SKY-DIVING FOR SCIENCE: A NOVEL TECHNIQUE FOR RAPID VERTICAL TRANSECT METHANE SAMPLING

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Abstract

Methane has nearly 30 times the global warming potential of carbon dioxide on a 100-year timeframe, and so understanding methane emission sources, both point and fugitive, is critical to reducing its impact on the radiation budget.

Literature suggests that quantities of methane driven by surface emissions typically occur within the first few hundred metres above ground level. Satellite observations or downwind surface and aircraft surveys may not capture the complete methane profile in the atmosphere from anthropogenic sources, in particular between tens of metres to hundreds of metres above the Earth's surface. Consequently, additional research is required to understand the lower atmosphere vertical profile of methane in relation to nearby sources.

This study aims to address this research gap by using an AirCore atmospheric sampling system and Picarro Cavity Ring-Down Spectroscopy (CRDS) for the vertical transect measurement of methane in the atmosphere in New South Wales, Australia. Unlike previously studies, the AirCore sampling system is not employed using a balloon-parachute arrangement or an Unmanned Aerial Vehicle (UAV), but instead the AirCore will be fitted to humans completing parachute jumps, enabling rapid, targeted transects through the bottom few kilometres of the atmosphere.

The preliminary results of this study highlight the benefits of the skydiving sampling approach and indicate future improvements to be able to successfully monitor methane emissions from nearby anthropogenic sources. The results from this study will be used to better inform emission inventories of the sources and vertical distribution of methane in the atmosphere.

Keywords: Methane, AirCore, Sky-diving, Picarro, Measurements, Monitoring.

1. Introduction

1.1. Methane and carbon management

With an ongoing focus on reducing greenhouse gas (GHG) emissions, monitoring the sources and concentration of methane (CH_4) in the atmosphere is becoming increasingly important. The main reason for understanding CH_4 emission sources, both point and fugitive, is because CH_4 has between 27-30 times the global warming potential (GWP) of carbon dioxide (CO_2), expressed over a 100-year timeframe. Additionally, CH_4 emitted today lasts about a decade on average, which is much less time than CO_2 (USEPA, 2024). This means that there are potentially 'quick wins' in reducing the anthropogenic impacts on the climate through prioritising CH_4 emission reductions in the short-term.

1.2. Sources of methane

There are two major categories of sources of CH_4 in the environment. These are biogenic (natural) sources and anthropogenic (human) sources. For example, CH_4 naturally occurs as a by-product of microbial respiration. This typically requires saturated environments where microbes called methanogens can thrive. Ideal environments include wetlands, bogs and stagnant waters. Animals such as termites are another natural source of CH_4. Anthropogenic emissions of CH_4 include the anaerobic processes at landfills and sewage treatment plants. Direct industrial emissions typically occur due to fugitive releases of coal seam gas (CSG) from either CSG extraction or open cut and underground coal mining activities. Another large source of anthropogenic CH_4 is agricultural practices via ruminant digestion processes or rice paddies.

The percentage contribution for removal of CH_4 from the atmosphere is estimated as being 90% associated with hydroxyl radical ($OH^{\bullet}$) decay processes, 4% from soil oxidation via bacteria, with oxygen and chlorine radicals (from sea-air interactions) contributing the remainder (Chai et al. 2016).

Figure 1 depicts the main sources and sinks of CH_4 in the environment.

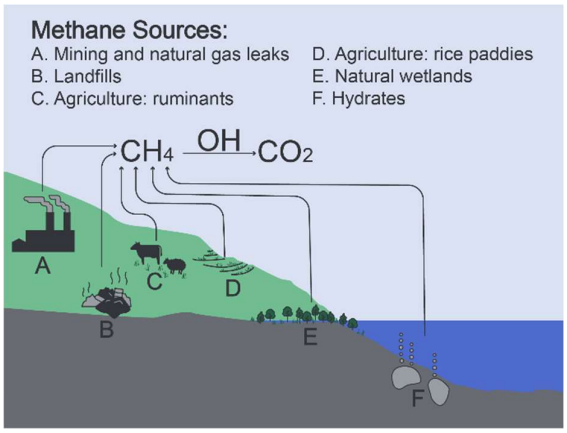

Figure 1. Sources and sinks of CH_4 in the environment.

1.3. Methane distribution in the vertical

Most of the material variability in GHG concentrations occurs within the Planetary Boundary Layer (PBL), and therefore targeting this lowest layer of the troposphere (typically up to between 1,000 to 3,000 m above ground level) is valuable for capturing the extent of plumes from emission sources, as well as small scale (temporal and spatial) variability.

A variety of different techniques exist for measuring CH_4 including satellite observations and downwind CH_4 surveys using in-situ techniques or aircraft. Existing surface measurements of CH_4 have shown considerable variability (Golston *et al*., 2017). In comparison, aircraft measurements have also shown enhanced CH_4 concentrations at 150 m (Neininger *et al*., 2021). The disparity in the literature between vertically resolved methane data may be because they are obtained in clean environments, have varying proximities to CH_4 sources, and that aircraft measurements do not reach low altitudes (less than 150 m) (Golston *et al*., 2017, Neininger *et al*., 2021).

One limitation to the existing measurement techniques available, is that they do not capture the CH_4 emission profile between tens of meters and 150 m in the atmosphere where significant amounts of CH_4 are expected be present when measurements are taken nearby to an emission source. This introduces an important data gap for fugitive emission sources, and therefore presents a need to accurately understand the vertical profile of fugitive CH_4 concentrations in the vicinity of both natural and anthropogenic sources.

1.4. Measuring methane in the atmosphere

AirCores are an innovative gas sampling system that were first developed by the United States National Oceanic and Atmospheric Administration (NOAA) in 2010 (Karion et al., 2010). The AirCore consists of stainless-steel tubing of sufficiently small diameter (typically 1/8-inch outer diameter) and varying length that allows for retention of profiles of the atmosphere (Labrèche et al, 2020).

The AirCore passively samples using changes in ambient pressure through the atmosphere (Karion et al., 2010). The Aircore has an opening at one end, and is closed at the other, allowing for the depressurisation of the AirCore's interior in its ascent to higher altitudes (Karion et al., 2010). The Aircore is then refilled in its descent as it samples the ambient gas as pressure is increased (Tans, 2022). This new ambient sample gas is subsequently introduced into the AirCore coil as it descends through the atmosphere, sampling at a rate that is dependent upon the fall speed, pressure difference and position aloft as it re-equilibrates (Tans, 2022).

The AirCore sampling system has become an important tool in assessments of atmospheric CH_4 and trace gas concentrations in the last decade (Baier 2023). The method's ability to take in situ measurements of columns of the Earth's atmosphere make it ideal for assessing spatial distribution of gases, in particular GHG gases (Asher et al., 2021).

The AirCore's characteristic of a slowly diffusing interior that allows for the subsequent analysis of the gas sample using a trace gas analyser, makes them a cost- and time-efficient and low management atmospheric sampling procedure (Baier 2023).

1.5. Conventional AirCore deployment

Weather balloons are commonly used to deploy AirCores (Baier et al. 2023), whilst more expensive methods include the use of unmanned aerial vehicles (UAVs) and within conventional aircraft (Tong et al. 2023).

Balloon deployment is not considered as reliable or accurate as UAV and aircraft-based sampling, however. Results can be subject to lapses in spatial resolution due to high recovery times and increased chance of equipment failure (Labrèche et al, 2020).

Weather balloon deployment typically requires extensive regulatory approvals. For example, in Australia, strict Civil Aviation Safety Authority (CASA) rules mean that balloon-borne AirCores can usually only be launched in remote areas. This provides a logistical challenge, and removes some of the utility of the method, for example in validating column measurements around anthropogenic GHG sources.

Given the passive nature of sampling, there is little control over where the payload descends. This means the potential for both long retrieval times and lost payloads.

A schematic showing the AirCore sample collection method using balloons is provided in Figure 2.

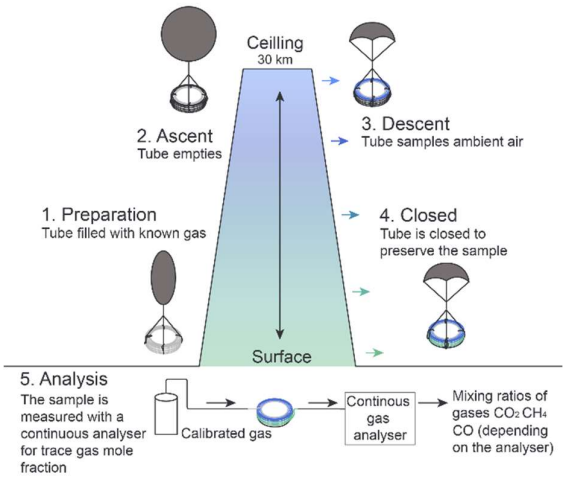

Figure 2. Schematic showing balloon-based AirCore sampling.

1.6. Aims and Objectives

Given the limitations of conventional AirCore deployment detailed above, there is motivation to explore alternative sampling methods.

This study aims to use AirCore sampling system for profiling the PBL, using commercial skydivers. Established literature on using skydivers as a mode of AirCore sampling is currently non-existent. However, the sampling principles for other airborne methods are applicable (Karion et al. 2010).

The use of commercial skydivers means that AirCores can be located at a reasonable distance to anthropogenic CH_4 sources without extensive approvals. The method is also cost effective relative to aircraft deployment, and allows for the targeted sample trajectory, controlled by the sky diver.

The sky diving deployment also minimises the time required to retrieve and close off the AirCore upon arrival at ground level. This avoids diffusion and dilution processes that occur during the recovery period, and therefore, increases the accuracy of column measurements.

2. Methodology

The current study uses an established AirCore monitoring technique used for vertical transect measurements of methane in the atmosphere.

2.1. Study locations

The areas selected for this evaluation are of interest in terms of CH_4 generation due to their proximity to several potential anthropogenic sources.

Picton is in the Southern Highlands region of New South Wales (NSW), Australia, an area of both historical and current coal mining activity and CSG extraction. For example, the monitoring location is approximately 12 km west of the Appin coal mine, 8 km southwest of Tahmoor colliery, and within a 30 km radius of the (now historical) Camden Gas (CSG) Project, along with several other underground coal mining locations (Dendrobium, Wollongong Coal), as shown in Figure 3.

The Hunter Valley (Elderslie) sampling location is close to a beef processing facility (and associated anaerobic wastewater treatment) as well as being located less than 30 km to the east of the major open cut coal mining of the Hunter Valley, NSW, as shown in Figure 4.

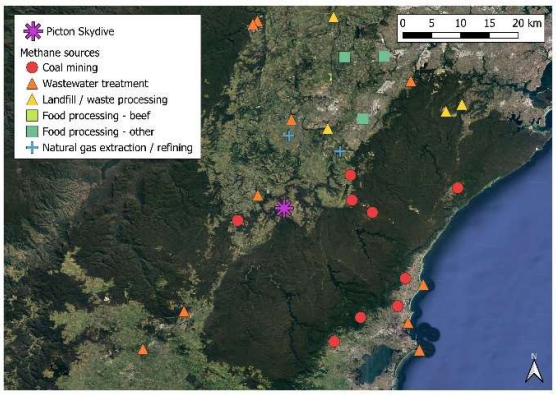

Figure 3. Picton location relative to potentially significant anthropogenic CH_4 sources.

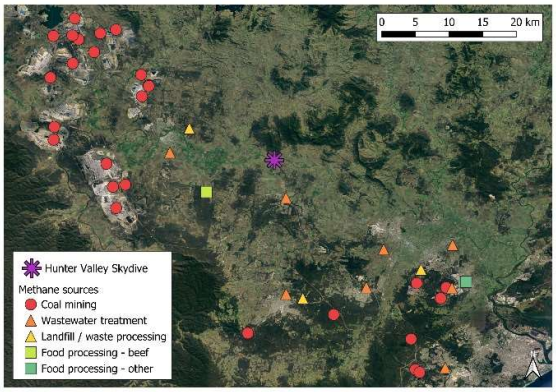

Figure 4. Hunter Valley location relative to potentially significant anthropogenic CH_4 sources.

2.2. Skydiving for science

A standard AirCore is packaged into a secure payload, which is attached to the front of the skydiver, as shown in Figure 5.

Figure 5. AirCore payload (A) deployed on skydiver (B).

The AirCore used here consists of 150 m of 1/8 inch stainless steel tubing, coated with SilcoNert™ to minimise the stickiness of sample to the tubing walls. The AirCore is pre-conditioned by flowing dry air of constant concentration through the coil into the analysis instrument until sufficiently low H_2O is measured, and the CO_2 and CH_4 mole fractions show minimal variability.

For this work, the maximum limits used to determine when the flow sample is constant and dry are as follows: H_2O below 0.010% (100 ppm), CO_2 variability (standard deviation) below 0.01 ppm and CH_4 variability less than 0.1 ppb. Both ends of the AirCore are then closed, and the pre-conditioning gas is also therefore used as a fill gas for the AirCore.

The AirCore is then placed into the payload (Figure 5), where it is strapped to prevent movement. One end of the tube is left protruding from the payload, to serve as the sample inlet. A Global Positioning System (GPS) is also secured within the payload to measure the position, pressure and temperature. Further information about position, height of plane exit, and total dive time is available from the skydiver's personal logging equipment. Once the skydiver has fitted their parachute, the payload is securely fastened to the diver. The diver opens the inlet upon plane take-off, creating a sample tube that is open at one end. As per the standard AirCore technique, the AirCore tube equalises pressure with its surroundings during the plane's ascent, causing partial evacuation of the AirCore.

Once the skydiver leaves the plane, the reverse process happens, with the pressure equalisation causing the AirCore to refill with air from the location of the diver. The narrow cross-section of the tube ensures minimal molecular diffusion, maintaining the pressure-dependent nature of the sample, which maps to the vertical location from which the air entered the tube.

Upon landing, the inlet of the AirCore is closed, preserving the sample. The payload is removed from the skydiver and transported to a GHG analyser for analysis.

2.3. Sample analysis

The measurement instrument used to derive mole fractions of CH_4 is a Picarro™ Cavity Ring-Down Spectrometer (CRDS) (Crosson, 2008). The CRDS uses a laser tuned to a characteristic wavelength for methane and shines this through the gas sample to be analysed.

For the measurements completed, a Picarro™ G2201-i analyser was used in high precision mode. The instrument measures $^{13}CH_4$, $^{12}CH_4$ (both in high range and high precision mode), $^{13}CO_2$, $^{12}CO_2$ and H_2O.

To provide a sample for analysis, the gas flows through a small internal cavity, or cell, within the CRDS. The relevant laser is shone into the cavity to detect the outgoing signal and hence the concentration of each species. The CRDS relies on maintaining a controlled temperature and pressure within its measurement cavity to reduce variations in concentration that can occur due to pressure and/or temperature changes. Prior to commencing measurements, the CRDS is allowed to stabilise to a standard temperature and pressure.

The same analyser is used during AirCore preparation to assess stability. Prior to analysis, the sample lines and connections outside the AirCore are flushed with gas of constant composition to once again achieve stability in CO_2 and CH_4 mole fractions, and low H_2O. Once that is achieved, the valves at either end of the AirCore are opened, and the sample pushed through, once more by a gas of constant composition and low H_2O. The end of the AirCore closest to the analysis instrument corresponds to the closed end of the AirCore during sampling. This ensures that the remaining fill gas within the AirCore is pushed through the analyser first. The arrival of the sample can be identified by a change from that constant composition. The end of the sample Is likewise identified by a return to a constant composition. The push and fill gases are of sufficiently different composition in one or more of the analysed gases to make this identification clear.

2.4. Mapping concentration to vertical profile

To map the analysed concentrations to a pressure-based profile, an approximate linear relationship between pressure during sampling (the skydive) and the time of analysis is assumed, and the analysed sample mapped to the corresponding pressures during the skydiver's descent. This assumption will

require revision to account for the different sampling rates in freefall and when the skydiver is under canopy.

At the start and end of the profile, there is a period of mixing between the fill gas and push gas, respectively, and the sample. To account for this, we implement a mixing model over the period of mixing.

The mole fraction of the fill/push gas is determined by the average of the 15 seconds prior to the sample mixing in. Over the period of mixing, and contributions of the constant gas and the real-world sample are assumed to vary. We investigate a model for this mixing, starting with a simple linear model. The linear model provides an excellent correction when the ratio of sample to constant gas is high, however breaks down closer to the ends of the sample when the sample contribution is low.

Figure 6 provides an example of the mixing effect at the start and end of the measured profile. The blue points indicate the original Picarro measurements, and the orange points are the corrected measurements accounting for mixing.

In this case the original 30 seconds and final 10 seconds (highlighted in grey in Figure 6) are excluded from the measured profile due to potential biases from the influence of the push and fill gases. Future work will focus on implementing the mixing model detailed by Tans (2022).

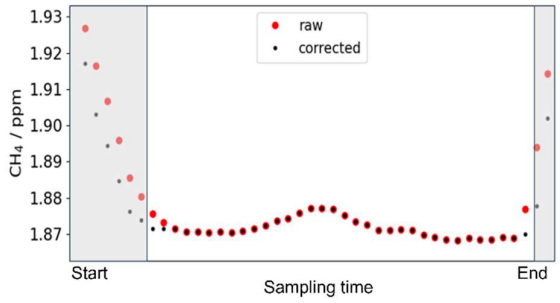

Figure 6. The raw and corrected CH_4 profile across the sampling period on 19 June 2024 at Picton. The mixing effect at the start and end of the measured profile is highlighted in grey.

2.5. Sampling summary

AirCore samples were collected at the Picton location on 21 May, 28 May and 19 June 2024.

Sampling occurred at the Hunter Valley (Elderslie, NSW) location on 23 June 2024, when three separate skydive samples were collected.

The distance aloft covered within the samples mean that the bottom 30-40% of the atmosphere, and all of the PBL, is covered. A summary of the six samples completed to date is provided in Table 1.

Table 1. Summary of jumps / AirCore samples. Each drive takes approximately two to four minutes from the plane exit.

Date (plane exit time)	Location	Max. altitude (m)	Min. pressure (hPa)
21/5/24 (10:42)	Picton	2,900	704
28/5/24 (10:35)	Picton	4,100	594
19/6/24 (10:32)	Picton	4,000	592
23/6/24 (12:50)	Elderslie	4,500	572
23/6/24 (15:01)	Elderslie	4,600	562
23/6/24 (16:16)	Elderslie	4,600	571

3. Results and discussion

The CH_4 profiles collected during the first two skydives using the AirCore sampling system are excluded from the results due to instrument and sampling errors.

Examples of the CH_4 profiles for the two skydive sites are presented in Figure 7 and 8.

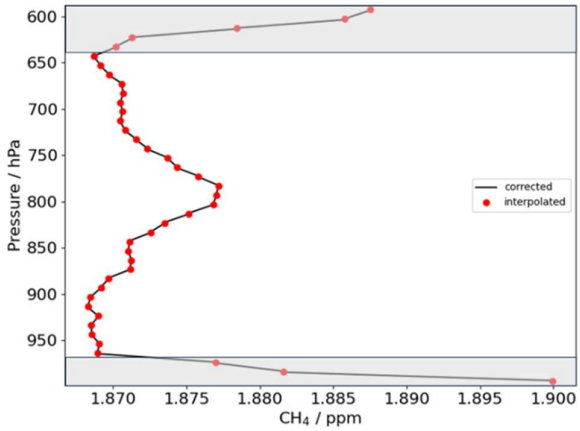

Figure 7. The AirCore derived CH_4 profiles for 19 June 2024 skydive at Picton.

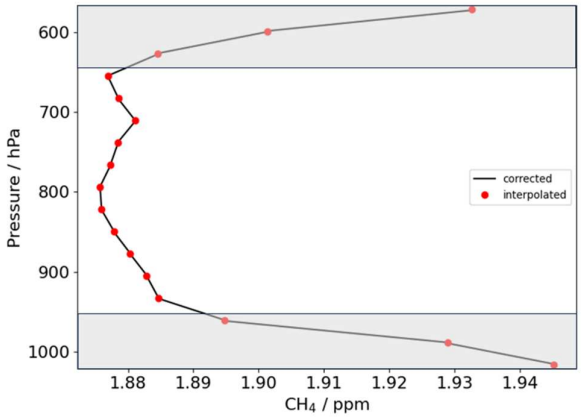

Figure 8. The AirCore derived CH_4 profiles for 23 June 2024 skydive at Elderslie.

For the reliable samples collected to date (four in total) there is limited variability between the CH_4 profiles.

The reasons for the lack of variability between the profiles may be due to the practical constraints of availability of the skydiver and suitable weather conditions. For example, on the sampling date in the Hunter Valley, winds were predominantly easterly meaning the sample was taken upwind of the largest local CH_4 sources, namely the Upper Hunter Valley coal mines.

4. Next Steps

4.1. Skydives and wind conditions

To date, considerations such as prioritising wind directions when known CH_4 sources are upwind have not been factored into the experimental design.

Consequently, future work will aim to prioritise conducting skydive sampling when wind conditions are favourable for the major CH_4 sources which are upwind.

For example, northwesterly wind conditions in the Hunter Valley, where potential emissions plumes from the Hunter Valley coalfields (shown in Figure 3) would be transported towards the skydive zone. Such wind vectors are common in spring and summer. Conversely, sampling at Picton, target days will comprise of easterly winds when emissions from the Appin underground coal mine (shown in Figure 4) may be transported towards the skydive zone.

4.2. Sampling

Future sampling will focus on trying to improve the sample resolution by reducing the flow rates through the Picarro.

4.3. Total column measurements

Co-located total column measurements at the skydive zone using a portable solar adsorption spectrometer (EM27/SUN) will complement the measurements at Wollongong's Total Carbon Column Observing Network (TCCON) site. The two column measurements can provide a differential upwind and downwind pair to quantify any emissions occurring between the instruments, whilst the skydive AirCore profiles will help identify where in the column any enhancements might occur.

4.4. Additional skydive sites

Discussions are currently taking place with the Commonwealth Science and Industrial Research Organisation (CSIRO) to complete additional sampling campaigns in Victoria, Australia.

5. Conclusions

The current study leverages the established AirCore sampling technique, typically deployed using a balloon-parachute arrangement, aircraft or UAVs.

This study adopts a novel approach by fitting an AirCore sampling system to skydivers whilst completing parachute jumps, enabling rapid, targeted transects through the bottom few kilometres of the atmosphere.

The results presented are preliminary but provide an understanding of the suitability of the AirCore's technique to provide vertical atmospheric CH_4 profiles.

Future measurements will aim to coincide with wind conditions when known major sources of CH_4 are upwind to further understand the distribution of this important GHG in the vertical atmospheric profile.

Acknowledgments

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