 67% attributed to natural gas distribution, with the remainder attributed to a landfill site. Through a combination of approaches Lamb et al. (2016) also report the dominance of natural gas leakage for non-biogenic sources for Indianapolis, with inventory estimates too low by a factor of between 3.5 and 6.9. In Boston, natural distribution losses are reported to account for greater than 60% of the regional methane flux with emission rates more than double inventory estimates (McKain et al. 2015). Through a nationwide measurement campaign, Lamb et al. (2015) report losses of

methane from local distribution systems 36% to 70% lower than the 2011 EPA emission inventory. This difference was attributed to improvements of infrastructure and leak detection and repair, while the role of methodological differences was not discounted, as the US inventory is based on factors derived from 1990. There is, however, a wide variation of the contribution of distribution losses to the total emission flux for different cities. For London in 1996 Lowry et al (2001) estimate that ~80% of methane was of biogenic origin (landfill) and that fossil fuels accounted for 20%. Natural gas distribution was reported at the dominant fossil fuel source with a negligible contribution to non-biogenic emissions of less than 5% from motor vehicle emissions.

Mitchell et al. (1990) noted the implications of under reporting emissions from this source with respect to climate change and energy policy. O'Shea et al (2014) report a methane emission region flux for greater London a factor 3.4 larger than the UK National Atmospheric Emissions Inventory. An early independent estimate of leakage from the UK natural gas distribution of 2 to 5% by Wallis (1990) was also higher than the inventory estimate of less than 1%. The need for improved reporting and control of natural gas leakage was clearly articulated by Mitchell (1993) and Lowry et al (2001). Zazarri et al (2015) note that the rapid identification, and therefore remediation of leaks, is now possible with the advent of fast response instruments for measurement of methane. Using fast response methane instruments Gallagher et al. (2016) surveyed natural gas leaks within three US cities. They noted an order of magnitude difference of leak densities with Manhattan, New York, the highest with 4.25 leaks/mile. The leak density for Manhattan is similar to that reported for previous surveys of Boston (Phillips et al. 2013) and Washington, DC (Jackson et al. 2014). Gallagher et al. (2015) relate lower leak rates to pipeline improvements, including the replacement of older materials, and highlight the benefits of pipeline replacement programs. The possibility of significance of post meter losses was highlighted by Wennberg et al. (2012) as one possible cause for the higher than anticipated methane levels in Los Angeles, US. Wunch et al. (2009) postulated a distribution leakage rate loss of 1 to 3% in addition to emission inventory estimated pipeline losses.

7.1.3 Regulation of onshore oil and natural gas emission sources in the United States

In the US there is a complex interwoven set of relationships between the public, regulators, and developers for regulation of emission from the O&G energy sector. Development may take place on private, county, state or federal land with permitting of anywhere from a few to many thousands of wells. For significant actions on federal land, the National Environmental Policy Act (NEPA) usually requires an Environmental Impact Statement (EIS). Air quality is a primary consideration and potential impacts are

estimated by the modelled influence of predicted emissions on ambient air quality. Regulatory agencies will only permit activities so long as they are able to predict that compliance with National Ambient Air Quality Standards (NAAQS), regulated through the Clean Air Act, will be achieved. Compliance with NAAQS is defined by data collected at air quality monitoring stations. When an area is designated as "nonattainment", more stringent permit conditions and increased scrutiny of emission inventories are generally imposed. A nonattainment area is a locality where air pollution levels persistently exceed one or more NAAQS. With respect to the O&G industry, this scrutiny is placed on emissions that affect ozone and nitrogen dioxide (Livovitz et al. 2013). Given that ozone is a secondary pollutant (not emitted directly but formed from atmospheric reactions), the emission and distribution of precursors such as VOC and NO_x are important to know before one can understand the influence of O&G emissions on air quality (McDuffie et al. 2016).

In 2012, US EPA issued important amendments to existing air quality regulations that apply to the O&G industry. The Clean Air Act requires US EPA to develop new source performance standards (NSPS) for industrial categories that cause, or significantly contribute to, air pollution that may endanger public health or welfare (US EPA 2012). Another regulatory program specified by the Clean Air Act, the National Emission Standards for Hazardous Air Pollutants (NESHAP), was reviewed at the same time. Together, the revised rules and regulations were implemented in 2015. The 2012 revision regulates a number of upstream processes not addressed previously, including: well completions and recompletions, compressors, pneumatic controllers, and storage containers. Notably, the rule requires Reduced Emissions Completions (REC) or, alternatively, flaring for most fractured wells. REC are predicted to greatly decrease emissions compared to pit storage of liquids and venting of gas to the atmosphere. While emissions from well completions are known to vary, REC are highly effective at eliminating waste of valuable products. At the US national scale, these control measures are predicted to reduce annual methane emissions by 1.0 to 1.7 million short tons, HAP emissions by 12,000 to 20,000 tons and VOC emissions by 190,000 to 290,000 tons. If fully implemented, VOC emissions from newly fractured wells should be reduced by 95%. Reduced methane emissions, although not directly addressed by the rule, are viewed as a "co-benefit". REC are now required in the US for both new wells and those that are recompleted or worked over to improve production. VOC emission reductions from REC, combined with reductions from other equipment have both economic and environmental benefits (US EPA 2012).

Considerable variation among US states remains for permitting guidelines, reporting requirements, leak testing and leak detection and repair (LDAR) programs. Some states

are acting to strengthen US EPA rules. In the US state of Colorado regulations proposed in November 2013 by the Colorado Public Health and Environment Department are designed to further reduce risks to human health from exposure to pollution. The comprehensive set of rules set stringent requirements to monitor, control, and reduce methane and VOC emissions. Particular attention is given to uncontrolled emissions of VOC, including BTEX, from glycol dehydration units. New units have limits on emission rates and allowable minimum distance from buildings and designated outside activity areas. The Colorado rules (regulation 7) should further reduce air pollution emissions over and above mandatory NSPS and NESHAP rules (CPDHE 2014). Federal agencies are now updating regulations and these recent regulatory actions are discussed in section +-

7.1.4 Onshore unconventional oil and natural gas development in Europe

In the global context, most interest for shale gas development has been expressed within Europe. However, to date only Poland and the United Kingdom (UK) have performed exploratory shale gas extraction, notwithstanding existing unconventional tight sandstone development in Germany. Preliminary pilot hydraulic fracturing has also been performed at the Tompa prospect in Southern Hungary (Shaoul et al. 2012). Preliminary indications are that extensive shale gas resources are present in Europe. The most promising resources have been identified by the US EIA (2011) within France (180 Tcf), Poland (187 Tcf), and Norway (83 Tcf). Technically recoverable reserves of 20 Tcf were also identified within the UK. These reserves are considerably smaller than the 1275 Tcf and 862 Tcf estimated for China and the US, respectively, but are still potentially significant (US EIA 2011). In 2012, the European Commission published three reports that addressed the potential risks and benefits of O&G development using hydraulic fracturing, with a focus on shale gas (Broomfield 2012; Foster and Perks 2012; Pearson et al. 2012). These reviews provided supporting information for development of a European Union (EU) regulatory policy for shale gas development. This policy was published in January 2014 (European Commission 2014). Prior to publication the regulatory framework for shale gas development was seen as critical for determining if investment will proceed and whether the environment and public health will be protected (Kibble et al. 2012). Rather than propose new legislation, the European Commission published a non-binding recommendation, communication, and impact assessment on the exploration and production of hydrocarbons using hydraulic fracturing in the EU. The adoption of a framework directive setting specific requirements for exploration and extraction remains a possibility. An EU wide regulatory framework would ensure a consistent regulatory environment for developers throughout the EU, while reassuring the public that it has clear and transparent environmental protection.

Although some EU member states are interested in developing their shale gas resources, others are not. Most notably, France has banned hydraulic fracturing. However, widespread activity is pending for the UK, with a strategic environmental assessment that aims to provide guidance in advance of additional onshore O&G licensing (UK DECC 2013). While proposed new licensing areas covers 50% of the UK, the proposed shale gas development for the entire UK is predicted at an upper limit of only 2880 wells. Experience in the US has, however, shown that initial estimates can be low compared to actual number of wells drilled (US BLM 2008).

Three recent reports in the UK have, to varying degrees, considered the air quality impacts of unconventional O&G. (UK DECC 2013; The Royal Society of Engineering 2012; MacKay and Stone 2013). The possibility of significant negative impacts was noted for greenhouse gas emissions. However, the UK's strategic environmental assessment (UK DECC 2013) did not directly address or even refer to air pollutants such as VOC, ozone or benzene. Human health impacts were not expected to be significant. In the US, policy is starting to catch up with development activity (US EPA 2012). The accelerated pace of drilling across the US has led to an ongoing reviews of Federal and State regulations related to O&G activities. A mandatory, clear, predictable, and coherent approach for the EU was not adopted in January 2014 (Broomfield 2012). Reporting protocols, assessment methods, and regulatory programs developed in the US could still be applied in the EU and elsewhere (Forster and Perks 2012; OECD 2012).

7.2 DISCUSSSION

7.2.1 Air quality research for the onshore oil and natural gas exploration and production sector in the United States

Through the development of quantification methods for estimating fluxes from basins and individual facilities, researchers in the US have improved the estimation of emission rates from O&G sources. The apparent disconnection of emission inventories, both for reporting and air quality modelling purposes, from measurement based studies remains problematic. A prime example is the inability of photochemical grid models, managed by the Wyoming Department of Environmental Quality, to replicate wintertime ozone episodes in the UGRB. While a similar problem for the Uintah Basin was resolved by Ahmadov et al. (2015) with significant revisions of model inputs (VOC increased by a factor of 2 and NO_x reduced by a factor of 4) a similar approach has not been applied in Wyoming. While respecting estimates of uncertainty, reconciliation between inventory calculations and measurement estimates is possible. This requires that inventory calculations are based upon accurate activity information, with suitable emission factors, and that measurement surveys are statistically representative of the area or activity considered.

At the basin scale research teams couple a variety of methodologies in order to develop an understanding of O&G emissions in the context of the particular situation. The Uintah basin, Utah has become a focus for research due to the relatively high emissions reported by Karion et al. (2013). In 2015 the JAGUAR project was performed, in conjunction with the SONGNEX project, to confirm that this basin had relatively high leakage and also to better understand particular O&G emission sources. This project was a collaboration between Utah State University (static monitoring station), University of Utah (balloon measurements and mobile surveys on roadways), Purdue University (airborne surveys and basin flux), Carnegie Mellon University (tracer release for large facilities), and the University of Wyoming (well pad Gaussian plume). Data validation is now completed and a number of publications are in preparation. A key finding is confirmation that the Uintah basin appears to have a higher leakage rate of CH_4 than other basins in the US (Robertson et al. 2017). Surveying of emissions from well pads revealed that a number of high emitting facilities accounting for a disproportionate amount of emissions. In the Uintah Basin gathering lines were also identified as significant, with a number of line breaks identified from surveying approaches.

By contrast to the JAGUAR study that was conducted independent from developers in 2015 a project entitled "Reconciling Basin-scale Top-down and Bottom-up Methane Emission Measurements for Onshore Oil and Gas Development" (RPSEA) was performed in full collaboration with developers. This project, funded by the US

Department of Energy, was designed to provide a definitive methodological approach for estimation of emission rates at the basin and facility scale for O&G sources. This project aims to develop independent estimates of methane emissions using top-down and bottom-up measurement approaches, at both the basin and facility level, and then to compare these estimates. This approach with independent sets of researchers for basin flux or mass balance (NOAA GMD), tracer release (Aerodyne) and Gaussian back trajectory (University of Wyoming). At the facility scale top-down approaches will be contrasted with bottom up approaches that measure emissions using source-level measurement techniques (e.g., high-flow meters). At the basin scale, the mass balance estimate will be compared with a bottom up estimate that is derived from scaling of facility scale measurements. The latter may include other appropriate estimates from similar studies where gaps exist in the types of facilities assessed. The final implementation of the project was within the Fayetteville shale, a dry gas play within a midcontinent US basin, predominately in the State of Arkansas. This development, with two major players, has an active multi-year LDAR program. A key preliminary finding is that the basin appears to have a lower leakage rate from O&G development than other basins in the US. And while the distribution is not as severe as other basins, a small number of high emitting facilities account for a disproportionate amount of emissions (Robertson et al. 2017).

The significant advances in understanding of the methane and VOC emission from O&G sources has relied heavily upon fast response and expensive equipment that requires highly trained research scientists. In the US there are ongoing research and development efforts for new instrument platforms, to support operators and regulators, such as the US Department of Energy's Advanced Research Projects Agency-Energy (APRA-E) program. A wide variety of projects funded through the ARPA-E's Methane Observation Networks with Innovative Technology to Obtain Reductions (E-MONITOR) program are supported as potential disruptive technologies that will transform the field of methane measurement and detection. The E-MONITOR program aims to deliver low-cost sensing systems that assist with reducing leaks anywhere in the natural gas system. As part of this effort, Colorado State University is developing a field test site that will simulate various aspects of the natural gas supply chain. This national research facility will allow researchers to test a wide range of technologies for enhanced methane sensing. The development of lower cost, novel and innovative technologies to monitor leaks is the key driver for this program. And given the high number of active and proposed O&G wells in the US, and concerns regarding unwanted losses impacting revenue streams and the environment, the development of "innovative technologies to cost-effectively and accurately locate and measure methane emissions associated with

natural gas production" is anticipated as an important avenue for meeting US climate action plan goals (ARPA-E, 2016).

There are a number of calls for proposals within the US for continued evaluations of the O&G sector. The US Department of Energy is supporting a number of on-going and proposed initiatives, including the University Coalition for Basic and Applied Fossil Energy Research and Development (UCFER) led by the National Energy Research Laboratory (NETL). The UCFER coalition aims to support research including reconciliation of emission rates for the O&G sector. Higher uncertainty with respect to emission factors is noted by NETL for water handling, hydraulic fracturing flow back, gathering pipelines, abandoned wells and distribution networks. Research funding in the US seeks to reduce the uncertainty of and improve the accuracy of the reporting of emissions from the O&G energy sector.

7.2.2 Regulatory approaches for the onshore oil and natural gas exploration and production sector in the United States

The US Climate Action Plan: Strategy to Reduce Methane Emissions published in March 2014 directed the US EPA to determine the best approach to control emissions from the O&G sector (US State Department, 2014). In April 2014 the US EPA published a series of technical white papers on significant emission sources and proposed mitigation techniques. The five white papers cover: Compressors; Emissions from completions and ongoing production of hydraulically fractured oil wells; Liquids unloading; Leaks; and Pneumatic devices. The first three white papers considered sources that were not covered under the US EPA's 2012 NSPS for VOCs (US EPA, 2012). The white paper for compressors notes that vented emissions typically increase with time due to degradation of mechanical components. Such emissions are greatly reduced with effective leak detection and repair (LDAR) programs. While some US states, e.g. Colorado, have requirements for reduced emission completions that avoid loses due to venting of gas, most US States do not. The white paper for completions outlines mitigation techniques that can be more widely applied to reduce emissions from this source throughout the US. Liquids' unloading refers to the processes that remove accumulated liquids in the well bore that reduce the flow of gas to the surface. This white paper presents approaches for mitigating the loss of VOC and methane from venting. The white paper for leaks covers the vast infrastructure systems that are required for production, processing, storage and transmission of natural gas. The white paper for pneumatic devices refers to controllers and pumps driven by high-pressure natural gas. These devices are widespread and release gas either continuously or with valve movements. A full regulatory impact assessment was published in May 2016 (US EPA 2016a).

In August 2015, the US EPA proposed updating the 2012 NSPS and in May 2016 a final NSPS rule (US EPA, 2016b) was published along with permitting rules: Source Determination Rule (US EPA, 2016c). A draft Federal Implementation Plan for Indian Country (US EPA, 2015c) is still under review. The final NSPS 2016 rule took account of ~900,000 public comments. In addition to these rules an initial Information Collection Request was published in March 2016 (US EPA, 2016d) with an additional request published in September 2016 (US EPA, 2016e). In August 2016 a voluntary request for information on innovative technologies to detect, measure and mitigate emissions was published (US EPA 2016f). Together the US climate Action Plan and the Clean Air Act aim to further reduce emissions of methane, VOCs and air toxics from new, reconstructed and modified sources while providing greater certainty regarding permitting requirements. The Source Determination Rule is important as it clarifies when multiple pieces of equipment and activities must be deemed a single source when determining whether major source permitting programs apply, or not. O&G developments are often associated with multiple minor sources that individually are small, but when conglomerated can account for a significant proportion of emissions. This rule defines adjacent equipment and activities that are under common control as a single source. To be considered as a single source adjacent equipment and activities are required to be located on the same site or sites that share equipment and are within ¼ mile of each other. While the Source Determination Rule applies primarily to permits issued by the US EPA, State and local permitting authorities may also adopt these changes.

The Information Collection Requests (US EPA 2016d; US EPA 2016e) are designed to gain information to inform the development of future regulations to further reduce emissions from existing sources. In recent years, through Greenhouse Gas Reporting Provisions and numerous studies, emissions from existing sources are often significantly higher than previously understood. As such further information is essential to reduce knowledge gaps and inform effective development of standards for existing sources. The inclusion of existing sources would result in a considerable increase of facilities that would be required to meet emissions standards. The latter is significant as older sources are regulated under less strict emission requirements. Information is requested on how emission controls are, or can be, configured together with the associated costs. And given the diversity of operations and set-up, operators are required to provide detailed facility descriptions that are representative of their onshore operations. Information is requested from each segment of the natural gas supply chain with reporting through Greenhouse Gas Reporting tools. The broad request for information by the US EPA includes natural gas venting that occurs as part of existing processes or maintenance work, e.g. well and pipeline blow-downs, equipment malfunctions and flashing emissions

from storage tanks. These sources were highlighted as the estimated level of emissions is poorly understood.

The 2016 NSPS defines costs and benefits for emission reductions, and in addition to covering hydraulically fractured wells, considers other activities including processing, transmission and storage that were not included the 2012 NSPS rules. Proposed emission limits for methane can be achieved using technologies that are cost effective, and readily available. And given that the types of sources controlled by 2012 NSPS for VOC, these controls already affect methane emissions. Besides applying to additional sources, an important new requirement is that owners and operators must find and repair leaks. While many operators have long established LDAR programs, up to now this has been a voluntary action, unless required by the State. Surveys are now required on a fixed schedule of twice a year at well pads and four times a year at compressor stations (gathering, boosting and transmission). In addition to using optical gas imaging operators are allowed to use alternative approaches.

A number of emission reduction requirements are being phased in, e.g. "Green completions" (also known as Reduced Emission Completions) to allow operators time to meet the new NSPS 2016 requirements. Reduced emission completions are estimated to reduce emissions of methane and VOC by 95% (US EPA 2016a). The main benefits by 2025 of the NSPS rules are the anticipated pollution emission reductions: 510,000 short tons of methane; 210,000 short tons of VOCs; and 3,900 short tons of air toxics. While the benefits of the reduction of VOC and air toxics were not quantified the US EPA estimates that climate benefits of $690 million outweigh the estimated costs of $530 million (US EPA 2016a). The final NSPS 2016 rule is designed to provide a pathway for operators to show that any current State requirements are comparable. The emission control requirements for State programs vary considerably across the US. Some States, e.g. Wyoming and Colorado, have already instigated emission control requirements. Under the Clean Air Act, States have the authority to regulate air emissions from sources within their boundaries, provided their requirements are not weaker than Federal rules. The US EPA final Control Technology Guidelines for reducing VOC emissions from existing sources in ozone nonattainment areas and for States in the Ozone Transport Region (US EPA 2016g). States will have to address sources contained in these guidelines as part of their State Implementation Plans (SIPs) for meeting National Ambient Air Quality standards. These guidelines are not regulations, and as such do not impose any legal requirements rather, they provide recommendations for state and local air agencies as they develop emissions limits for sources in their jurisdictions in order to meet Reasonably Available Control Technology (RACT) requirements. Together these US EPA actions (NSPS rule, Permitting rules and Control Technology Guidelines) are

designed to ensure that the US Administration meets its goal of cutting methane emissions from the O&G sector by 40 to 45% below 2012 levels by 2025.

The reductions from the new NSPS 2016 rule will be supplemented by those from the US EPA's new Natural Gas STAR: Methane Challenge Program and through actions by other Federal agencies, e.g. the Downstream Initiative and the One Future Initiative (US EPA, 2016h). Further engagement between regulators and developers are anticipated as rules are developed for existing sources. Further Federal regulation of the O&G sector proposed by the Bureau of Land Management (BLM) are under legal challenge (US BLM, 2016). The BLM is proposing to update its regulations to reduce emissions, and waste of natural gas, from flaring, venting, and leaks from O&G production on public and Indian lands. The proposed revisions update rules that are outdated with respect to current technological approaches in the industry. The Mineral Leasing Act requires that the BLM ensures that developers use "all reasonable precautions to prevent waste of oil or gas." Currently there is no upper limit on how much gas can be flared. And as such, some oil-rich developments, e.g. Bakken shale in North Dakota, flare huge quantities of gas due to a lack of natural gas delivery pipelines. While flaring is preferable to venting, combustion efficiencies are dependent upon a number of factors including wind speed (Leahey et al. 1995). The proposal would phase in progressively stricter limits over a period of three years. It is anticipated that production operators could meet the new requirements through a number of approaches: expand capture and delivery of natural gas to existing infrastructure; add alternative on-site capture infrastructure; and/or slow production until adequate infrastructure for transport of gas is achieved. The proposal for reducing flaring also requires metering when volumes reach 50 Mcf/day. The BLM proposal also includes provisions for pre-drilling planning for gas capture, as currently there is no such requirement. Before drilling a well, operators would need to prepare a waste minimisation plan that would be submitted with a Drilling Permit Application. Proposed BLM Leak detection requirements are similar to those proposed through the US EPA NSPS 2016 rules, and are already required in Wyoming and Colorado. A further provision is providing guidelines and requirements to reduce of venting of natural gas. Operators would be required to replace "high bleed" pneumatic controllers with "low bleed" versions within one year. Operators would also be required to replace suitable pneumatic pumps with solar powered pumps, and/or to re-route pump flow to control devices. Operators of new wells would no longer be allowed to vent to the atmosphere. And unloading liquids should follow best management approaches to reduce emissions. Control of emissions from completions is similar to US EPA NSPS 2016 rules and requires that gas is captured, used, flared or re-injected rather than released to the atmosphere. If the US EPA NSPS 2016 rules proceed then the BLM rule would apply to conventional

wells as the former only applies to hydraulically fractured well completions and re-completions.

The US Inventory of Greenhouse Gas Emissions and Sinks: 1990-2014 published in February 2016 provides a platform for better reconciliation of emission estimates with atmospheric observations (US EPA, 2016i). This inventory contains a number of updates from new and improved data that has become available through the US EPA's Greenhouse Gas Reporting Program and from numerous studies conducted by government, academic and industry organisations. The publication in 2015 of Greenhouse Gas Reporting Data by the US EPA for data up to 2014 has provided a benchmark for subsequent revision and improvement (US EPA, 2015d). This supports the goals outlined in the US Administration's 2014 Methane Strategy to improve the completeness, quality, accuracy and transparency of data, while improving the ability of regulators and other stakeholders to better understand emissions, trends and air quality impacts.

7.3 CONCLUSIONS

- Research and regulatory oversight of emissions from the O&G energy sector in the US are primarily driven by impacts upon climate change, human health and ozone production.

- Research projects led by a wide variety of organisations in the US are improving our understanding of emission sources within the entire lifecycle of unconventional O&G resources, namely: Production; Gathering and Processing; Transmission; and Distribution.

- While there is progress through field validation campaigns emission inventories managed by State agencies are often unable to replicate actual emission amounts derived from atmospheric measurements.

- Future O&G related air quality research in the US is anticipated to focus upon emission sources with the greatest uncertainty, namely: water handling; hydraulic fracturing flow back; gathering pipelines; abandoned wells; and distribution networks.

- A wide variety of measurement based approaches are being applied in the US to support emission detection and quantification efforts. The US Department of Energy's Advanced Research Projects Agency-Energy (ARPA-E) program is actively developing disruptive technologies to transform the field of methane measurement and detection.

- O&G emissions in some Western US states have caused pollution episodes that have led to the US EPA to declare some areas as not meeting (nonattainment) the National Ambient Air Quality Standard for ozone.

- The US Climate Action Plan: Strategy to Reduce Methane Emissions published in March 2014 directed the US EPA to determine the best approach to control emissions from the O&G sector.

- In May 2016 through provisions of the Clean Air Act the US EPA updated New Source Performance Standards for the O&G industry. These actions, together with permitting rules are anticipated to significantly reduce emissions.

- US EPA actions are designed to ensure that the US Administration meets it's goal of cutting methane emissions from the O&G sector by 40 to 45% below 2012 levels by 2025.

- Further Federal regulation of the O&G sector proposed by the Bureau of Land Management (BLM) are under legal challenge. The BLM is proposing to update its regulations to reduce emissions, and waste of natural gas, from flaring, venting, and leaks from production on public and Indian lands.

- The US Inventory of Greenhouse Gas Emissions and Sinks is being improved through updates from new and improved data that has become available through the US EPA's Greenhouse Gas Reporting Program and from numerous studies conducted by government, academic and industry organisations.

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List of abbreviations, acronyms and definitions

A_i	intercept: regression parameter of Eq. 1
Ace	acenaphtene
Acethln	acenaphthylene
Anth	anthracene
ARPA-E	Advanced Research Projects Agency-Energy
$B_{i,j}$	regression parameter of Eq. 1
BLM	Bureau of Land Management
BTEX	Benzene, Toluene, Ethyl-Benzene, m,p,o-Xylenes
C_4-C_6	Hydrocarbons from 4 to 6 carbon number
C_{5+}	Hydrocarbons with chain of 5 carbons or longer
C_{11}-C_{22}	Hydrocarbons from 11 to 22 carbon number, Heavy Hydrocarbons.
$C_{i,j}$	concentration of a pollutant, j, at the sampling point, i.
CC	Converse County
CH_4	Methane
CO_{2GMD}	Carbon dioxide
e	Napierian number
EC	European Commission
EDGAR	Emission Database Global Atmospheric Research
EIA	Energy Information Administration
EIS	Environmental Impact Statement
E-MONITOR	Methane Observation Networks with Innovative Technology to Obtain Reductions
EPA	Environmental Protection Agency
EU	European Union
Flu	fluorene
Fluthn	fluoranthene
GAO	Government Accountability Office
GMD	Global Monitoring Division
GPS	Global Positioning System
H	High (production level)
HAPs	Hazardous Air Pollutants
HHCs	Heavy Hydrocarbons
JAGUAR	Joint Air and Ground based Uintah Basin Air emissions Reconciliation project
JRC	Joint Research Centre
L	Low (production level)
LDAR	Leak testing and Leak Detection and Repair
M	Medium (production level)

Mcf	Millions cubic feets
1+2MePhe	1+2-methyl-phenanthrene
3+4MePhe	3+4-methyl-phenanthrene
MSD	Mass Spectrometer Detector
NAAQS	National Ambient Air Quality Standards
NESHAP	National Emission Standards for Hazardous Air Pollutants
NETL	National Energy Research Laboratory
NMHC	Non Methane Hydrocarbons
NOAA	National Oceanic and Atmospheric Administrations
NO_x	Nitrogen oxides
NPL	National Physical Laboratory
NPL project	Normally Pressured Lance project
NSPS	New Source Performance Standards
OECD	The Organisation for Economic Co-operation and Development
O_3	Ozone
O&G	Oil and Gas
PAHs	Poly Aromatic Hydrocarbons
P_i	Gas, oil or water production for a particular buffer well (i)
Phe	phenanthrene
PMF	Positive Matrix Factorisation
PoD sampler	Pocket Diffusive Sampler
Pyr	pyrene
REC	Reduced Emissions Completions
SONGNEX	Studying the Atmospheric Effects of Changing Energy Use in the US at the Nexus of Air Quality and Climate Change
Tcf	Trillions cubic feets
TDU	Thermal Desorber Unit
UB	Uintah Basin
UBWOS	Uintah Basin Winter O_3 Study
UCFER	University Coalition for Basic and Applied Fossil Energy Research and Development
UGJF	Upper Green Jonah Field
UGPA	Upper Green Pinedale Anticline
UGRB	Upper Green River Basin
UT	Utah
US	United States
VOCs	Volatile Organic Compounds
WDEQ	Wyoming Department of Environmental
WSD	World Geodetic System

Z   Zero to marginal (production level)

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