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

Final Report for GISERA Project G5

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

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Important disclaimer

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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 March 2018. The Inquiry recommended that baseline monitoring of methane emission 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, and to community concerns regarding potential methane fugitive emissions by the onshore gas industry, 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 Northern Territory. Specifically, the study area covered the exploration permits that lay within the Beetaloo Sub-Basin (EP76, EP98, EP117, EP161 EP167, EP168 and EP169) and was guided by the gas operators Pangaea, Origin and Santos along tracks that were trafficable by four-wheel-drive at the time of the surveys, and spans across areas that are likely to be explored in the next 6-12 months.

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. 2013; Day et al. 2014; Day et al. 2016a; Day et al. 2017a; Day et al. 2017b; Day et al. 2015; Day et al. 2016b; Day et al. 2010; Etheridge et al. 2016; Feitz et al. 2018; Luhar et al. 2018; Ong et al. 2017) 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 this project (GISERA Project G5) is to provide:

- baseline 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 that may have been found.

It is important to recognise that *concentration* is a measure of the abundance of methane in the 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.

At the start of the project, gas exploration activities were projected 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, robust and quickly deployable methods to obtain as comprehensive as possible background atmospheric 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. In addition, fixed monitoring towers may not provide an accurate picture of regionally important sources because of the static and limited footprint of the towers. In addition, consideration was also given to the logistics of operating in remote regions, which at that time was also prohibitively expensive and unreliable and hence may lead to the potential of loss of data. However, as technology progress and the costs decreases, fixed monitoring methods will be considered in later projects.

In this report, we synthesise the finding of three mobile survey campaigns conducted over a six month period between July 2018 to February 2019, capturing the dry, fire and wet season, respectively.

In summary, based on the surveys conducted, we found

- The background atmospheric methane concentrations in the region are close to the global average methane concentrations;
- The main sources detected were grazing cattle, townships, a section of above-ground gas pipeline and associated valves, fires, termites and wetlands;
- The largest source of emissions were grazing cattle but generally, the sources detected were small;
- The main sinks for the region is likely to be soils;
- Seasonal variation were observed around natural sources such as fires, termites, wetlands and soils;
- These natural sources are also the source of the main areas of uncertainty; and,
- No geological seeps were identified during the surveys.

Surveys of atmospheric methane concentrations within the Beetaloo Sub-Basin region were made using mobile surveys with gas analysers mounted in a four-wheel-drive vehicle. Although methane was the principal gas of interest, carbon dioxide and ethane were also measured during the first campaign 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 multiple 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 in the first mobile survey campaign to assess its potential for use in remote location fixed monitoring stations in future monitoring programmes.

The vehicle surveys covered a total of approximately 14850 km. Specifically, 5500, 5300 and 4050 km respectively for the first, second and third mobile survey campaigns along trafficable roads and tracks. For each campaign, between 200 and 600 km were driven each day. The road and environmental conditions were best during the first campaign although 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 permit areas. Fewer ground tracks were covered in the second and third campaign as the conditions of some tracks did not allow access on them. In addition to the tracks, targeted surveys were made at plugged and abandoned or suspended petroleum wells and water bores. A total of 11 plugged and abandoned or suspended petroleum wells, and 25 water bores were visited (or were close to the survey track) at least once during the three mobile survey campaigns. A pipeline riser of the Daly Waters to McArthur River Gas Pipeline adjacent to the Carpentaria Highway was also surveyed for the presence of methane during each of the three campaigns.

The average atmospheric methane concentration across the survey area ranged from 1.80 to 1.82 ppm (dry basis) with a standard deviation ranging from 0.013 to 0.019 ppm. This background atmospheric methane concentration is close to the normal background atmospheric concentrations of approximately 1.8 ppm expected in rural or natural areas (see, for example, Ong et al., 2017). Overall, the majority of methane concentrations recorded during the three mobile survey campaigns were within this average concentration ± 2 standard deviations. Isolated pockets of slightly elevated methane concentrations were observed in some areas; the sources of these were identified as:

- grazing cattle
- townships
- a section of above-ground gas pipeline and associated valves
- fires
- termites
- wetlands

Most of the elevated methane levels were detected in the vicinity of grazing cattle. The methane elevations were quite small, with the maximum concentrations less than 2.9 ppm (i.e. about 1.1 ppm above background). Moreover, the concentration peaks were measure only for a short duration since the survey vehicle quickly moved past the source, reflecting the small size of these sources. Estimating the amount of methane emitted from a dispersed source such as open range cattle is difficult using mobile ground surveys. Instead, we used an alternative approach based on the aggregated carrying capacity for the stations that fall within the Beetaloo sub-basin region together with published methane emission factors for Australian cattle. This yielded an emission rate of approximately 7,402,160 kg CH_4 yr^{-1} across the Beetaloo Sub-basin. To put this into context this, this figure is 0.28% of the total methane emission from cattle report as part of the National Greenhouse Gas Inventory for the year 2017 (NGGI 2018). As a side note, the total methane emission from cattle in the Northern Territory represents 2.32% of the Australia's total methane emission for that same year.

Some other instances of elevated methane concentrations of between 1.85 to 2.09 ppm were recorded at townships along the Stuart Highway during all three mobile survey campaigns. 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. Emission rate estimates were not made for these sources during these campaigns, but because of the very small population density, are expected to comprise a minor component of the total methane budget of the survey region.

No elevated methane concentrations were detected at the petroleum wells and water bores that were specifically investigated during this field campaign. Moving into the future, it is likely that gas exploration will commence in the next 6-12 months in the survey region covered by this report. Before drilling activities begin, there is an opportunity to extend the baseline mesurements with a comprehensive soil methane baseline acquired around the well pad areas to capture the natural background methane emission of the surrounding area. Some studies conducted in the USA have suggested that the hydraulic fracturing process could potentially be a significant source of methane. However, in Australia, there is currently a gap in understanding of the emissions likely to be produced during this operation. Therefore, it would be important to collect methane measurements throughout the hydraulic fracturing operations to understand the emissions related to the hydraulic fracturing operations and related flow back. In addition, it may be useful to install remote monitoring stations close to the well pad at this stage according to recommendations 9.3 of the Scientific Inquiry. With the small footprint of most remote monitoring stations, the optimal useage is likely to be in such application, that is, close to gas infrastructure to provide continuous monitoring of the operations of the infrastructure. However, this has to be weighed against the cost and uncertainties of the towers.

During the first mobile survey campaign, above average methane concentrations which were highly correlated with ethane concentrations were detected approximately 10 m from the fence adjacent to a pipeline riser of the Daly Waters to McArthur River gas pipeline. It must be also be noted that this is a small leak and falls under the threshold of "reportable leak" as defined as by the Northern Territory 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 5000 ppm. The high correlation between the methane and ethane indicates that the methane is of a thermogenic nature and likely to be from the natural gas in the pipeline. A second visit to the same pipeline riser area confirmed the presence of above average methane concentrations that were approximately 0.070-0.080 ppm above the background value. Following the report of this leak, a service provider to the owner of the pipeline at the request of the Department of Primary Industry and Resources investigated it and found that during their monthly inspection no leak had been detected. However, an additional survey was initiated on 8th January 2019, which detected a small leak that originated from a grease nipple on a ball valve. The nipple cover was tightened as it was found to be only finger tight. A gas level reading was again taken 150mm from the nipple which indicated there was no leak present. An additional step has now been included in the valve check sheet, which requires that all grease nipple covers be tightened. This pipeline riser was again visited during the third mobile survey campaign where, the emission rate from this facility was estimated and found to be approximately 43.80 kg CH_4 yr^{-1}.

To put this into context, this emission rate is 60-80 % of the average of 54.75 to 73.00 kg CH_4 yr^{-1} produced per head of cattle.

Fires were minor methane sources recorded during the first and second mobile survey campaigns. All the fires measured were spatially small sources, and above average concentrations of up 0.30 ppm above background were recorded. Although elevated methane concentrations were recorded for other spatially larger fires or smoke plumes observed during the surveys, access to the plume limited the estimation of emission rate for the fire. For such spatially large fires where it is typically challenging to access the plumes via mobile survey, methods which provide the spatial comprehension incorporating spatial data such as remote sensing could potentially provide more accurate quantification of emission rates. Additionally, the contribution of methane from fires is transient and seasonal, suggesting that methods which can systematically collect temporal measurements remotely to capture these variations is required. Therefore, it would be useful to investigate methods which make use of remote sensing technology which may include expanding on existing methods which employ traditional optical satellites data and, also investigate new satellites that specifically measure greenhouse gases such as methane, carbon dioxide and carbon monoxide such as the European Space Agency's Sentinel-5P.

Wetlands are well-known but not well-quantified sources of CH_4. Some estimates suggest that wetlands are sources of methane and are estimated one of the largest sources of global methane emission (Saunois et al. 2016). A survey of the wetlands surrounding the Mataranka and Bitter Springs was conducted to understand the contribution of this source of methane to the survey area. However, elevated methane concentrations were not detected along the access routes during any of the three mobile survey campaigns. Further investigations into the spring area during the first mobile survey campaign did record above average levels. In addition the very low ethane concentration and the lack of correlation between the methane and ethane measurements indicated that the source of the methane is likely to be biogenic (i.e. unrelated to natural gas). The lack of methane recorded during this survey could be a function of the season as found by other studies indicating that methane emissions were higher in the wet seasons compared to the dry seasons. It may be useful to revisit this source of methane by walking into the spring area with a more portable methane analyser during the wet season in future, if access to the spring area may be available. Although elevated ambient methane concentrations were not observed adjacent to wetland areas during these surveys, surface flux measurements made on soil and inundated areas confirmed that methane was being emitted from the sites, especially during the wet season. Estimating the total regional methane emission rates from such transient wetlands, however, is a complex an challenging task and at these stage we are unable to make a reasonable estimate of emissions to the Beetaloo Basin.

Termites are a source of methane, but their contribution to the global budget is one of the most uncertain. In the Beetaloo Sub-Basin, they are a widespread methane source, but their emissions are subject to large seasonal variations. Measurements made during the third mobile survey campaign in the wet season clearly showed methane emissions, yet no emissions were detected from a selection of termite mounds during the first mobile survey campaign in the dry season. These observations are consistent with findings that seasonal variations govern the methane fluxes. The fluxes related to termite mounds were up to 3.5-fold greater in the wet season as compared to the dry season. Using emission factors found from previous work, the total emission in the Beetaloo Sub-Basin was estimated to be approximately 900,000 kg CH_4 yr^{-1}.

Soils are known to be methane sinks rather than a source. Using emission factors developed from previous work, the soil sink for the Beetaloo Sub-Basin was estimated to be more than 4,000,000 kg CH_4 yr^{-1}. Clearly, developing a detailed and accurate methane budget for the region is a complex task and needs to account for both natural and other sources as well as the strong seasonal effects on these sources.

The work undertaken as part of this project (GISERA Project G5) as part of this project is an important step in the application and research of improved methane measurement and monitoring methodology. The results provide a comprehensive baseline of background atmospheric methane emissions across the Beetaloo Sub-Basin study area against which the impact of unconventional gas development can be assessed.

1 Introduction

This project (GISERA Project G5) specifically addresses the first component of Recommendation 9.3 of the Northern Territory Government's 'Scientific Inquiry into Hydraulic Fracturing' Final Report that refers to the measurement and monitoring of 'methane (CH_4) concentrations' before the granting of exploration approvals and production activities by gas companies (Andersen et al. 2018).

Concentration and flux

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)). In this report, all concentrations will be reported in ppm. 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). In this report, where flux had been estimated they will be reported in kg yr^{-1}. Both concentrations and flux 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 background atmospheric concentration levels of CH_4 in areas of the Beetaloo Sub-Basin (outline of Sub-Basin shown on Figure 1 in red) that are of interest for exploration (exploration permit areas in green on Figure 1) and identify sources for locations where elevated CH_4 levels are found and, where applicable and feasible, quantify the fluxes related to these sources.

This final report documents the findings of three surveys conducted over between July 2018 – February 2019 where CH_4 concentrations in the region were measured using gas analysers mounted in a four-wheel-drive vehicle. The three surveys traversed a total of almost 15,000 km with each campaign covering between 5,500 and 4,050 km. During the surveys CH_4 and carbon dioxide (CO_2) measurements were acquired continuously along the route. In the first survey, ethane (C_2H_6) measurements were also collected.

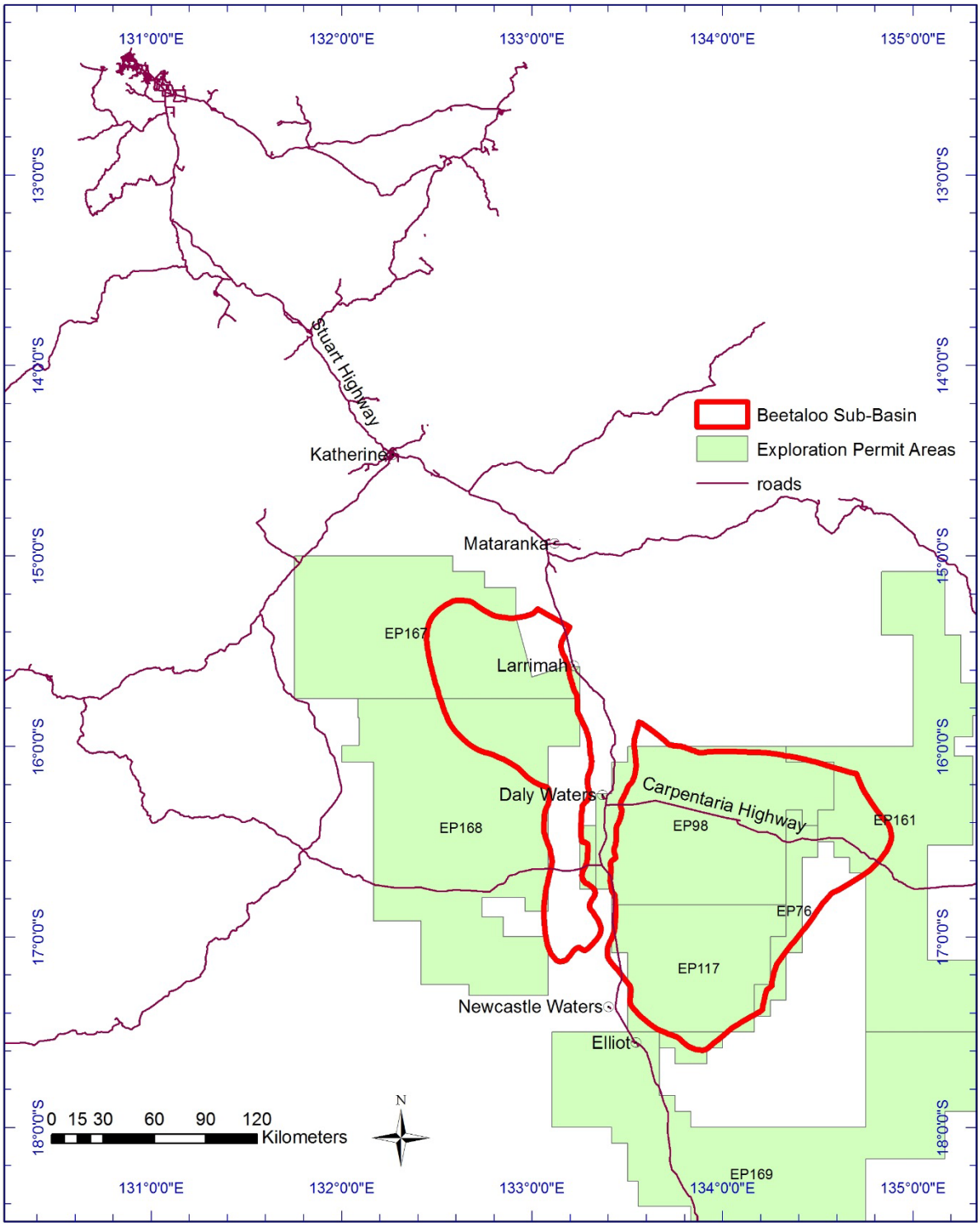

Figure 1: Map of the study area where the mobile surveys were conducted. Outline of the Beetaloo Sub-Basin (red polygon) and the exploration permits within the Sub-Basin (green).

2 Experimental Methods

CSIRO and other research organisations have been actively conducting research into CH_4 emissions from the unconventional gas industry over many years (Day et al. 2013; Day et al. 2014; Day et al. 2016a; Day et al. 2017a; Day et al. 2017b; Day et al. 2015; Day et al. 2016b; Day et al. 2010; Etheridge et al. 2016; Feitz et al. 2018; Luhar et al. 2018; Ong et al. 2017). 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; fixed flux towers (e.g. eddy covariance methods); bulk atmospheric concentration measurements; and 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 (Luhar et al. 2018).

In the current project, however, time was limited by the need to complete the first set of monitoring measurements before the 2018/2019 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 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 rate 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. That is, repeat access can be constrained due to changed conditions.

3 Instrumentation

The mobile surveys were 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.

The first mobile survey campaign used all three instruments; the second and third campaign used only the PICARRO and LGR analysers as the AERIS analyser was being used for developments of a remote monitoring station and hence was not available during the survey periods.

All three instruments are capable of reliably detecting changes in CH_4 concentration as low as 0.002 ppm and have 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. However for the mobile surveys, there were insufficient length of time and quantities of CH_4 to provide 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 because

- it's molecular mass (26) is close to air;
- naturally occurring C_2H_2 is very low (~1 ppbv);
- it decomposes in atmosphere relatively quickly with a half life of ~13 days;
- it is readily available and inexpensive; and,
- it has strong absorption bands in the spectral regions of the gas analysers (near infrared).

The AERIS analyser measures CH_4 but also simultaneously measures ethane (C_2H_6). The presence of C_2H_6 may indicate thermogenic sources (Yacovitch et al. 2014).

All the instruments are also designed to measure the water content of air so that CH_4, CO_2, C_2H_6 and C_2H_2 can be reported on a 'dry air' basis.

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 to ensure an accurate measurement is recorded. Positional data were combined with the gas concentration data to produce maps of CH_4 concentration across the study region.

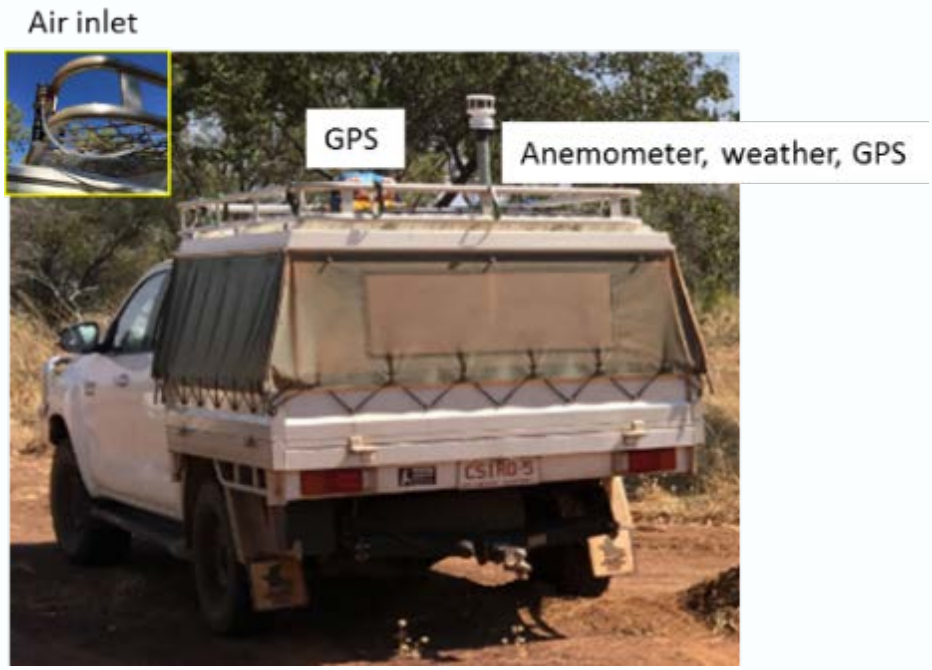

Figure 2: Survey vehicle showing the location of sampling lines, GPS and combined anemometer, GPS and weather station.

The survey vehicle is shown in Figure 2. 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 2) 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 analyser located at the tray of the ute (shown in 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.

During the first campaign, all three instruments were initially mounted in the covered tray of the vehicle as shown in Figure 3; 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 as shown on Figure 4, which successfully prevented this problem for the rest of that campaign as well as the other two campaigns.

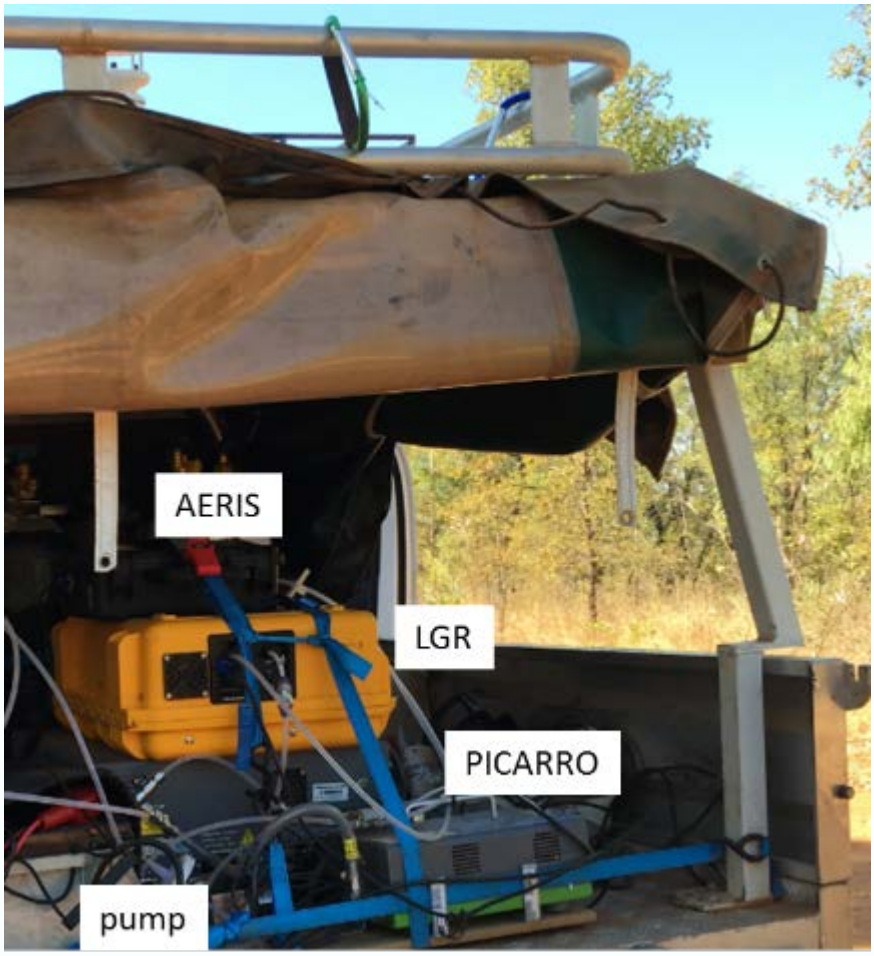

Figure 3: PICARRO, LGR and AERIS analysers mounted in the rear of the field vehicle before being relocated to the cabin.

Figure 4: PICARRO and LGR analysers mounted in the back seat of the field vehicle. Note that the back seat has been removed.

3.1 Flux Measurements

On some occasions during the surveys, CH_4 emission flux measurements were attempted where possible; specifically, emissions measurements were made at a gas pipeline riser near the Carpentaria Highway which had previously been found to be emitting small amounts of CH_4, and at some termite mounds. Soil flux measurements were also made at some locations. The methods used for these measurements are briefly described below.

3.1.1 TRACER METHOD

In one approach to determine CH_4 emission rates, a stable tracer gas (such as acetylene) can be released at a known rate from the same location as the source of CH_4. Provided that the tracer gas is released sufficiently close to the CH_4 emission point and mixes into the CH_4 plume, the CH_4 emission rate can be readily

calculated by multiplying the acetylene release rate by the ratio of the CH_4 and acetylene concentrations downwind of the source.

3.1.2 PLUME TRAVERSES

In some circumstances, it is possible to estimate CH_4 emissions using a plume dispersion method. In this method, the CH_4 concentration profile in a plume originating from the source is measured downwind by performing traverses across the plume. The emission flux, *F*, may be estimated by integrating the CH_4 concentration enhancement, *C*, of the plume in the horizontal, *y*, and vertical, *z*, directions and multiplying by the average wind velocity, *u*.

$$F = u \int_{-y}^{y} \int_{0}^{z} C(y,z) dy dz \qquad \text{Equation 1}$$

Because concentration measurements are made only at ground level, the vertical dispersion must be estimated by reference to plume dispersion models such as the Pasquill-Gifford curves of σ_z (i.e. the standard deviation of the distribution of CH_4 concentration in the vertical direction) as a function of downwind distance under given atmospheric turbulence conditions. The vertical concentration profile of CH_4 within the plume is assumed to decrease from the ground level concentration with height according to a Gaussian distribution. However, because the vertical concentration is estimated based on plume modelling methods, the uncertainty of this approach may be significant.

3.1.3 FLUX CHAMBERS

Some measurements were made on various ground surfaces to determine soil CH_4 flux. For these measurements, a metal bucket approximately 30 cm in diameter with a total volume of about 9 L and an area of coverage of 0.07 m^2 was placed on the ground at each sampling point (see Figure 5 of the flux chamber in operation). The chamber was connected to the LGR CH_4 analyser in the field vehicle via a ¼" nylon tube and the CH_4 concentration within the chamber, *C*, continuously measured over a period of several minutes. The flow rate of the sample stream from the flux chamber to the analyser was approximately 100 mL min^{-1}, which was returned to the chamber via a second tube (Figure 3).

The CH_4 emission flux, *F*, was calculated according to Equation 2:

$$F = \frac{dC}{dt} \times \frac{V}{A} \qquad \text{Equation 2}$$

where *V* is the volume of the chamber, *dC/dt* is the rate of change in the CH_4 concentration over time, *t*, and *A* is the area of the surface covered by the chamber.

Figure 5: Surface flux chamber used for measuring soil CH_4 emissions. This chamber was also used for measuring CH_4 emissions from some termite mounds. The particular measurement was taken on saturated soils.

4 Data Processing

The main data processing performed on the data collected for all the mobile survey campaigns include:

- applying appropriate calibration factors;
- correction of the measured concentration data to account for humidity;
- 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 was acquired simultaneously.

Before commencing the first mobile survey campaign, 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 to 102 ppm (similar to the maximum CH_4 concentration likely to be measured during the field campaigns). At the time of the calibration, all instruments were linear over this range.

Table 1. Reference gas compositions used for calibration checks on the PICARRO and LGR analysers.

	Methane (ppm)	Acetylene (ppm)	CO_2 (ppm)	Oxygen (%)	Nitrogen
Reference 1	1.02	1.00	-	20.8	Balance
Reference 2	3.02	-	496	20.9	Balance
Reference 3	4.07	3.97	-	20.9	Balance
Reference 4	10.2	-	1010	20.9	Balance

It should be noted that although moisture concentration data are not strictly required for the CH_4 and CO_2 analyses, the varying levels of moisture present in the atmosphere affect the concentrations of these species over long and short timeframes. Hence, to allow direct comparison with data collected under different humidity conditions, the CH_4 and CO_2 concentratons are often reported on a 'dry air' basis (i.e. the CH_4 and CO_2 are corrected to a moisture content of 0 %). Methane and CO_2 concentrations reported from global monitoring stations (e.g. the CSIRO Cape Grim station in Tasmania) are generally reported on a dry basis.

The second and third survey was conducted under more extreme weather conditions with higher temperatures and larger variations in humidity than during the first survey made in July and August 2018. The temperature variations were mitigated with the installation of the instrumentation in the cabin of the vehicle. However, the high humidity levels were found to have a significant impact on the measurements. Both the PICARRO and LGR have the ability to measure water vapour. Ideally, these measurements which would have been acquired simultaneously with the CH_4 and CO_2 concentrations would be used to correct for humidity impacts. However, investigation of the water vapour measurements found that the sensor on the PICARRO was not functioning properly and was recording spurious levels. This meant that humidity impacts were not properly accounted for in the PICARRO data. This impact is illustrated in Figure 6, where the CH_4 concentration recorded by the instruments is plotted as a function of the measured water vapour concentration. The right graph shows that the water vapour concentrations recorded by the PICARRO were very low and inconsistent with the humid conditions; many of the values were also negative or very close to zero. There also appears to be four discrete sets of data indicating that there may be four different sets of estimations for water vapour. In comparison, the water vapour measurement to the CH_4 concentration for the measurements recorded by the LGR as shown on the left graph, the water vapour values were as would have been expected from the weather conditions and there was a clear linear relationship between the concentration of CH_4 and water vapour.

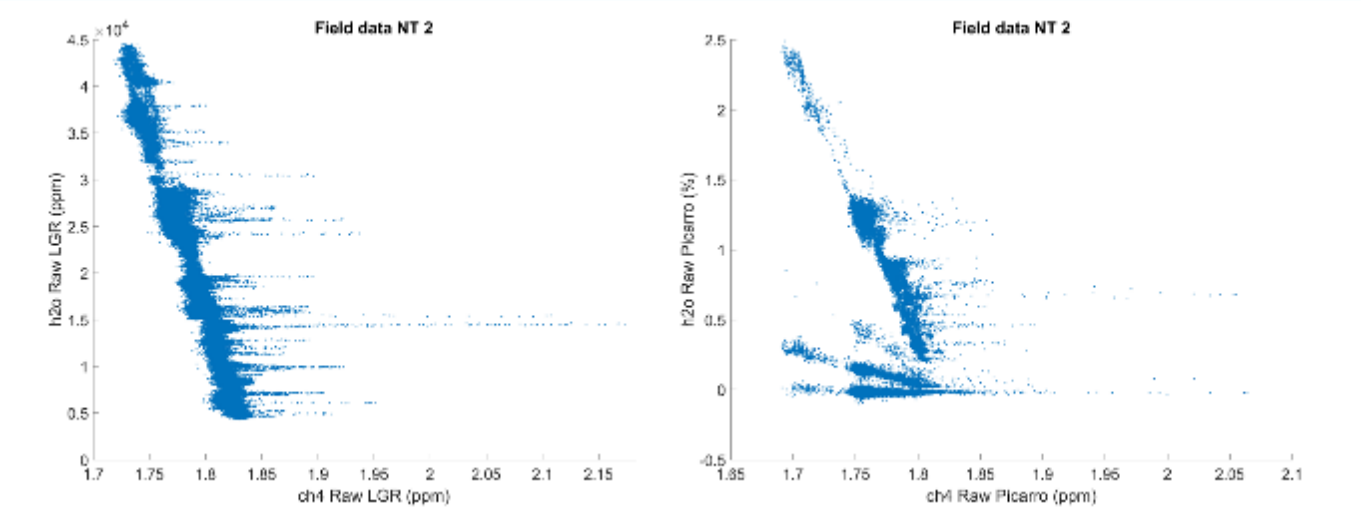

Figure 6: Left: Raw LGR CH_4 concentration plotted against the LGR H_2O vapour measurementsmeasurements. Right: Raw PICARRO CH_4 concentration plotted against the H_2O vapour measurements Note that the H_2O vapour is measured in percent water for the PICARRO and in ppm for the LGR.

As the humidity impact was significant, it was important that these impacts were removed. Examination of the water vapour measurement recorded by the LGR and illustrated on the left graph in Figure 6 indicate that the water vapour and the CH_4 measurements were well correlated and hence are unlikely to be distorted by other instrumental effects. Therefore, a correction was implemented where the water vapour measurements from the LGR was used to correct the PICARRO data using the simplified equation below. The same equation was also applied to the LGR data to ensure that both datasets were corrected similarly.

$$CH4corrected = \frac{CH4raw}{(1 - H2O)}$$

Where $CH_{4corrected}$ is the corrected CH_4 concentration (ppm), CH_{4raw} is the raw CH_4 concentration (ppm), and H_2O is the water vapour concentration (%).

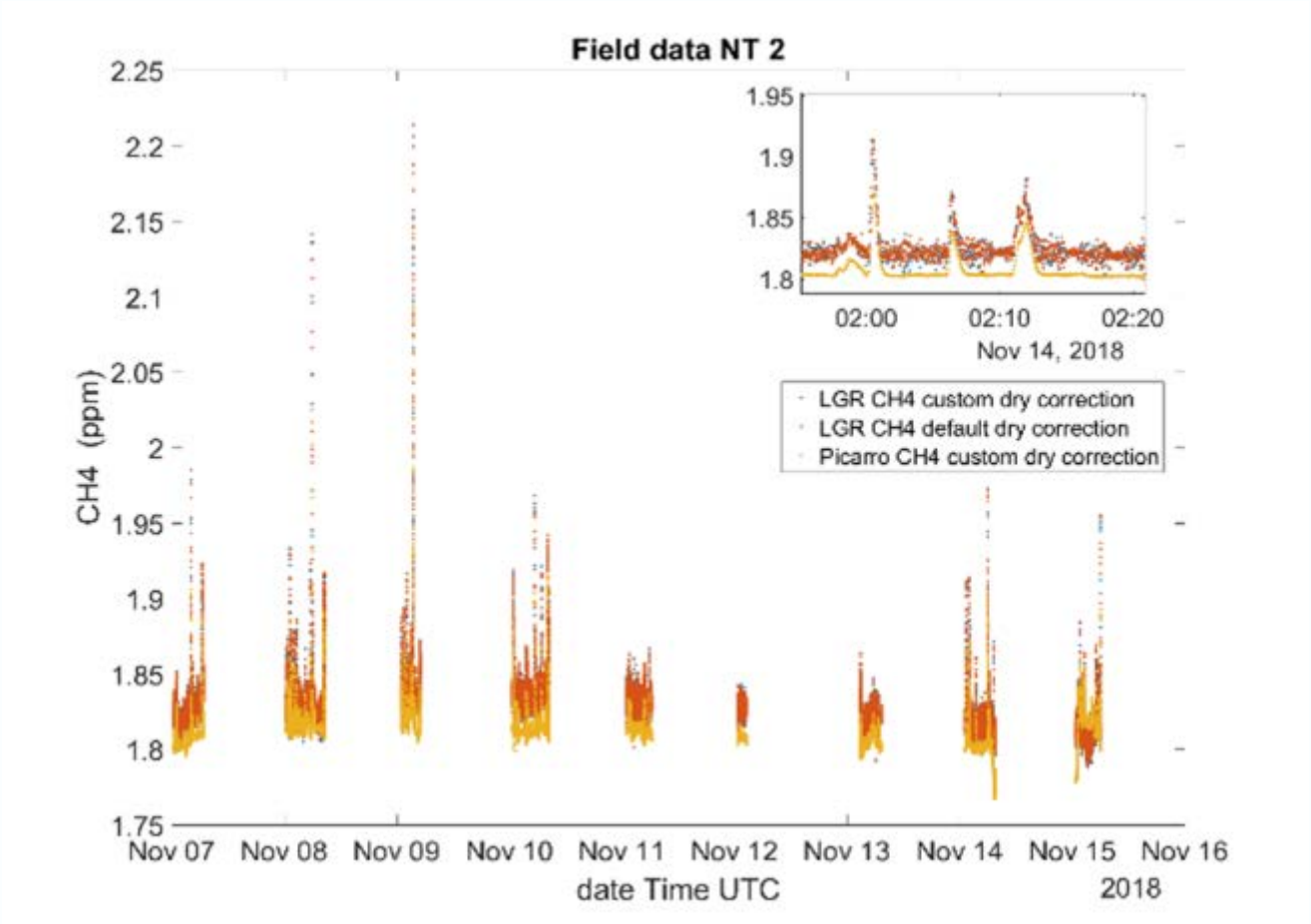

Figure 7: Corrected PICARRO (orange dots) and LGR (blue dots) CH_4 concentration using the customised method described above and automated dry CH_4 values provide by correction implemented by LGR (red dots). The top right hand inset graph shows a zoomed in view for a portion of a survey on 14th November 2018.

Figure 7 plots the results of the correction and shows that the humidity impact has been accounted for as indicated by similar average baseline values for all dates. Additionally, the graph shows that the customised correction produced similar results to the correction implemented by LGR.

Figure 8 shows the correlation between the PICARRO and LGR methane measurements. The figure illustrates the influence of humidity on the raw CH_4 measurement (plotted in blue) where the baseline level varied from 1.75 to 1.85 ppm. After the correction, this baseline remained constant at 1.85 ppm (plotted in orange).

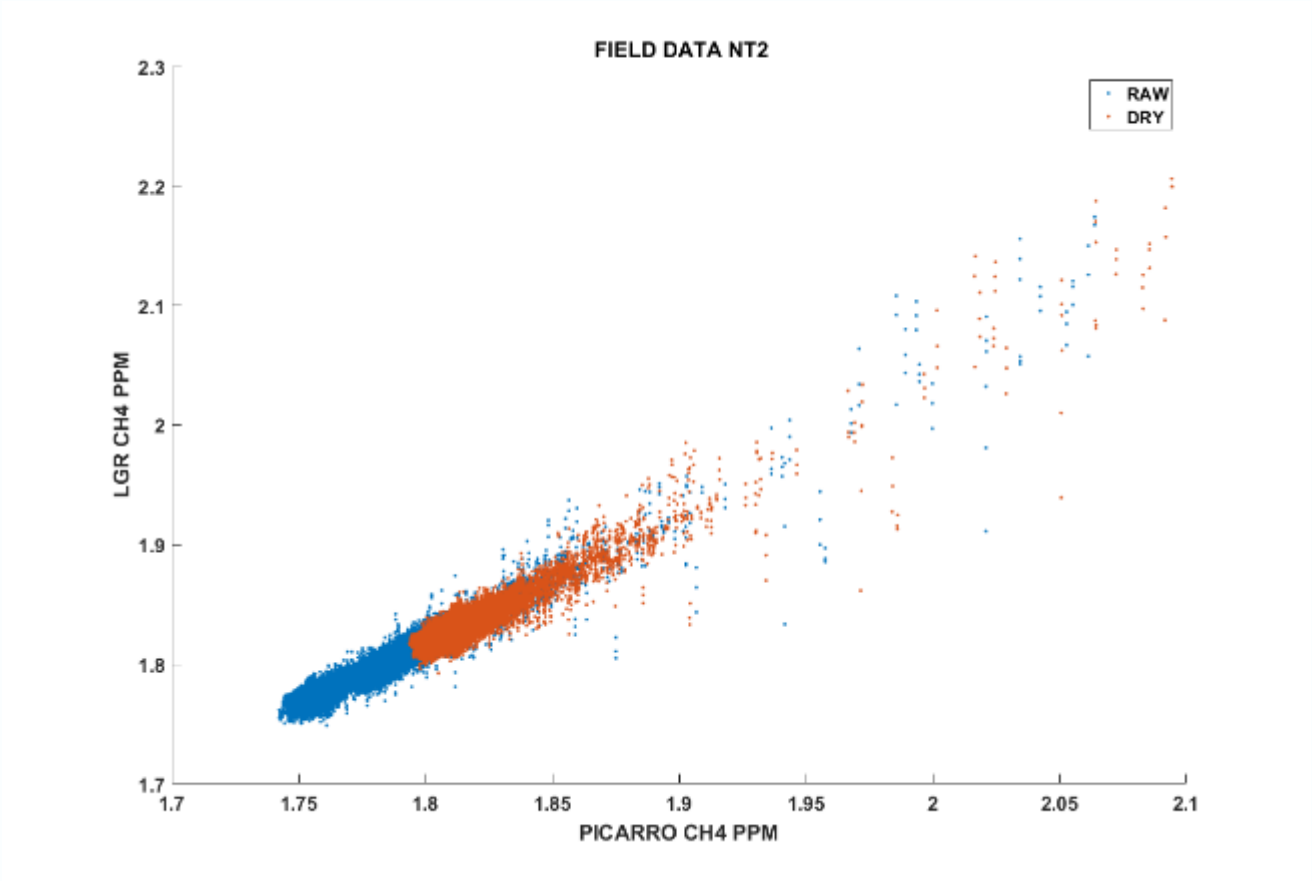

Figure 8: Correlation between the PICARRO and LGR raw (blue) and dry (corrected for humidity effects in orange) methane concentration measurements.

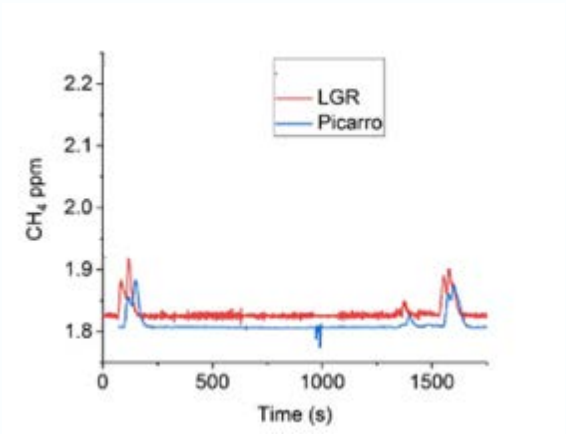

Figure 9: CH_4 detected by the PICARRO and LGR analysers before realignment for the response time difference.

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 Figure 9, where the response time difference related to emissions from a herd of cattle is illustrated. 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. 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 two GPS units used on some occasions, the GPS dedicated to the LGR failed to provide a signal on some occasions. Hence it was necessary to link the LGR measurements to the PICARRO's GPS. This was performed by synchronising the time stamp on the LGR data to the other GPS's time stamp.

5 Results

Three mobile survey campaigns totalling almost 15,000 km were conducted between July 2018 to February 2019. In the first mobile survey campaign representing the dry period, 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' permit areas (shown on Figure 1) 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. 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 exploration permit areas.

The second mobile survey campaign was conducted over a 9-day period between 6th – 15th November 2018 covering 5,300 km representing the fire season. To enable direct comparisons between the mobile survey campaigns, the tracks predominantly replicated the ones traversed during the first mobile survey campaign except for some areas across the Origin exploration permit areas which were not trafficable due to previous rain. However, the area that was covered was sufficient to represent the area which will be explored in the future for Origin (Kernke, pers. comm.).

The third and final mobile survey campaign was conducted over a 7-day period during the wet season between 30th January and 5th February 2019. As anticipated, heavy rain and deep water across roads meant that there were less trafficable tracks than in the other seasons and hence a lesser number of kilometres; specifically, 4050 km was traversed during this survey. Nevertheless, despite the reduced distance, the survey covered as much as trafficable the routes traversed during the first and second set of surveys; the main areas where traversing was not possible was on the Origin and Pangea exploration permit areas.

Figure 10, Figure 11 and Figure 12 provide the maps of the tracks traversed and the CH_4 concentration measurements acquired by the PICARRO analyser during the three survey campaign. Table 2 summarises the average, median, standard deviation and maximum CH_4 concentration values measured during the three mobile survey campaigns. The average CH_4 concentration measured by the PICARRO during the surveys ranges from 1.82 to 1.80 ppm (dry basis) with a standard deviation ranging from 0.013 to 0.019 ppm. For comparison, the CH_4 concentration measured at the CSIRO Cape Grim atmospheric monitoring station for the months of August 2018, November 2018 and February 2019 are shown in Table 3. It indicates comparable CH_4 concentrations were recorded at Cape Grim to the CH_4 concentrations measured during the mobile survey campaigns in the Beetaloo Sub-Basin. Cape Grim is located on Tasmania's west coast (40.68°S, 144.69°E), one of the three premier Baseline Air Pollution Stations in the World Meteorological Organization-Global Atmosphere Watch (WMO-GAW) network. (https://www.csiro.au/en/Research/OandA/Areas/Assessing-our-climate/Latest-greenhouse-gas-data). As a guide, these CH_4 concentrations are equivalent to or below the global CH_4 level of 1.85 ppm (Dluugokencky 2018). Slightly higher (0.02 ppm more) average CH_4 concentration values were recorded by the LGR. These larger CH_4 concentration values are consistent with the comparisons to the reference gas measurements, which indicated that the LGR registered slightly higher values than the PICARRO, and potentially the faster response time of the LGR compared to the PICARRO.

The maximum CH_4 concentration measured with the PICARRO analyser during the mobile survey campaigns ranges from 2.09 to 2.31 ppm. These maxima were mostly attributed to grazing cattle close to the survey vehicle. The maxima recorded by the LGR is slightly higher because of the differences in 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 differences between the two analysers.

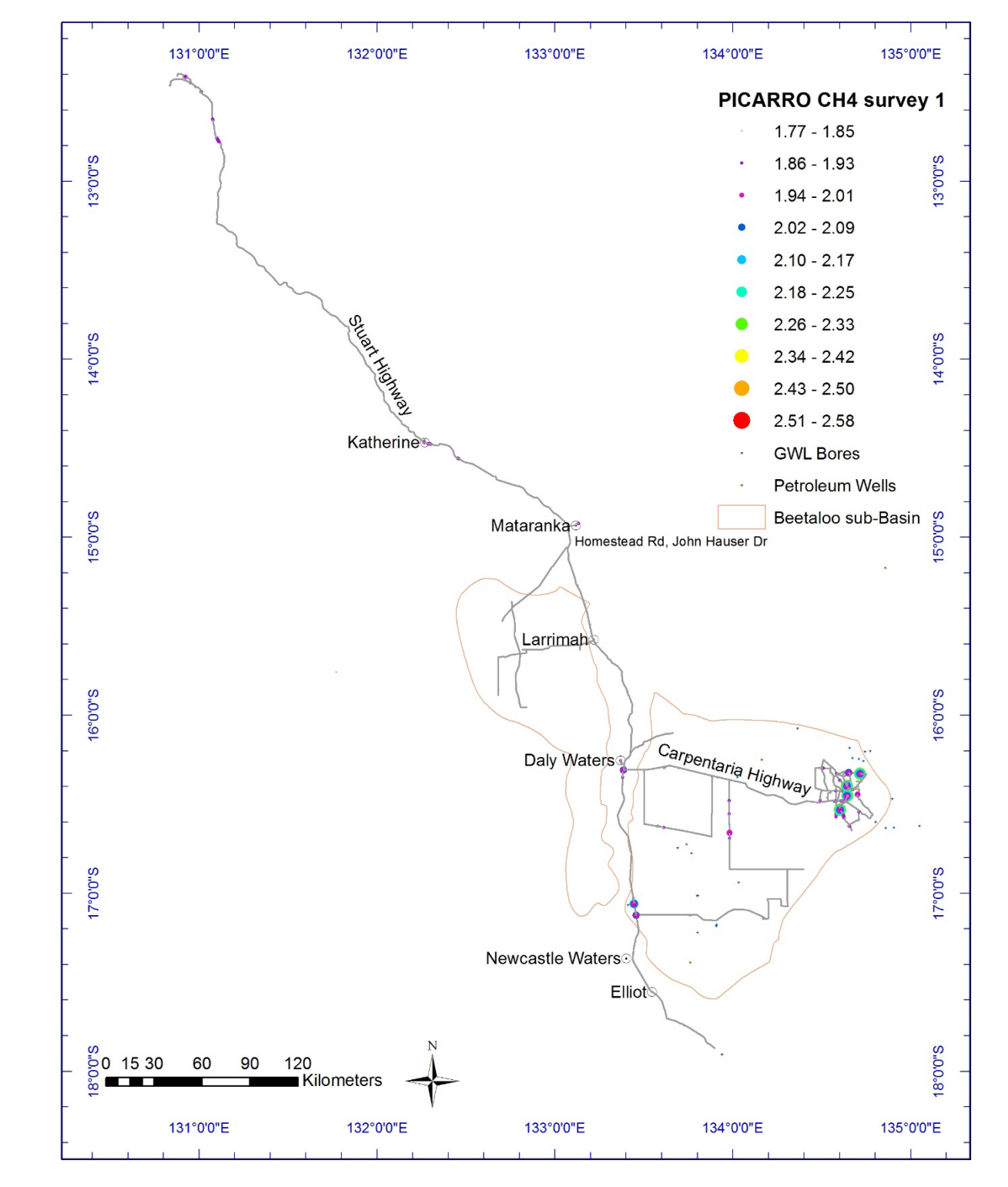

Figure 10: CH_4 concentration measured using the PICARRO analyser along tracks and roads across the Beetaloo Sub-Basin during the first mobile survey campaign conducted during 29th July – 10th August 2018.

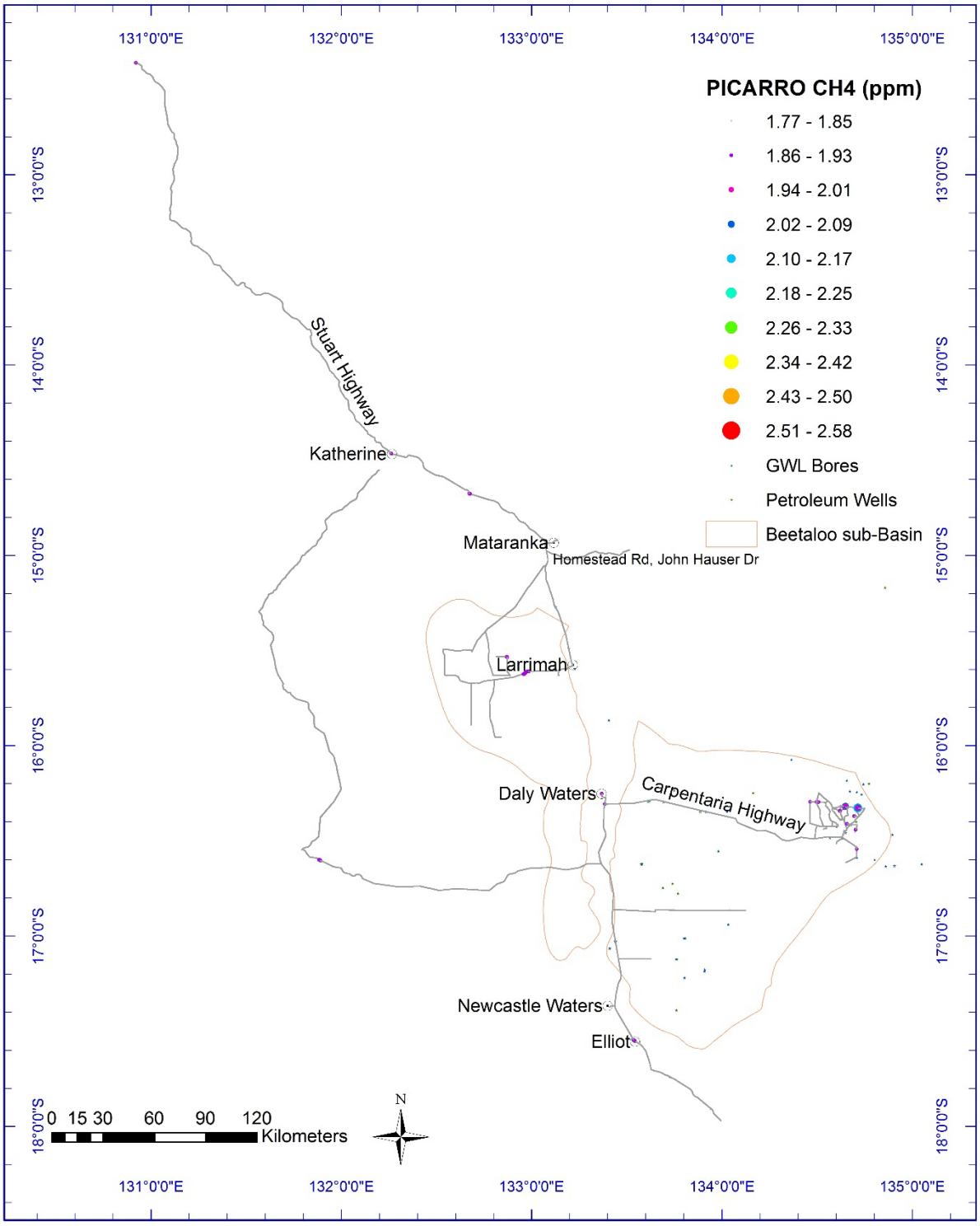

Figure 11: CH_4 concentration measured using the PICARRO analyser along tracks and roads across the Beetaloo Sub-Basin during the first mobile survey campaign conducted during 6th – 15th November 2018.

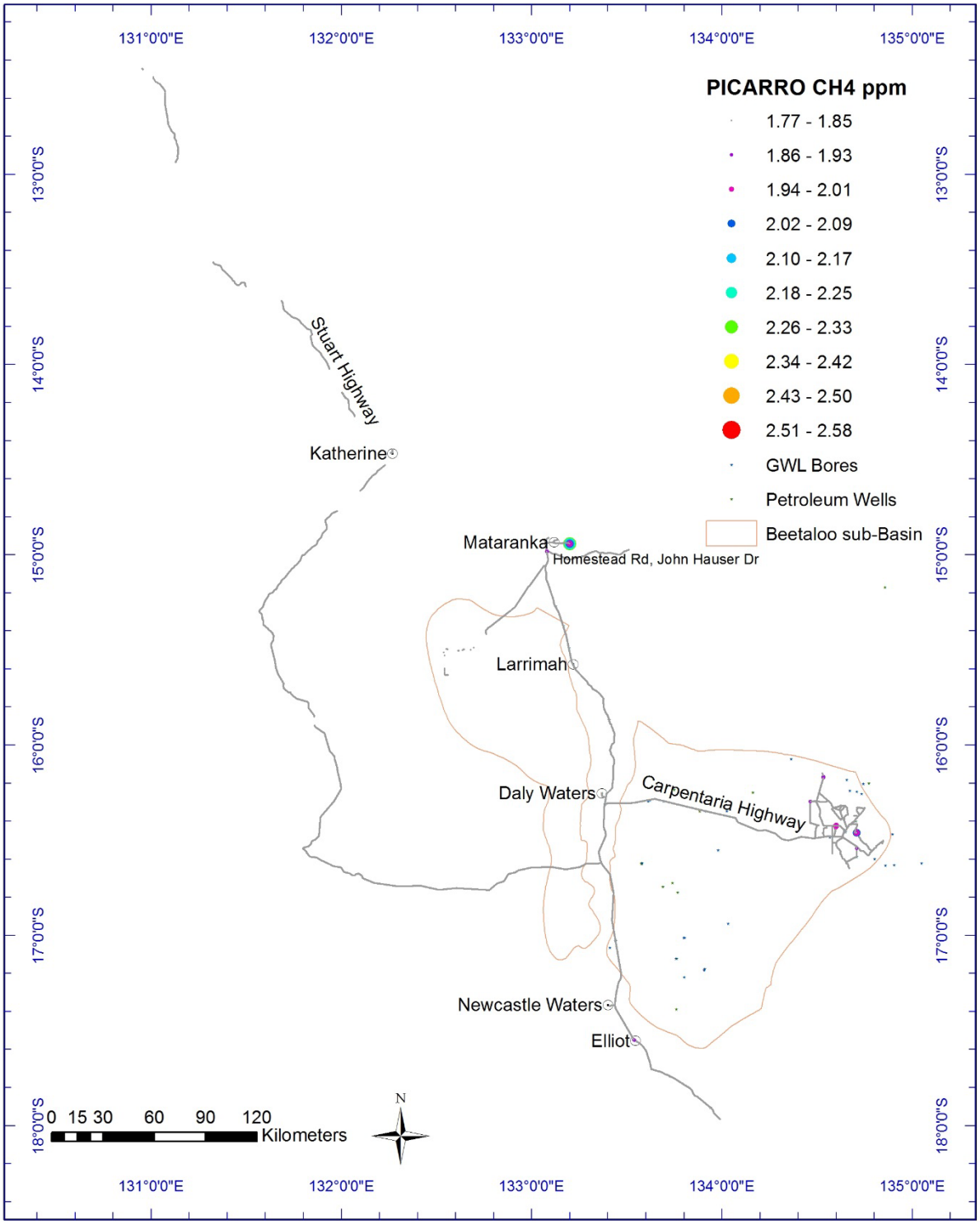

Figure 12: CH_4 concentration measured using the PICARRO analyser along tracks and roads across the Beetaloo Sub-Basin during the first mobile survey campaign conducted during 30th January and 5th February 2019.

Table 2: The average, median, standard deviation and maximum CH_4 concentration values measured during the three mobile survey campaigns conducted during 29th July – 10th August 2018, 6th – 15th November 2018 and, 30th January and 5th February 2019.

	Campaign 1 (LGR)	Campaign 2 (LGR)	Campaign 3 (LGR)	Campaign 1 (PICARRO)	Campaign 2 (PICARRO)	Campaign 3 (PICARRO)
Average (ppm)	1.839	1.827	1.808	1.817	1.811	1.796
Median (ppm)	1.835	1.826	1.807	1.813	1.811	1.795
Standard deviation	0.019	0.013	0.017	0.018	0.012	0.019
Maximum (ppm)	2.604	2.206	2.920	2.310	2.094	2.297

Table 3: Methane concentration measured at Cape Grim for the months of August 2018, November 2018 and February 2019.

August 2018	November 2018	February 2019
1.826	1.820	1.798

Similar distributions of CH_4 concentration were seen between the three mobile survey campaigns. That is, most of the area was within the average CH_4 concentration ± 2 standard deviations, but there were pockets of elevated CH_4 concentrations.

GRAZING CATTLE

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 concentrations detected along the west and east of the Stuart Highway for all the campaigns, as shown in Figure 10, Figure 11 and Figure 12. For example, the largest number of elevated concentrations were detected furthest west on the Santos exploration permit areas on each of the campaigns at the Tanumbirini cattle station; most can directly be attributed to herds of cattle passing along the tracks travelled.

TOWNSHIPS

The other main sources of elevated CH_4 concentrations detected were townships. Elevated CH_4 concentrations were recorded at Katherine, Mataranka and Daly Waters along the Stuart Highway on the first campaign (Figure 10). Elevated CH_4 concentrations were recorded again at these townships in the second campaign and also for Elliot (Figure 11). Elevated CH_4 concentrations were observed again on the third campaign (Figure 12). In all of these cases, the levels observed were only slightly above background, suggesting that the emission rates from the sources are low. The elevated CH_4 concentrations were likely due to anthropogenic sources at townships such as fuelling stations, seepages of natural gas from domestic or commercial usage, vehicles or sources such as sewage/waste treatments and landfills.

PETROLEUM BORE HOLES

During the three mobile survey campaigns, 11 plugged and abandoned and suspended wells were visited (or were close to pastoral tracks where measurements were made) at least once during the three mobile survey campaigns. These wells were visited as most of the well completion reports indicated that gas was detected which indicate that they may be potential sources of CH_4. Details of these wells, their EP number, locations, depth, whether the prospective formation were intersected, whether gas was detected and, when they were visited or passed are shown in Table 3. No elevated CH_4 concentrations were detected at any of the well sites visited.

Table 4: List of plugged and abandoned and suspended petroleum wells visited during the survey.

Well ID/Name	EP #	Latitude, Longitude (locations mostly are shown on Figure 9)	Depth (m)	Intersection with prospective formation (K=Kyalla, V=Velkerri) (m)	Hydrocarbon shows in Well Completion Report (WCR)	Comments	Visited during campaign number
Wyworrie 1	EP 167	15°22'31"S,132°43'56"E	1385	Yes (K + V)	Yes, gas	Plugged and abandoned (PNA)	1, 3
Tarlee 1	EP 168	15°57'16"S,132°50'23"E	1335.5	Yes (K + V)	Yes, gas	PNA	1, 2
Tarlee 2	EP 168	15°53'32"S,132°41'4"E	1180	Yes (K + V)	No mention in WCR	PNA	1, 2
Tarlee S3	EP 167	15°37'55"S,132°49'36"E	1650	Yes (K + V)	No mention in WCR	PNA	1, 3
Birdum Creek 1	EP 167	15°37'50"S,133°8'35"E	1935	Yes (K + V)	No mention in WCR	PNA (see Figure 12)	1, 2, 3
Kalala S1	EP 98	16°17'38"S,133°36'49"E	2619	Yes (K + V)	Yes, gas	Suspended	1, 2, 3
Amungee	EP 98	16°20'50"S,133°53'4"E	2609	Yes (K + V)	Yes, gas	Suspended	1, 2, 3
West Beetaloo 1	EP 117	17°7'14"S,133°45'42"E	3165	Yes (K + V)	Yes, gas	Suspended (see Figure 13)	1, 2 (spikes related to vehicle)
Shenandoah 1	EP 98	16°37'11"S,133°34'44"E	2703	Yes (K + V)	Yes, gas	PNA	1, 2
Burdo	EP 23	16°15'4"S,134°30'37"E	1239	Yes (K)	Yes, gas + oil	PNA, historical 1980s bore	1, 2, 3
Tanumburini 1	EP 161	16°23'57"S,134°42'14"E	3946	Yes (K + V)	No oil, but gas reported	Drive along pastoral track close to bore?	1, 2, 3

Figure 13: CH_4 measurements were acquired at the Birdum Creek plugged and abandoned well site.

Figure 14: Collecting CH_4 and wind measurements at the West Beetaloo 1 suspended well.

WATER WELLS

In addition to the petroleum wells, CH_4 concentration measurements were made at (or close to) 25 water bores during at least one of the mobile survey campaigns. Details of the water bores, including the location and when they were visited or was close to the survey track are shown in Table 5.

Elevated concentrations were not detected in the vicinity of most of these bores. At a small number of these bores (marked with [1]* denoting the first campaign and [2]* denoting the second campaign in Table 5), elevated values were detected, but there were also cattle close by. It was not possible to discriminate between CH_4 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 (hundreds at each locations), cattle are likely to be the dominant influence. Elevated CH_4 concentrations were consistently recorded over a period of five days close to the Daly Waters Motel in the first mobile survey campaign (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 CH_4 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.

A revisit of the site during the second survey also recorded elevated CH_4 concentration at this site, which was similarly small. However, no CH_4 was detected during the third survey. This may be due to the differing wind conditions and, potentially a smaller number of occupants during the wet season.

Table 5: Water bores investigated.

Water Bore Name	Latitude, Longitude (locations mostly are shown on Figure 10)	Comments	Visited during campaign number
1. Water Bore close to Tarlee 2	15°57'21"S,132°39'5"E	Measurement collected at bore close to Tarlee 2.	1
2. Jabiru	16°57'25"S, 134°18'22"E	Measurement collected at bore at Amungee Station (see Figure 15)	1
3. Motel Bore# RN24618	16°18'28"S, 133°23'9"E	Measurement collected approximately 500 m from a bore at Daly Waters Motel during campaign 1	1, 2, 3
4. Unnamed water bore at Beetaloo Station	16°41'27"S, 132°58'52"E	Measurement collected close to the bore	1, 3
5. RN029012	15°16'16"S, 133°8'32"E	Drive along Stuart Hwy close to the bore	1, 2
6. RN038810	15°22'24"S, 133°9'55"E	Drive along Stuart Hwy close to the bore	1, 2
7. RN038811	15°29'23"S, 133°11'42"E	Drive along Stuart Hwy close to the bore	1, 2
8. RN028082	15°35'43"S, 133°13'34"E	Drive along Stuart Hwy close to the bore	1
9. RN029013	15°16'16"S, 133°8'32"E	Drive along Stuart Hwy close to the bore	1
10. RN005942	15°16'16"S, 133°8'32"E	Drive along Carpentaria Highway close to bore along Carpentaria Highway	1, 2
11. RN005764	15°17'35"S, 133°36'44"E	Drive along Carpentaria Highway close to bore along Carpentaria Highway	1, 2
12. RN5844	15°20'51"S, 133°54'54"E	Drive along Carpentaria Highway close to bore along Carpentaria Highway	1, 2

13.	RN38109	16°33’8”S, 133°58’48”E	Drive along pastoral tracks close to the bore	1
14.	RN037655	16°28’58”S, 134°33’59”E	Drive along pastoral tracks close to the bore	1, 3
15.	RN033608[1*2*]	16°27’22”S, 134°38’53”E	Drive along pastoral tracks close to the bore	1, 2, 3
16.	RN039693	16°29’9”S, 134°38’11”E	Drive along pastoral tracks close to the bore	1, 2, 3
17.	RN038179	16°25’28”S, 134°36’7”E	Drive along pastoral tracks close to the bore	1, 2, 3
18.	RN008101[2*]	16°24’57”S, 134°40’27”E	Drive along pastoral tracks close to the bore	1, 2, 3
19.	RN007659[1*2*]	16°19’42”S, 134°42’48”E	Drive along pastoral tracks close to the bore	1, 3
20.	RN033671[1*2*]	16°23’8”S, 134°38’1”E	Drive along pastoral tracks close to the bore	1, 3
21.	RN031244	16°35’14”S, 134°42’27”E	Drive along pastoral tracks close to the bore	1, 3
22.	RN38818	133.941119 -17.907635	Stuart Hwy	2, 3
23.	RN38817	133.721827 -17.743166	Stuart Hwy	2, 3
24.	RN38815	133.443144 -17.030469	Stuart Hwy	2, 3
25.	RN031244	16°35’14”S, 134°42’27”E	Drive along pastoral tracks close to the bore	2, 3

Figure 15: Methane measurements were collected at the Jabiru water bore at Amumgee Station.

PIPELINE RISER

During the first mobile survey campaign, 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, above average concentrations up to approximately 2.3 ppm CH_4 were detected at a pipeline riser adjacent to tracks covered on the Tanumburini station (see the photo in Figure 16). The measurements were collected next to the fence approximately 10 m from the pipeline. This pipeline riser also had several valves attached. Figure 17 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.

Figure 16: Photo of the above ground section on the Daly Waters to MacArthur River gas pipeline where above average CH_4 values were found. The geographic location of the site is 16.5437°S, 134.7087°E.

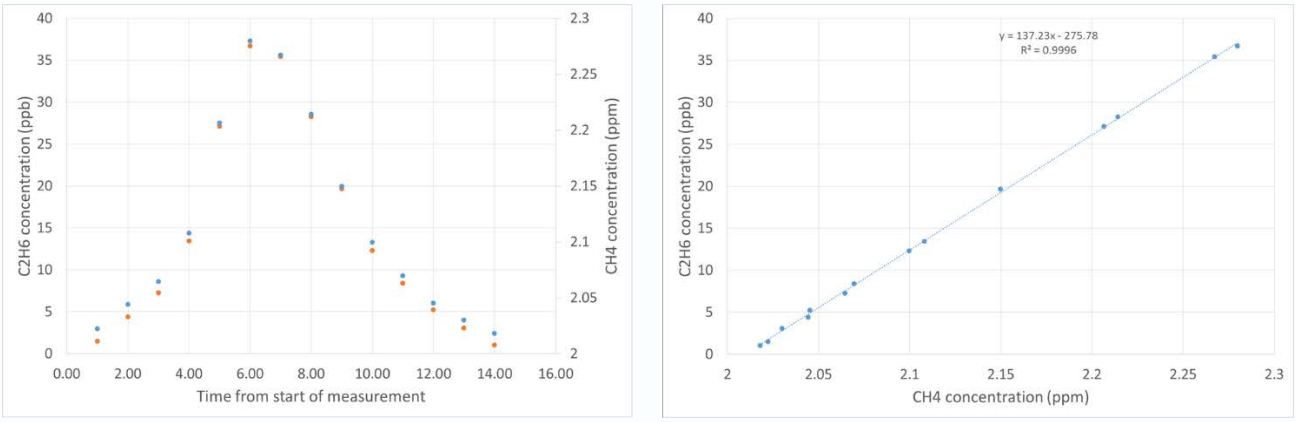

Figure 17: Left: CH_4 and C_2H_6 concentration measured at an above ground section of the Carpentaria gas pipeline. Right: Correlation between the CH_4 and C_2H_6 concentrations measured at the gas pipeline.

A revisit of pipeline riser was made specifically during the second mobile survey campaign, and data were collected around the site. The measurements made during this survey confirmed the presence of elevated CH_4 levels at the site. The results are shown in Figure 18, where the maximum elevated concentrations above the background are 0.07-0.08 ppm. These CH_4 concentrations measured at about 10 to 15 m downwind from the facility (unable to get closer due to the fence) during these visits were low, suggesting that the source of CH_4 was minor. Therefore, no further investigations were performed on that occasion to measure the emission rate because the elevated levels were small, equivalent to the levels detected in the region for cattle.

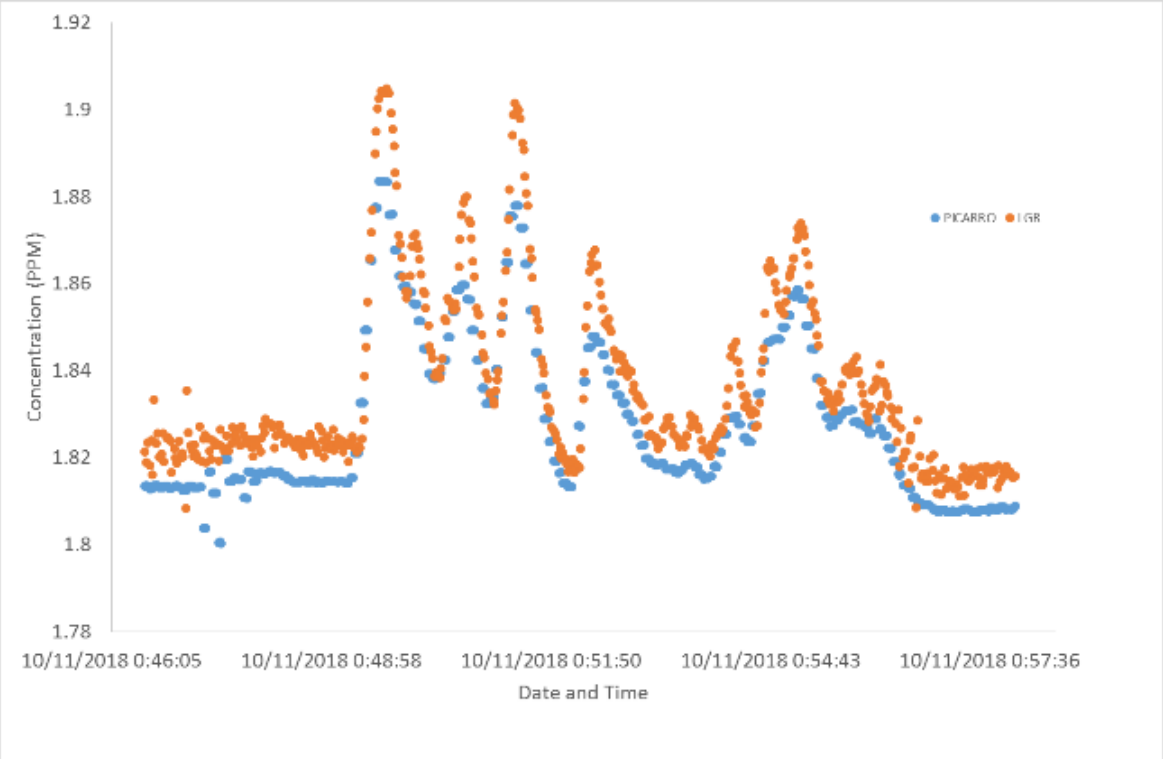

Figure 18: PICARRO and LGR CH_4 concentrations detected around a section of the Daly Rivers to MacArthur River pipeline.

In the third mobile survey campaign, we attempted to quantify the pipeline riser CH_4 source, initially using the acetylene tracer method described in Section 3.1.1. This method has been previously shown to yield accurate results when applied to CSG wells and other CH_4 sources (Day et al., 2016). However, because the pipeline riser was inside a fenced enclosure, we were unable to position the acetylene outlet near the likely source. In some cases, this can be mitigated by conducting concentration measurements further downwind, but the presence of thick vegetation restricted measurements to no more than about 20 m from the source, which was insufficient to allow acceptable mixing of the tracer acetylene with the source CH_4 plume.

Instead, we used the plume traversing method (Section 3.1.2) to estimate the emissions rate from the pipeline riser. The conditions for this method were favourable at the time of the visit with a steady breeze of about 3 m s^{-1} and consistent direction prevailing. Six circuits over about a 10-minute period were made around the fence about 15 m from the source. Like previous surveys, the peak CH_4 concentrations were very low, with the maximum concentration measured during the traverses about 0.06 ppm above background. The results of the traverses are plotted in Figure 8, where the CH_4 peaks detected during the traverses are clearly visible.

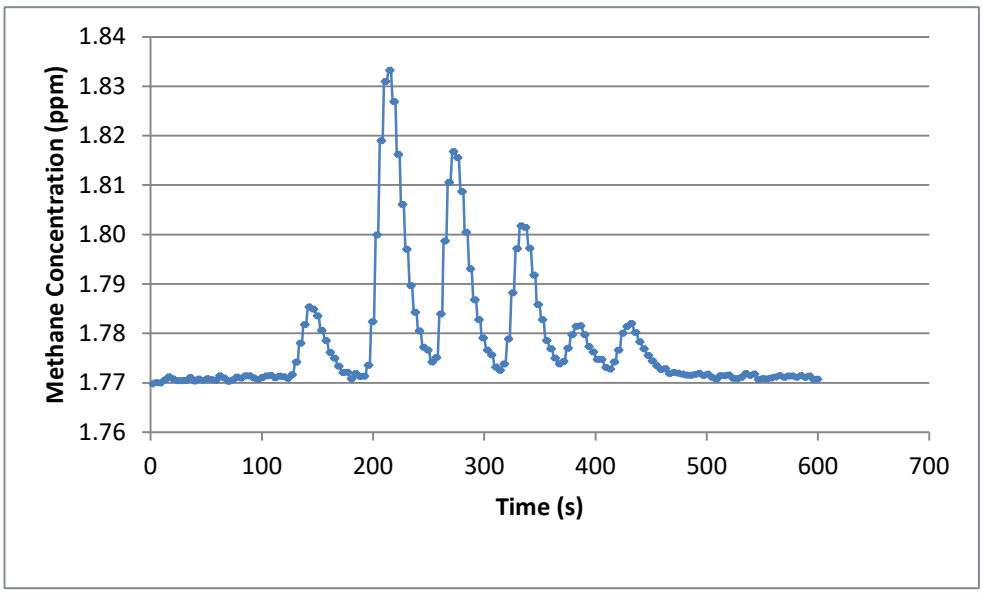

Figure 19. Methane concentration measured during six mobile traverses around the pipeline riser enclosure. The peaks measured during each traverse are clearly visible with the maximum concentration (second traverse) slightly above 1.83 ppm or less than 60 ppb above background.

The average emission rate estimated from the six individual traverses was approximately 43.8 kg CH_4 yr^{-1}.

FIRE

The other elevated CH_4 concentrations that are visible on the regional scale displayed in Figure 10 and Figure 11 can be attributed to fires. For example, in Figure 10, two sources that were located north of Newcastle Waters along the Stuart Highway. These fires 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 on Figure 20. 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 21. 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 21). 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 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 in Figure 22 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 20: Burnt grass at the side of a road.

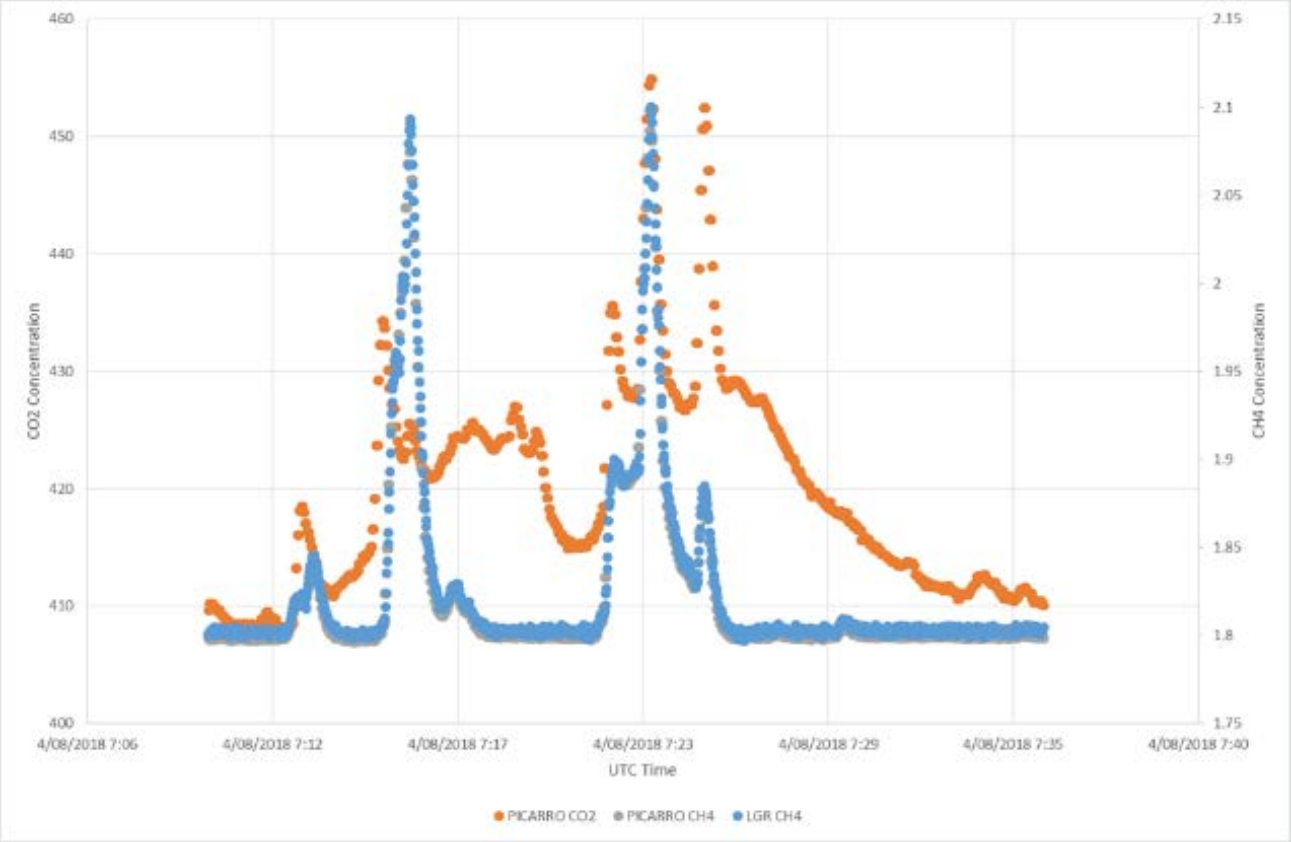

Figure 21: Concentration of CH_4 and CO_2 detected by the PICARRO and LGR analysers at a roadside grass fire on 4 August 2018.

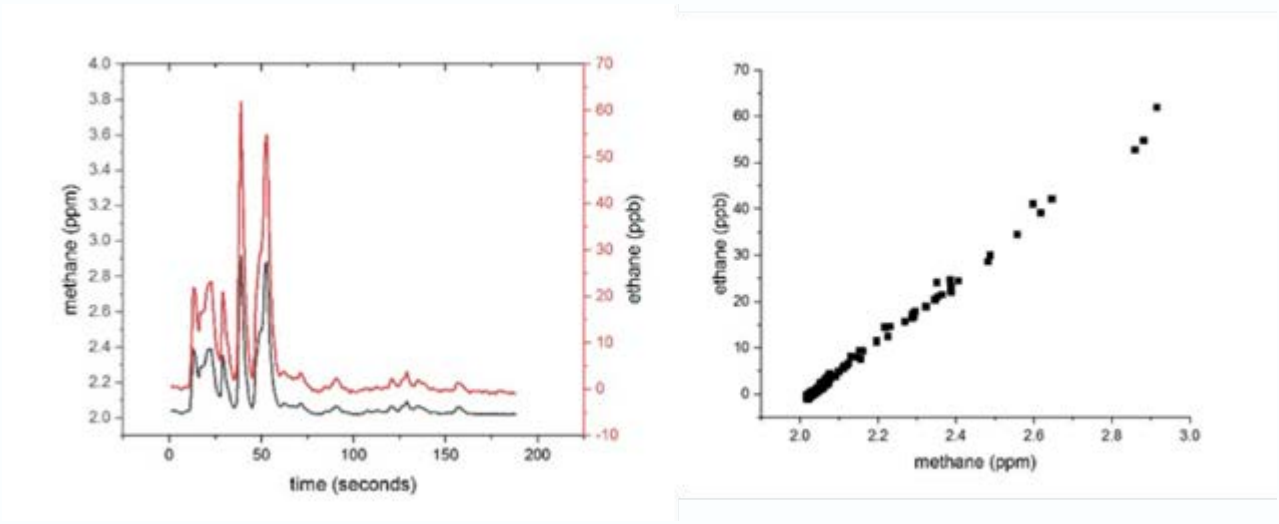

Figure 22: Left: CH_4 and C_2H_6 detected at roadside grass fires on 4 August 2018. Right: Correlation between CH_4 and C_2H_6.

Fires were also detected on the second mobile survey campaign. An example can be seen in Figure 11, where a grass fire close to the roadside on Larrimah Western Creek Road was detected on the 14th November 2018. This fire was observed for an extended period while the vehicle was travelling on the side of the road. The concentration of CH_4 detected by the PICARRO and LGR is shown in more detail in Figure 23. There is a good correlation between the concentrations measured by both analysers and the maximum value detected was close to 1.98 ppm detected by the LGR.

CO_2 was also detected and showed similar trends to the CH_4 (also in Figure 23). Unlike the previous fires detected in the first mobile survey campaign, there is a better correlation between the CH_4 and CO_2, suggesting that both gasses are derived from the same source.

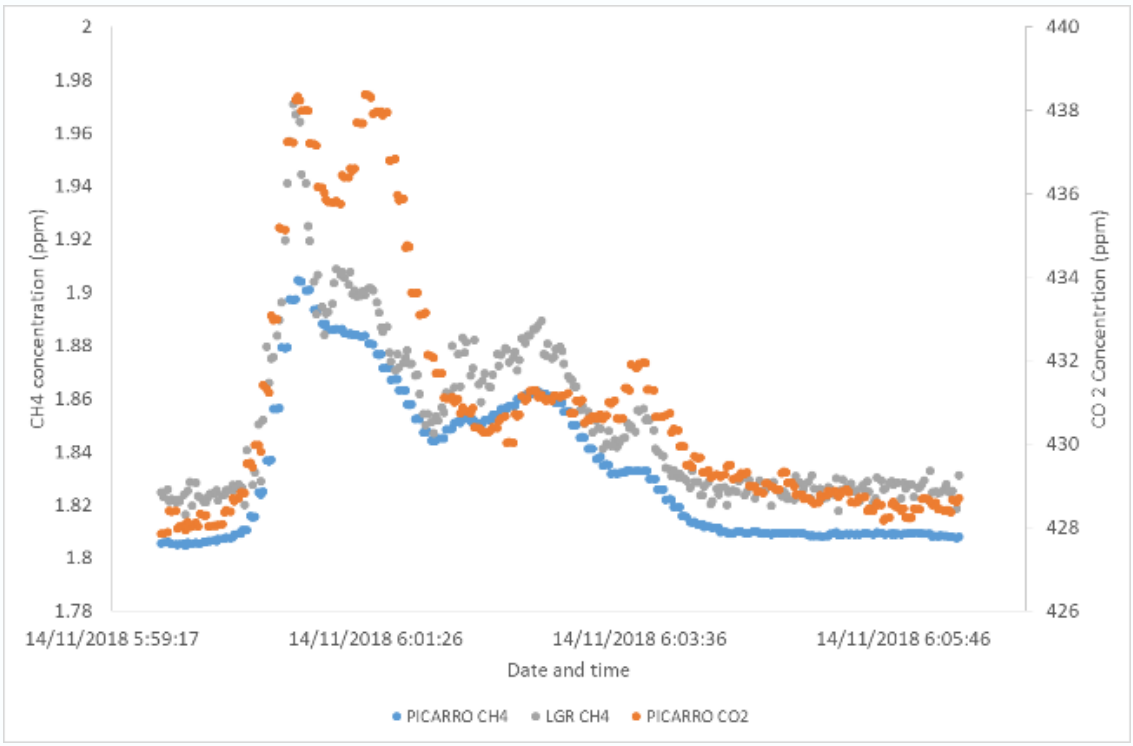

Figure 23: Concentration of CH_4 and CO_2 detected by the PICARRO and LGR analysers at a roadside grass fire on 14th November 2018.

Although the second mobile survey campaign was conducted during the fire season, only one fire was sufficiently close to the survey tracks to be detected although smoke plumes were observed. 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 better quantification of flux. Examination of fire scars mapped using remote sensing produced by Northern Australia Fire Information (NAFI) for the period of the survey showed a fairly large fire scar mapped in the vicinity confirming the presence of a fire. This is shown in Figure 24, which plots the CH_4 concentration measured by the LGR analyser overlaid with the fire scars mapped from remotely sensed data during the survey period. Further examination of Figure 24 indicates that there were additional areas where elevated CH_4 concentrations detected by the LGR coincided with fire scars mapped from remote sensing. Specifically, these were 1) the elevated CH_4 concentrations detected south of Elliot, and, 2) the area close to the cross road between Buntine and Buchanan Hwy. Cross checking with the CO_2 concentrations measured by the PICARRO analyser shown in Figure 25 shows that elevated CO_2 were also detected at these locations. Therefore, it is likely that fires were present as mapped with remote sensing.

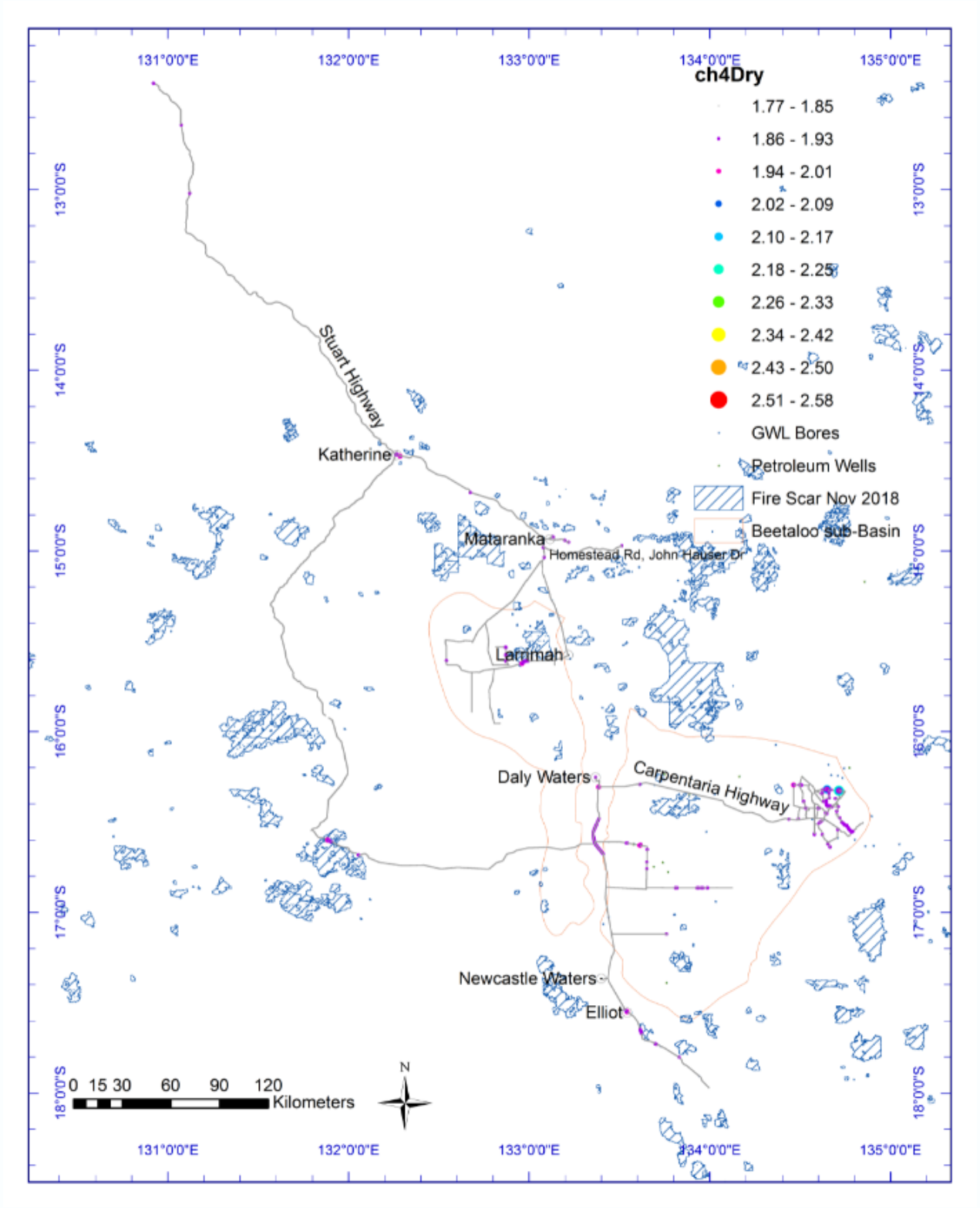

Figure 24: Methane concentration measured using the LGR analyser along tracks and roads across the Beetaloo sub-basin. The cross hatched area shows fire scars mapped from remotely sensed data extracted from Northern Australia Fire Information (NAFI).

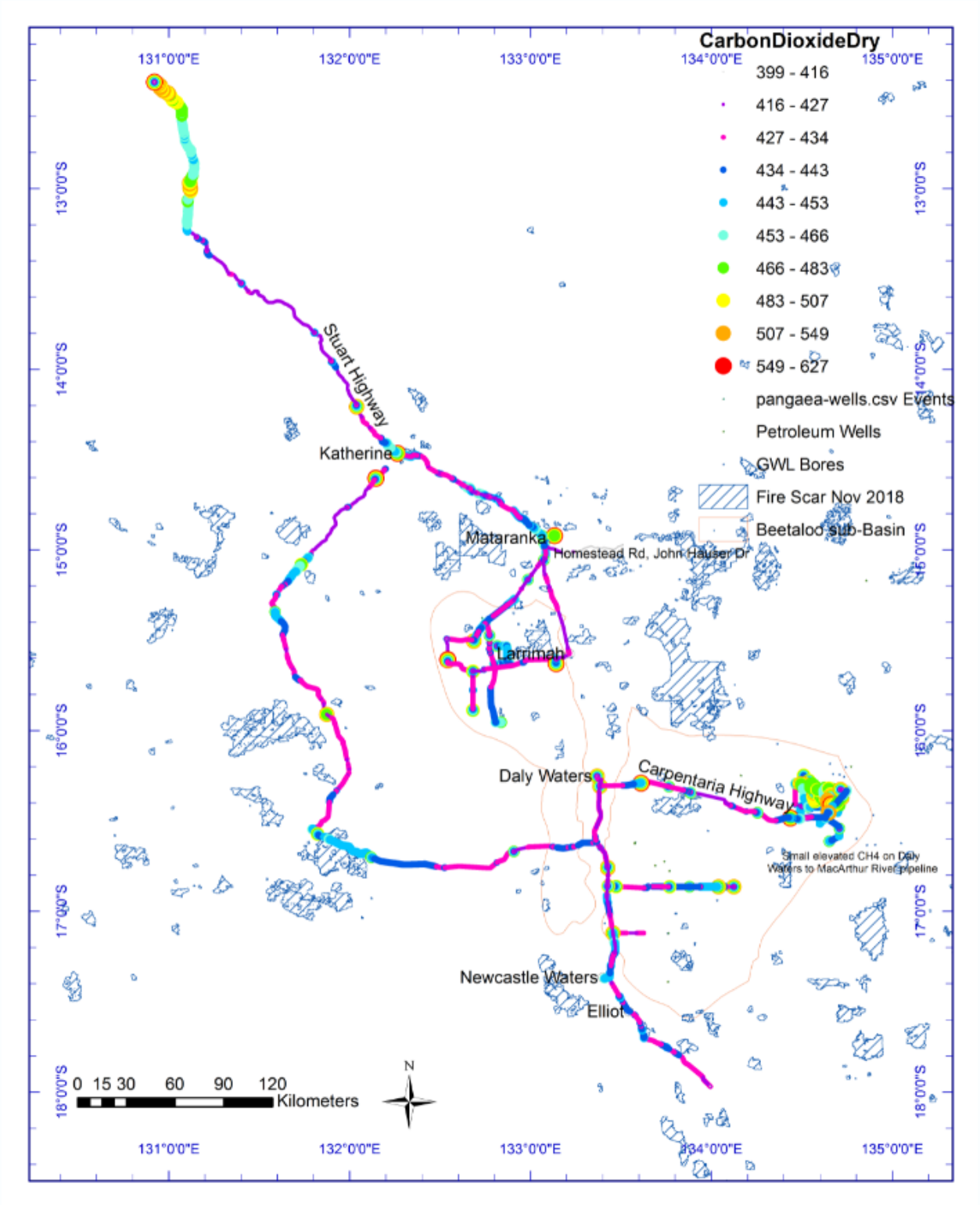

Figure 25: Carbon dioxide concentration measured using the PICARRO analyser along tracks and roads across the Beetaloo Sub-Basin. The cross hatched area shows fire scars mapped from remotely sensed data extracted from Northern Australia Fire Information (NAFI).

WETLANDS

Natural sources of CH_4 expected in the area are wetlands and natural geological sources such as springs. During the first mobile survey campaign, CH_4 concentration measurements were collected 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 0.30 ppm above background were detected at Mataranka Springs (see the photo of the location in Figure 26) as shown on Figure 27. Above average CH_4 concentrations were also detected at Rainbow Springs. For both of the CH_4 elevated concentration recorded, the CH_4 concentrations were not correlated to the C_2H_6 concentrations. The concentration of C_2H_6 were below 0.01 ppm (Figure 27).

Figure 26: 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 and uncorrelated, the sources are likely to be of a biogenic nature (Yacovitch et al. 2014).

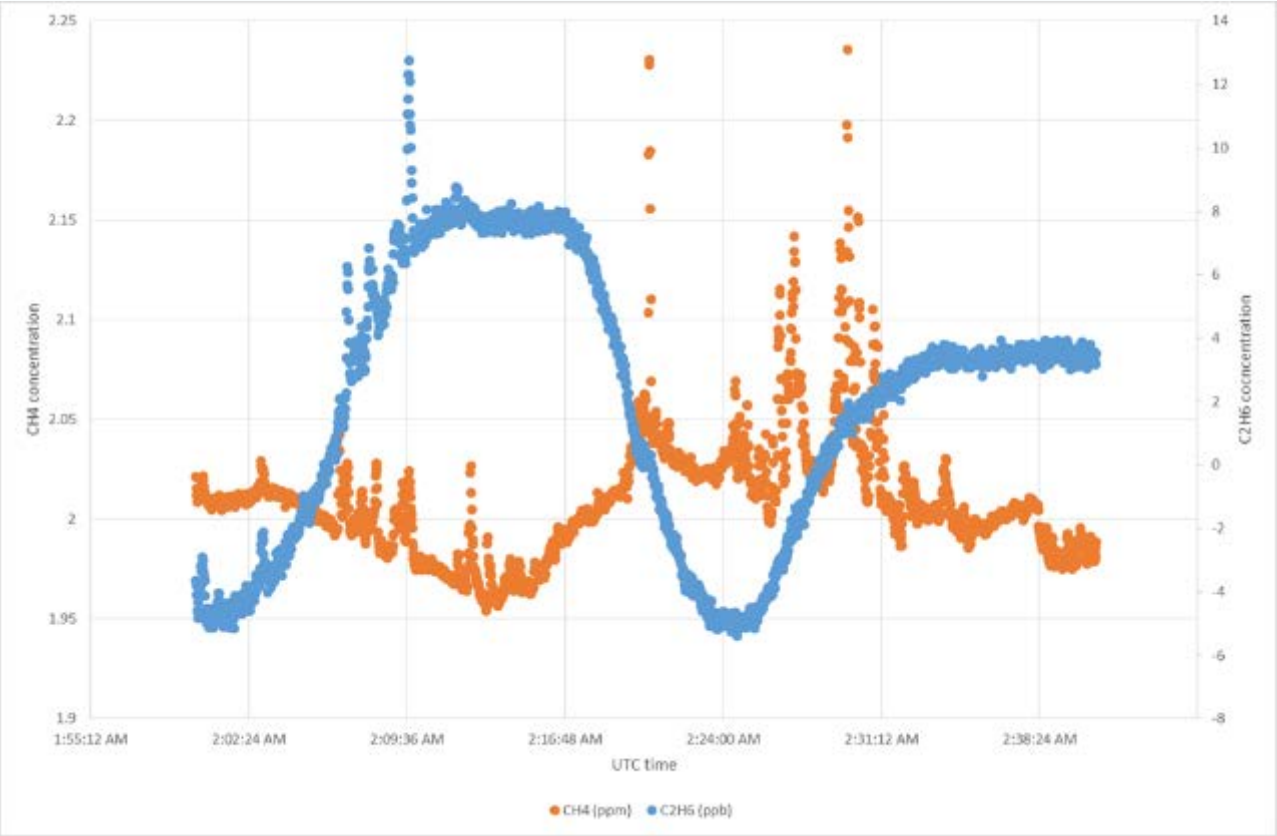

Figure 27: CH_4 and C_2H_6 concentrations recorded at Mataranka Springs.

A revisit of the wetlands along the public roads Homestead Rd and John Hauser Rd to determine the possible presence of CH_4 from the wetlands area around Bitter Springs and Mataranka Springs were undertaken in the second and third campaign. No elevated concentrations were detected along these roads on both occasions. The AERIS analyser was not available during these two later campaigns measurements were not collected near the springs as in the first survey.

TERMITES

The other likely natural source of CH_4 in the region are termites. A total of 8 termite mounds were measured during the first mobile survey campaign. 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 28) to a large mound approximately 1 × 1.6 m in size similar to the one shown in Figure 29. 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 30. This figure also shows the concentration of C_2H_6, which in contrast is very low (approximately 0.008 ppm). The CH_4 and C_2H_6 concentrations measured in the mound are plotted in the right hand graph in Figure 29, indicating that there is no apparent correlation between these measurements. This lack of correlation makes sense since termites are a biological source of CH_4.

Figure 28: Small termite mound measured with a 9-litre flux chamber.

Figure 29: 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).

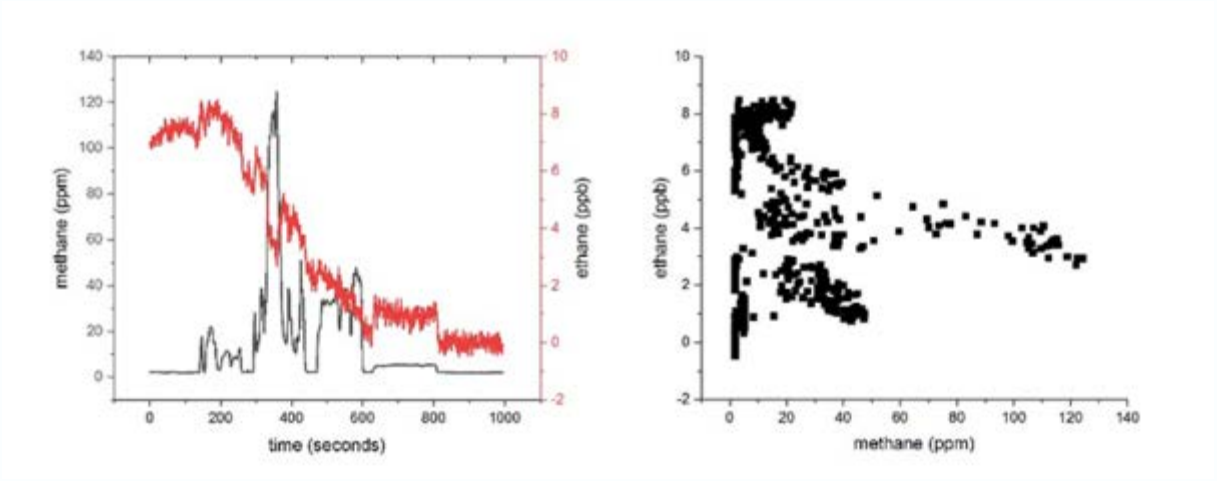

Figure 30: Left: Concentration of CH_4 and C_2H_6 recorded by the AERIS analyser when tubing was inserted into a large termite mound. Right: Concentration of CH_4 plotted against C_2H_6 indicating that the two gasses were not correlated.

The lack of emissions found in the first mobile survey campaign was consistent with seasonal variability of emissions, and hence further measurements of CH_4 emissions were attempted during the third mobile survey campaign conducted during the wet season.

Measurements were made at three small termite mounds that could be fully enclosed in the 9 L flux chamber. In each case, CH_4 emissions were found. An example of the CH_4 concentration change over time within the flux chamber during one experiment is shown in Figure 31.

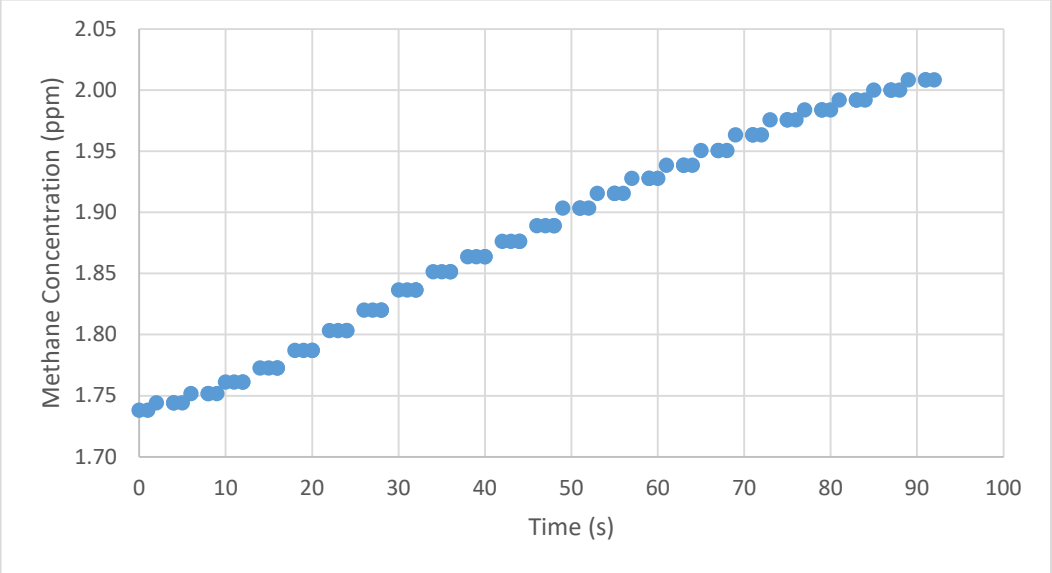

Figure 31. Methane concentration as a function of time inside the flux chamber during measurement of emissions from a termite mound.

The CH_4 emissions measured on these mounds were reasonably consistent, ranging from 2.7 to 4.1 mg CH_4 per m^3 per day with an average value of 3.3 mg CH_4 per m^3 per day (note that the units used here are in terms of the mound volume rather than the area covered by the flux chamber as in other chamber results). There was also a strong CO_2 emission from the termite mounds averaging at approximately 1200 mg CO_2 per m^3 per day. Other studies have found similar rates of both CH_4 and CO_2 emissions in the Northern Territory (Jamali et al. 2011a) and in other parts of the world (Seiler et al. 1984).

As noted previously, flux measurements made at the time of the first mobile survey campaign during the dry season yielded virtually no CH_4 emissions from a termite mound in the survey region. Jamali et al. (2011a) conducted an extensive study of CH_4 emissions from termites in the Northern Territory and reported a

ninefold difference in emission rates between the wet and dry seasons. This strong seasonal effect is consistent with our results.

SOIL EMISSIONS

Soil CH_4 fluxes were measured at eight sites throughout the third mobile survey campaign, and the results are summarised in Table 6 (in units of mg CH_4 m^{-2} day^{-1}). The last four measurements shown were made around a large stagnant water body on the Tanumburini Station (Figure 31).

Figure 32. Ground surface flux measurements near a body of stagnant water.

Table 6. Soil CH_4 fluxes measured during the survey.

Site Surface Description	Methane Emission Flux (mg CH_4 m^{-2} day^{-1})
Grassed edge of track – damp soil	-2.3
In free water on the grassed verge (see photograph in Figure 3)	-1.4
Dry ground without vegetation	-3.8
Dry ground without vegetation	0.5

Grassed edge of a large stagnant water body; Location 1	98.0
Grassed edge of a large stagnant water body; Location 2	5.1
Grassed edge of a large stagnant water body; Location 3	23.3
Stagnant water body – in the water	113

The small sample of measurements shows a significant range of CH_4 fluxes with the highest associated with the stagnant water body. Some of the fluxes were negative (i.e. indicating that CH_4 is being consumed from the atmosphere). This is consistent with other measurements made in Australia and other parts of the world (e.g.(Dalal et al. 2008; Day et al. 2016a; Day et al. 2015; Ong et al. 2017) and is due to the presence of microorganisms capable of oxidising CH_4 in aerated soils.

Measurements made near the stagnant water were significantly higher with the maximum emission rate of 113 mg CH_4 m^{-2} day^{-1}. This is not unexpected since wetlands are well known to produce CH_4 due to anaerobic microbial activity. This particular site was not a permanent wetland and for much of the year would be dry; it is likely, therefore that emissions from the site would also be much lower.

Although the CH_4 fluxes from the wetland examined during this survey were significantly higher than other non-wetland areas, the highest emissions were generally less than 0.1 g CH_4 m^{-2} day^{-1} which was usually not sufficient to produce measureable elevated ambient CH_4 concentrations in the vicinity. However, the extent of wetlands during the wet season means that CH_4 from this source is likely to be a significant source of the CH_4 in the Sub-Basin, although the magnitude will be highly dependent on seasonal conditions.

Figure 33 shows the CO_2 concentrations recorded by the PICARRO analyser for the first mobile survey campaign. This figure is representative of CO_2 concentration and their sources. That is, 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 the 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 first mobile survey campaign and the second and third mobile survey campaigns were confounded by CO_2 produced by nearby vehicles.

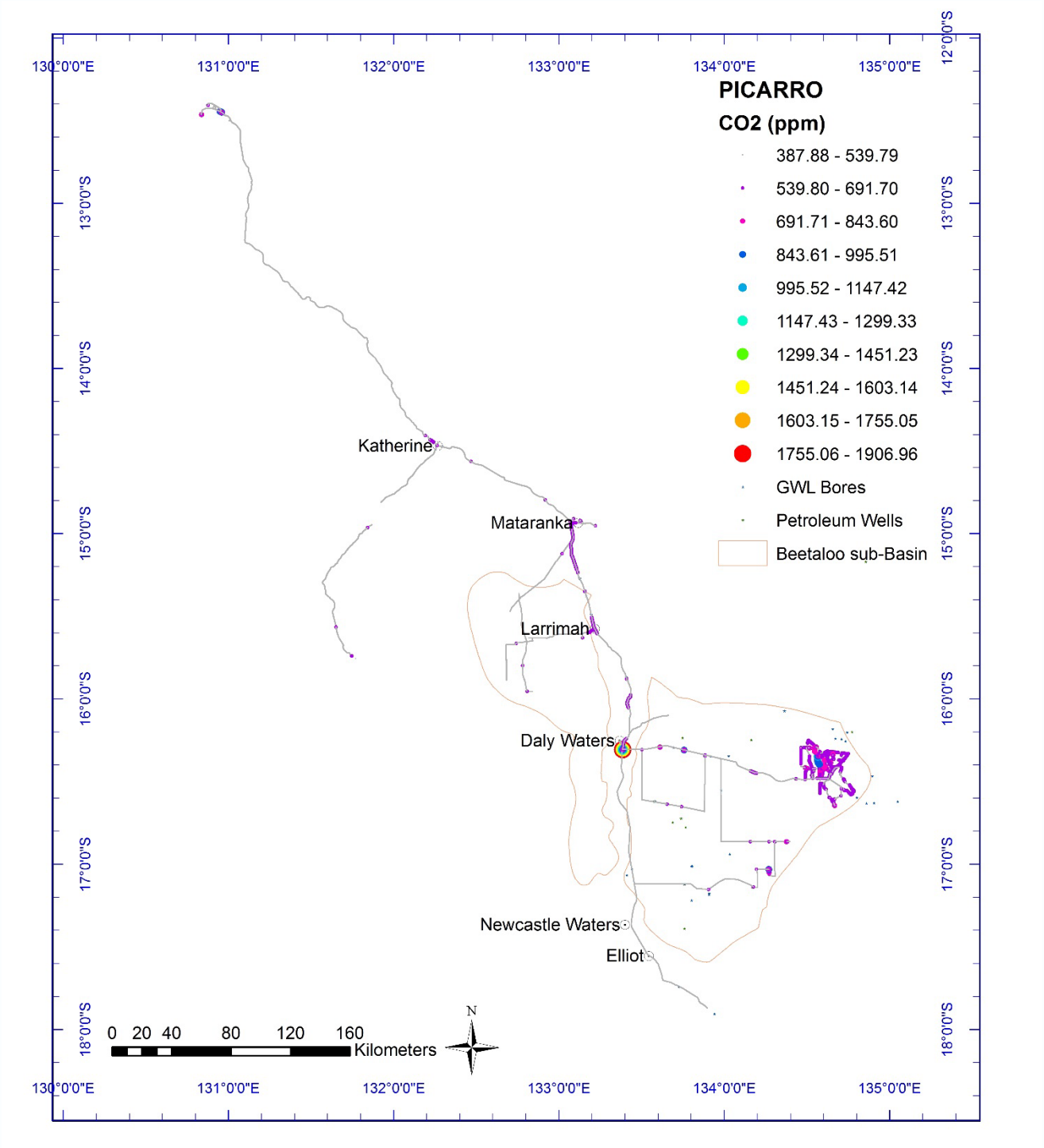

Figure 33: Carbon dioxide concentration measured by the PICARRO analyser during the first mobile survey campaign conducted during 29th July – 10th August 2018.

6 Discussion and Conclusions

Overall, the majority of elevated CH_4 concentrations recorded in the survey area for all the mobile survey campaigns were related to cattle. This is in line with the fact that cattle production is currently the dominant industry in the area and hence is likely to be the dominant 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 and estimate from those measurements the emission rates 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 in the survey area. 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 calculate the emission rates from these dispersed sources quantitatively. In the case of the Beetaloo Sub-basin the average of the emission factors from Charmley et al. (2016), specifically 63.88 kg yr^{-1} per beast was used for the estimation. In addition, the aggregate of the carrying capacity for all the stations that fall within the Beetaloo Sub-basin totalling 115,876 cattle was sourced from NTCA (2019) and was used together with the emission factor to provide an estimated total emission of 7,402,159 kg CH_4 yr^{-1} from this source. Note that the carrying capacity is not necessarily the actual numbers in the stations and will vary according to a number of factors including climatic conditions.

Secondary sources of elevated CH_4 concentrations of between 1.85 to 2.09 ppm were recorded at townships along the Stuart Highway for all three mobile survey campaigns. These concentrations are similar to values recorded at other towns or cities and are believed to be related 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.

No elevated CH_4 concentrations were detected at the plugged and abandoned petroleum wells and most of the water bores that were specifically investigated. Moving into the future, it is likely that gas exploration will start in the next 6-12 months. Before drilling activities begin, there is an opportunity to extend the baseline measurements to include a comprehensive soil CH_4 baseline around the well pad areas to capture the natural background CH_4 emission of the surrounding area. Studies have been conducted in the USA to estimate the emissions related to the hydraulic fracturing process (Howarth et al. 2011; O'Sullivan and Paltsev 2012) which indicate that these operations could be a significant source of CH_4. However, in Australia, there is currently a gap in understanding on the emissions that is likely to be produced during this operation. Therefore, it would be important to collect CH_4 measurements throughout the hydraulic fracturing operations to understand the emissions related to the hydraulic fracturing operations and related flow back. In addition, it may also be useful to install remote monitoring stations at this stage according to the Scientific Inquiry recommendations 9.3 to provide continuous monitoring of the operations and infrastructure. With the small footprint of most remote monitoring stations, the optimal useage is likely to be in such application, that is, close to gas infrastructure to provide continuous monitoring of the operations of the infrastructure. However, this has to be weighed against the cost and uncertainties of the towers.

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.

During the first mobile survey campaign, above average CH_4 concentrations which were highly correlated with C_2H_6 concentrations were detected approximately 10 m from the fence adjacent to a pipeline riser 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. A second visit to the same pipeline riser area confirmed the presence of above average CH_4 concentrations that were approximately 0.07-0.08 ppm above the background value. Following the report of this leak, a service provider to the owner of the pipeline at the request of the Department of Primary Industry and Resources

investigated it and found that during their monthly inspection no leak had been detected. However, an additional survey was initiated on 8th January 2019, which detected a small leak that originated from a grease nipple on a ball valve. The nipple cover was tightened as it was found to be only finger tight. A gas level reading was again taken 150 mm from the nipple which indicated there was no leak present. An additional step has now been include in the valve check sheet, which requires that all grease nipple covers be tightened. This pipeline riser was again visited during the third mobile survey campaign where, the emission rate from this facility was estimated and found to be approximately 43.8 kg yr^{-1}. To put this into context, this emission rate is 60-80 % of the average of 54.75 to 73.00 kg CH_4 yr^{-1} produced per head of cattle (Charmley, 2016; Tomkins, 2011). It must be also be noted that this is a small leak and falls under the threshold of "reportable leak" as defined as by the Northern Territory 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 5000 ppm (DNRM 2018). In addition to leaks, some pneumatic devices such as valve controllers, pressure regulators, etc. operate off natural gas pressure and are designed to vent small amounts of CH_4 during normal operation. A possible explanation for the CH_4 observed at the pipeline riser is the operation of such devices at the times of the site visits.

Fires were minor sources recorded during the first and second mobile survey campaigns. All the fires measured were spatially small sources, and above average concentrations of up 0.30 ppm 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).

Although elevated CH_4 concentrations were recorded for other spatially larger fire or smoke plumes were observed from the survey vehicle, 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 (eg. a total of ~1520 km^2 was estimated to be burnt in the Beetaloo Sub-Basin areas in 2018), methods which provide the spatial comprehension incorporating spatial data such as remote sensing could potentially provide more quantitative quantification of emission rates. Additionally, the contribution of CH_4 from fires is transient and seasonal, suggesting that methods which can collect the measurements remotely to capture these variations are required. Therefore, it would be useful to investigate methods such as those developed by Russell-Smith et al. (2009) using remote sensing technology and consider the method as a base template for further development specifically for quantification of CH_4. In addition, satellites that specifically measure greenhouse gases such as CH_4, CO_2 and carbon monoxide (CO) would be worth investigating. For example, the European Space Agency's Sentinel-5P (Veefkind et al. 2012) measure CH_4 and CO concentrations with a spatial resolution of 7 km may provide the ability to quantify and discriminate the fires from other sources of CH_4.

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 during any of the three mobile survey campaigns. Further investigations into the spring area during the first mobile survey campaign did record above average levels. As termite is a biogenic source of CH_4, it is not surprising that there is a lack of correlation between the CH_4 and C_2H_6 measurements. 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). Unfortunately, the AERIS analyser used in the first survey to walk into the spring area was not available during the third mobile survey campaign conducted during the wet season. In future, if access to the spring is available during the wet season, it may be useful to collect CH_4 measurements from this source in this season.

Termites are one of the sources of CH_4, but their contributions to the global budget are one of the most uncertain. In the Beetaloo Sub-Basin, they are a widespread CH_4 source, but their emissions are subject to large seasonal variations. Measurements made during the third mobile survey campaign in the wet season

clearly showed CH_4 emissions, yet no emissions were detected from a selection of termite mounds during the first mobile survey campaign in the dry season consistent with findings that seasonal variations govern the CH_4 fluxes and the fluxes related to termite mounds were up to 3.5-fold greater in the wet season as compared to the dry season (Jamali et al. 2011a). With the measurements acquired at the Beetaloo Sub-Bain during the third mobile survey in the wet season, it is not possible on the basis of these few measurements to quantify termite emissions but previous work has estimated CH_4 emissions in northern Australia to be 0.24 kg CH_4-C ha^{-1} yr^{-1} or 0.32 kg CH_4 ha^{-1} yr^{-1}(Jamali et al. 2011b). Using this emission factor, the total emission in the Beetaloo Sub-Basin, which is approximately 28,000 km^2, is estimated to be approximately 900,000 kg CH_4 yr^{-1}.

Methane emissions from saturated soils were also detected during the third mobile survey campaign during the wet season. Given that such surfaces are very extensive during the wet these emissions, although the measurement made during this survey indicated that they are low, they are likely to account for significant CH_4 emissions to the atmosphere across the sub-basin. However, for much of the year, these areas will be dry and may likely become CH_4 sinks rather than sources. Previous studies by Jamali et al. (2011b) estimated that soil uptake of CH_4 was 1.14 kg CH_4-C ha^{-1} y^{-1} (1.52 kg CH_4 ha^{-1} yr^{-1}), which is nearly five times the amount of CH_4 emitted by termites. If this emission factor was used to estimate the soil sink for the Beetaloo Sub-Basin, the emission sink would be approximately 4,200,000 kg CH_4 yr^{-1}. Clearly, developing a detailed and accurate CH_4 budget for the region is a complex task and needs to account for both natural and other sources as well as the strong seasonal effects on these sources.

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