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Pre-Exploration Measurement and Monitoring of Background Landscape Methane Concentrations and Fluxes in the Beetaloo sub-Basin, Northern Territory

Dry Season Baseline Methane Concentrations:

Interim Report for GISERA Project G5

Cindy Ong, Matthew Myers, Mederic Mainson, Bruce Maney and Stuart Day

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Interim Report for GISERA Project G5 - Pre-Exploration Measurement and Monitoring of Background Landscape Methane Concentrations and Fluxes in the Beetaloo sub-Basin, Northern Territory

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Executive Summary

The Scientific Inquiry into Hydraulic Fracturing in the Northern Territory submitted its final report to the Northern Territory Government in August 2018. Among other things, the Inquiry recommended that baseline monitoring of methane be undertaken ahead of the granting of exploration approvals for unconventional gas. Specifically, Recommendation 9.3 of the Inquiry's final report states:

> *That baseline monitoring of methane concentrations be undertaken for at least six months prior to the grant of any further exploration approvals. In areas where hydraulic fracturing has already occurred, the baseline monitoring should be undertaken at least a year prior to the grant of any production approvals.*

In response to this recommendation, the Northern Territory Government commissioned CSIRO through GISERA to conduct baseline methane monitoring in the Beetaloo Sub-basin which is a prospective gas production region in the NT.

Measuring and monitoring methane emissions in natural gas regions is currently an active area of scientific research throughout the world and as yet there is no standard method. In recent years, a significant amount of research has been conducted in the coal seam gas regions of Queensland and NSW (Day et al., 2015; Day et al., 2016; Ong et al., 2017; Etheridge et al., 2018) to help develop appropriate methodology and includes the establishment of fixed monitoring stations, mobile surveys using gas analysers mounted in vehicles or aircraft, and other ground-based measurements.

The aim of the project described in this interim report is to provide:

- background landscape concentrations of methane in the Beetaloo Sub-basin and
- investigate methane emission rates (fluxes) and identify the sources of any elevated methane levels found.

This work is another important step in the application and research of improved methane measurement and monitoring methodology.

It is important to recognise that *concentration* is a measure of the abundance of methane in air, usually defined in terms of the proportion of the total volume it accounts for in air (units are often parts per million, ppm, or parts per billion, ppb). Emission rate or *flux* is defined as the rate of flow of methane from the source. The emission rate may be expressed as a volumetric flow (in m^3 per unit time) or mass flow (in g or kg per unit time). Both concentration and emission rate data are required for baseline studies since areas of elevated methane concentrations help in locating and identifying sources while emission rates yield the amount of methane being released to the atmosphere.

This project (GISERA Project G5) represents the first step of a larger methane baseline programme with the objective of acquiring baseline measurement and monitoring data of methane concentrations across a larger area of the Beetaloo Basin. This work provide a comprehensive baseline of background methane emissions across this landscape against which the impact of unconventional gas development can be assessed.

Gas exploration activities are due to begin seeking approvals in the sub-basin early in 2019. Due to this short timeframe, the first task of Project G5 required the most practical and quickly deployable methods to obtain as comprehensive as possible background methane concentrations across the central region of the Beetaloo sub-basin over the six-month period from July 2018. This meant that initial monitoring relied mainly on mobile ground surveys using a vehicle equipped with suitable methane analysers. The establishment of fixed site monitoring techniques is more challenging and requires a significantly longer lead time to install reliable monitoring equipment in remote locations. However, fixed monitoring methods will be considered in later stage projects.

In this report, we present the finding of the first surveys conducted over a 12-day period during the dry season between the 29th July and the 10th August 2018. Additional surveys are planned for the region in November 2018 for the fire season and January to cover the wet season.

Surveys of atmospheric methane concentrations within the Beetaloo Sub-basin region were made using mobile surveys with three gas analysers mounted in a four-wheel-drive vehicle. Although methane was the principal gas of interest, CO_2 and ethane were also measured during many of the surveys to assist with identifying the source of emissions. Ethane in particular is useful for identifying emissions from gas production facilities or natural methane seeps since it is often a minor component of natural gas. The use of three analysers also provided a level of redundancy in the event of equipment failure in the remote and harsh operating conditions of the survey region. In addition, one of the instruments (the AERIS analyser), a newly developed commercially available analyser, has a compact design and low power consumption. The AERIS was trialled to assess its potential for use in remote location fixed monitoring stations in future monitoring programmes.

The vehicle surveys covered approximately 5,500 km on trafficable roads and tracks during the 13-day survey period. Between 200 and 600 km were driven each day. Many of the tracks were very rough and often obstructed by vegetation which limited access. In these areas of the survey region, the methane concentration data were spatially sparse compared to other areas where vehicle access was better. Most of the surveys were conducted on pastoral land, crown land and exploration leases, however, targeted surveys were made at 11 plugged and abandoned or suspended petroleum wells, and 21 water bores. A section of the Daly Waters to McArthur River Gas Pipeline adjacent to the Carpentaria Highway was also surveyed for the presence of methane.

Overall, the majority of methane concentrations recorded during the surveys were within the range of 1.77 to 1.85 parts per million (ppm) which is close to normal background concentrations of approximately 1.8 ppm expected in rural or natural areas (see for example, Ong et al., 2017). Isolated pockets of slightly elevated methane concentrations were observed in some areas; the sources of these were identified as:

- grazing cattle
- urban areas
- small grassfires
- a natural spring
- a section of above-ground gas pipeline and associated valves.

Most of the elevated methane levels detected were in the vicinity of grazing cattle. The methane elevations were quite small, with the maximum concentrations less than 2.6 ppm (i.e. about 0.8 ppm above background). Moreover, the concentration peaks were of short duration since the vehicle quickly moved past the source reflecting the small size of these sources. Although individual cattle represent a minor source of methane (about 100-200 g CH_4 per beast per day, up to a maximum of 300 g/day), collectively, cattle production is a significant contributor of methane. Estimating the amount of methane emitted from a dispersed source such as open range cattle is difficult using ground surveys. Instead we used an alternative approach based on herd numbers for the Amungee, Beetaloo, Barkley and Tanumburini stations within the region and published methane emission factors for Australian cattle. This yielded an emission rate of approximately 40 kg CH_4 min^{-1} across the entire survey region.

Some other instances of elevated methane concentrations of between 1.85 to 2.09 ppm were recorded at townships along the Stuart Highway. These levels are similar to concentrations measured in other towns and cities, and usually relate to the use of natural gas, vehicle emissions, fuel storage, landfills and wastewater treatment activities associated with urban areas. Flux estimates were not made for these sources during this campaign but because of the very small population density, are expected to comprise a very minor component of the total methane budget of the survey region.

Grass fires were found to be sources of methane during the survey with methane concentrations between about 2.1 and 2.8 ppm measured on two occasions. These observations, which were only slightly above background, were made immediately adjacent to the roadside fires indicating the fires were small sources of methane. Methane emission rates from large fires (which are common in the region at some times of the

year) are likely to be more significant but challenging to quantify using mobile surveys. It is often not possible to fully access fire plumes as is required for some quantification methods that rely on plume dispersion characteristics. Further investigations into alternative methods that provide better spatial coverage incorporating remote sensing data could potentially provide more quantitative flux estimates for bushfires. This is currently being investigated for future work.

A survey of the wetlands surrounding the Mataranka and Bitter Springs was conducted to understand the contribution of this potential natural source of methane to the survey area. No elevated methane concentration was detected along access tracks leading to the springs, however, slightly increased methane levels approximately 0.3 ppm above background (i.e. 2.1 ppm total methane concentration) were observed close to running water at the spring itself. Ethane was not detected at this site suggesting that the methane emission was of biogenic origin. Further measurements of this site are planned for the wet season to determine if emissions show seasonal variability.

Slightly elevated methane concentrations up to approximately 2.3 ppm were detected about 10 m downwind of some valves on an above-ground section of the Daly Waters to McArthur River gas pipeline. Ethane was also detected at this location (maximum concentration was 0.035 ppm), which was highly correlated with methane indicating that the methane was indeed from natural gas and that the emission source was most likely the pipeline or valves. Flux measurements of the probable source were not possible at the time of the survey due to restricted access (the site was fenced off) but the very low levels of methane detected would suggest the emission rate was low. No other methane sources were detected along this length of pipeline. Nevertheless, it is still important to collect more comprehensive measurements closer to the pipeline during future surveys to provide a comprehensive baseline of such potential sources of methane.

No elevated methane concentrations were detected at the petroleum wells and water bores that were specifically investigated during this field campaign.

Finally, a series of measurements were made at eight termite mounds ranging in size from about 20 cm to 1.6 m tall. Termites are known to be sources of methane in tropical regions; however, no elevated methane concentrations were detected near the mounds during these surveys. Some internal measurements of methane levels within the mounds were conducted and found to be relatively high with concentrations up to 100 ppm, yet this did not appear to be escaping from the mounds. Despite the apparent lack of methane emissions from the small sample of termite mounds on this occasion, the results are consistent with previous studies which has found the methane emissions were low during the dry season but substantially higher emissions during the wet season. Further work will be required to better understand the methane emission characteristics of termite mounds in the Beetaloo sub-basin and their seasonal variation. This will be investigated during further survey planned over the course of the project.

1 Introduction

The Northern Territory Government's 'Scientific Inquiry into Hydraulic Fracturing' Final Report requires that methane measurement and monitoring occur before the advent of exploration and production activities by gas companies (Andersen et al. 2018). Specifically, Recommendation 9.3 of the Final Report requires:

> '*That baseline monitoring of methane concentrations be undertaken for at least six months before the grant of any further exploration approvals. In areas where hydraulic fracturing has already occurred, the baseline monitoring should be undertaken at least a year before the grant of any production approvals.'*

This project (GISERA Project G5) specifically addresses the first component of this Recommendation that refers to the measurement and monitoring of 'methane concentrations' before the granting of exploration approvals.

For clarity, concentration is in this report mean a measure of the abundance of gas (in this case methane (CH_4)) in the air, defined as the proportion of the total volume it accounts for (units are parts per million (ppm) or parts per billion (ppb)). Flux or emission rate is defined as the rate of flow of gas per unit time (for example in m^3 per unit time on a volumetric basis or in g or kg per unit time on a mass basis). Both measurements are required to be able to provide a baseline and quantify the natural and anthropogenic CH_4 emissions, identify where these background emissions are occurring and how much CH_4 is being released to the atmosphere.

The objective of this project is to quantify the landscape background concentration levels of CH_4 and identify sources for locations where elevated CH_4 levels are found and, where applicable, quantify the fluxes related to these sources.

This interim report documents the findings of the first survey conducted over a 13-day period between 29th July – 10th August 2018 where CH_4 concentrations in the region were measured using gas analysers mounted in a four-wheel-drive vehicle. A total of 5,500 km were traversed during this survey, and CH_4 measurements were acquired continuously along the route. Data collected during this survey provides an indication of the background CH_4 concentrations during the dry season of 2018, sources of CH_4 emissions and some preliminary estimates of the source fluxes.

2 Experimental Method

CSIRO and other research organisations have been actively conducting research into CH_4 emissions from the unconventional gas industry over many years. Techniques now available for monitoring and quantifying emissions include mobile surveys using vehicles or aircraft equipped with CH_4 monitoring instruments in combination with plume modelling or tracers; and, fixed flux towers (e.g. eddy covariance methods), bulk atmospheric concentration measurements or remote sensing technology combined with inverse atmospheric transport models. Some methods such as those using inverse modelling and fixed monitoring stations, require a substantial investment and long lead-time to establish, making them more suited to longer monitoring campaigns. A system using two fixed stations and modelling methods was used over a three-year period to monitor regional emissions in the Surat Basin in Queensland as part of a GISERA project (Etheridge et al., 2018).

In the current project, however, time was limited by the need to complete the first set of monitoring measurements before the wet season. Moreover, exploration applications are expected from industry early in 2019. Consequently, more rapidly deployable methods were necessary for this study and therefore mobile surveys were the principal method used during this stage of the project.

Mobile survey methods are one of the most widely used, reliable and well-developed techniques for undertaking baseline measurements of landscape CH_4 concentrations and fluxes and have been used in Australia, the United States and United Kingdom (Ong et al., 2017; LTE, 2007; Phillips et al., 2013; Zazzeri et al., 2015). Deployment of mobile surveys over time allows for accurate monitoring of CH_4 emission concentrations and fluxes under conditions which preclude immediate deployment of *in situ* monitoring stations. Mobile surveying can also be advantageous in remote regions of Australia where difficult access and lack of reliable power and communication networks make installation of long-term fixed monitoring stations challenging.

Mobile survey methods employ high sensitivity CH_4 analysers suitable for accurately measuring small changes in ambient CH_4 concentrations and have proven effective for the quantification of a range of CH_4 sources including gas wells (both abandoned and operational), gas processing facilities, landfills, wastewater plants, water bores and natural geological seepages in studies completed by CSIRO in Queensland, New South Wales, Western Australia and Victoria (Day et al., 2015; Day et al., 2016; Ong et al., 2017).

The instruments deployed in this study were based on cavity ring down spectroscopy (CRDS), off-axis integrated cavity output spectroscopy (OA-ICOS) and direct absorption tunable diodes – these systems are described in more detail in Section 3 of this report.

Apart from the speed at which mobile vehicle surveys can be conducted in a region, there are several other advantages with the technique. First, there is the capacity to travel over many thousands of kilometres to enable broad-scale measurement programs to be undertaken. Second, the mobility of the system also allows detailed surveys of areas to be conducted to locate and identify CH_4 sources. Thirdly, in some cases, emission flux estimates may be made if the ground concentration data are combined with local meteorological data and a simple plume dispersion model. This approach was used to measure CH_4 emissions from coal seam gas wells in Queensland and NSW (Day et al., 2014).

Disadvantages, however, include the fact that for vehicle based surveys, monitoring is limited to trafficable roads and tracks and, like most ground based monitoring methods, surveys are dependent upon favourable wind conditions (i.e. the vehicle must be downwind of the source). Also surveys are periodic, not continuous so do not capture long term temporal variations in emissions.

3 Instrumentation

This survey was performed using three CH_4 analysers:

- Picarro G2301 $CO_2/CH_4/H_2O$ analyser, which is a cavity ringdown spectrometer.
- Los Gatos Research (LGR) Ultraportable $C_2H_2/CH_4/H_2O$ Methane/Acetylene Gas Analyzer (acetylene can sometimes be used as a tracer gas in experiments designed to measure emission rates from some methane sources). The LGR is an OA-ICOS system.
- AERIS Technologies PICO analyser, utilising tuneable diodes.

All three instruments are capable of reliably detecting changes in CH_4 concentration as low as 2 ppb and have the high levels of stability necessary for mobile operation (e.g. Crosson, 2008).

As well as measuring CH_4, the Picarro instrument simultaneously measures CO_2 concentrations. This instrument also has the capability of measuring the ratio of $^{13}C/^{12}C$ in both CH_4 and CO_2. Isotopic ratios can in some cases provide information on the origin of the source of CH_4, provided concentrations are sufficient for reliable measurements.

The LGR measures CH_4 and C_2H_2 (acetylene) simultaneously. Acetylene is often used as a tracer when determining emission fluxes from some sources, but this method was not used during this field trip. If suitable CH_4 sources are located during subsequent campaigns the tracer method may be employed to measure fluxes.

The AERIS analyser measures CH_4 but also simultaneously measures ethane (C_2H_6) which is often present in unconventional gas and hence the presence of C_2H_6 may indicate thermogenic sources (Yacovitch et al. 2014).

Each analyser had its own dedicated GPS receiver (Garmin 18X, Hemisphere R330 DGPS and Gill Maximet GMX500, respectively) to enable positional data to be recorded simultaneously with the gas concentration data. Local meteorological data (wind speed and direction, temperature and humidity) were measured with the Gill Maximet GMX500 Compact Weather Station fitted to the top of the field vehicle (Figure 1). Note that wind speed and direction measurements were made only when the vehicle was stationary. Positional data were combined with the gas concentration data to produce maps of CH_4 concentration across the study region.

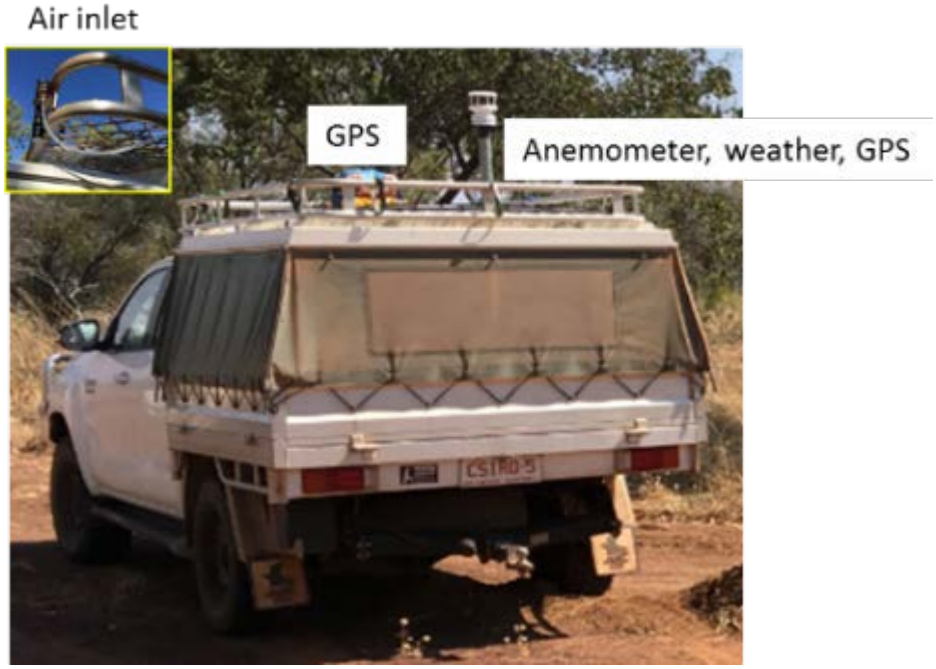

Figure 1: Survey vehicle showing the location of sampling lines, GPS and combined anemometer, GPS and weather station.

The survey vehicle is shown in Figure 1. The GPS for the LGR analyser were located on the roof rack at the front middle of the vehicle, and the Gill combined anemometer, weather station and GPS were also located on the roof rack at the front right-hand side of the vehicle. The inlet of the sampling line was located on the roof rack (see insert on the top left of Figure 1) on the front left-hand side of the vehicle. The location of the inlet was such that it was not affected by the vehicle's exhaust while travelling. The same sampling line was used for all three analysers and during surveying air was drawn from the front of the vehicle to the rear of the vehicle via a single pump. The air stream from the inlet line was then distributed via three separate lines to each analysers located at the tray of the ute (shown on Figure 2). The distances between the three distribution lines were small and the length of lines between the intake and each analyser was approximately equal.

All three instruments were initially mounted in the covered tray of the vehicle as shown in Figure 2; however due to high ambient temperatures (~ above 30 °C on some days), the instruments tended to overheat in the enclosed canopy during the first three days of operation. The analysers were therefore relocated to the rear seat in the vehicle's cabin for subsequent surveys, which successfully prevented this problem during those days.

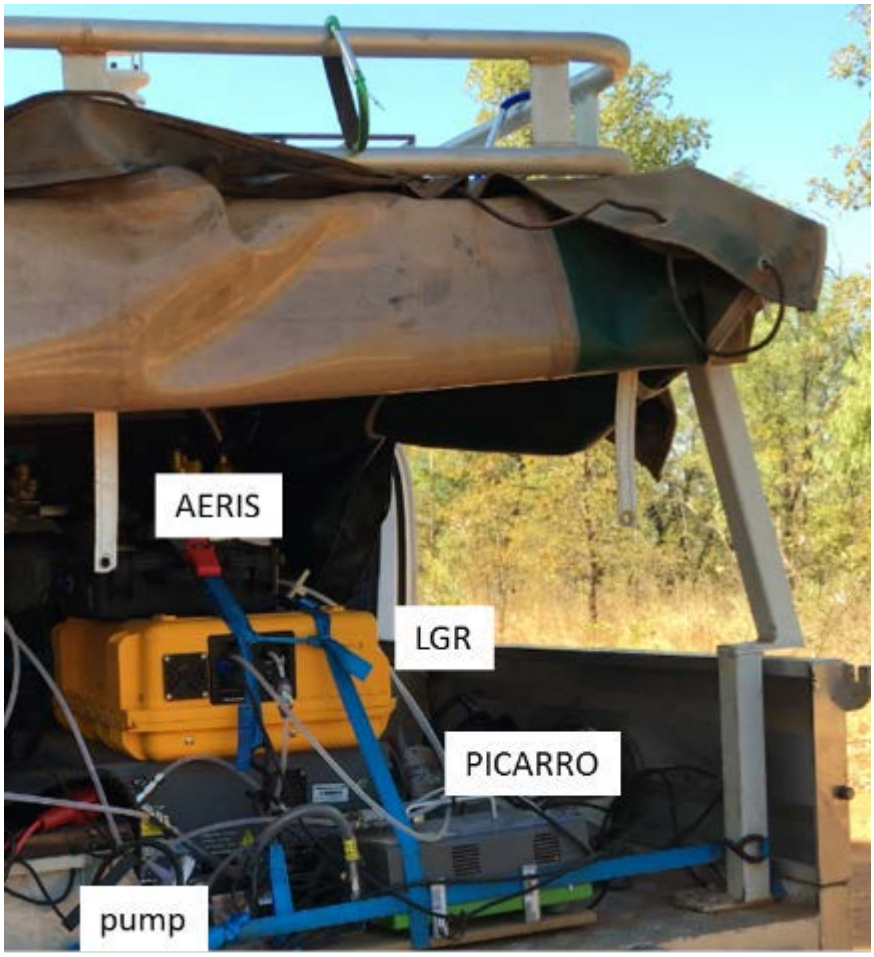

Figure 2: PICARRO, LGR and AERIS analysers mounted in the rear of the field vehicle before being relocated to the cabin.

4 Data Processing

The main data processing performed on the data collected during this survey include:

- correction of the measured concentration data to account for daily drift in the analysers;
- alignment of each of the respective data to a common reference to account for the different response time of each analyser; and
- where there were gaps in one of the dedicated GPS for one of the analysers to fill these in with the GPS data that were acquired simultaneously.

Before commencing the field programme, each analsyer was subjected to a multi-point calibration using up to five reference gasses in the CSIRO Kensington laboratories in Perth. The calibration standards contained CH_4 at concentrations ranging from approximately 1.8 ppm (close to ambient air concentration) up 102 ppm (similar to the maximum CH_4 concentration measured during the field campaign). All instruments were linear over this range.

As well as the laboratory calibrations, each instrument was checked at least once each day during the field surveys to account for any drift. These performance checks were made using two reference standards (containing nominally 4 and 20 ppm CH_4). Concentration data from each day were then corrected for drift by applying a correction factor scaled from the daily performance checks against the two reference gasses and assuming that instrument response remained linear over the measurement range (Equation 1).

$$yy = mmmm + cc \qquad \text{Eq 1}$$

In Equation 1, *y* is the corrected concentration, *m* is the gradient determined from the calibration line derived from the two daily calibration reference gasses, *x* is the measured (raw) concentration, and *c* is the intercept from the calibration line.

Table 1 and Table 2 below presents the gradients (m) and intercepts (c) for the daily calibration equations determined for the LGR and the PICARRO analysers. The table shows that both instruments were stable during the survey after the first three days as indicated by the small between change in the gradient and intercept between each consecutive days. In the first three days, large differences were experienced by both analysers due to the high temperatures in the tray of the ute where the analysers were located. The maximum temperatures during the period of the survey ranged from 26.8 – 31.8°C and the relative humidity ranges were 16-62% at 9 am and 9-30% at 3 pm (BOM 2018a, b). However, the temperatures under the closed canopy of the ute were significantly higher so that the analysers were sometimes operating above their maximum normal operating temperatures. For example, the temperature of the gas recorded from the LGR was significantly higher than the maximum recommended operating temperature of 45 °C (see Figure 3). The relocation of the analysers to the back seat mitigated this temperature effect, and this configuration will be used in future surveys.

Table 1: Gradients (m) and intercepts (c) for each date for the straight line calibration equations of the form y = mx + c applied to the LGR data where y = the adjusted measurements and x is the raw measurements. Note that calibration data were acquired only for one of the first three days.

LGR Date	29/07/ 2018	30/07/ 2018	31/07/ 2018	1/08/ 2018	2/08/ 2018	3/08/ 2018	4/08/ 2018	5/08/ 2018	6/08/ 2018	7/08/ 2018	8/08/ 2018	9/08/ 2018
CH4 m	1.000	1.000	1.000	1.005	1.002	1.006	1.006	1.006	1.005	1.005	1.006	1.007
CH4 c	0.006	0.006	0.006	-0.003	-0.004	-0.003	-0.005	-0.004	-0.001	-0.002	-0.006	-0.004

Table 2: Gradients (m) and intercepts (c) for each date for the straight line calibration equations of the form y = mx + c applied to the PICARRO data where y = the adjusted measurements and x is the raw measurements.

PICARRO Date	29/07/2018	31/07/2018	1/08/2018	2/08/2018	3/08/2018	4/08/2018	5/08/2018	6/08/2018	7/08/2018	8/08/2018	9/08/2018
CH4 m	0.981	0.989	0.989	0.989	0.989	0.989	0.988	0.989	0.989	0.989	0.988
CH4 c	0.020	-0.022	0.015	0.013	0.015	0.013	0.015	0.014	0.016	0.014	0.011
CO2 m	0.927	0.938	0.935	0.934	0.935	0.934	0.934	0.934	0.932	0.935	0.934
CO2 c	26.417	20.398	23.585	24.187	23.505	24.134	24.294	24.097	24.804	23.781	23.996

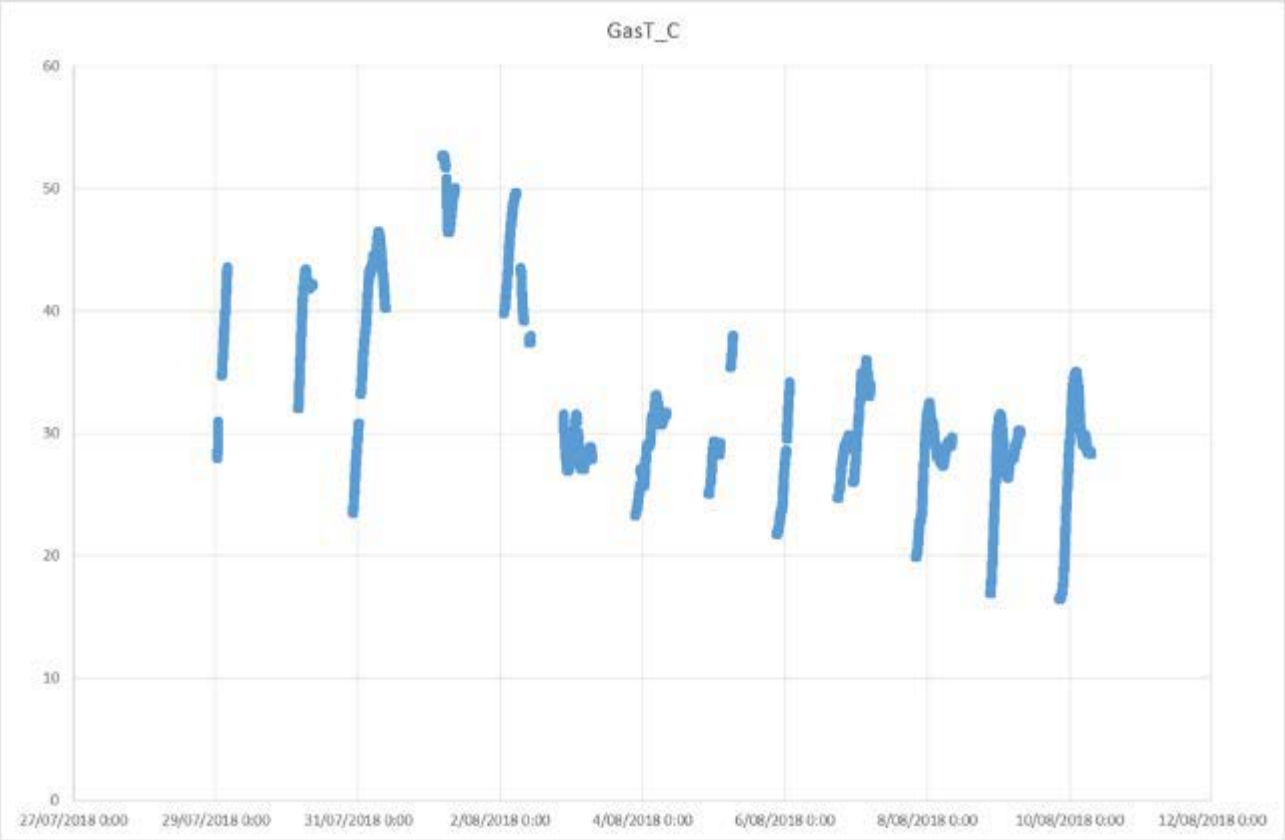

Figure 3: Gas temperatures recorded by the LGR during the survey period. Note that the units of the y axis are in °C.

The response time of each analyser is a function of the size of the sample cell within the instrument, the flow rate of the gas into the analyser which in turn is a function of the pump capacity and size of the inlet valve into the analyser. An example of the differing instrument response time is shown in the Results section of this report (see Figure 9). As a consequence, when the survey vehicle was moving, CH_4 peaks detected by each instrument appeared to be spatially offset. Time corrections were applied to each instrument's results to ensure CH_4 peaks were properly aligned.

Although there were three GPSs used, on some occasions, the GPS dedicated to the LGR failed to provide a signal. Hence it was necessary to link the LGR measurements to the other GPS units. This was performed by synchronising the time stamp on the LGR data to the other GPS's time stamp.

5 Results

A total of approximately 5,500 km was traversed during the period 29th July – 10th August 2018. Typically, between about 200 and 600 km was covered each day. The area of coverage was guided by the gas operators Pangaea, Origin and Santos along tracks that were trafficable by 4WD at the time of the survey, and spans across areas that are likely to be explored in the next 6-12 months. The routes traversed were predominantly on private tracks on pastoral leases across the respective gas operators' leases and lay mainly within the Beetaloo sub-basin. It is important to note that the tracks were not of equal spatial densities across the entire area. For example, across the pastoral leases closest to the Stuart Highway towards the East and West, the tracks were spatially sparse and many of the tracks were covered by substantial amounts of regrowth.

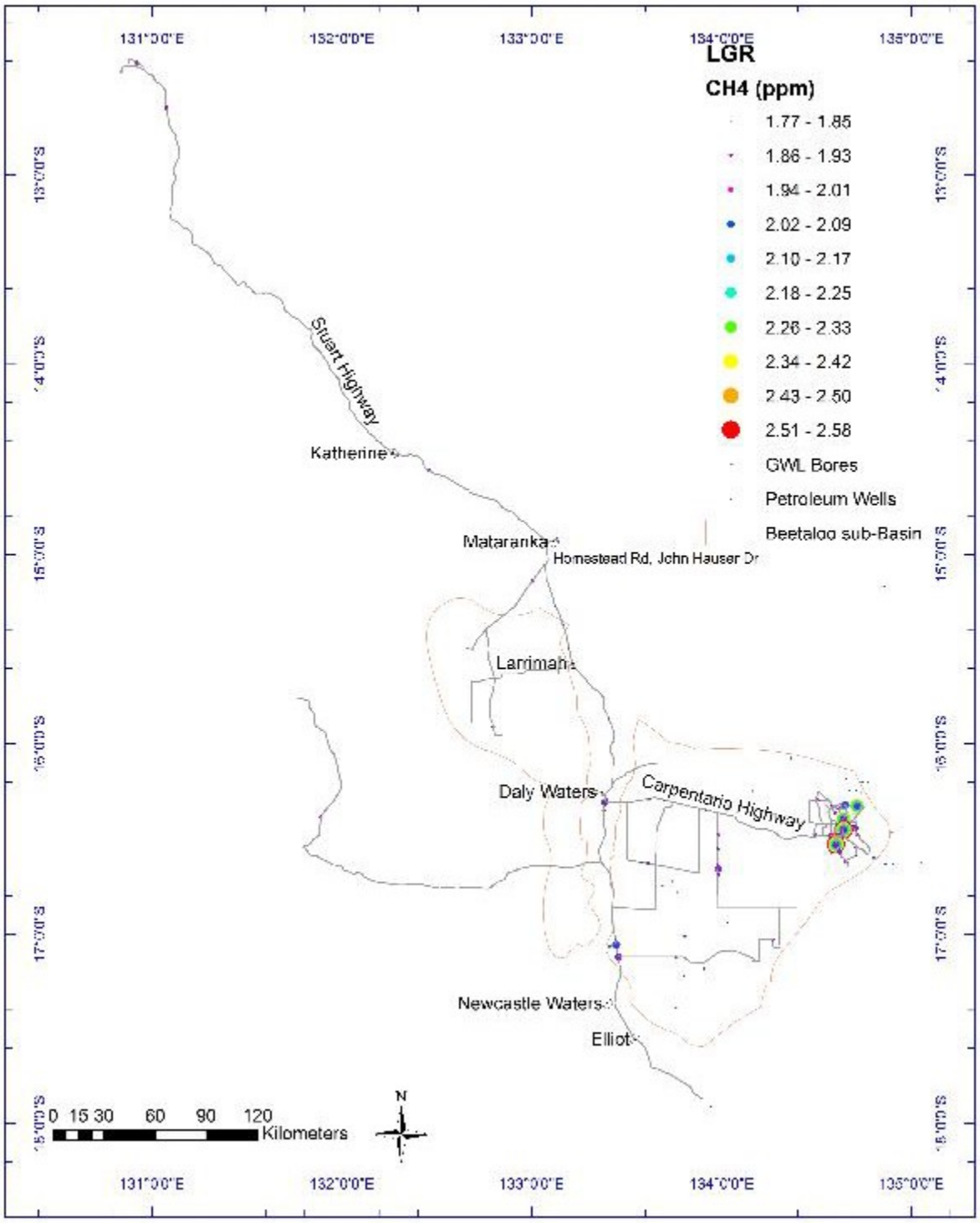

Figure 4: CH_4 concentration measured using the LGR analyser along tracks and roads across the Beetaloo sub-basin.

In contrast, the tracks were spatially denser across the areas further west along the Carpentaria Highway. Many of the tracks were also similarly obscured by regrowth. In addition to private tracks, where possible, data were collected along public roads such as state and national highways surrounding the lease areas.

Figure 4 shows a map which represents a summary of the CH_4 concentration measurements collected by the LGR across the area traversed during the entire survey. The CH_4 concentration (in ppm) is colour coded, and the markers are sized relative to the gas concentrations. The bulk of the data shows a concentration level between 1.77-1.85 ppm. These concentrations would be representative of the natural ambient/background

levels across the study area. As a guide, these values are equivalent to or below the global CH_4 level of 1.85 ppm (Dluugokencky 2018).

Across the survey, there were pockets of elevated concentrations of CH_4 measured. The main sources of these elevated levels were predominately cattle, townships and to a small degree fire.

Throughout the survey, most of the elevated concentrations measured could be attributed to cattle when the vehicle passed herds of cattle on the side of the tracks travelled. This was the case for the bulk of the elevated levels detected along the west and east of the Stuart Highway. For example the largest number of elevated concentrations were detected furthest west on the Santos lease area, each of which can directly be attributed to herds of cattle passed along the tracks travelled.

The other main sources of elevated CH_4 concentrations detected were townships as indicated at Katherine, Mataranka and Daly Waters along the Stuart Highway (Figure 4). The sources of these elevated values were likely to be attributed to commonly known anthropogenic sources at townships such as fuelling stations, seepages of natural gas from domestic or commercial usage, vehicles or sources such as sewerage/waste treatments and landfills. For example, at Daly Waters, the main sources of the elevated levels may have been due to the fuel station and the sewage system, although we were not able to confirm this at the time of the survey.

The other elevated CH_4 concentrations that are visible in the regional scale displayed in Figure 4 can be attributed to fires. These two sources located north of Newcastle Waters along the Stuart Highway were detected on the 4th of August 2018 and were small grass fires on the side of the highway similar to the burnt area shown in the photograph Figure 7. They were detected while the vehicle was stationary on the side of the highway allowing the small plumes to be detected. The concentration of CH_4 detected by the PICARRO and LGR is shown in more detail in Figure 5. There is a good correlation between the concentrations measured by both analysers.

CO_2, which is the main gaseous composition of bushfires (Urbanski et al. 2008) was also detected and showed similar trends to the CH_4 (also in Figure 5) However, there was not a good overall correlation indicating that CO_2 concentrations are not necessarily a good surrogate for CH_4. The lack of correlation may be due to incomplete combustion zones within the fire which produce CH_4 (and maybe C_2H_6) whereas complete combustion does not produce any minor gasses (only CO_2 and water). In addition, as the fire is on the side of a road any passing vehicles would contribute to the total CO_2 detected.

In addition to CO_2 and CH_4, another major gaseous composition of bush fires is ethane (C_2H_6) (Urbanski et al. 2008). The AERIS analyser was able to detect C_2H_6 at these fires, which was possibly a product of incomplete combustion. The left graph on Figure 6 shows the concentration of CH_4 and C_2H_6 detected at one of these fires. The chart on the right plots the relationship between CH_4 and C_2H_6 indicating that the two gaseous compositions detected were highly correlated.

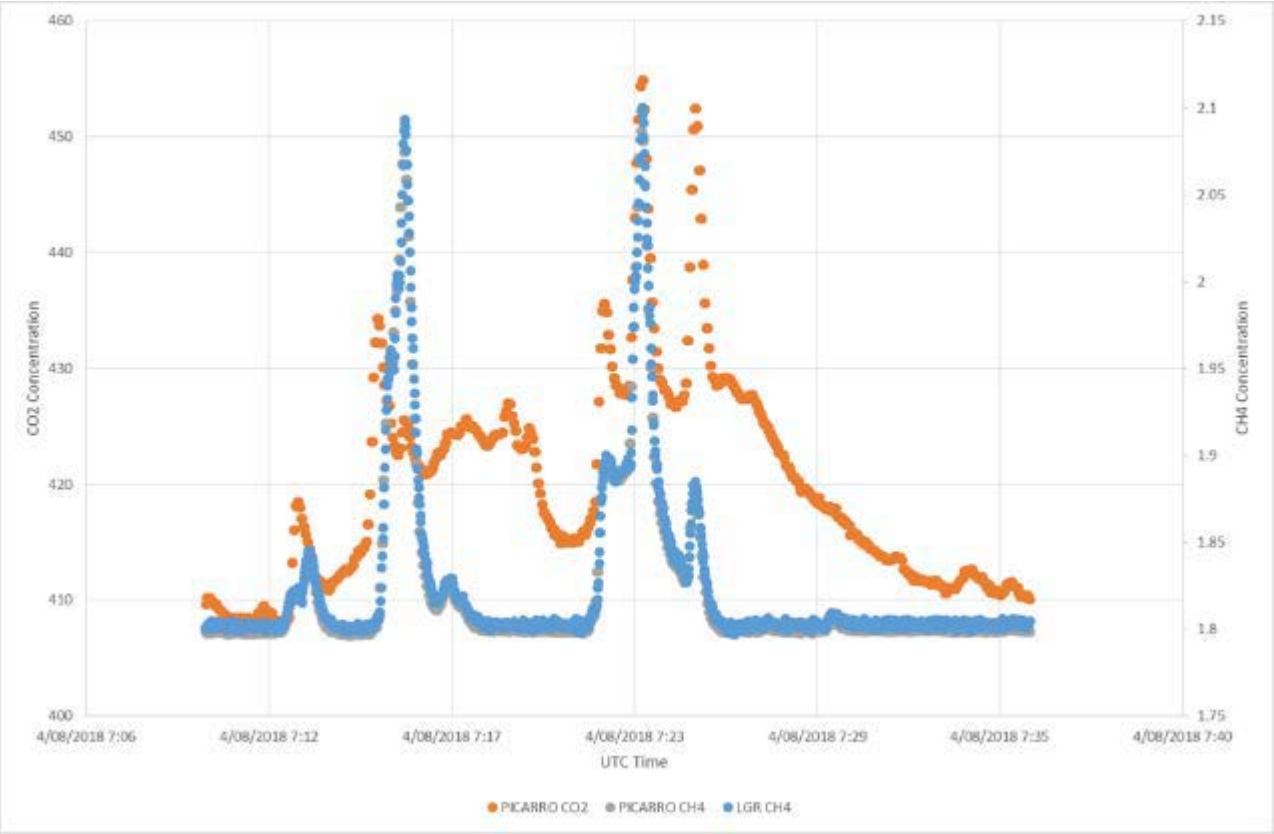

Figure 5: Concentration of CH_4 and CO_2 detected by the PICARRO and LGR analysers at a roadside grass fire on 4 August 2018.

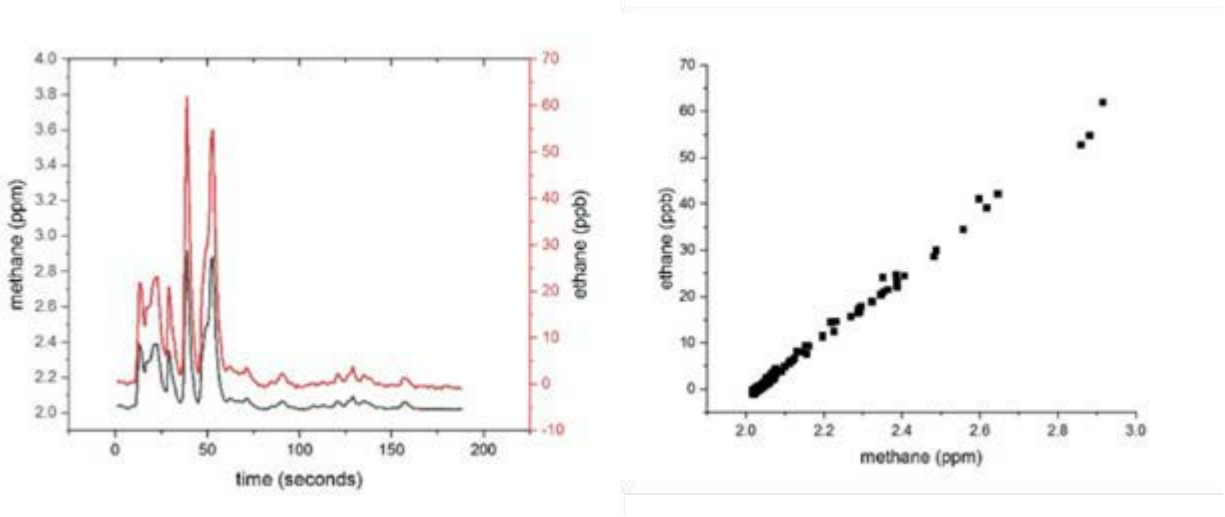

Figure 6: Left: CH_4 and C_2H_6 detected at roadside grass fires on 4 August 2018. Right: Correlation between CH_4 and C_2H_6.

Figure 7: Burnt grass at the side of a road.

Another fire detected during the survey on 8 August 2018 was a larger bushfire and methane was detected for an extended period of half an hour along the Stuart Highway. During this period, small elevated values of approximately 300 ppb above the background (i.e. total CH_4 concentration was approximately 2.1 ppm) were detected as shown in Figure 8. The bushfire was nearby and the conditions during this period were hazy.

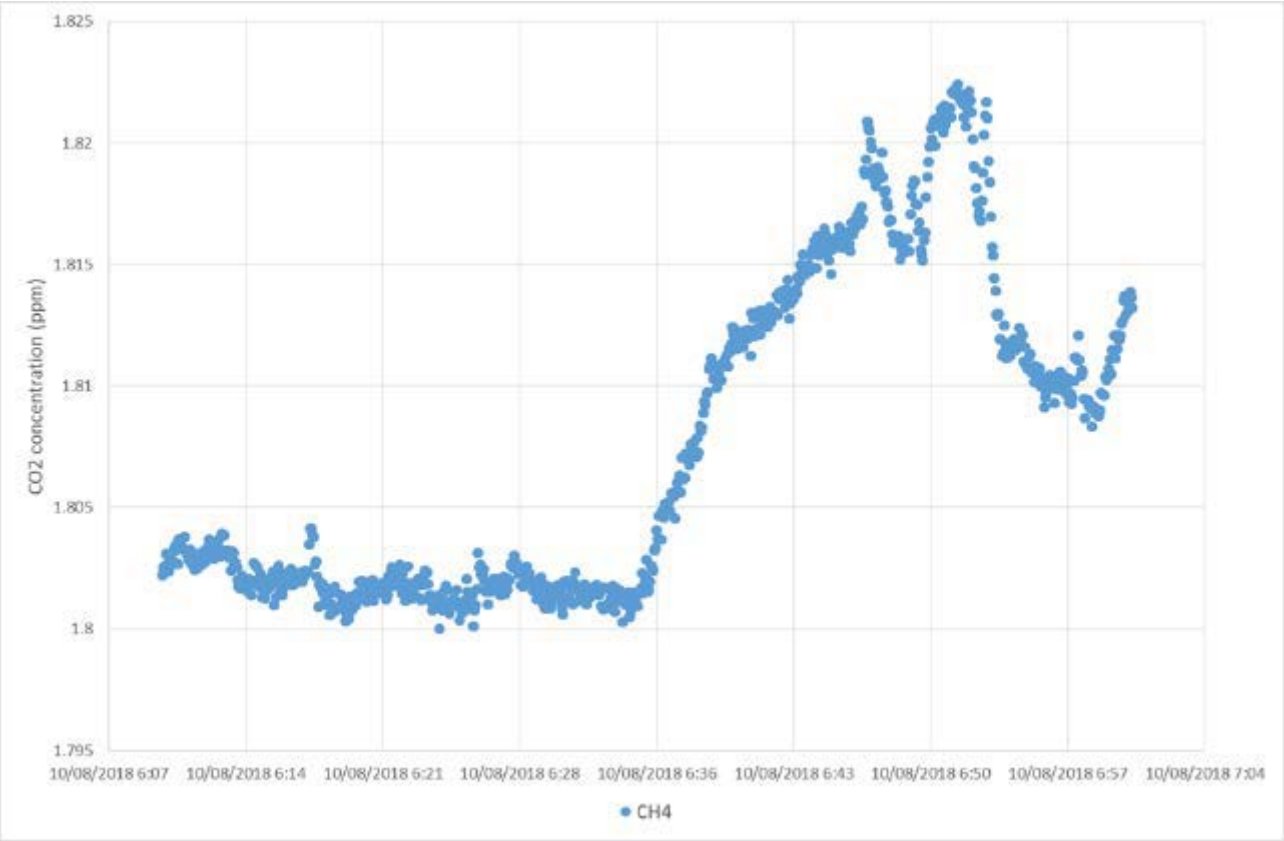

Figure 8: CH_4 concentration measured by the PICARRO analyser while driving across a bushfire plume(s) on 8 August 2018.

Figure 10 shows the CH_4 concentration measurements collected by the PICARRO analyser during the survey. There is good agreement between Figure 4 (LGR results) and Figure 10 where similar background concentrations were detected and areas where the sources were static such as at extended periods at townships, similar concentrations were measured by both analysers. However, for a more dynamic source such as those produced by cattle, the concentrations vary by a small amount. For example, the highest value detected throughout the survey can be attributed to cattle at the far western side of the survey at 2.6 ppm as detected by the LGR analyser. At this location, the PICARRO detected a peak value of 2.3 ppm. This difference is a function of the different response time of the LGR and PICARRO, and, the dynamic nature of the plumes. In addition, the moving vehicle is another factor contributing to the small 300 ppb difference.

The response time difference related to a herd of cattle is illustrated in Figure 9. This example illustrates that the Picarro (blue line) has a slightly slower response time to the LGR (red line) but in general the profiles are similar. The AERIS (black line) shows a somewhat different profile due to its very fast response time that allows higher resolution of transient CH_4 peaks.

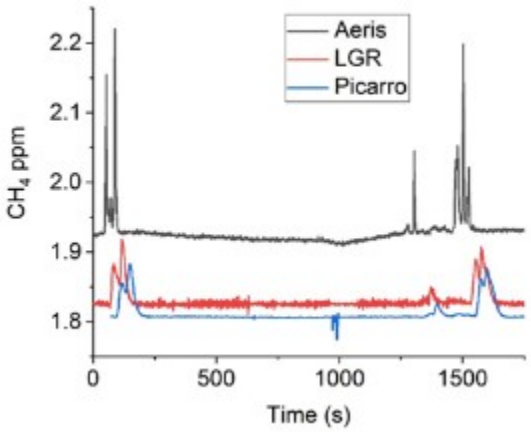

Figure 9: CH_4 detected by the PICARRO, LGR and AERIS analysers before realignment for respond time difference.

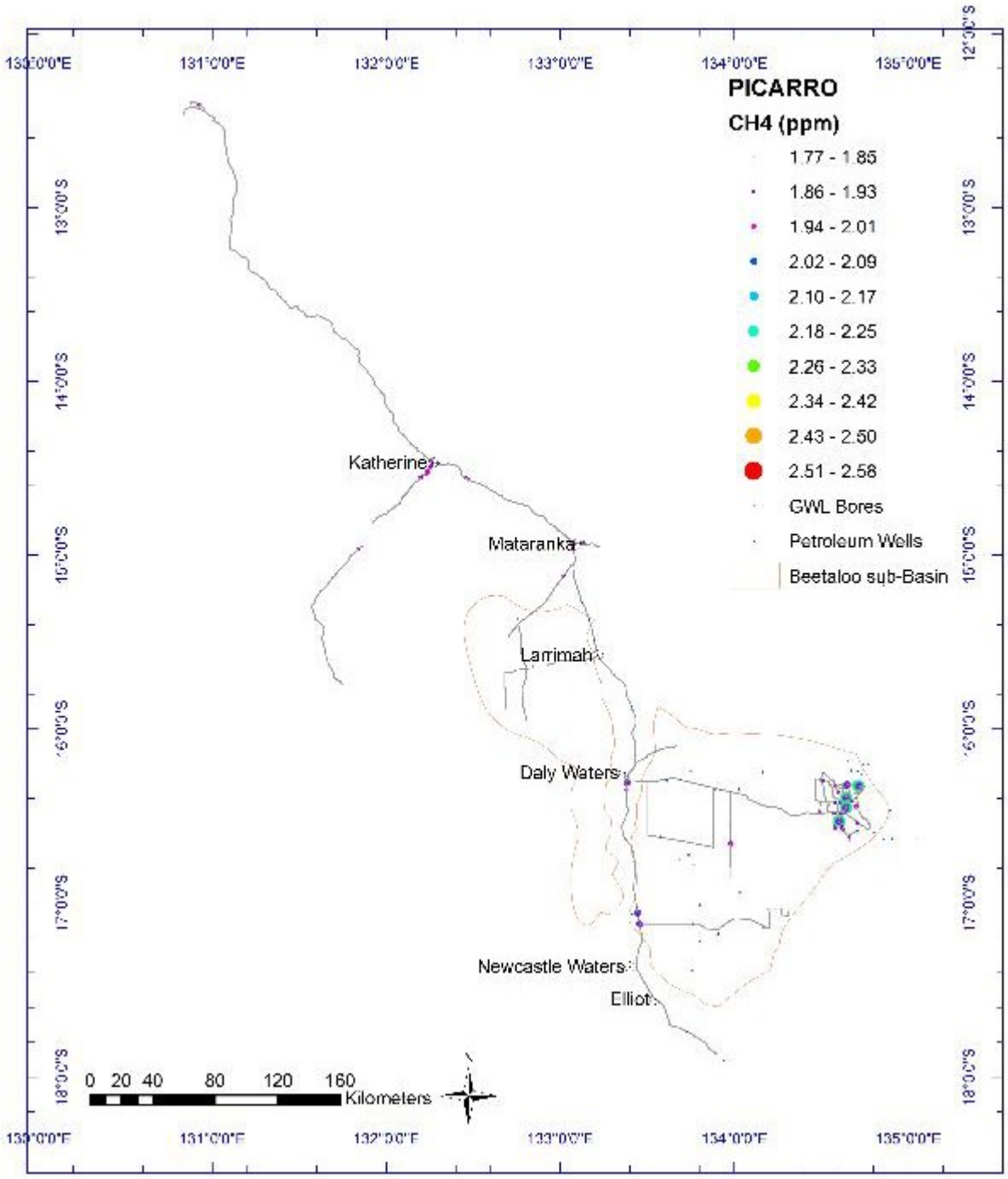

Figure 10: Methane concentration measured using the PICARRO analyser along tracks and roads across the Beetaloo sub-basin.

During the surveys, a total of 11 plugged and abandoned and suspended wells which are potential sources of methane, were visited (or were close to pastoral tracks where measurements were made). Details of these wells are shown in Table 3. No elevated CH_4 concentrations were detected at any of the well sites visited.

Table 3: List of plugged and abandoned and suspended petroleum wells visited during the survey.

Well ID/Name	Latitude, Longitude (locations mostly shown on Figure 4, Figure 10 and Figure 20)	Comments
1. Wyworrie 1	15°22'31"S,132°43'56"E	Plugged and abandoned (PNA)
2. Tarlee 1	15°57'16"S,132°50'23"E	PNA
3. Tarlee 2	15°53'32"S,132°41'4"E	PNA
4. Tarlee S3	15°37'55"S,132°49'36"E	PNA
5. Birdum Creek 1	15°37'50"S,133°8'35"E	PNA (see Figure 11)
6. Kalala S1	16°17'38"S,133°36'49"E	Suspended
7. Amungee	16°20'50"S,133°53'4"E	Suspended
8. West Beetaloo 1	17°7'14"S,133°45'42"E	Suspended (see Figure 12)
9. Shenandoah 1	16°37'11"S,133°34'44"E	PNA
10. Burdo	16°15'4"S,134°30'37"E	PNA, historical 1980s bore
11. Tanumburini 1	16°23'57"S,134°42'14"E	Drive along pastoral track close to bore?

Figure 11: CH_4 measurements were acquired at the Birdum Creek plugged and abandoned well site.

Figure 12: Collecting CH_4 and wind measurements at the West Beetaloo 1 suspended well.

In addition to the petroleum wells, methane concentration measurements were made at (or close to) 21 water bores during the survey. Details of the water bores are shown in Table 4.

Elevated concentrations were not detected in the vicinity of most of these bores. At a small number of these bores (marked with * in Table 4), elevated values were detected, but there were also cattle close by. It was not possible to discriminate between methane produced by the cattle and those that may be seeping from the water bores. However, as the size of the herd was usually quite large, cattle is likely to be the dominating influence. Elevated concentrations were consistently recorded over a period of five days close to the Daly Waters Motel (marked with #). As this bore is only approximately 250 m away from a septic tank, it is likely that the septic tank is a confounding influence. In addition, the fuel station is also less than 500 m away from the water bore which may have influenced the results.

Although elevated methane concentrations were measured, the elevation above background in the vicinity of less than 1 ppm (generally below 2 ppm total concentration measured) collected approximately 500 m from the well did not warrant flux measurements to be undertaken.

Table 4: Water bores investigated

Water Bore Name	Latitude, Longitude (locations mostly shown on Figure 4, Figure 10 and Figure 20)	Comments
1. Water Bore close to Tarlee 2	15°57'21"S,132°39'5"E	Measurement collected at bore close to Tarlee 2.
2. Jabiru	16°57'25"S, 134°18'22"E	Measurement collected at bore at Amungee Station (see Figure 13)
3. Motel Bore#	16°18'28"S, 133°23'9"E	Measurement collected approximately 500 m from bore at Daly Waters Motel
4. Unnamed water bore at Beetaloo Station	16°41'27"S, 132°58'52"E	Measurement collected close to bore
5. RN029012	15°16'16"S, 133°8'32"E	Drive along Stuart Hwy close to the bore
6. RN038810	15°22'24"S, 133°9'55"E	Drive along Stuart Hwy close to the bore
7. RN038811	15°29'23"S, 133°11'42"E	Drive along Stuart Hwy close to the bore
8. RN028082	15°35'43"S, 133°13'34"E	Drive along Stuart Hwy close to the bore
9. RN029013	15°16'16"S, 133°8'32"E	Drive along Stuart Hwy close to the bore
10. RN005942	15°16'16"S, 133°8'32"E	Drive along Carpentaria Highway close to bore along Carpentaria Highway
11. RN005764	15°17'35"S, 133°36'44"E	Drive along Carpentaria Highway close to bore along Carpentaria Highway
12. RN5844	15°20'51"S, 133°54'54"E	Drive along Carpentaria Highway close to bore along Carpentaria Highway
13. RN38109	16°33'8"S, 133°58'48"E	Drive along pastoral tracks close to the bore
14. RN037655	16°28'58"S, 134°33'59"E	Drive along pastoral tracks close to the bore
15. RN033608*	16°27'22"S, 134°38'53"E	Drive along pastoral tracks close to the bore
16. RN039693	16°29'9"S, 134°38'11"E	Drive along pastoral tracks close to the bore
17. RN038179	16°25'28"S, 134°36'7"E	Drive along pastoral tracks close to the bore
18. RN008101	16°24'57"S, 134°40'27"E	Drive along pastoral tracks close to the bore
19. RN007659*	16°19'42"S, 134°42'48"E	Drive along pastoral tracks close to the bore

20. RN033671*	16°23’8”S, 134°38’1”E	Drive along pastoral tracks close to the bore
21. RN031244	16°35’14”S, 134°42’27”E	Drive along pastoral tracks close to the bore

Figure 13: CH_4 measurements were collected at the Jabiru water bore at Amumgee Station.

One of the sources of natural sources of CH_4 expected in the area are wetlands and natural geological sources such as springs. A survey was conducted along the public roads: Homestead Rd and John Hauser Rd to quantify the possible CH_4 from the wetlands area around Bitter Springs and Mataranka Springs. No elevated concentrations were detected along these roads. However, the locations of the springs were some distance from the road. As the AERIS analyser is less than 3 kg, a walking survey was undertaken with it along the path to Mataranka Springs. No elevated values were detected along the paths into Mataranka Springs, but elevated CH_4 concentrations of up to 300 ppb above background were detected at Mataranka Springs (Figure 14) and small elevated CH_4 concentrations were also detected at Rainbow Springs. This was not correlated to the C_2H_6 concentrations which were below 10 ppb (Figure 15).

Figure 14: Location where CH_4 measurements were recorded with the AERIS analyser.

Previous research indicates that where the CH_4 and C_2H_6 ratios are low or uncorrelated, the sources are likely to be of a biogenic nature (Yacovitch et al. 2014).

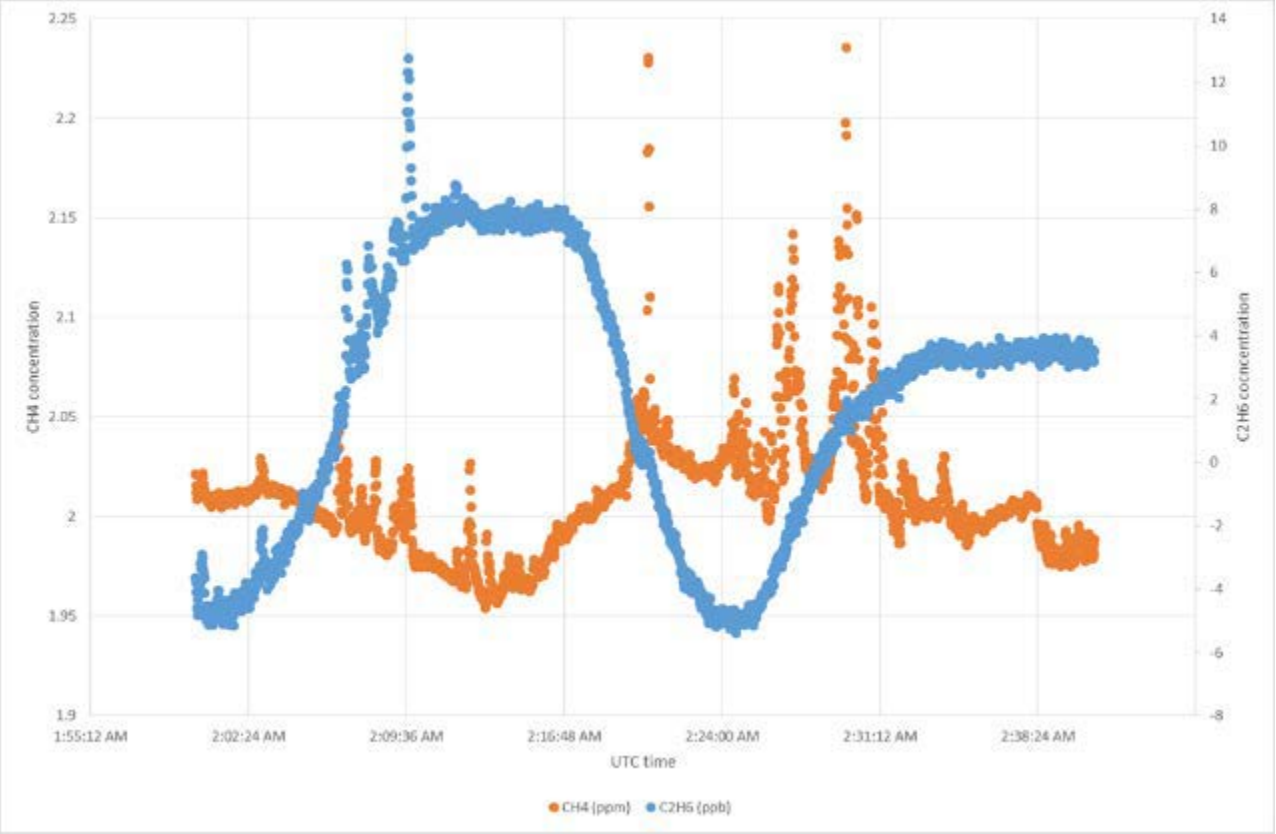

Figure 15: CH_4 and C_2H_6 concentrations recorded at Mataranka Springs.

The other likely natural source of CH_4 are termites. A total of 8 termite mounds were measured during the survey. These were located across the survey area; close to Mataranka Spring, close to Daly Waters and at Tanumbirini Station and ranged in size from a small enough to fit a 9 litre flux chamber (see Figure 16) to a large mound approximately 1 × 1.6 m in size similar to the one shown on Figure 17. For mounds small enough to fit into a 9 or 20-litre flux chamber, measurements were made with the mounds enclosed within the flux chamber. With larger mounds, the flux chamber was attached to the side of the mounds. No, or very small, elevated concentrations were recorded for all the mounds when the flux chamber was used. However, larger concentrations were measured when the tubing was inserted into the side of the mounds. The highest value measured when this was done was in excess of 100 ppm as shown on the left chart in Figure 18. This figure also shows the concentration of C_2H_6 which in contrast is very low (approximately 8 ppb). The CH_4 and C_2H_6 concentrations measured in the mound are plotted in the right hand graph in Figure 18 indicating that there is no apparent correlation between these measurements. The lack of correlation suggests that the CH_4 is unlikely to be thermogenic in origin.

Figure 16: Small termite mound measured with a 9-litre flux chamber.

Figure 17: Large termite mound measured at the side with a 20-litre flux chamber (middle photo) and with the tubing inserted into the side of the mound (left photo).

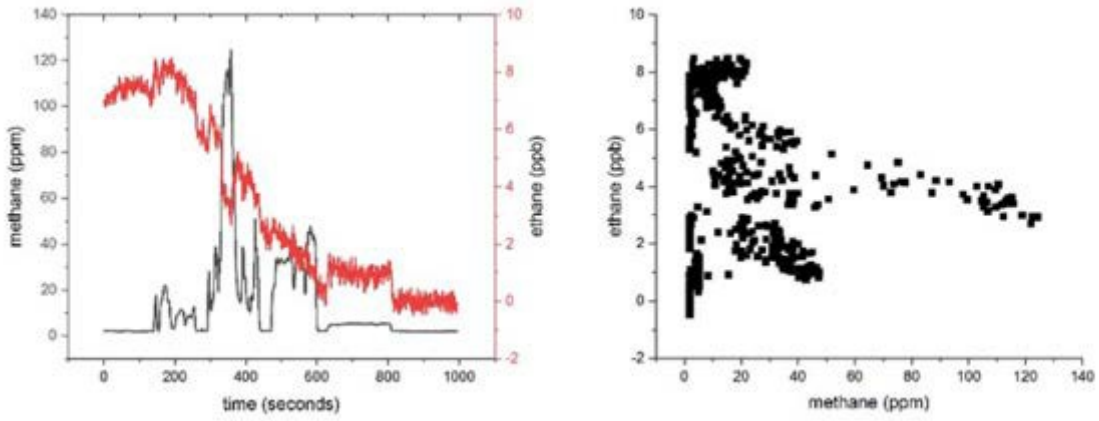

Figure 18: Left: Concentration of CH_4 and C_2H_6 recorded by the AERIS analyser when tubing was inserted into a large termite mound. Right: Concentration of CH_4 plotted against C_2H_6 indicating that the two gasses were not correlated.

During regional survey, CH_4 measurements were also made alongside a section of the underground Daly Waters to McArthur River Gas Pipeline as it is adjacent to the Carpentaria Highway. For all of the survey along the highway, no elevated CH_4 concentrations were detected. However, small elevated concentrations up to approximately 2.3 ppm CH_4 were detected at an above ground section of the pipeline adjacent to tracks covered on the Tanumburini station. The measurements were collected next to the fence approximately 10 m from the pipeline. This section of pipeline also had several valves attached. Figure 19 shows the concentrations of CH_4 and C_2H_6 measured by the AERIS analyser on the left and the right-hand graph shows the high correlation between the CH_4 and C_2H_6 indicating that the source of the CH_4 is likely to be from the natural gas in the gas pipeline. It is possible that the small amount of CH_4 detected near the pipeline was leaking from one of the valves close to the measurement location.

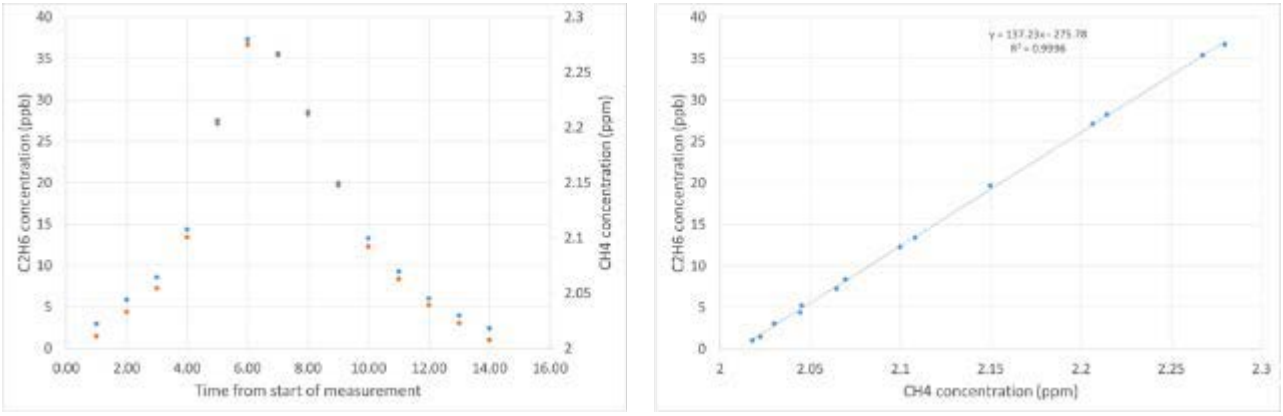

Figure 19: Left: CH_4 and C_2H_6 concentration measured at an above ground section of the Carpentaria gas pipeline. Right: Correlation between the CH_4 and C_2H_6 concentrations measured at the gas pipeline.

Figure 20 shows the CO_2 concentrations recorded by the PICARRO analyser for the entire survey. Generally, the majority of the elevated concentration can be attributed to CO_2 produced at townships or vehicle-related CO_2 while driving behind another vehicle. This can be clearly seen on the road away from Larrimah and leading to Mataranka where there exhaust from other traffic were vehicles in front of the survey vehicle resulted in higher CO_2 concentrations. Additionally, the largest concentrations were measured at Daly Waters, which is a major truck stop. Although it is sometimes possible to attribute specific sources of CO_2, such as the fires on the side of the road described above, many of the elevated CO_2 levels encountered across the survey, were confounded by CO_2 produced by nearby vehicles.

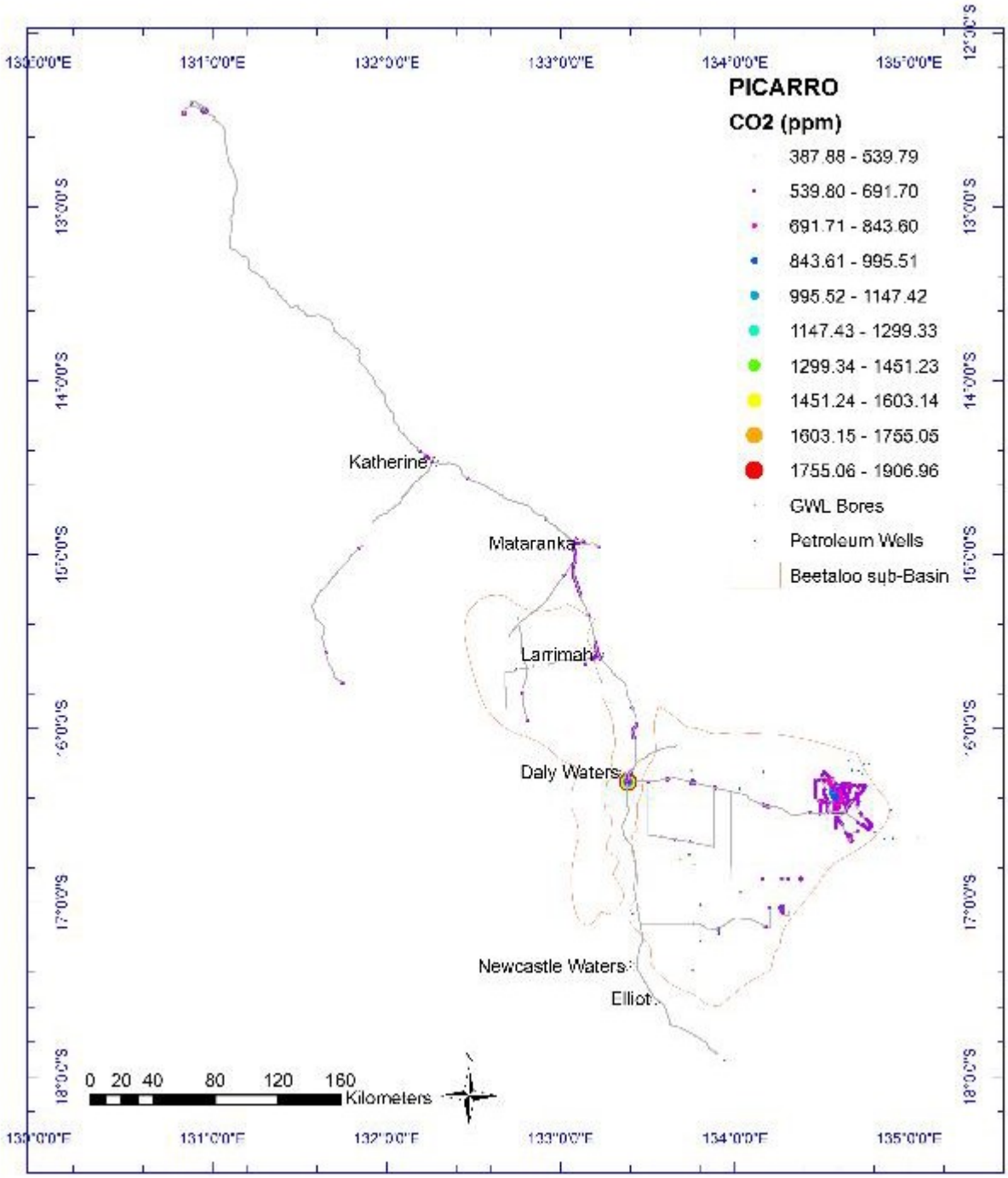

Figure 20: Carbon dioxide concentration measured by the PICARRO analyser during the survey.

6 Discussion

Overall, the majority of elevated CH_4 concentrations recorded in the survey area for the dry season of 2018 was related to cattle. This is in line with the fact that cattle production is currently the dominant industry in the area and is the main source of CH_4. Mobile surveys can be used to detect these sources and where there is a large compound full of cattle as in the case of a feedlot, it is possible to collect measurements related to flux for such feedlots (Day et al., 2016). In the case of the survey area in the Beetaloo sub-basin, the cattle were free ranging, and there were no feedlots observed. In such cases, emission factors related to cattle have been well established (Charmley et al. 2016; Eady et al. 2016; Navarro et al. 2016; Tomkins et al. 2011) and can be used to quantitatively calculate the flux from these dispersed sources. Therefore, where the estimated herd numbers were provided (Wear, pers. comm., Edwards, pers. comm.) the fluxes from the herds were calculated as below on Table 4 using the average emission factors from Charmley et al. (2016).

Table 5: Estimated flux for known number of herds across the Beetaloo sub-basin.

Station	Herd Size Estimated	Estimated Flux (g/min)
Amungee	70,000 breeders	8190
Beetaloo	120,000 breeders	14040
Barkley	100,000 breeders	11700
Tanumburini	50,000 breeders	5850

Secondary sources of elevated CH_4 concentrations of between 1.85 to 2.09 ppm were recorded at townships along the Stuart Highway. These concentrations are similar to values recorded at other towns or cities and are believed to relate to multiple factors including domestic and industrial natural gas usage, landfills, sewage and waste treatment. For example, the concentrations recorded at the CSIRO Kensington site in Perth is typically approximately 1.9 ppm and at the CSIRO Newcastle site between 2-3 ppm, although these show significant temporal variation due to changing atmospheric conditions.

Fires were minor sources recorded during the survey. Two of the fires measured were spatially small sources, and small elevated concentrations of up 300 ppb above background were recorded. The ability to record C_2H_6 and its correlation to CH_4 provided an additional insight which could potentially be used to discriminate CH_4 related to fire and the type of fires. The ratio of CH_4 to C_2H_6 could potentially be a useful indicator to distinguish between biogenic and thermogenic CH_4 source as found by (Yacovitch et al. 2014). Additionally, this ratio also relates to conditions such as the biomass and temperature of the fire (Urbanski et al. 2008).

Elevated CH_4 concentrations were recorded for another spatially larger fire, but access to the plume limited the estimation of flux for the fire. For such large fires where it is typically challenging to access the plumes via mobile survey and are usually spatially large, methods which provide the spatial comprehension incorporating spatial data such as remote sensing could potentially provide more quantitative quantification of flux. Therefore, although the method currently does not specifically estimate CH_4 emissions, it would be useful to investigate methods such as those developed by Russell-Smith et al. (2009) and consider the method as a base template for further development specifically for quantification of CH_4.

No elevated CH_4 concentrations were detected at the plugged and abandoned petroleum wells and most of water bores that were specifically investigated. Although elevated values were detected close to a small number of water wells, these were proximal to other potential CH_4 sources like cattle or other more significant sources such as fuel station and septic tank. Hence it was not possible in this handful of instances to conclusively determine the source of the CH_4 detected.

Wetlands are a well-known but not well-quantified sources of CH_4 estimated to be one of the largest sources of global CH_4 emissions (Poulter et al. 2017; Tian et al. 2016; Zhang et al. 2017). A survey of the wetlands surrounding the Mataranka and Bitter Springs was conducted to understand the contribution of this source of CH_4 to the survey area. However, elevated CH_4 concentration was not detected along the access routes. Further investigations into the spring area did record small elevated levels. The lack of correlation between the CH_4 and C_2H_6 measurements indicated that the source of the CH_4 is likely to be biogenic. The lack of and low concentrations recorded during this survey could be a function of the season as found by other studies indicating that CH_4 emissions were higher in the wet seasons compared to the dry seasons (Grand and Gaidos 2010).

Termites are one of the sources of CH_4, but their contributions to the global budget are one of the most uncertain. Measurements collected across eight termite mounds of a wide range of sizes from less than 20 cm to 1.6 m height found that little or no CH_4 was emitted on the outside walls of the termite mounds. The lack of CH_4 is broadly consistent with findings that seasonal variations govern the CH_4 fluxes and the fluxes related to termite mounds were 3.5-fold greater in the wet season as compared to the dry season (Jamali et al. 2011), although it is acknowledged that further work is required to confirm this. High CH_4 concentrations were measured only when tubing was inserted into a large termite mound indicating that there was CH_4 produced in the mound but not emitted into the atmosphere.

Small elevated CH_4 concentrations which were highly correlated with C_2H_6 concentrations were detected approximately 10 m from the fence adjacent to an above ground section of the Daly Waters to McArthur River gas pipeline. The high correlation between the CH_4 and C_2H_6 indicates that the CH_4 is of a thermogenic nature and likely to be from the natural gas in the pipeline. Since the elevated concentrations were detected close to a valve, it is likely that it was related to a leak in the valve. However, it must be noted that this is a small leak and may fall under the threshold of “reportable leak” as defined as by the Queensland Government’s Code of Practice that, at a measurement distance of 150 mm immediately above (and downwind) of the source, gives a sustained reading of greater than 10,500 ppm (DNRM 2018). Despite this, it may still be important to collect more comprehensive measurements closer to the pipeline for future surveys to provide a more comprehensive baseline of such potential sources of CH_4.

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